1 **Mercury and Methylmercury Incidence and Bioaccumulation in Plankton**
2 **from the Central Pacific Ocean**

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Abstract

Open ocean samples of phytoplankton and zooplankton were collected from the central Pacific on board the R/V Kilo Moana during October of 2011. The cruise traveled from Hawaii to Samoa, progressing through a High Nutrient Low Chlorophyll (HNLC) zone, and an equatorial upwelling region. Phytoplankton samples were size fractionated into 0.2-5 μm , 5-20 μm , and >20 μm samples. Methylmercury concentrations were $2.91 \pm 2.58 \text{ pmol g}^{-1}$ (wet weight) for the overall <200 μm size fractions, and highest around the HNLC region. Phytoplankton bioconcentrations factors (logBCFs) averaged to 5.69 ± 0.98 and were higher than values found for coastal regions. Both %MeHg ([MeHg]/[Hg]) and logBCF values indicated that the lowest size fraction had the largest fraction of the HgT as MeHg, signifying enhanced accumulation of MeHg into smaller organisms. Zooplankton vertical net tows were completed from depths of 200 m up to ~10 m. Zooplankton samples were analyzed for carbon, nitrogen and sulfur in addition to Hg and MeHg at size fractionations of 0.2-0.5 mm, 0.5-1.0 mm, 1.0-2.0 mm and occasionally >2.0 mm. Zooplankton abundance and MeHg concentrations both peaked at stations 3 and 5 (upwelling region). The %MeHg in the organisms was highest in the >2.0 mm size class, displaying MeHg bioaccumulation for increasing zooplankton sizes. Separate day and night net tows were collected at stations 3 and 5 in order to investigate differences due to diurnal migration of zooplankton. There were higher concentrations of MeHg for all sizes in night collections of zooplankton at station 5, but no discrepancy for station 3. These results represent some of the few measurements of Hg and MeHg at the base of the open ocean food chain, and are significant as they represent the instigation of bioconcentration into the base of marine food webs.

Key Words: Bioaccumulation; Plankton; Methylmercury; Inorganic Mercury; Pacific Ocean

Introduction

Mercury (Hg) is a substantiated global concern due to the high bioavailability of its organic form methylmercury (MeHg), which is a damaging neurotoxin (Clarkson and Magos, 2006), and highly bioaccumulative in aquatic food chains (Chen et al., 2008; Sunda, 2012). Mercury emissions have increased above natural environmental levels due to anthropogenic practices, such as high temperature combustion, artisanal gold mining and cement manufacturing (National Research Council, 2000). Due to its long atmospheric residence time, Hg has the ability to travel throughout the world, impacting remote regions such as the open ocean (Lamborg et al., 2014; Mason et al., 2012; Mason et al., 1994). Oceanic fish represent a critical source of MeHg into diets, as most humans consume seafood in higher quantities than freshwater fish (Sunderland, 2007; Chen et al., 2008). Nevertheless, little research has been undertaken at mercury's point of entry into open ocean marine food webs, primarily due to the fact that it is difficult to measure MeHg uptake and trophic transfer under natural conditions, as open ocean concentrations of inorganic Hg and MeHg are extremely low (<1 pM for MeHg) (Mason et al., 2012; Gill and Fitzgerald, 1985; Lamborg et al., 2014). As a result, uncertainty still exists related to the behavior of Hg and MeHg in lower trophic levels of the food chain in marine ecosystems. Likewise, the exact processes which drive and control natural bioaccumulation and trophic transfer are still unresolved for the open ocean.

It is imperative to fully comprehend the transport of Hg and MeHg into the lower oceanic trophic levels, as these reflect the inception of exposure. Algae accrue nutrients, including essential metals (e.g. iron, zinc) and unessential elements (e.g. Hg, lead, arsenic) from the

surrounding media, and also release compounds that influence metal speciation. Phytoplankton population and composition therefore has a significant impact on the levels and availability of trace metals in marine waters. The remineralisation of algae during sinking and bloom degradation also impact metal speciation and availability. The resultant formation of natural complexes influences uptake of many metals by biota. Prior research suggests that phytoplankton MeHg uptake occurs primarily via passive diffusion of neutral complexes across a cell membrane (Pickhardt and Fisher, 2007; Gorski et al., 2006; Fisher and Reinfelder, 1995; Lawson and Mason, 1998; Mason et al., 1996). Passive uptake of both species could also result after sorption of Hg and MeHg to cell surfaces (Fisher, 1985).

However, the possibility also exists that phytoplankton assimilate Hg and MeHg via active or facilitated transport. Active uptake of MeHg remains unspecified. It is probable that MeHg is likewise accumulated when cells acquire organic compounds which are strongly bound to MeHg, such as cysteine or other thiols (Moye et al., 2002; Lawson and Mason, 1998), as has been found to occur in bacteria (Ndu et al., 2012; Schaefer et al., 2011). Pickhardt and Fisher (2007) demonstrated that live cells accumulated more MeHg than dead cells in freshwater phytoplankton, suggesting possible uptake via active processes. This was not the case for inorganic Hg. Because MeHg permeates into cellular cytoplasm rather than being bound with the cellular membrane (Mason et al., 1996; Pickhardt and Fisher, 2007), transfer and accumulation into the marine food web is greater for MeHg than for inorganic Hg (Reinfelder and Fisher, 1991; Mason et al., 1996). Subsequently, MeHg is bioconcentrated in phytoplankton and transferred via consumption through the food chain to herbivorous zooplankton, planktivorous fish, and eventually larger predators, with the fraction of MeHg amplifying at each trophic level (Foster et al., 2012; Kim et al, 2004; Berntssen et al, 2003). Therefore, it is crucial

to quantify Hg and MeHg movement from ocean waters into primary and secondary consumers, as this is where the principal biomagnification occurs (Pickardt and Fisher, 2007; Mason, 2002; Le Faucher et al., 2014).

Relatively little data have been published on natural concentrations of Hg in coastal ocean plankton (Foster et al., 2012; Hammerschmidt et al., 2013; Mason et al., 2012), and there is even less information concerning the open ocean. In addition, there has been little study of the concentrations of MeHg in different size fractions of plankton, except for those published recently by Hammerschmidt et al. (2013). Furthermore, quantified amounts of MeHg transfer have not been established for primary (phytoplankton) and secondary producers (zooplankton) for open ocean populations. Even though coastal ecosystems represent a large fraction of overall productivity for marine systems, the global expanse of the open ocean, and in particular the more productive regions such as the equatorial Pacific, warrant consideration in terms of importance towards Hg and MeHg bioaccumulation. This study presents measurements of size fractionated Hg and MeHg concentrations for open ocean phytoplankton and zooplankton. The results of this study indicate that concentrations in zooplankton increase with organism size and that the bioaccumulation factor for phytoplankton decrease as suspended particle concentration increase. Determining these values have helped to establish levels of Hg and MeHg at the base of the open ocean food chain, at the onset of marine bioaccumulation. Additionally, these data provide information to managers and modelers concerned with the bioaccumulation of MeHg into open ocean fish consumed by humans.

Methods

Sample collection

Phytoplankton and zooplankton samples were collected along a transect from Oahu, Hawaii to Apia, Western Samoa, with a brief stopover at the Island republic of Kiribati (Figure 1). The cruise occurred on the R/V Kilo Moana during October 3-23, 2011. The cruise transect traveled through a High Nutrient Low Chlorophyll (HNLC) zone (~station 3) and the highly productive equatorial upwelling region (station 5). Eight (out of twelve) stations were sampled for plankton, and Table 1 details the coordinates for each station sampled. Two stations, station 3 and 5, were occupied for a 3-day period, which yielded the opportunity to collect multiple temporal zooplankton samples, and compare concentrations in night and day zooplankton collections. Separate day and night net tows were collected in order to investigate any differences due to the possible occurrence of diurnal migrating zooplankton. Water and plankton were sampled using trace-metal clean techniques.

Phytoplankton (seston) samples were amassed from known volumes of water collected out of Go-Flo bottles at the depth of the chlorophyll maximum of each station (Table 1). Phytoplankton was concentrated on acid-cleaned polycarbonate filters for various size fractionations by filtering seawater through 0.2 μm , 5 μm , and occasionally 20 μm filters. Potential occurrence of zooplankton in these fractions was impeded using a 200 μm mesh shield prior to filtering. Volumes filtered ranged from 0.5-2 L for the smallest fraction to 2-5.5 L for the larger fractions. Given this fractionation, phytoplankton size classes will be referenced as <5 μm (for 0.2-5 μm fractions), 5-20 μm and >20 μm . The 20 μm fraction represents plankton of >20 μm but <200 μm . Samples were collected at each size fraction for fluorescence, which was measured on board using a handheld Turner aquafluor fluorometer. In order to assess phytoplankton concentrations on a mass basis, biomass was calculated using both the chlorophyll

and phaeopigment measurements, assuming a constant carbon:chlorophyll ratio of 100 for the region (Taylor et al., 2011; Claustre et al., 1999; Wang et al., 2013). These values were further converted into biomass by assuming 40% carbon in biomass. The calculated phytoplankton biomass values were scaled by a factor of 3 to account for biomass from sources other than phytoplankton, such as bacteria, inorganic particles, fecal pellets and other detritus (Church, 2008; Libes, 1992). Thus, the reported concentrations reflect those of the different seston sizes. These concentration values are comparable to those collected by others using size fractionation approaches (e.g. Hammerschmidt et al., 2013).

Water profiles were collected at each station using trace metal clean techniques and a trace metal-free rosette. They were analyzed on board for mercury speciation concentrations, including total Hg and dissolved gaseous Hg (elemental and dimethylmercury). Samples were also collected and analyzed for nutrients, dissolved oxygen, temperature and salinity by the geochemistry group at the Woods Hole Oceanography Institute (Munson, K., 2014; Lamborg et al, 2014).

Zooplankton collections were completed using a vertically towed 200 μ m mesh opening/closing net, hauled on a Kevlar line, from depths of 200 m up to ~10 m. The net was closed at ~10 m below the keel of the ship in order to minimize potential vessel contamination. Zooplankton size fractionations were separated by using multiple acid-cleaned nylon mesh screens on a bench covered with plastic sheeting. Zooplankton was divided into sizes of 0.2-0.5 mm (200-500 μ m), 0.5-1.0 mm, 1.0-2.0 mm and >2.0 mm. Zooplankton size fractions were divided into multiple subsamples when large biota volumes (i.e. Station 5) permitted. Subsamples of the zooplankton fractions for carbon, nitrogen and sulfur determinations (C, N, S)

were also collected on pre-combusted 25 mm GF/F filters to assess cellular content of these nutrients.

Analytical

Samples were extracted in acid cleaned 15 ml centrifuge tubes with dilute 4.51 M HNO₃ (7 ml total) in a 60 °C water bath for ~12 hours prior to analysis (Hammerschmidt and Fitzgerald, 2005). For MeHg measurements, sample aliquots were neutralized using 10 N potassium hydroxide, and buffered with 4 M ammonium acetate (800 µL) to maintain the optimum pH of 4.9 in a sparging flask containing ~100 mL ultra pure (18.2 MΩ) water. Sodium tetraethylborate (200 µL; 1% w/v) was added to form derivative Hg species. Volatile ethylated derivatives were purged onto a Tenax trap using ultra high purity nitrogen gas (N₂). The sample was then thermally desorbed from the Tenax, releasing and separating the ethylated Hg derivatives via isothermal gas chromatography (GC). Methylmercury samples are ultimately quantified by Tekran 2500 cold vapor atomic fluorescence (CVAFS), based on the methylethylmercury peak area using an external calibration curve ($r^2 > 0.998$). There was not sufficient volume to perform substantial replicate sample analysis; however, standard addition and other QA/QC practices were incorporated into the analytical protocols to ensure measurements accuracy and precision. The RSD based on repeat analysis of recovered matrix standards was 6.7%. The detection limit was 0.016 pmol MeHg.

In order to completely extract all Hg for total analysis, brominemonochloride (BrCl, 40µl; 0.2% v/v) was added to the remaining 4.51 M HNO₃ sample aliquot, which was further diluted with 18.2 MΩ water, and digested >24 hours. Hydroxylamine hydrochloride (20 µl; 0.1% v/v) was then added to reduce any remaining BrCl prior to Hg analysis. A sample aliquot

was added to a sparging flask filled with ~ 100 mL 18.2 MΩ water to which tin chloride reductant (100 µl; SnCl₂) was added to reduce ionic Hg. The solution was then purged with N₂ to transfer elemental Hg onto a trap containing gold-coated beads (dual amalgamation). Once the trap was heated, it released gaseous Hg⁰ which was measured using Tekran 2500 CVAFS. The concentration was determined using an external calibration curve ($r^2 > 0.999$). Replicate analyses were completed when possible, and standard addition and other QA/QC practices were integrated into the analytical practices to ensure accuracy and precision. The RSD for total Hg based on repeat standard recovery was 2.9%, and the detection limit was 0.091 pmol.

Chlorophyll concentrations were determined on board as described above. Zooplankton samples collected on 25 mm GF/F filters were dried at ~60 °C for approximately 36 hours, and wrapped in tin foil capsules. These dried samples were then analyzed on a Fisons Instrument EA 1108 CHNS elemental analyzer for C, N and S measurements using an external standard calibration curve ($r^2 > 0.999$). Sulfur measurements were only completed for samples from Station 5. The precision of the measurements were ± 2 µmol, and the detection limit was 0.4 µmol for the EA analysis.

Results

Phytoplankton

Chlorophyll concentrations were highest in the <5 µm fraction at all stations and were >90% of the estimated seston biomass, confirming the dominance of small plankton in this ocean region. Chlorophyll concentrations were somewhat higher but comparable to other recent measurements in the region (e.g. Taylor et al., 2011), as expected given that sampling was at the

chlorophyll maximum at each station. The ratio of chl a/pheopigment (Chl/Phaeo) was between 1 and 2.5, with the higher ratios, signifying less degradation, being at the equatorial and near equatorial stations. The estimated suspended particulate concentrations for the <200 μm fraction ranged from 0.5 to 1.6 mg/L and was highest at Stations 5 and 12, and lowest at Stations 7-9. The higher concentration at Station 12 may appear surprising given that it was furthest from the equator in the Southern Hemisphere; however, this station was relatively close to the island of Samoa. Seston MeHg concentrations were highest in the 5-20 μm and >20 μm size fractions at stations 1, 3, and 6, and did not exceed 10 pmol g-biomass⁻¹ (wet weight) for the remaining size classes. The <5 μm size fraction exhibited the highest concentration at station 1 (8 pmol g-biomass⁻¹ (wet weight)), and decreased throughout the cruise transect from north to south, with relatively minor variations in concentration. As the <5 μm fraction dominated the biomass, the overall MeHg concentrations for the <200 μm seston fraction was similar, ranging up to 8.2 pmol g⁻¹ wet weight and was highest at stations 1 and 3 and lowest at station 12 (Figure 2a).

Inorganic Hg values were more variable, with highest concentrations for each station always in the >20 μm (58-1437 pmol g-biomass⁻¹ (wet weight)) or 5-20 μm (20.3-830 pmol g-biomass⁻¹ (wet weight)) size classes, and lowest concentrations (1.31-31.4 pmol g-biomass⁻¹ (wet weight)) were in the <5 μm fraction throughout the transect (Figure 2b). The extremely high value for >20 μm at station 3 (1437 pmol g-biomass⁻¹ (wet weight)) could be a result of mass estimation error due to the particularly low biomass compromising the calculation for HgT. Thus, it was concluded that this value represents an outlier and was not included in the figure. For the combined fractions (<200 μm) the concentrations ranged from 6 to 60 pmol g-biomass⁻¹ (wet weight).

As a potential explanation for some of the measured higher HgT values, there was an inverse relationship between HgT concentration in the various seston size classes and the Chl/Phaeo ratio in the fraction. For samples where the ratio was >1, reflecting actively growing biomass, the HgT concentration was lowest. The highest HgT concentrations were found for samples where the ratio was <1 (indicating relatively degraded biomass). In all cases this was for the largest size fractions. Furthermore, the Chl/Phaeo ratio was >1, and mostly >2, for every <5 µm fraction, suggesting that this fraction represented actively growing plankton biomass. There was some relationship between MeHg in seston and the Chl/Phaeo ratio, but it was less apparent than for HgT.

Notably, the percentage as MeHg (%MeHg; [MeHg]/[HgT]) in the seston was always highest in the <5 µm phytoplankton size class for each station (Figure 3a). Only stations 7 and 8 had %MeHg values that were under 20% for the <5 µm size class, while all partitions for the 5-20 µm and >20 µm size classes had values below 20% for %MeHg. Considering the overall <200 µm fraction, the %MeHg ranged from 3 to 29%, comparable to values found in other studies (<2 to 15%; Mason et al., 2012; Hammerschmidt et al., 2013), and similar to that found for suspended particulate during the cruise (up to 25% for the upper water column; Munson, 2014). As shown in Figure 3a, the %MeHg concentrations for phytoplankton were higher than in the water (Figure 3c; Munson, 2014), which is expected given that MeHg is accumulated more readily into phytoplankton over inorganic Hg (e.g. Mason et al., 1996). Moreover, the %MeHg in the dissolved fraction is relatively high for the stations occupied during the cruise compared to other ocean regions (Mason et al., 2012; Munson et al., 2015). Thus, the higher %MeHg in the seston of this study appears credible.

Ratios of the concentration in an organism to what was in the water column (Bioconcentrations factors; BCFs (L/kg)) were also calculated for all fractions at each station of the cruise transect. The water to phytoplankton step is the greatest concentration step in the trophic transfer of MeHg. The logBCFs ranged from ~3.8-7 for HgT, and ~4.1-6.8 for MeHg for all <200 μm size classes (Table 2). There was a slight trend in increasing logBCF values with increasing seston size class for HgT, but no trend with size class was evident for MeHg. Specifically, the HgT logBCF values were almost always greater than the MeHg log BCF values for both the >20 μm and 5-20 μm size fractions, but the majority of <5 μm fractions had larger MeHg log BCF values. The measured filtered HgT concentrations for the depths of the chlorophyll maximum were <0.2 pM in many instances in this study (Munson et al., 2015). These relatively low values, compared to previous measurements (0.5-2 pM; Mason and Fitzgerald, 1993; 0.3-1.5 pM Laurier et al., 2004 for stations around 25 °N) may be a reason for the higher estimated values in this instance. The higher logBCF for MeHg in the <5 μm fraction compliments the result that the highest %MeHg value for phytoplankton was always higher for the <5 μm size class. Average logBCF's for the entire seston (<200 μm fraction) ranged from 4.4 to 6.2.

Zooplankton

The C:N values for all size classes of zooplankton ranged from 4.4 to 8.5, representing a large distribution in cell nutritional content. The Redfield ratio for C:N is 6.6, therefore higher values indicated consumption of more degraded material or N limitation, as N is typically recycled faster than C. Theoretically, smaller C:N values could indicate that zooplankton species were consuming more nitrogen and possibly eating higher on the food chain, as C:N values

decrease in higher trophic level consumers. However, there was no consistent relationship between size class and C:N values for the transect (Figure 4a). In terms of other parameters measured, there was a correlation for sulfur content and MeHg for Station 5 (mmol:pmol; $r^2 = 0.5102$; $p = .0006$; $n=12$) where S was measured (Figure 5). However, no correlation was found with S and HgT (mmol:pmol; $r^2 = 0.0359$; $n=12$).

Total Hg concentrations for zooplankton size fractions ranged from 5 - 135 pmol g-biomass⁻¹ (wet weight; Figure 4b), while MeHg concentrations ranged from ~1 - 17 pmol g-biomass⁻¹ (wet weight; Figure 4c). Zooplankton abundance and MeHg concentrations both peaked at stations 3 and 5 (in the upwelling region), and lowest MeHg concentrations were found at the open ocean stations 7 through 12 (2 – 7 pmol g-biomass⁻¹ (wet weight)). The overall %MeHg in zooplankton was greatest at stations 1, 3, 5 and 7, and consistently increased with increasing size class (Figure 3b). There was variability in zooplankton HgT and MeHg concentrations throughout the transect, which is consistent with previous investigations (Hammerschmidt et al, 2013; Foster et al, 2012; Chen et al, 2009). Multiple measurements at each station agreed within the variability for HgT replicate measurements (± 0.3 -11.3 pmol g-biomass⁻¹) and MeHg (± 0.06 -2.2 pmol g-biomass⁻¹), although there were larger deviations at some stations (Figure 4).

No distinctive difference emerged between day and night zooplankton collections for HgT for all size classes at stations 3 (T-test, $p=0.608$, $n=14$) and 5 (T-test, $p=0.714$, $n=34$). There was also no difference for MeHg concentrations for all sizes of station 3 (T-test, $p=0.561$, $n=14$). On the other hand, all size fractions at station 5 contained higher concentrations of MeHg for night collections of zooplankton (T-test, $p=0.0001$, $n=34$). These results were consistent when each size class was evaluated separately. Therefore, it appears that there is no major

impact on HgT concentrations in diel migrating zooplankton for this study. However, migration could influence the MeHg concentrations for zooplankton. The results suggest that zooplankton residing deeper in the water column around the productive upwelling zone of station 5 have higher concentrations.

Bioconcentration factors for zooplankton were also calculated based on water concentrations for each station, and are reported in Table 3. The logBCF values range from 3.7-6.1 for HgT and 4.1-6.5 for MeHg. The logBCFs exhibit a much smaller range than those calculated for phytoplankton, as the overall average was ~5.3. Values are consistent with, though slightly higher than, previously reported numbers, as coastal and estuarine MeHg logBCFs range from 3.8 to 5.3 (Mason et al., 2012). The slightly elevated values are consistent with the higher BCF values found for phytoplankton. Average logBCF values for all size classes are greater for MeHg than HgT at station 1, though conversely all other stations exhibit the opposite tendency. Overall no clear trends emerged based on station or size fractions measured for zooplankton logBCFs. As zooplanktons are believed to acquire Hg and MeHg primarily from their feeding activities, the lack of variation is not unexpected.

Discussion

Phytoplankton

Previous studies in estuarine and coastal waters have reported logBCF values for MeHg from 3.3 to 4.2, lower than found in this study. Hammerschmidt et al. (2013) found that their values for MeHg logBCF increased from low values around 3.6 to 4.0 to values up to 5.0 at low suspended solid (TSS) concentrations, with the increase becoming apparent at TSS

concentrations of <1 mg/L. As estimated TSS concentrations for the equatorial Pacific during our cruise were mostly <1 mg/L, it is apparent that the logBCF's reported here are consistent with those found in their study off the western North Atlantic Ocean shelf. In addition, concentrations found here (0.5-8.2 pmol/g wet wt) are overall higher than those reported by Hammerschmidt et al. (2013) for their seston at the lower TSS concentrations (0.3-2 pmol/g). Hammerschmidt and Bowman (2012) estimated a concentration for mixed layer seston at the Pacific Ocean intercalibration SAFE site of 4 pmol/g wet wt, which is equivalent to a logBCF of 5.3 based on their measured filtered MeHg concentrations, and analogous to values reported here. Overall, our results and the other limited data for the open ocean suggest that relative concentrations of MeHg in open ocean phytoplankton are higher than that of coastal waters, though differences are less than an order of magnitude between coastal and offshore regions.

Higher concentrations for the open ocean, where plankton biomass are lower, are consistent with the idea that concentrations decrease as biomass increases, and growth rates increase (i.e. growth dilution; Pickhard et al., 2004; Hammerschmidt et al., 2013; Driscoll et al., 2012). It is evident that phytoplankton concentrate more Hg and MeHg from the water in the oligotrophic open ocean, suggesting that the MeHg may be more bioavailable in offshore waters. The higher MeHg and Hg concentrations in the larger sized classes of phytoplankton around the HNLC region could be a consequence of those plankton accruing more Hg and MeHg as they attempt to accumulate the limited essential metals (i.e. iron, zinc) of that section. Additionally, there is the potential for enhanced bioaccumulation in offshore waters due to carbon recycling through the biomagnifications of the microbial loop.

A further difference between coastal and offshore waters is the dissolved organic carbon (DOC) concentrations and composition, which is lower, more biologically derived, and less

degraded for subtropical and equatorial Pacific waters ($<80\ \mu\text{M}$; Taki and Suzuki, 2001) compared to coastal and shelf environments (Hansell and Carlson, 2002). These waters likely bind the Hg and MeHg less strongly than terrestrial-derived and humic-rich organic matter, increasing their bioavailability to plankton, as demonstrated by Schartup et al. (2015) when comparing coastal and offshore waters of the North Atlantic shelf.

Another explanation for the relatively high MeHg BCFs is that the area is dominated by small microbes and phytoplankton ($<5\ \mu\text{m}$), which have previously been shown to accumulate higher MeHg than larger organisms. Kim et al. (2012) found MeHg logBCF values, estimated from their reported volume concentration factors and organism size data, to range from 5.1 for the cyanobacteria *Chroococcus minutus* to 3.9 for a large diatom *Stephanopyxis palmeriana*. Overall, the bioconcentration factor increased as cell size decreased. In freshwater, Pichhardt and Fisher (2007) found a similar range in values for the logBCF, from 5.0 for the cyanobacteria *Synechocystis* to 4.3 for larger algal species. Through bioaccumulation modeling Driscoll et al. (2012) showed that the concentration in small phytoplankton ($<3\ \mu\text{m}$) could be up to 5 times that of larger species, a result consistent with culture studies. These differences are equivalent with values found in this study.

The %MeHg in phytoplankton size classes offer interesting insights into possible bioconcentration mechanisms. Methylmercury has been found to reside predominantly in the cytoplasm ($>60\%$) and bind to the proteins within cells (Wu and Wang, 2011; Mason et al., 1996), while inorganic mercury binds preferentially to the outer membranes of cells ($>90\%$), and is thus assimilated at a less efficient rate (Reinfield and Fisher, 1991). Hence, the surface to volume ratio of phytoplankton may be the factor influencing %MeHg for phytoplankton, as smaller phytoplankton have a greater surface to volume ratio, and therefore their MeHg is more

bioavailable for accumulation into zooplankton relative to HgT. That appears to be the case with our measurements, as the <5 μm size class has greatest %MeHg throughout the transect, even though that size class has the lowest concentrations of MeHg. Alternatively the highest %MeHg in that fraction could be an indication of Hg being methylated by bacteria which would be part of the <5 μm fraction. Lehnherr et al. (2011) demonstrated methylation within the high chlorophyll region in the Arctic Ocean, thus it is possible that it could also be occurring in the equatorial open ocean region. Unfortunately no microbial identification was undertaken during this cruise, so differentiation of potential methylating microbes between size fractions remains unknown for this region. However, based on incubation experiments using unfiltered waters, Munson (2014) found methylation at all stations in the chlorophyll maximum zone, suggesting that in situ methylation may account for the higher %MeHg in the <5 μm fraction.

Nutrients and dissolved oxygen water measurements did not bestow insights into factors controlling the variability of phytoplankton Hg and MeHg. However, there were correlations between MeHg concentrations in the phytoplankton and the corresponding water %MeHg, total Hg and MeHg concentrations. Peaks in the %MeHg in the plankton were at stations 3 and 9, which actually had relatively low levels of measured %MeHg in the water column (Figure 3c). This may suggest that MeHg uptake into phytoplankton and particulate export reduced the locally dissolved water MeHg concentrations for these sites. Alternatively, the low TSS and likely lower DOC at these sites resulted in higher bioaccumulation. Hammerschmidt et al (2013) reported no correlation between dissolved MeHg and MeHg in microseston (<200 μm), and currently few studies have been able to demonstrate strong relationships between marine water column MeHg and phytoplankton MeHg (Luengen and Flegal, 2009; Sunda, 2012). This markedly reflects the fact that one major driver of MeHg bioavailability in oxic surface waters is

DOC concentration, which likely does not vary in a consistent manner with dissolved MeHg. Freshwater studies have shown that the bioconcentration of MeHg decreases with the DOC concentration based on studies using DOC extracted from numerous locations (Luengen et al., 2012). Variations in DOC potentially accounts for the lack of correlation in the present study.

Zooplankton

Reduced sulfur resides in cellular protein, and is known to bind very strongly to Hg and MeHg through thiol ligands (Ravichandran, 2004; Yoon et al., 2005). The weak association with S indicates that diet and other factors are additionally important to zooplankton MeHg bioaccumulation, and predominantly more so for HgT bioaccumulation for these organisms. Chen et al (2008) noted that physical and ecological processes were more important than sources regarding MeHg transfer and bioaccumulation in marine food webs among estuarine and coastal regions. Additionally, laboratory experiments by Lawson and Mason (1998) showed that while dissolved speciation of MeHg impacted assimilation into phytoplankton, trophic transfer to zooplankton was not influenced by water column speciation. Our results from the analysis of open ocean plankton further strengthen this conclusion.

Only Station 1 appeared to display an increase in MeHg concentrations and corresponding decrease in HgT concentrations for increasing size classes. For the rest of the stations, this trend of increasing bioaccumulation with larger size fractions of zooplankton was inconsistent. Though the measured concentrations were variable from station to station, the %MeHg for zooplankton was relatively continual across the transect. The smallest size class (<0.5 mm) consistently had the lowest %MeHg. Furthermore, the %MeHg for the larger size classes amplified with increasing size fractions (Figure 3b). This trend was most prominent at

station 1, which was the station closest to Hawaii, and station 5, the upwelling zone. The greater %MeHg in the largest zooplankton is suggestive that these animals could be consuming smaller zooplankton rather than phytoplankton, or phytoplanktons which contain more %MeHg.

Due to the commonly higher MeHg concentrations in sub-thermocline waters compared to the mixed layer (Mason et al., 1990; Munson et al., 2015; Mason et al., 2012) it was hypothesized that zooplankton living at depth would have higher concentrations than those at the surface. The results obtained here suggest that their vertical migration and feeding at the surface mitigates any influence of residing in deeper waters during the day for station 3. This could also be a reflection of the relatively consistent MeHg concentrations through the water column at station 3. Nonetheless, the distinctly higher concentrations of MeHg in night collections of zooplankton at station 5 could be demonstrating this effect. The highest MeHg concentration for water was found below ~600 m at station 5, much deeper than zooplankton collection for this study. However, zooplankton can swim hundreds of meters within several hours (Brierley, 2014; De Robertis et al., 2003), which could be sufficient for animals residing in the deeper, high MeHg waters to have migrated into nighttime capture.

Water concentrations of HgT and MeHg were fairly low throughout the transect (Munson et al., 2015). As noted above, there was a slight increase in both the MeHg and %MeHg for station 5 (>0.05 pM and >10%, relatively), which corresponds to the increase in zooplankton %MeHg at that station (Figure 3b). While there was an additional %MeHg enhancement measured in all size classes of phytoplankton at station 5, there were additional peaks in stations 3 and 9 as well for the <5 μ m phytoplankton size class, which did not further correspond with water or zooplankton trends. However, these results should not be over-interpreted, considering

that phytoplankton were collected from only one depth, while zooplankton are likely feeding throughout the mixed layer.

Cumulative biomass collected for each tow at station 5 was substantially more abundant than any other station, which was a striking visual indication of productivity for the upwelling region of the equatorial Pacific. Considerable labile dissolved organic material can be produced via 'sloppy feeding' by zooplankton (Steinberg et al, 2004), in addition to ample fecal material and discharge resulting from turnover of dissolved organic carbon through the microbial loop (Møller and Nielsen, 2003). The presence of zooplankton has been shown to increase microbial productivity, likely as a result of increased derivatives of labile dissolved material (Peduzzi and Herndl, 1992). Algal decay has also been shown to release MeHg in natural waters (Luengen and Flegal, 2009), thus it is possible that ruptured and processed algae particles can further contributed to enhanced water MeHg within the mixed layer. As a result, it is feasible that the abundant zooplankton in this region of Station 5 could have contributed to amplified MeHg within the deeper water column by providing a significant input of processed and labile organic matter raining down from the surface ocean. This could potentially fuel microbial methylation in the deeper waters, especially below the oxygen rich euphotic zone. Higher sub-thermocline MeHg concentrations are found in the upwelling region, and mass balance estimations indicated it is mostly derived from in situ production, rather than due to transfer with sinking particulate (Sunderland et al., 2009; Mason and Fitzgerald, 1993). These potential sources of MeHg are evident in the large peak of %MeHg in the water column at the surface and directly deep below the prolific upwelling zone in Figure 3c (Munson, 2014).

This research reports on the first attempt at quantifying Hg and MeHg in different phytoplankton and zooplankton size fractions in the open ocean, a region where plankton are

relatively scarce compared to the more scrutinized coastal environment. Overall the results reported provide important clues into the sources and uptake of Hg and MeHg into phytoplankton, and transfer to zooplankton, as well as how plankton size impacts these relationships in the open ocean. However, results presented here have also highlighted the many gaps and questions that plankton Hg and MeHg bioaccumulation research needs to further investigate. Future research and modeling studies should build on these initial results to further elucidate open ocean Hg and MeHg cycling at the base of the marine food chain.

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Figure Captions

Figure 1. Cruise track starting from Honolulu, Hawaii to Apia, Samoa, with a brief stopover in the Island Republic of Kiribati. Stations which were sampled for plankton are noted on the map, and locations are detailed in Table 1.

Figure 2. (A) Methylmercury (MeHg) and (B) total mercury (HgT) concentrations (pmol/g wet wt.) for phytoplankton collected in the eastern central Pacific. Size classes are noted as <5 μm for the measured 0.2-5 μm fractions, 5-20 μm , >20 μm , and <200 μm for the entire phytoplankton average for each station.

Figure 3. (A) Percent of phytoplankton biomass as MeHg (%MeHg; [MeHg]/[Hg]) displayed for each size class, for each of the stations. (B) The percent of zooplankton biomass as MeHg demonstrated for each size class, for each station sampled. This graph includes all multiple vertical net tows completed at each station, as coordinated along the cruise tract. (C) The concentration of %MeHg in the water column for the cruise transect, displayed in depth from surface to 1000 m. Stations are approximately aligned, and numbered along the horizontal axis for each section. Station 5 (at equator) represents an equatorial upwelling zone, and displays some of the highest %MeHg in the biomass, as well as deeper within in the water column. Water column data for (C) was taken from Munson (2014), and used with permission of the author.

Figure 4. (A) The carbon: nitrogen (C:N) values for each zooplankton size class collected during the cruise. Size classes were divided as sizes of 0.2-0.5 mm (200-500 μm), 0.5-1.0 mm,

1.0-2.0 mm and > 2.0 mm. **(B)** Total mercury (HgT) and **(C)** methylmercury (MeHg) concentrations (pmol/g wet wt.) for multiple zooplankton size fractions at each station. Error bars indicate standard deviation values for duplicate subsamples for each size fraction, when measured. Stations 3 and 5 had multiple net tows for different days, including different tows at day and night time, and for separate days at the 3 day stations. Tows collected on a second day of the station 3 and 5 day/night samples are noted with 'b' after the station number.

Figure 5. Linear correlation (and 95% confidence intervals) of elemental sulfur (mmol/g dry wt.) to methylmercury (pmol/g wet wt.) in zooplankton of all size classes at station 5. Linear correlation is significant ($r^2 = 0.5102$; $p = 0.0006$; $n = 12$), and indicates a correlation with organic S, presumably within proteins, and MeHg concentration. There was no correlation for elemental sulfur (mmol/g dry wt.) with total mercury (pmol/g wet wt.) in zooplankton ($r^2 = 0.0359$; $n = 12$).

Table Captions

Table 1. Coordinates for plankton stations, and chlorophyll maximum depth sampled for phytoplankton, during the central Pacific research cruise.

Table 2. Bioconcentrations factors (logBCFs) calculated for each phytoplankton size class collected throughout the cruise transect. LogBCFs were calculated for both HgT and MeHg levels for each size class filtered.

Table 3. Bioconcentrations factors (logBCFs) calculated for each zooplankton size class, including separate day and night stations, collected throughout the cruise transect. LogBCFs were calculated based on measured water concentrations for each station ($\log\text{BCF} = \text{zooplankton concentration} / \text{water concentration}$). Both HgT and MeHg values were calculated for each size class measured.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Table 1. Coordinates for plankton stations, and chlorophyll maximum depth sampled for phytoplankton, during the central Pacific research cruise.

Station #	Longitude (°W)	Latitude (°S)	Chl Max. depth (m)
1	154.40	-17.00	115
3	156.00	-8.00	80
5	157.87	-0.36	40
6	160.77	3.50	75
7	162.61	5.96	75
8	165.36	9.25	79
9	167.56	12.00	60
12	173.10	15.00	125

Table 2. Bioconcentrations factors (logBCFs) calculated for each phytoplankton size class collected throughout the cruise transect. LogBCFs were calculated for both HgT and MeHg levels for each size class filtered.

Station #	LogBCF HgT <i>0.2-5μm</i>	LogBCF MeHg <i>0.2-5μm</i>	LogBCF HgT <i>5-20μm</i>	LogBCF MeHg <i>5-20μm</i>	LogBCF HgT <i>>20μm</i>	LogBCF MeHg <i>>20μm</i>
1	4.46	5.60	5.64	5.90	5.65	5.72
3		6.12	6.44	6.75	7.05	6.74
5	4.48	4.53	5.45	5.18	5.91	5.10
6	5.30	5.11	6.16	4.56	6.98	6.10
7	5.50	4.72	6.63	5.77		
8	5.20	4.88	6.74	5.42		
9	4.04	5.40	6.67	5.14		
12	3.77	4.33	6.16	5.36		

Table 3. Bioconcentrations factors (logBCFs) calculated for each zooplankton size class, including separate day and night stations, collected throughout the cruise transect. LogBCFs were calculated based on measured water concentrations for each station (logBCF = zooplankton concentration/water concentration). Both HgT and MeHg values were calculated for each size class measured.

Station	LogBCF HgT	LogBCF MeHg	LogBCF HgT	LogBCF MeHg	LogBCF HgT	LogBCF MeHg	LogBCF HgT	LogBCF MeHg
	<i>0.2-0.5 mm</i>	<i>0.2-0.5 mm</i>	<i>0.5-1.0 mm</i>	<i>0.5-1.0 mm</i>	<i>1.0-2.0 mm</i>	<i>1.0-2.0 mm</i>	<i>>2.0 mm</i>	<i>>2.0 mm</i>
1	4.00	4.54	4.05	4.79	3.87	4.88	3.77	5.02
3 day	5.22	5.51	5.24	5.76	5.10	4.48		
3b day	5.17	4.91	5.40	5.20	6.03	6.18		
3 night	4.96	5.14	5.27	5.37	4.77	5.57	5.42	4.24
3b night	5.80	6.53	5.51	5.85	5.54	5.30		
5 day	5.53	4.11	5.37	4.43	5.64	4.59	4.98	4.43
5b day	5.60	4.23	5.56	5.16	6.01	4.46	5.73	5.40
5 night	5.63	5.41	5.83	5.55	5.46	5.22	6.11	5.87
5b night	5.65	5.09	5.86	4.35	5.53	5.28	5.78	4.42
6	5.67	4.68	5.86	4.44	5.78	4.15		
7	5.92	5.26	5.98	5.58	5.83	5.36		
8	5.26	4.74	5.15	4.04	5.10	4.52		
9	5.66	5.00	5.23	4.60	5.46	4.81		
12	5.57	4.85	4.84	4.93	5.32	4.84		