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ARTICLE *in* ENVIRONMENTAL SCIENCE & TECHNOLOGY · MARCH 2013

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Methylmercury Accumulation in Plankton on the Continental Margin of the Northwest Atlantic Ocean

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Supporting Information

ABSTRACT: Accumulation of monomethylmercury (MMHg) by plankton is a key process influencing concentrations of this toxic mercury species in marine food webs and seafood. We examined bioaccumulation and biomagnification of MMHg in microseston and four size fractions of zooplankton on the continental shelf, slope, and rise of the northwest Atlantic Ocean. The bioaccumulation factor (BAF, L/kg) for MMHg in microseston averaged $10^{4.3\pm0.3}$ among 21 locations, and concentrations were unrelated to those in colocated, filtered surface water. Instead, concentrations and the BAF of MMHg in microseston were related inversely with total suspended solids in surface water, a proxy for planktonic biomass at these remote locations. MMHg was biomagnified by a factor of 4 from microseston to zooplankton, and both concentrations of MMHg and the fraction of total mercury as MMHg increased with larger size fractions of zooplankton. These results suggest that the initial magnitude of MMHg uptake into pelagic marine food webs is influenced by the degree of primary production in surface waters and propagated up through large zooplankton. Accordingly, biological productivity, in addition to inputs of MMHg to surface waters, must be considered when predicting how MMHg bioaccumulation will vary spatially and temporally in the ocean.

INTRODUCTION

Humans are exposed to the neurotoxin monomethylmercury (MMHg) primarily by eating seafood.[1,2] While MMHg is present in seawater at exceedingly low concentrations (0.02−0.2 pM),[3,4] its extraordinary degree of bioaccumulation and biomagnification in marine food webs can result in predatory fishes, including some species consumed by humans, having wet-weight tissue concentrations that are 10^6−10^7 fold greater than those in water.[5−7] Such concentrations may be toxic to the fish themselves[8] and pose a health risk to human consumers.[9,10] Indeed, consumption of MMHg-contaminated seafood has been linked to an increased risk of fetal neurodevelopmental problems in as many as 5−10% of U.S. women of childbearing age.[11]

Mechanisms of bioaccumulation and biomagnification of MMHg and their variability in marine food webs are poorly understood given their importance in affecting wildlife and human exposure.[4] Most studies of MMHg bioaccumulation in marine ecosystems have focused on piscivorous fishes and coastal regions of known Hg contamination. There have been no comprehensive, multi trophic-level investigations of MMHg in the open ocean.[3] Nearly all that is known about MMHg biomagnification in marine environments has resulted from studies in estuaries and coastal embayments.[5−7,12,13] These studies have suggested that the bioconcentration of MMHg from seawater by living organisms in the microseston size fraction (i.e., phyto-, bacterio-, and microzooplankton; 0.2−200 μm) is the most important bioaccumulation step in marine food webs.[4,6,14] Microseston are hypothesized to concentrate MMHg from seawater by passive uptake of the CH_3HgCl^0 ion pair, which reacts with intracellular ligands and accumulates in the cytoplasm.[14] In Long Island Sound, for example, MMHg is magnified by $10^{4.2}$ from water to microseston.[6] The increase between water and biota is often expressed as a bioaccumulation factor (BAF; L/kg), which is the concentration in biota (wet-weight basis) divided by that in water. Bioaccumulation factors for MMHg between seawater and marine microseston can range from $10^{3.4}$ to $10^{5.3}$ (ref 15), with additional biomagnification of 2−10× per increasing trophic level[4,6] as a result of dietary uptake and assimilation.[16] Accordingly, concentrations in higher trophic levels within a food web can be influenced by the degree to which MMHg is initially bioconcentrated from water by microseston.

There is scarce information related to the accumulation of MMHg in lower trophic levels of marine food webs, in particular phyto- and zooplankton.[4] Little is known about the spatial and temporal distribution of MMHg in plankton, the factors that influence MMHg levels in microseston, and the degree to which it is bioconcentrated and biomagnified within the planktonic community. The objective of this study was to address some of these important information gaps by examining MMHg in surface water, microseston, and size-fractionated zooplankton sampled from a broad region on the continental margin of the northwest Atlantic Ocean.

Received: November 28, 2012
Revised: March 5, 2013
Accepted: March 14, 2013
Published: March 14, 2013

MATERIALS AND METHODS

Sampling. Microseston and zooplankton were sampled at multiple locations on the continental margin of the northwest Atlantic Ocean in August 2008, September and October 2009, and July 2010 (Figure 1). Most stations were occupied only once, although both microseston and zooplankton were sampled during each cruise at Station 9 to examine temporal variability. Sampling locations were selected to span a range of planktonic productivity[17] and a presumed range of aqueous MMHg exposures. In situ production and mobilization from sediments is hypothesized as the major source of MMHg to food webs on the continental shelf and upper slope (<1000 m depth),[18,19] whereas methylation in the water column is hypothesized to be a dominant source in open-water environs.[4] Sampling locations also were selected to be representative of those near shore and on the continental shelf, slope, and rise as well as above the abyssal plain. Bathymetric depths among the stations ranged from 35 m (Station 1, Cape Cod Bay) to 4600 m at Station 17.

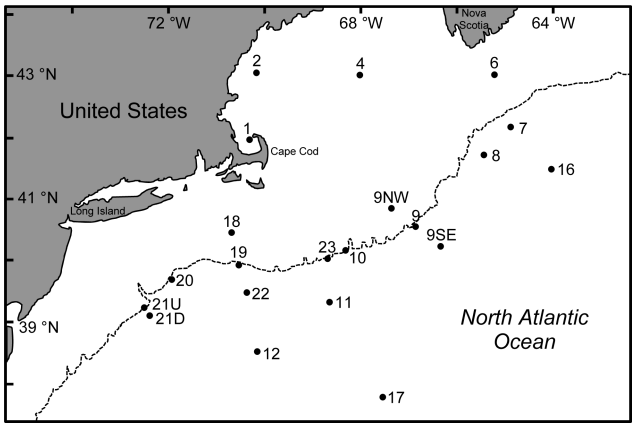

Figure 1. Plankton sampling stations on the continental margin of the northwest Atlantic Ocean. The dashed line delineates the location of the shelf break. Station coordinates and water depths are in Supporting Information, Table S1.

Seawater and microseston were sampled with trace-metal clean techniques from the upper 60 m of the water column,[20] which included the chlorophyll maximum (located between 20−50 m depth). At least two water depths were sampled at each station: one between 8−20 m and the deeper chlorophyll maximum. Water was collected with trace-metal clean Go-Flo bottles suspended from a nonmetallic hydrowire. Microseston, which was operationally defined by size (0.2−200 μm) and assumed to result from autochthonous production at these remote locations, was concentrated from a known volume of water from the Go-Flo bottles onto 0.2 μm polycarbonate filters after the water was passed through an acid-cleaned, 200 μm nylon mesh screen to remove larger organisms. A separate aliquot of water from the same Go-Flo bottle was used to determine the concentration of total suspended solids (TSS) in the same size fraction.[21] Promptly after sampling, filtered (<0.2 μm) seawater was purged of volatile dimethylmercury and Hg^0 and analyzed for MMHg by gas-chromatographic cold-vapor atomic fluorescence (CVAFS) in a shipboard laboratory.[22] Filters loaded with material for MMHg and TSS analysis were stored frozen (≤−20 °C) until analysis on shore.

Zooplankton were sampled with a 200 μm mesh, trace-metal clean, opening/closing net towed vertically with nonmetallic hydrowire. The net was deployed closed, opened at depth (~15 m above bottom for shallow stations, 200−300 m water depth at deeper stations), and closed again after the up cast, about 10 m beneath the keel of the ship. Zooplankton were size fractionated with nylon mesh screens in a Class 100 clean bench on the research vessel. Size fractions included 200−500, 500−1000, 1000−2000, and >2000 μm. Not all size fractions of zooplankton were present in sufficient amounts for MMHg analysis at some locations. After size fractionation, zooplankton were rinsed onto acid-cleaned polycarbonate membrane filters with filtered seawater, frozen shipboard, and transported on dry ice to Wright State University. Zooplankton size fractions that appeared to have relatively small masses of material (<~5 mg) were divided evenly for analysis of both MMHg and mass of total carbon (TC).[23] This was done so that the mass of zooplankton analyzed for MMHg could be estimated from the TC content in the equally divided subsample. A relationship between the TC content (mg) and zooplankton dry mass (mg) was established empirically with a subset of organisms (TC = 0.34[dry mass] + 0.25; n = 21, r^2 = 0.82). The slope of the relationship was in good agreement with Redfield−Ketchum−Richards stoichiometry (0.36).[24]

MMHg Extraction and Analysis. MMHg was measured in zooplankton after digestion with 4.57 M HNO_3 in a 60 °C water bath for 12 h.[6] At Wright State University, frozen zooplankton (on membrane filters) were thawed briefly in a Class 100 clean bench, rinsed with reagent-grade water (resistivity, ≥ 18 MΩ·cm) to remove sea salt, and refrozen prior to freeze-drying. Masses of dried zooplankton digested for MMHg analysis were determined either gravimetrically, for larger samples, or by estimation from the amount of TC in an equally divided subsample and the empirical equation described above. MMHg in sample digestates was determined by gas-chromatographic, flow-injection CVAFS.[25,26] Filters loaded with microseston were digested and analyzed similarly;[6] there was no detectable interference from acid leaching of the polycarbonate membranes. Mass-normalized concentrations of MMHg in microseston were calculated by dividing the volumetric concentration of MMHg determined from the filter (pmol/L of water filtered) by the concentration of TSS (g dry/L). Dry-weight concentrations of MMHg in plankton were converted to a wet-weight basis, assuming that water content averages 95% in microseston and 90% in zooplankton.[27]

The accuracy of MMHg measurements in plankton was assessed by analyses of (i) procedural blanks and calibration standards subjected to the digestion process; (ii) certified reference materials from the National Research Council of Canada, lobster hepatopancreas (TORT-2) and fish protein (DORM-3); and (iii) procedural and analytical replicates of zooplankton samples. Standard solutions of MMHg were calibrated, following BrCl oxidation, by comparison to a Hg solution traceable to the U.S. National Institute of Standards and Technology (NIST). All measurements of MMHg in the two reference materials (n = 36 each) were within the certified ranges. The mean measured concentration in MMHg in TORT-2 was 785 pmol/g (certified range, 695−825 pmol/g) and that of DORM-3 was 1630 pmol/g (certified range, 1500−2060 pmol/g). Variance in procedural reproducibility (relative percent difference), measured by digestion and analysis of replicate subsamples, averaged 16% (n = 11), which was similar to analytical precision (mean = 11%; n = 28). Estimated method detection limits were about 0.05 pmol/g wet weight for a 100 mg sample of zooplankton and 0.2 pmol/g wet weight for

dx.doi.org/10.1021/es3048619 | Environ. Sci. Technol. 2013, 47, 3671−3677

a microseston sample concentrated from seawater containing 1 mg/L of TSS.

Total Hg Analysis. The 4.57 M HNO_3 digestates of zooplankton for MMHg analysis also were used to quantify total Hg in the same samples. Aliquots of the digestates were oxidized with BrCl for ≥12 h prior to addition of NH_2OH.[6] Sample Hg was reduced with $SnCl_2$ and measured by dual-Au amalgamation CVAFS.[28,29] The total Hg in 2010 microseston was calculated as the concentration difference between unfiltered and filtered water (pmol/L, passed through a clean capsule)[20] divided by the TSS concentration (g/L).[30]

Total Hg analyses were calibrated with aqueous Hg^{2+} solutions traceable to the U.S. NIST. As with MMHg, all measurements of total Hg in TORT-2 and DORM-3 (n = 21 each) were within the certified ranges: Total Hg in TORT-2 averaged 1310 pmol/g (certified range, 1050−1650 pmol/g) and that in DORM-3 averaged 1760 pmol/g (certified range, 1610−2210 pmol/g). Recovery of known Hg additions averaged 97 ± 4%. Procedural reproducibility (relative percent difference) averaged 4.6% (n = 10) and was comparable to analytical precision (mean = 3.8%, n = 28).

Statistical Analysis. Parametric tests were used to compare data groups with normal distributions and equal variance, whereas nonparametric techniques were used when these criteria were not met. Appropriate posthoc tests were performed when differences were found. A Type I error (α) of 0.05 was used to judge the significance of all tests, which were performed with SigmaPlot (version 11 or 12.3).

RESULTS AND DISCUSSION

Microseston. MMHg bioaccumulates in microseston (0.2−200 μm) on the continental margin of the northwest Atlantic Ocean. Among all sampling locations and periods, the mean concentration (±SD) of MMHg in microseston was 0.70 ± 0.31 pmol/g wet weight (n = 25) and ranged from 0.11 to 1.3 pmol/g (Supporting Information, Table S1). The fraction of total Hg as MMHg averaged 6 ± 3% in microseston sampled in July 2010. These MMHg values are consistent with the range of concentrations and percentages observed in microseston in other marine systems, including coastal waters of the Bay of Fundy, Long Island Sound, Gulf of Lions, Jamaica Bay (New York Harbor), and the North Sea (MMHg = 0.3−2.5 pmol/g wet weight, MMHg/total Hg = 2−10%).[5,6,15,30−32] The fraction of total Hg as MMHg in microseston was much greater than the ratio in marine sediments (~0.5%),[18] implying accumulation of MMHg in planktonic organisms as opposed to equilibrium partitioning of Hg species with suspended detritus. The mean level of MMHg in microseston in the current study was not different from that measured at three sites within the study area in 2003 (0.86 ± 0.18 pmol/g).[18] Concentrations of MMHg in microseston were much greater than those in colocated, filtered seawater, which contained between 0.015 and 0.056 pM. The BAF for MMHg in microseston averaged $10^{4.3\pm0.3}$ among all stations. Concentrations of MMHg in microseston were unrelated to those in colocated, filtered surface water among all sampling locations (Spearman rank order, p = 0.63), although levels in surface water varied by less than a factor of 4 among sites. This suggests that factors other than aqueous concentration have an important influence on MMHg accumulation by microseston.

MMHg in microseston has been posited to reflect the nutrient status and associated biological productivity of marine ecosystems.[15] A relatively lower BAF is hypothesized for eutrophic waters because greater primary production will dilute the supply of MMHg among a larger pool of planktonic biomass, the so-called biodilution hypothesis.[33] BAFs for MMHg in marine microseston range from $10^{3.4}$−$10^{3.7}$ in mesotrophic marine systems (e.g., Bay of Fundy, North Sea, Belgian coast) to $10^{5.3}$ in oligotrophic waters of the open North Pacific Ocean.[15,34] The mean BAF for MMHg in microseston in the current study ($10^{4.3\pm0.3}$) is less than that in the subtropical North Pacific Ocean (primary production = ~70 g $C{\cdot}m^{-2}{\cdot}y^{-1}$) and consistent with a greater degree of primary production in the study area (~150−300 g $C{\cdot}m^{-2}{\cdot}y^{-1}$).[17]

Among locations on the continental margin, an apparent connection between MMHg in microseston and concentration of TSS is consistent with the biodilution hypothesis (Figure 2).

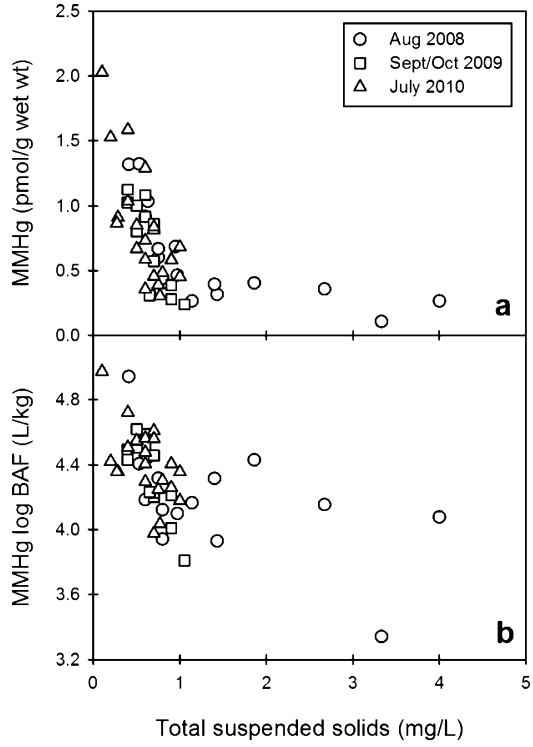

Figure 2. Concentrations and bioaccumulation factors (BAF) of monomethylmercury (MMHg) in microseston as a function of total suspended solids in surface water (upper 60 m) on the continental margin of the northwest Atlantic Ocean (Spearman rank order correlation, p-values <0.001 for both panels).

The concentration of TSS was used as a proxy for the amount of planktonic biomass in these surface waters, which are far removed from terrigenous inputs of material. The decrease of MMHg concentration with TSS, which was replicated during all three cruises, implies that greater algal biomass dilutes the amount of MMHg per gram of microseston (Figure 2a), as hypothesized by others.[33] Moreover, the corresponding decrease of MMHg BAF in microseston with greater productivity supports the hypothesis that differences in aqueous MMHg concentrations are not wholly responsible for variability of levels in microseston (Figure 2b). Indeed, both relationships in Figure 2 suggest that the kinetics of MMHg uptake by microseston are slow relative to cell division, which may explain the nonlinear responses of MMHg concentrations and BAFs in microseston at TSS levels greater than about 1 mg/L. The relationship between MMHg BAF and TSS (Figure 2b) is

dx.doi.org/10.1021/es3048619 | Environ. Sci. Technol. 2013, 47, 3671−3677

opposite of well-known positive correlations between the distribution coefficient of MMHg (K_D, L/kg) and the concentration of nonliving organic material in marine sediments.[35] The difference between the BAF and K_D relationships suggests that most MMHg associated with microseston was not a result of partitioning with detritus and supports our assumption that microseston was mostly living organisms at our sampling locations, although the composition of microseston was not determined. While biodilution of MMHg has been observed in experimental mesocosms,[33] temperate lakes,[36] and San Francisco Bay,[37] this is the first reported observation of this phenomenon in the ocean.

Zooplankton. MMHg biomagnifies during trophic transfer between microseston and zooplankton. Biomagnification results when one organism consumes another and the substance (e.g., MMHg) in the prey is retained in the consumer's body to a greater degree than the mass of organic material consumed, much of which is not assimilated into new biomass (i.e., respired, egested). Among all size-fractionated zooplankton samples (>200 μm; n = 78), the mean concentration of MMHg (2.8 ± 2.8 pmol/g wet weight) and fraction of total Hg as MMHg (15 ± 12%) were significantly greater than the average values in microseston (0.70 ± 0.31 pmol/g and 6 ± 3%; Mann−Whitney rank sum, $p < 0.001$). The mean biomagnification factor of 4× for MMHg between microseston and all zooplankton in this study is within the range of values observed in other coastal waters for which there is concentration information for both microseston and zooplankton, including the Long Island Sound (2.3),[6] Bay of Fundy (3.2),[31] and Gulf of Lions (6.3).[32]

MMHg levels in zooplankton on the northwest Atlantic margin are some of the lowest reported in marine systems (Table 1). Among known marine investigations, the average concentration of MMHg and fraction of total Hg as MMHg in all zooplankton fractions examined in this study are not different from those in Long Island Sound and the Gulfs of St. Lawrence and Lions, but less than those in the Southern and Arctic Oceans and coastal seas of Japan. One explanation for increased levels near Japan is that Minamata Bay and adjacent Yatsushiro Sea were impacted by mercury pollution from a chemical manufacturer during the 1930s to 1960s.[42] Greater concentrations of MMHg in zooplankton of the Southern Ocean may be attributed to atypically high levels of methylated Hg in surface waters.[43]

MMHg biomagnification within the zooplankton community also is suggested by concentration differences among size fractions (Figure 3). Although there was considerable variability within each size fraction, mean MMHg concentrations (±1 SD) in zooplankton increased with size: 200−500 μm = 1.3 ± 1.1 pmol/g wet weight, 500−1000 μm = 2.2 ± 1.6 pmol/g, 1000−2000 μm = 3.8 ± 3.1 pmol/g, and >2000 μm = 4.8 ± 3.7 pmol/g. All but the smallest zooplankton (200−500 μm) had MMHg concentrations significantly greater than those in microseston (Figure 3).

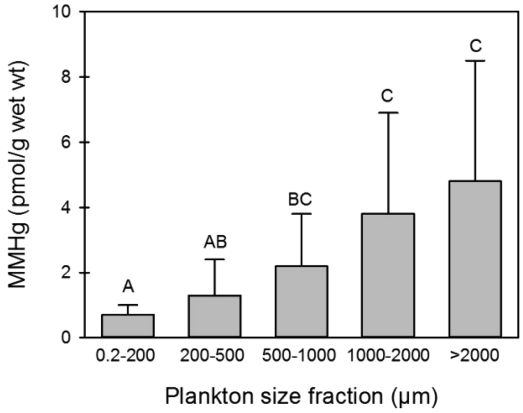

Figure 3. Mean (±1 SD) concentrations of MMHg in size fractions of plankton on the continental margin of the northwest Atlantic Ocean. Means with different letters are significantly different (Kruskal−Wallis ANOVA on ranks, Dunn's pairwise multiple comparison, $p < 0.05$).

The observed differences in mean MMHg concentration with zooplankton size may be attributed to larger animals being longer lived and having greater time to accumulate the contaminant. Long-lived copepods and euphasiids were the major zooplankton genera greater than 1000 μm. For example, zooplankton at stations on the shelf (e.g., Stations 2−6) were dominated by large *Calanus* sp. copepods, which can survive for more than a year by overwintering in deep waters.[44,45] Similarly, at the offshore stations on the slope and rise, the fraction greater than 2000 μm was comprised of large euphasiids, identified as either *Meganyctiphanes norvegica* or *Thysanoessa inermis*.[46] These krill species are capable of living for 1−3 y.[47,48] The longer lifespans of zooplankton in the larger size fractions may explain greater MMHg bioaccumulation. Alternatively, MMHg may be biomagnified when larger zooplankton prey on smaller animals. The offshore stations also had large (>2000 μm) predatory *Themisto* sp. amphipods, which prey on small copepods and euphasiids[49] and are themselves potential prey for marine birds.[50]

The hypothesized biomagnification within the zooplankton community is supported by an increase in the fraction of total Hg as MMHg with zooplankton size. The mean percentage of total Hg as MMHg (±1 SD) increased with zooplankton size: 200−500 μm = 7.1 ± 6.3%, 500−1000 μm = 14 ± 10%, 1000−2000 μm = 16 ± 12%, and >2000 μm = 26 ± 16%. Among size

Table 1. Spatial Comparison of Mean MMHg Concentrations (± 1 SD, Wet-Weight Basis) in Marine Zooplankton

location[a]	MMHg[b] (pmol/g)	MMHg/ total Hg (%)	n	reference
Minamata Bay (>95 μm, C)	79 ± 48	28 ± 26	4	38
Yatsushiro Sea (>95 μm, C)	29 ± 24	50 ± 19	11	38
Arctic Ocean polynya (>520 μm, M)	20 ± 15	70 ± 38	4	39
Seto Inland Sea (>95 μm, C)	16 ± 4	63 ± 3	4	38
Southern Ocean (>328 μm, C)	14 ± 5	26 ± 28	9	40
Tropical Pacific Ocean (>328 μm, M)	8.7 ± 5.5	45 ± 29	5	41
Long Island Sound (>200 μm, C)	5.5 ± 1.0		4	6
Gulf of St. Lawrence (>333 μm, M)	2.2 ± 1.0	13 ± 8	15	12
Gulf of Lions, inshore (>200 μm, M)	2.2 ± 0.9	21 ± 4	15	32
Northwest Atlantic margin (>200 μm, M)	2.8 ± 2.8	15 ± 12	78	this study

[a]Size fraction of zooplankton and whether samples were copepods (C) or mixed (M) zooplankton are in parentheses. [b]Wet-weight concentrations assume water content is 90% in zooplankton.[27]

dx.doi.org/10.1021/es3048619 | Environ. Sci. Technol. 2013, 47, 3671−3677

fractions, MMHg/total Hg percentages were significantly greater in zooplankton >2000 μm than in those in the 200−500 μm portion (Dunn's pairwise multiple comparison, $p < 0.05$). The fraction of total Hg as MMHg typically increases from prey to consumer because MMHg depurates at a slower rate than Hg^{2+} from the consumer,[16] and this is evident from the zooplankton. While MMHg levels increased with greater zooplankton size (Figure 3), mean concentrations of non-methylated Hg (i.e., total Hg minus MMHg) were not different among size fractions: 200−500 μm = 20 ± 13 pmol/g wet weight, 500−1000 μm = 17 ± 14 pmol/g, 1000−2000 μm = 21 ± 12 pmol/g, and >2000 μm = 15 ± 7 pmol/g (Kruskal−Wallis one way ANOVA on ranks, $p = 0.16$). The increase of both MMHg concentration and percentage of total Hg as MMHg between the smallest (200−500 μm) and largest (>2000 μm) size fractions suggests that zooplankton in these size fractions have different diets. However, measurement of stable nitrogen isotopes and taxonomic identification in the different size fractions would be needed to confirm dietary differences and biomagnification.

MMHg concentrations in all size classes of zooplankton were unrelated to levels in filtered water (Spearman, $p = 0.08−0.97$), which is consistent with zooplankton acquiring the contaminant mainly through their diet.[16] However, and among the four size fractions of zooplankton, only the 1000−2000 μm portion contained MMHg concentrations that were significantly correlated with concentrations in microseston, with an average biomagnification factor of about six for this size fraction (slope of regression; Figure 4). This modest relationship and lack of a correlation between MMHg levels in microseston and other zooplankton size fractions (Spearman rank order, $p = 0.10−0.93$) suggests that concentrations in discrete samples of microseston (from 2−3 depths in the mixed layer at each station) are not predictive of levels in zooplankton sampled with vertical net tows. This disconnection may result from either a sampling artifact or vertical migrations of zooplankton and feeding at depths other than those sampled for microseston.

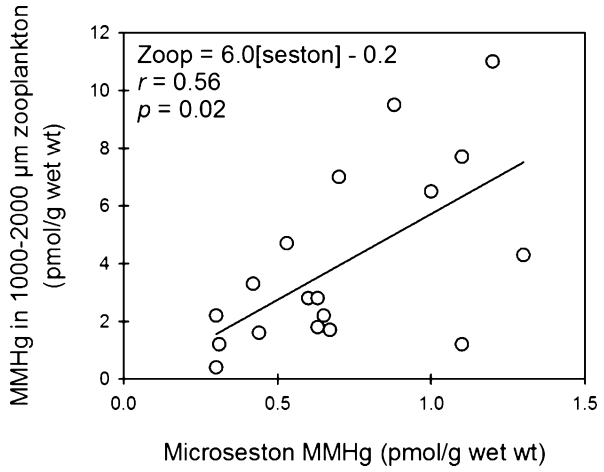

Figure 4. MMHg concentrations in 1000−2000 μm size fraction zooplankton related to those in microseston (0.2−200 μm).

Spatial and Temporal Variability. Proximity to the coastline and underlying sediments, two potential sources of MMHg to surface water, had no apparent influence on concentrations of MMHg in plankton. MMHg concentrations in each of the five size fractions of plankton were unrelated to either bathymetric depth of the sampling station or distance from the nearest coastline (Spearman rank order, $p > 0.05$). Moreover, MMHg concentrations in each of the size fractions were not significantly different between stations on the shelf (Stations 1, 2, 4, 6, 9NW, 18, 19; Figure 1) and those further off shore (Mann−Whitney rank sum, $p = 0.30−0.87$). These findings are not surprising given the complex hydrography of the region and that methylation in the water column may be a significant source of MMHg accumulating in pelagic biota.[4]

Although there were no clear geographical patterns of MMHg in plankton on the margin, there may be substantial temporal variability. Station 9 was sampled for all size fractions of plankton in each of the three study years (Supporting Information Table S1). Concentrations of MMHg in plankton varied among years (two-way ANOVA with size fraction nested within month, $p = 0.03$) and were significantly greater in July 2010 than August 2008 (Tukey multiple comparison, $p = 0.03$). Concentrations in September and October 2009 were not different from either of the other two sampling periods. It is unclear if such differences are a result of either interannual or seasonal factors; higher frequency sampling is needed to resolve how plankton MMHg concentrations vary over time. Temporal variation in plankton MMHg could result from the timing and size of blooms as well as the speciation, life history, and feeding patterns of the zooplankton. Changes in plankton MMHg are likely to influence concentrations in higher trophic levels.

Bioconcentration from seawater by microseston is the greatest magnification step of MMHg concentration in marine food webs. We found that microseston on the continental margin bioconcentrate MMHg by a factor of $10^{4.3\pm0.3}$, which is intermediate between BAFs observed in mesotrophic nearshore and oligotrophic open-ocean waters and in support of a hypothesized relationship between trophic status and microseston BAF.[15] Moreover, we observed connections between planktonic biomass and both MMHg concentration and BAF that imply algal biomass dilutes the amount of MMHg per gram of microseston. These findings suggest that the initial magnitude of MMHg uptake into pelagic marine food webs is influenced by the degree of primary production in surface waters. Accordingly, biological productivity, in addition to inputs of MMHg to surface waters, must be considered when predicting how MMHg bioaccumulation will vary spatially and temporally in the ocean, including as a result of climate change. Indeed, as the surface ocean warms, becomes more stratified, and experiences reduced primary production,[51] our results imply that MMHg concentrations in the planktonic food web could increase as a consequence of the biodilution phenomenon.

■ ASSOCIATED CONTENT

Supporting Information

Tabulated concentrations of MMHg in filtered seawater, microseston, and zooplankton. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank Kati Gosnell, Jenay Aunkst, Hans Dam, and captains and crews of the R/V *Endeavor* and *Oceanus* for help with plankton sampling. Katlin Bowman measured MMHg in filtered water. We are grateful to three anonymous reviewers for helpful comments that improved the manuscript. This research was supported by the Chemical Oceanography program of the U.S. National Science Foundation.

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