1-1-2015

Mercury (Hg) speciation in coral reef systems of remote Oceania: Implications for the artisanal fisheries of Tutuila, Samoa Islands

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Abstract

We investigated Hg in muscle tissue of fish species from three trophic levels on fringing reefs of Tutuila (14°S, 171°W), plus water, sediment and turf alga. Accumulation of total Hg in the herbivore Acanthurus lineatus (Acanthuridae, lined surgeonfish, (n =40)) was negligible at 1.05 (±0.04) ngg^{-1} wet-weight, (~65% occurring as methyl Hg). The mid-level carnivore Parupeneus spp. (Mullidae, goatfishes (n =10)) had total Hg 29.8 (±4.5) ngg^{-1} wet-weight (~99% as methyl Hg). Neither A. lineatus or Parupeneus spp. showed a propensity to accumulate Hg based on body size. Both groups were assigned a status of "un-restricted" for monthly consumption limits for non-carcinogenic health endpoints for methyl Hg. The top-level carnivore Sphyraena qenie (Sphyraenidae, blackfin barracuda, n =3) had muscle tissue residues of 105, 650 and 741ngg^{-1} wet-weight (100% methyl Hg, with increasing concentration with body mass, suggesting that S. qenie >15kg would have a recommendation of "no consumption".

Keywords

samoa, tutuila, fisheries, artisanal, implications, oceania, remote, systems, islands, reef, mercury, coral, speciation, hg

Disciplines

Arts and Humanities | Law

Publication Details

Morrison, R. John., Peshut, P., West, R. J. and Lasorsa, B. K. (2015). Mercury (Hg) speciation in coral reef systems of remote Oceania: Implications for the artisanal fisheries of Tutuila, Samoa Islands. Marine Pollution Bulletin, 96 (1/2), 41-56.

Mercury(Hg) speciation in coral reef systems of remote Oceania: Implications for the artisanal fisheries of Tutuila, Samoa Islands

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ABSTRACT

We investigated Hg in muscle tissue of fish species from three trophic levels on fringing reefs of Tutuila (14^{o} S, 171^{o} W), plus water, sediment and turf alga. Accumulation of total Hg in the herbivore *Acanthurus lineatus* (Acanthuridae, lined surgeonfish, (n=40)) was negligible at 1.05 (±0.04) ng g^{-1} wet-weight, (~65 % occurring as methyl Hg). The mid-level carnivore *Parupeneus spp.* (Mullidae, goatfishes (n=10)) had total Hg 29.8 (±4.5) ng g^{-1} wet-weight (~99 % as methyl Hg). Neither *A. lineatus* or *Parupeneus spp.* showed a propensity to accumulate Hg based on body size. Both groups were assigned a status of "un-restricted" for monthly consumption limits for non-carcinogenic health endpoints for methyl Hg. The top-level carnivore *Sphyraena qenie* (Sphyraenidae, blackfin barracuda, n=3) had muscle tissue residues of 105, 650 and 741 ng g^{-1} wet-weight (100% methyl Hg, with increasing concentration with body mass, suggesting that *S. qenie* >15 kg would have a recommendation of "no consumption".

Key words: Mercury; Bio-accumulation; Coral reefs; Remote ecosystems; Artisanal fisheries

1. INTRODUCTION

The recent finalisation of the Minamata Convention to address the deleterious environmental effects of mercury (Hg) marks a major step forward in the control of the uses and distribution

of an element with a history of significant human health and ecosystem impacts (Burke, 2013). This global agreement should see significant controls and reductions on the products and processes where Hg is used or released into the environment. These uses range from medical equipment to light bulbs to power stations and mining activities. Mercury and its compounds are known to cause brain and neurological disorders, as well as kidney and digestive system damage. Under this new global agreement the production and trade in Hg containing products such as, batteries, cosmetics, mercury dental amalgams and certain types of compact fluorescent lamps will be banned by 2020, and controls on emissions of Hg from coal-fired power stations, cement factories and small scale gold mining operations will be dramatically increased.

Mercury is ubiquitous around the globe and occurs in the environment in various speciated forms including Hg^0, inorganic Hg(I) and Hg(II) species, and a series of organic forms with RHgX (especially MeHgX) and Me_2Hg being the most common (Leopold et al, 2010, van Dam et al, 2011). This speciation facilitates Hg movement rapidly around the globe in the atmospheric and aquatic environments including movement into and within coral reef environments, leading to contamination in various locations. For example, anomalously high Hg concentrations have been identified in sediments in various locations along the Great Barrier Reef resulting from use in mining (Bowling Green Bay) and agricultural pesticides (Upstart Bay) (Brodie et al, 2012). In the Pearl River estuary (Shi et al, 2010) Hg concentrations decrease on moving seawards along the estuary. In Venezuela, Ramos et al (2009) linked increases of Hg in coral skeletons to rainfall and runoff from industrial sites, while in the Caribbean, Guzman and Garcia (2002) found elevated Hg concentrations in various components of the reef environment, and related these to burning and mining activities with some evidence of long distance movement.

Speciation studies have focussed more on Hg forms in biota. Bloom (1992) found that in fish and invertebrates Hg was present in tissues predominantly (>95%) as MeHg forms, while Bank et al (2007) found that in snapper species in the Gulf of Mexico, MeHg species accounted for ~97% of total Hg, and that position in the food chain and eating habits can influence the nature and extent of bioaccumulation. These studies indicate the importance of accurate MeHg species measurements. Leopold et al (2010) present a major review of speciation measurement methods in natural waters while Chakraborty et al (2014a) present review sediment speciation methods. Chakraborty et al (2014b) were also able to identify the

importance of organic carbon in sediments and in the overlying water on Hg sequestration in sediments in India.

While there has been a major body of work in the last 50 years collecting information about the distribution, transport and speciation of Hg in the environment (e.g., Shia et al., 1999; Mason and Sheu, 2002; UNEP, 2002; Nriagu and Becker, 2003; Fitzgerald et al., 2007; Selin, 2009; Lavoie et al., 2013; Ferris and Essington, 2014; Ganguli et al., 2014), data from developing country locations isolated/remote from major Hg sources are quite limited. In the South Pacific region, data on Hg are relatively uncommon. Denton et al (2006) found enhanced Hg concentrations in sediments in the Tanapag Lagoon in Saipan (Commonwealth of the Northern Marianas, and later (Denton et al, 2011) were able to show that a source of some problems (including Hg in local fish species) was a hospital incinerator that was used for all waste including batteries, thermometers and fluorescent bulbs. Recently Lee et al (2015) measured the Hg in mollusc tissues in hydrothermal vents in the Tonga Arc and found total Hg contents of 11-66 µg/g dry weight with MeHg species being <0.1% of the total. None of these studies, however, have examined in any detail the pathways of Hg transfer through trophic chains. The objective of this study was to address this deficiency by investigating Hg in components of the coastal marine environment in Tutuila (14^{o}17'S, 170^{o}41'W), the main island of the American Samoa group and the site of Pago Pago, the capital and main port. This is a globally remote location, being 4,200 km SSW of Honolulu, Hawaii, 2,900 km NNW of Auckland, New Zealand and 10,090 km W of Lima, Peru. Landmasses between Tutuila and these points of reference are limited to small oceanic islands, many of which are uninhabited, amid large expanses of open ocean; Tutuila is therefore, remote from any major global sources of pollution, unless pollutants have been transported atmospherically over long distances (Morrison et al., 2010).

Tutuila, the largest (137 km^2) and most populous (~56,000) island in American Samoa, is the centre of Territorial government and commerce (Wingert, 1981; US Department of Commerce, 2001). As the island is dominated by steep terrain, habitation is almost exclusively confined to the southern coastal plain and southern coastal fringe, with a few small remote villages scattered along the north coast. The isolation of Tutuila, the small population, and limited commercial development (especially along the north coast) has provided an opportunity to assess the behaviour of Hg in the coastal environment in a relatively unpolluted part of the central South Pacific Ocean. This study looked at the

concentrations and speciation of Hg in marine waters, coastal sediments, turfgrass as a food source for herbivores, and at several fish species, to develop an understanding of patterns and possible food chain bioaccumulation.

Tutuila is a rugged volcanic island with a generally wet tropical climate. The 200 km long coastline is extremely irregular with numerous small bays open to the sea, which lack bars or barrier reefs, and are subject to the sea conditions of exposed coasts. Pago Pago Harbour is one of the few protected areas and maintains most of its natural shoreline, except for some filled areas near the main port, which have been developed for commercial shipping activities and two fish canneries. Tutuila has no estuaries and most of the coastline is rocky with abrupt elevation changes immediately above breaking waves. Surface waters are limited to a few dozen perennial streams, most of which have short, steep reaches, and typically low base flows. Coral reefs are the dominant marine habitat for Tutuila near-shore waters, with 60% of the coastline occupied by narrow fringing reefs (Morrison et al, 2010).

2. MATERIALS AND METHODS

2.1. Study Sites

Since no data were available from which to discern patterns of Hg distribution among Tutuila coastal reef environments, the selection of representative study sites was initially based on local knowledge. Six marine bay sites (Figure 1) with similar fringing reef habitats were selected, with five sites (Aasu/Fagafue, Tafeu, Vatia/Amalau, Masausi, Alega) representing fringing reefs on exposed coasts associated with catchments having minimal or no anthropogenic disturbance (“un-impacted”), and one site (Loa) represented a fringing reef on a protected coast where there is extensive residential and commercial development in the catchment (“impacted”).

INSERT FIGURE 1 ABOUT HERE

General reef geomorphology was similar for all study sites. Typically, there is a back reef of narrow to moderate width (< 150 m), with moat, and a prominent reef crest composed primarily of crustose coralline algae. Seaward from the crest is a steep-faced fore reef that

terminates at depths of ~10-25 m at a shallow-grade talus sand slope. An exception to general reef geomorphology is Tafeu Cove, which has a poorly developed reef flat structure that extends just a few metres seaward from the near-vertical mountain sides of the bay's catchment, with no moat and only an intermittent reef crest. One or two small perennial streams discharge into each bay. All study sites represented healthy and robust tropical reef habitats with species-rich assemblages of scleractinian corals, benthic and cryptic fauna, and fish. A brief description of each study site is presented in Table 1.

INSERT TABLE 1 ABOUT HERE

2.2. Sampling

2.2.1. General Aspects

All field samples for marine sediment (n=192), marine water (n=36), turf algae (n=30) and fishes (n=120) were collected mainly by the second author with assistance from technical personnel in the field and at the on-island laboratory; fishes were collected by technical divers under direct supervision. All species identification and body measurements for fish were completed as soon as possible after collection.

All sampling equipment was stringently acid-cleaned at the principal off-island analytical laboratory, and shipped and stored at the on-island staging laboratory in strict accordance with ultra-clean, trace elements field protocols. Collection, handling and shipping for all field samples were in accordance with the "clean hands, dirty hands" trace elements protocol, based on EPA Method 1669 (US EPA, 1996b). Chain of custody was maintained for transport of all samples from the on-island laboratory to the analytical laboratories, and all samples were analysed within method specified holding times, unless otherwise noted.

2.2.2. Marine Waters

Marine water samples were collected at Loa, Vatia and Aasu on three separate occasions during July, August, and October 2007. Two samples for Hg analyses and one sample for TSS analysis were collected from each of two sampling stations within each bay for each sampling event. Each sample was collected by hand in a clean 1 L Teflon® bottle using SCUBA at a depth of ~8 m, and ~25 m seaward from the fore reef. After collection, sample bottles were immediately stored on ice in the dark until return to the on-island laboratory.

At the on-island laboratory, marine water samples for Hg analyses were preserved with 0.2% low-Hg H_2SO_4 (18 mol L^{-1}, J.T. Baker Instra-Analyzed®) and stored in the dark until shipment to the analytical laboratory (usually < 1 week). Marine water samples for TSS analysis (not acid preserved) were stored at 4°C in the dark until shipment to the analytical laboratory.

2.2.3. Marine Sediments

Thirty two sediment samples were collected from each site; sampling and handling details are given in Morrison et al. (2010) using certified trace-metal clean, pre-labelled wide-mouthed 250 mL glass jars, as supplied by the analytical laboratory. The sediments were mainly coarse-grained coral dominated materials with small inclusions of volcanic materials and minimal (< 5%) contents of silt + clay. Samples were frozen immediately after collection, and were stored at -80°C until analysis.

2.2.4. Biota

2.2.4.1. Turf Algae Collection

Collection stations for turf algae were spaced at ~30 m intervals based on a selected transect length that approximated the transect length for marine sediment collections, and which encompassed the length of reef where herbivorous fish were collected. All turf algae patches sampled were qualitatively selected for similarity in colour, texture, density of cover, and growth height. Samples were collected in a depth range of 5-10 m at all locations, which encompassed the grazing range of the herbivorous fish. The extent of grazing pressure was determined on the basis of clear and apparent excessive growth, compared to the obviously grazed stands. Un-grazed turf was selected over grazed turf to facilitate collection of adequate tissue mass for ultra-trace elements analyses, since grazed turf was too closely cropped for efficient collection.

Turf algae were collected by dislodging turf-covered reef substrate by hand or with a small stainless steel hammer. Small- to medium-sized substrate projections of ~15-20 cm in length, with a length-width ratio of approximately 4:1, were targeted over more compact geometries. Long, thin projections facilitated easy removal from the reef matrix, improved handling and storage of samples, and maximised surface area to volume ratio to maximise tissue mass. Once the turf-covered substrate was free from the reef mass, the piece was waved vigorously

in the water column to dislodge any fine sediment that might be entrained within the base of the algal matt or within the holdfast weave. This procedure reduced the probability that fine sediments would be taken up for analysis when algae were removed from the substrate during laboratory processing. After collection, turf covered pieces were carried to the surface, immediately rinsed gently with deionised (DI) water, placed in double zip-seal bags, and placed on ice in the dark for transport to the on-island laboratory. At the on-island laboratory algae samples were stored in the dark at 4^{o}C (to avoid rupture of cells and thus potential loss of Hg from tissue resulting from freezing) until shipment to the analytical laboratory.

2.2.4.2. Herbivorous Fish Collection (surgeonfish, *Acanthurus lineatus*)

Twenty *A. lineatus* specimens were collected from the Loa, Vatia and Aasu Bays, consistent with the study sites for marine water and turf algae. In the field, surgeonfish were collected by hand spear within two hours after dusk, using SCUBA. All specimens were speared in the head to avoid puncture of muscle tissue or the gut. After spearing, each specimen was returned to the boat for euthanisation in an ice bath. Fish were then rinsed with DI water, and placed in labeled zip-seal bags on ice for transport to the on-island laboratory, where they were identified to species, measured for standard length and fork length, and weighed. Whole fish were frozen at -20^{o} C for storage at the on-island laboratory until shipment to the analytical laboratory.

2.2.4.3. Carnivorous Fish Collection (goatfish, Mullidae spp.)

Due to availability, Mullidae collections did not target a single species. Goatfish in American Samoa are represented by 13 species, none of which appear in large schools, nor consistently among all reef habitats (Wass, 1984; P. Brown, US National Park Service, pers. comm.). Moreover, there is considerable variability among species' feeding behaviours, with about half the species each in daytime and night-time feeding regimes (Myers, 1999; Randall, 2005). All Mullid specimens were collected from the same area where sediment samples were collected; a total of 20 Mullidae spp. were collected from each marine bay for the Loa, Vatia and Aasu study sites, consistent with study sites for marine water, turf algae, and herbivorous fish. Mullidae specimens were collected, handled, and stored as described above for *A. lineatus*.

2.2.4.4. Carnivorous Fish Collection (barracuda, *Sphyraena qenie*)

Collections for *S. qenie* were targeted for late-afternoon to dusk, when barracuda are known to disperse from the shelter of the reef to feed (Myers, 1999). Range, time, and method of capture were based on local knowledge and experience of the barracuda catch in Tutuila waters. Minimum numbers of field samples or specific sampling locations were not specified for *S. qenie*, due to the inherent vagaries of off-shore trawling. Three *S. qenie* were taken on trawled lures from small sport-fishing craft along the southeast coast of Tutuila, within 1 km of the shore. After body measurements and weight were taken, a 20 g (approximate) sample of boneless and skinless *S. qenie* muscle tissue was removed from the thickest part of the dorsal muscle, using a clean dissection scalpel. Tissue samples were frozen at -20^{o} C at the on-island laboratory until shipment to the analytical laboratory.

2.3. Laboratory Analyses

All laboratory analyses for Hg for all environmental matrices were conducted by the Battelle Marine Sciences Laboratory, Sequim, Washington, USA. Mercury compounds in marine water were analysed using cold vapour atomic fluorescence (CVAF), with analytical results reported as ng L^{-1}. All results for solid matrices were reported as ng g^{-1} or μg g^{-1} dry-weight. For tissues, Hg compounds were analysed using CVAF and were reported as ng g^{-1} wet-weight. Table 2 summarises instrumentation, analytical method, and detection limit (DL) information for each environmental matrix investigated. Summaries of analytical quality assurance and quality control (QA/QC) results are presented in Peshut (2009).

2.3.1. Mercury in Marine Water

Water samples were analysed for THg and MeHg as using CVAF and US EPA Methods 1631 E and 1630 respectively (US EPA, 2001a, 2002). Detection levels achieved were 0.188 ng L^{-1} for THg and 0.0188 ng L^{-1} for MeHg. All reagents used in the analyses were verified to have very low Hg content or were purified prior to use to ensure negligible reagent blank concentrations. One acid blank and one field blank were prepared – the acid blank consisted of a ~25 mL aliquot of H_2SO_4 taken from the 500 mL original acid container. The field blank consisted of a sample bottle filled in the field from DI water provided by the analytical laboratory.

For TSS, 1.0 L of un-preserved marine water sample was vacuum filtered on pre-weighed nominally 0.45 μm glass fibre filter membranes. Membranes were dried to constant weight at 60^{o} C. Weight differential was calculated to obtain the final reported result (Method SM2540D, Rice et al., 2006). The DL achieved was 0.1 mg L^{-1}.

INSERT TABLE 2 ABOUT HERE

2.3.2. Mercury in Marine Sediments

Sediment analysis methods are summarised in Table 2. MeHg was analysed on wet sediment and the concentration was converted to a dry-weight basis using % moisture data. Following MeHg analysis, samples were freeze-dried and homogenised to a fine powder in a ball mill prior to digestion for analyses of THg.

The CVAF techniques used for Hg analyses in marine sediments (EPA 1631E, EPA 1630) were similar to methods used for Hg marine water samples, and differed only in the digestion of the solids matrix for analysis. For sample digestion for THg analysis, 5 mL of aqua regia (4 HCl: 1 HNO_3) was added to a 0.2 g aliquot of homogenised sample in a glass vial and allowed to digest at room temperature for 24 h. After digestion, 16 mL of a solution of BrCl (0.07 mol L^{-1}) in DI water was added to destroy any remaining organics. Following digestion, Hg^{2+} in an aliquot of the sample was reduced to Hg^{0} by the addition of $SnCl_2$ solution to the purge vessel. For MeHg, sample extraction was in accordance with procedures described for solids by Bloom et al. (1997). Total organic carbon (TOC) analyses were carried out using high temperature combustion following ATSM method D4129-82 (ATSM, 1982).

2.3.3. Mercury in Coral Reef Biota

Analyses for THg and MeHg in biological tissue used the same atomic fluorescence techniques used for marine water samples, and differed only in sample preparation and the digestion of the tissue matrix for analysis (Table 2).

Algae samples were stored at 4^{o} C in complete darkness until processing. Under trace metal clean room conditions, algae were carefully removed from the substrate by mechanical

extraction with stainless steel forceps. Care was taken to avoid forceps contact with substrate. To the extent practicable, turf algae material was extracted from the substrate in a manner that avoided fine sediment entrained within the matt base. A 5.0 g sample of algal tissue was retained from each field sample for analysis. Whole surgeonfish and goatfish, and a 20 g sample of barracuda muscle tissue, were received frozen at the analytical laboratory and were stored at -20^{o} C until sample processing. For the whole fish, a 5.0 g skinless tissue sample was removed from the thickest part of the fish dorsal muscle for analysis. For the barracuda sample, a 5.0 g sample was removed from the field sample. Tissue samples were freeze-dried and homogenised to a fine powder in a ball mill prior to digestion.

For THg, samples were digested in 70% HNO_3/30% H_2SO_4 and analysed in accordance with Method 1631E (US EPA, 2002). For MeHg, sample digestion was in accordance with procedures described for tissue by Bloom (1989). Following digestion, quantification of THg and MeHg in tissue samples followed procedures for marine waters.

2.4. Statistical Applications for Evaluating Hg in Environmental Matrices

For parametric data sets, analyses of variance (ANOVA) and Student-Newman-Keuls (SKN) tests were used for statistical comparisons. For non-parametric data sets, Kruskal-Wallis ANOVAs and either Student-Newman-Keuls means rank test or Dunn procedure were used. All parametric and non-parametric statistical analyses were limited to single-factor comparisons.

Simple (two variable) linear regressions were used to evaluate functional relationships between Hg in muscle tissue and body weight for fish. A significance level of α=0.05 was selected for all statistical analyses.

Bio-accumulation factors (BAFs) were calculated along the herbivorous and carnivorous trophic gradients, with marine water as the common reference point. A BAF for each trophic gradient was calculated for the Aasu, Vatia and Loa sites. The un-impacted sites of Aasu and Vatia were also combined for a BAF calculation.

The herbivorous trophic gradient included:

- Marine water » Turf algae » *A. lineatus* muscle tissue.

The carnivorous trophic gradients included:

- Marine water » Mullidae spp. muscle tissue;
- Marine water » *S. qenie* muscle tissue.

The BAF for each trophic step was calculated as a simple quotient, using the step as numerator and the previous step as denominator. Mean concentrations for THg and MeHg in each matrix were used for BAF calculations.

2.5. Assessment of Human Health Risks from Consumption of Tutuila Reef Fish

a. Basis for risk-based consumption limits

Risk-based fish consumption limits for MeHg for surgeonfish and goatfish were prepared in accordance with US EPA guidance (US EPA, 2000). Consumption limits determine the allowable number of fish-meals that can be consumed over a given time period (month), based on the concentration of MeHg found in the target tissue. Methyl Hg is not known to be carcinogenic, so non-cancer health endpoints were assessed using a hazard quotient of 1 (US EPA, 2000). US EPA has established an interim reference dose (RfD) of 1×10^{-4} mg kg^{-1} d^{-1} for MeHg based on data on neurological effects in children who had been exposed to Hg *in utero* (Marsh et al., 1987). This RfD was considered scientifically sound and is supported by human and animal studies (ATSDR, 1999; US EPA, 2000).

b. Calculation of risk-based consumption limits for Methyl Hg

Two equations were used to calculate consumption limits based on non-cancer health endpoints for MeHg. Equation 1 is used to establish the maximum allowable daily rate of ingestion based on the concentration of MeHg in fish muscle tissue. Equation 2 is used to establish number of fish meals allowed for consumption during a given one-month period, so not to exceed the average daily intake limit for MeHg. Consumption limits for MeHg in fish muscle tissue are based on a 0.227 kg (8-ounce) meal size, and a body weight of 70 kg.

Equation 1. Non-cancer health endpoints:

$CR_{lim} = (RfD)(BW)/(C_m)$

where:

CR_{lim} = Consumption Rate, maximum allowable limit (kg d^{-1})

RfD = Reference Dose (mg of MeHg per kg of body weight per day)

BW = Body Weight (kg)

C_m = Concentration of MeHg in fish muscle tissue (mg kg^{-1} wet-weight)

Equation 2. Consumption limits:

Meals per month = $(CR_{lim})(T_{ap})/(MS)$

where:

CR_{lim} = Consumption Rate, maximum allowable limit (kg d^{-1})

MS = Meal Size (kg fish $meal^{-1}$)

T_{ap} = Time averaging period (365.25 d y^{-1} ÷ 12 mo y^{-1} = 30.44 d mo^{-1})

One consumption limit table was prepared for goatfish and one for surgeonfish for each study location where fish were collected. Tables were prepared based on mean MeHg in muscle tissue of fish from respective sites. If the average concentration of MeHg fell between two consumption limits, the more conservative consumption rate was selected.

3. RESULTS AND DISCUSSION

3.1. Mercury in Marine Water at Tutuila

Marine water samples were analysed to provide a reliable value for Hg in reef waters, to serve as a reference datum for calculation of BAFs. Correction factors to account for potential contamination from Hg in acid used for sample preservation, or from contaminated sampling equipment, were negligible. For all sites, THg occurred at trace levels, < 4 times the DL in all samples (Table 3) with Methyl Hg occurring at < 5 times the DL for all but one sample at Loa (Table 4).

INSERT TABLES 3 AND 4 ABOUT HERE

All Hg in marine water data met assumptions of ANOVA, except for marginal departures in homogeneity of variance for the Aasu MeHg (p=0.028), and normal distribution for Loa MeHg data (p=0.043). ANOVA was therefore used to evaluate differences in THg and MeHg concentrations in marine water among the three study sites. There was a significant difference among sites for THg (p=0.013) and for MeHg, (p=0.007), attributable in each case

to higher Hg levels at Loa, the impacted Harbour site. There was no difference in Hg in marine water between the north-shore un-impacted sites of Aasu and Vatia (p>0.05).

A non-parametric method was used to confirm differences for Hg among sites because of the relatively small sample size per site (n=12), and the marginal violations of assumptions for ANOVA for MeHg data. In agreement with the parametric test, the Kruskal-Wallis test showed a significant difference among sites for THg (p=0.026) and for MeHg (p=0.010), attributable to higher Hg levels at Loa, and no difference in marine water Hg between the un-impacted sites of Aasu and Vatia (p>0.05). The slightly elevated levels of Hg in Loa surface waters compared to Aasu and Vatia, probably result from significantly reduced mixing and exchange of surface waters in the Harbour compared to the exposed north shore (US ACOE, 1979).

When results for Aasu and Vatia are combined (n=24), mean THg was 0.322 ±0.016 ng L^{-1} (0.206-0.485 ng L^{-1}), and mean MeHg was 0.029 ±0.004 (nd-0.065 ng L^{-1}), in good agreement with historical and recent data for THg in open surface waters across the northern and southern Pacific Ocean. No reliable data were found for MeHg in open ocean environments from historic or contemporary studies. Gill and Fitzgerald (1987) reported THg in marine water from a range of data sets that included: offshore stations in the Tasman Sea beyond the influence of North Island of New Zealand upwelling, with mean THg of 0.56 ±0.08 ng L^{-1} (0.40-0.72 ng L^{-1}, n=4); along the 160^{o} W meridian between Hawaii and Tahiti, with mean THg of 0.27 ±0.06 ng L^{-1} (0.17-0.44 ng L^{-1}, n=5); and, also along the 160^{o} W meridian, mean THg of 0.42, 0.44 and 0.40 ng L^{-1}, (for all values, n=4, standard errors and data ranges not given). From a 1990 equatorial Pacific cruise, Mason and Fitzgerald (1991) reported mean THg in near-surface waters in the range of 0.15-0.35 ng L^{-1}. Laurier et al. (2004) reported THg in North Pacific surface waters for four cruises in the western, central, and east-northeast sectors from 1980-2002. In the mixed surface layer (~ 75 m), overall mean THg was 0.28 ±0.07 ng L^{-1} for the 1980 cruise, 0.11 ±0.07 ng L^{-1} for the 1986-1987 cruises, and 0.13 ±0.05 ng L^{-1} for the 2002 cruise. If near-surface samples only are considered (~10 m) for the 2002 cruise, mean THg between Japan and Hawaii ranged from 0.15-0.40 ng L^{-1}, in excellent agreement with results for Tutuila reef waters (depth ~ 8 m).

Total suspended solids (TSS) were investigated for marine water primarily to provide a qualitative assessment of Hg phase in solution (dissolved or adsorbed) for Tutuila reefs.

Concentrations of TSS appear consistent among study sites (Table 5). Relatively elevated mean TSS for the un-impacted north-shore site of Aasu is likely attributable to the anomalously high readings for MW1-1a and 3a, since all other values for Aasu and other sites lie within the fairly narrow range of ~0.75-1.75 mg L^{-1}. All MW1 samples for all sites were collected on the same day, within a few hours of each other, and under similar sea and weather conditions. It is possible that a localized event introduced an unaccountable bias in the MW1 samples for Aasu. If the anomalous values are removed from calculation of the mean, the resultant 1.14 mg L^{-1} for Aasu is in good agreement with the other sites, and the overall mean for all sites is 1.03 mg L^{-1}, in excellent agreement with limited historical data for TSS in Tutuila near-shore waters of ~1 mg L^{-1} (CH2M Hill, 2007; DiDonato et al., 2007).

In clear waters (<2 mg L^{-1}) the particle-water partition coefficient for Hg is $>10^6$, and Hg would be expected to occur mainly in the <0.45 μm filter-passing "dissolved" phase (Choe et al., 2003). Partitioning of Hg between dissolved and particulate phases (>0.45 μm) has been quantified by a particle-water partition coefficient (K_D) applied to estuarine waters (e.g., Choe et al., 2003) though no application of this method was found for tropical reef waters. Colloid-bound Hg as part of the "dissolved" phase Hg was first identified by Wallace et al. (1982) and Santschi (1982) and may occur as one-half or more of Hg not bound to large particles (>0.45 μm) (Stordal et al., 1996; Choe et al., 2003). For this study, unfiltered reef water samples were analysed for Hg, so a determination of colloid-bound Hg and truly dissolved Hg could not be made. At the range of TSS found for Tutuila reef waters, dissolved Hg (<0.45 μm, filter-passing) is expected to predominate in the reef water column.

INSERT TABLE 5 ABOUT HERE

MeHg occurred in Tutuila near-shore marine water as 7.6%, 10.3 %, and 12.3 % of THg, for Aasu, Vatia, and Loa, respectively, ~20-30 times higher than the percentage MeHg in Tutuila rainfall (~0.4%, Peshut, 2009). Enrichment of MeHg as a proportion of Hg in Tutuila near-shore surface waters compared with rainfall input is potentially attributable to a terrestrial source, since significant *in situ* methylation of Hg in the marine water column is inconsistent with the biological methylation theory (Weber, 1993; Benoit et al., 1999; Weber, 1999) based on the relatively high sulfate concentration in seawater (~2700 mg L^{-1}). Another important factor that may further limit methylation in the clear, oligotrophic waters of the tropical coral

reef is the availability of appropriate methyl donors, such as, fulvic or humic acids (Celo et al., 2006).

Potential limitations on methylation in tropical reef waters, and enriched MeHg in the water column, leads to the suggestion that a sustained terrestrial input (humic-bound Hg) may be important as a source for MeHg to Tutuila reef systems. As discussed below, elevated Hg in Tutuila reef sediments compared with expected values for marine sediments also suggests a sustained terrestrial input of Hg to Tutuila reefs. Careful further study is needed to determine the relative roles of direct Hg inputs to the water column via stream discharges, or Hg inputs to overlying water via a sediment pathway.

3.2. Mercury in Tutuila Marine Sediments

There was a clear progression of increased THg and MeHg in basin sediments from east to west among the five un-impacted sites (Tables 6 and 7), except for a slight, and insignificant, decrease at Vatia. There was a sharp rise in THg for the impacted Harbour site of Loa, but only a marginal increase in MeHg at Loa compared with un-impacted sites. Among un-impacted sites, the pattern for Hg in basin sediments is consistent with the pattern for TOC (r^2=0.564, in line with Chakraborty et at, 2014), and consistent with THg and TOC concentrations found in stream suspended solids entering each bay (Peshut, 2009).

The redox horizon in marine sediments (data not reported) was documented to validate that sediment samples were collected consistently from oxic conditions among all study sites (sampling depth limited to ~6 cm). Additionally, a distinct oxic/anoxic boundary layer indicated sediment stability, which showed that study design criteria were met for collecting from accumulated and stable sediment deposits that were not subject to significant disturbances by current or wave action.

INSERT TABLES 6 AND 7 ABOUT HERE

Statistical comparisons of Hg in sediments indicated a significant difference in sediment THg between Loa and all un-impacted sites (p<0.001). For MeHg, Loa was significantly higher than Masausi, Alega, Vatia, and Tafeu, (p<0.05), but was not significantly different from Aasu (p~0.50). Higher concentrations of THg in Loa reef sediments compared to the un-

impacted sites, but similar patterns for concentrations of MeHg among all sites, supports the concept that anthropogenically-derived, non-point source Hg contributes to elevated Hg levels at the impacted Harbour location. As expected, there was a fairly strong association between MeHg and THg among un-impacted sites (r^2=0.61), but the mean MeHg:THg ratio among un-impacted sites was 2.1 % (1.7-2.8%), which is somewhat high for marine surficial sediments, which are typically <1 % MeHg (Kannan and Falandysz, 1998; Bloom et al., 1999; Wasserman et al, 2002; Sunderland et al., 2006). The ratios found for Tutuila reef sediments are more representative of freshwater systems (Gilmour et al., 1998; Morel et al., 1998). Elevated ratios for MeHg in Tutuila reef sediments may be additional evidence for a sustained and significant terrestrial input of MeHg to Tutuila reef systems.

3.3. Total Organic Carbon in Tutuila Marine Sediments

The pattern for TOC in marine sediments follows the pattern for THg and MeHg unerringly for the un-impacted sites, but differs for the Loa Harbour site (Table 8). Extensive disturbance of the Loa catchment area through engineered fill for structures, construction of impervious pavement, and the distribution of beach sand for landscaping, has almost certainly altered the composition of eroded materials at Loa compared with the un-impacted catchment areas. Celo et al. (2006) and Chakraborty et al (2014) discuss the importance of organic carbon compounds as receptors of Hg in the terrestrial environment, and as part of the transport and transformation pathways (donors for organic ligands in methylation processes) and this may indicate that Hg in Tutuila's aquatic environment is of significant terrestrial origin.

INSERT TABLE 8 ABOUT HERE

3.4. Mercury in Tutuila Coral Reef Biota

3.4.1. Mercury in Turf Algae

This component aimed to quantify Hg species in this exclusive food source for the herbivorous surgeonfish *Acanthurus lineatus*, to calculate Hg accumulation factors for turf algae referenced to water column, and to provide intermediate resolution of BAFs for the herbivorous trophic gradient on Tutuila reefs. Mercury in turf algae varied little among the

un-impacted north-shore sites and the impacted Harbour site (Tables 9 and 10), with similar concentrations for Aasu and Loa turf, and only slightly lower levels for Vatia. There was no significant difference in turf algae THg among study sites ($p>0.05$). For Vatia, turf algae MeHg was lower than the other sites, and was below the DL for all samples. Data for MeHg in turf algae did not meet assumptions for ANOVA, but the non-parametric Kruskal-Wallis Test showed that all sites were significantly different from each other for MeHg, ($p<0.001$). It was recognised that mean MeHg values showed relatively small differences, and were near the limits of analytical capabilities, which precludes a robust interpretation of differences in MeHg in turf algae among reef sites.

INSERT TABLES 9 AND 10 ABOUT HERE

Of considerable interest regarding Hg movement through environmental compartments on remote Tutuila, was the unexpected magnitude of accumulation found between the water column and turf algal tissue. Accumulation factors for THg for marine water » turf algae were similar for all sites, at $10^{4.6}$, $10^{4.5}$ and $10^{4.5}$ for Aasu, Vatia, and Loa, respectively while accumulation factors for MeHg were marginally more variable. As discussed later, the principal bioaccumulation step for Hg in the herbivorous trophic uptake pathway investigated here is from the water column to turf algae tissue, and that for both THg and MeHg, a less significant ($10^{\pm1}$) step occurs between turf algae and muscle tissue of the herbivorous *Acanthurus lineatus*.

The pattern of accumulation between water column Hg and algal tissue Hg, and between algae and fish tissue Hg for Tutiuila reefs, is fundamentally similar to that described by Mason et al. (1995) for bio-accumulation steps for water, phytoplankton, and fish, where the greatest accumulation ($\sim10^5$) takes place between the abiotic and biotic compartments, via phytoplankton primary production. Ionic Hg species prevail in marine water (Bloom and Crecelius, 1983; Gill and Fitzgerald, 1985; Gill and Fitzgerald, 1987; Fitzgerald and Mason, 1996), and given the large accumulation factors between water and algal tissue found here, it is reasonable to suppose that dynamics of active transport of Hg across cellular membranes in coral reef turfs are similar to those shown for phytoplankton and macroalgae (Morris and Bale, 1975; Eide et al., 1980; Mason et al., 1995; Mason et al., 1996).

High accumulation factors for Hg in Tutuila turf algae are noteworthy because these algae, as the source of most primary production on the reef, are grazed extensively by a great variety of marine herbivores, with biomass turnover ~4-12 days (Carpenter, 1985; Klumpp et al., 1987). Magnification of Hg in turf algae of at the levels found here, consistent across three widely separated reef systems, appear remarkable given the ephemeral nature of turf algae on the reef, and indicate a highly efficient mechanism for Hg uptake from reef waters via the primary production pathway on coral reefs. These findings challenge conventional notions that sediments are the principal sink for Hg pollution, and the principal source for Hg uptake in aquatic food webs.

On coral reefs, turf algae and accumulated basin sediments are distinct habitats, and likely form independent bases for feeding regimes. Hg may enter the food web independent of sediment-associated pathways through uptake by biota that feed on turf algae. The convergence of sediment-based and turf-based uptake pathways in top-trophic predators that feed directly or indirectly on prey from both feeding regimes, could reasonably be expected to be manifested in an increased body burden of Hg. This may help explain the unexpectedly high levels of Hg found in the top-tropic reef predator *Sphyraena qenie*, even though these fish were of relatively small size and occupied pristine reef environments.

Efficiency of Hg uptake in the turf algae pathway and the potential importance of primary production as a major magnification step for Hg on coral reefs is evident when concentrations of Hg in algae are compared with reef sediments. On the basis of dry-weight, turf algae had consistently higher THg (2-20 fold) and MeHg (10-40 fold) than marine sediments. An enriched MeHg:THg ratio for turf algae compared with sediments suggests the possibility of preferential sequestration of Hg in turf algae, or turf algae as a biotic methylation site in the marine environment. These findings suggest that a significant proportion of Hg that accumulates in reef biota has its origin through the turf algae pathway.

3.4.2. Mercury in Reef Fish

3.4.2.1. Acanthurus lineatus

For *A. lineatus*, all specimens collected were used for study purposes (Aasu n=20, Vatia n=20, Loa n=20). For the Mullids, only specimens of *Parupeneus cyclostomus* were used for

comparisons among study sites (Aasu n=5, Vatia n=5, Loa n=11) because an evaluation of inter-species variability of weight and muscle tissue Hg showed that meaningful comparisons could likely not be made with multiple species data. There was no difference in THg and MeHg from *A. lineatus* among the un-impacted sites of Vatia and Aasu (p>0.05), with ~60-65 % of Hg in muscle tissue occurring as MeHg (Table 11). Values for THg, MeHg, %MeHg, and weight, appear consistent for fish among the un-impacted sites. Because of the high site fidelity of these fish (Craig, 1996) and the distance between the study sites (~8 km), the probability of overlapping populations of *A. lineatus* between Vatia and Aasu was considered negligible. With regard to Hg in herbivorous fish, these sites were interpreted as independent, and highly representative of un-impacted coral reef systems in remote Oceania.

Muscle tissue THg and MeHg for *A. lineatus* from Loa were significantly higher (p<0.001) than for fish from Aasu and Vatia. The six-fold increase for THg in *A. lineatus* from the Loa site compared with Aasu and Vatia fish, the corresponding order of magnitude increase for MeHg and the increased MeHg:THg ratio (~95%), combined with the overall smaller size of Loa fish, is not easy to explain. There was no difference in turf algae THg among the un-impacted and impacted sites, and only slight differences in turf algae MeHg among sites, so differences of Hg in food source for *A. lineatus* cannot be associated with differences in muscle tissue Hg among un-impacted and impacted sites.

INSERT TABLE 11 ABOUT HERE

Sediment Hg at Loa was significantly higher than the Aasu and Vatia sites, and may offer a plausible explanation for the differences in muscle tissue Hg for *A. lineatus* among the Loa and un-impacted sites. Field observations confirmed that the turf algae matt entrains fine sediment and dissection of fish showed that full stomachs from *A. lineatus* contained a large (un-quantified) amount of sediment material. If it is assumed that scavenging of Hg from the water column deposits Hg to turf matts in a manner generally similar to deposition of Hg in basin sediments, then increased uptake of Hg via ingestion of entrained turf sediment while feeding could explain the higher levels of Hg in muscle tissue of *A. lineatus* at Loa.

A. lineatus from all sites showed no relationship between body weight and Hg accumulation in muscle tissue (r^2 for all sites <0.0036), although accumulation was much greater in fish from Loa than from Aasu and Vatia. The data from the Aasu and Vatia sites suggest that Hg accumulation reaches a limit in fish from un-impacted environments. The Loa data suggest

that if physiological functions limit accumulation of Hg in coral reef fish, these are easily overwhelmed by an increase in Hg above normal background levels. Accumulation patterns for Hg displayed for *A. lineatus* were found to be similar for the benthic-feeding carnivorous Mullids from Aasu and Vatia (see below).

3.4.2.2. Mullidae spp.

Mullids feed extensively on invertebrate benthic infauna (mainly worms, mollusks, and crustaceans) primarily in sediments below the lower reef margin (Wahbeh, 1992; Myers, 1999; Kulbicki et al., 2005), and were considered to be potentially good indicators of Hg sequestration and uptake from the sediment compartment of remote coral reef environments. Parametric (ANOVA) and non-parametric (Kruskal-Wallis) methods agreed for comparisons among sites for Hg in Mullid muscle tissue. There was no difference in THg or MeHg in *P. cyclostomus* muscle tissue among un-impacted sites ($p>0.05$), but fish from the impacted Harbour site had significantly higher THg and MeHg ($p<0.001$) than fish from Aasu or Vatia. MeHg:THg was more variable for *P. cyclostomus* from Loa (81-100%) than for Vatia (88-100%) and Aasu (93-100%), but did not appear to be associated with fish body weight at any study site.

INSERT TABLE 12 ABOUT HERE

INSERT FIGURES 2 AND 3 ABOUT HERE

Results from Loa compared with un-impacted sites indicate that sediments are an important factor in Hg uptake in coral reefs; directly for benthic feeding carnivores, and collaterally for turf algae grazers exposed to entrained fine sediments. Four other species of Mullids collected for this study (*P. multifasciatus*, n=19; *P. crassilabris*, n=8; *P. barberinus*, n=4; *Mulloidichthys vanicolensis*, n=8) showed similar patterns of Hg accumulation to *P. cyclostomus*. Similar to accumulation patterns seen in *A. lineatus*, the Mullids displayed limits to accumulation of Hg in the un-impacted habitats (Aasu and Vatia). *Parupeneus cyclostomus* show minimal evidence of a link between muscle Hg and body weight (Figures 2 and 3); when all Mullids collected for this study are included in the weight versus muscle tissue analysis, the pattern remains consistent.

3.4.2.3. Sphyraena qenie

Muscle tissue Hg from the top-trophic carnivore *Sphyraena qenie* (blackfin barracuda) showed a consistent pattern of increased concentration with body weight, unlike reef fish from lower trophic levels (Table 13), and was expectedly higher than for *A. lineatus* and the Mullids. The magnitude of difference, however, was surprising, given the relatively small size of these predator fish, and the pristine environment where they were captured. The larger fish greatly exceeded the US EPA screening level of 0.3 parts-per-million (US EPA, 2000), and could warrant a public health concern for consumption, if popular in the local diet.

INSERT TABLE 13 ABOUT HERE

Regression analysis showed a strong relationship between Hg in muscle tissue and whole body weight for *S. qenie* (r^2=0.98), but was not shown to be statistically significant (p ~0.08), probably due to small sample size. Results for Hg in *S. qenie* are important primarily as an indicator of the Hg flux in the remote Tutuila aquatic environment. Public exposure to Sphyraenids in American Samoa is minimal, as they are taken frequently on a trawled lure by sport-fishing craft, but the numbers of coastal-going sport craft that trawl in American Samoa are low, and barracuda are not a preferred sport fish and are generally released (P. Peshut, pers. obs., 2001-2008).

At the top trophic level of near-shore marine habitats, Sphyraenids are potentially good indicators of Hg prevalence in remote ecosystems, and other large top predators on coral reefs such as jacks and trevallys (Carangidae) and snappers (Lutjanidae) may show similar accumulation patterns. This limited data set indicates that Hg in these relatively small reef predators from un-impacted waters is quite high by world health standards, and that there is no attributable source of Hg except for background environmental levels of Hg in this remote southern hemisphere location.

3.5. Bio-Accumulation Factors

Bio-accumulation factors (BAFs) were calculated for marine water » turf algae » muscle tissue for *A. lineatus*, and marine water » muscle tissue for the Mullids and *S. qenie* (Tables 14 and 15). Factors calculated for MeHg should be interpreted with caution. The data, at the

limit of analytical capabilities (generally ≤5 times DL), is considered sound, but an indeterminable degree of uncertainty is acknowledged.

INSERT TABLES 14 AND 15 ABOUT HERE

BAFs for turf algae were similar for THg and MeHg, and for MeHg, about an order of magnitude less than reported values for phytoplankton (~$10^{5.5}$), the nearest counterpart to turf algae found for comparison in the literature (Mason et al., 1995). As noted earlier, if the DL for MeHg in turf algae for Vatia is used for BAF calculation rather than ½ DL as reported and used by convention, then the BAF is $10^{4.1}$ for MeHg in Vatia turf. No BAF data were found in the literature for comparison with *A. lineatus* and the Mullids. BAFs for *S. qenie* agreed well with the range of 10^6-10^7 reported for top-trophic fish. The increase of approximately one order of magnitude for MeHg BAFs compared with THg BAFs for *A. lneatus*, *P. cyclostomus*, and *S. qenie*, agrees well with studies that show greater trophic transfer efficiency for MeHg than THg (e.g., Mason et al., 1995).

Studies on trace metals and algae show that passive and active transport of dissolved Hg species across cellular membranes is the prevalent mechanism for metals uptake, and has been shown to be particularly efficient for divalent metals, with little or no release back to the water column once metals are incorporated in tissue (Morris and Bale, 1975; Eide et al., 1980). Uptake of Hg from the water column by diffusion across cellular membranes in phytoplankton and diatoms has been established as the principal magnification step in aquatic food webs, ~10^5, (Mason et al., 1995; Mason et al., 1996), and similarly, turf algae-associated Hg is probably limited to cellular uptake from the water column (Morris and Bale, 1975; Eide et al., 1980).

3.6. Human Health and Mercury in Tutuila Coral Reef Fish

Among the general population, health risks from exposure to Hg are attributed principally to MeHg through seafood consumption (e.g., US EPA, 1997; ATSDR, 1999; Mergler et al., 2007). Mercury species are not known to be carcinogenic (ATSDR, 1999; US EPA, 2000) so non-cancer health endpoints, primarily with regard to neurotoxicity (Dales, 1972; Chang, 1977; Castoldi et al., 2001) were used as the basis for risk assessment.

Consumption limits in fish-meals per month, based on MeHg concentration in fish tissue, were prepared in accordance with US EPA (2000) guidelines (Table 16). As public health determinations, consumption limits are typically conservative, a common practice in the public health fields (ATSDR, 1999; US EPA, 2000). Consumption limits based on national or regional standards have recognized limitations, and the applicability of values presented in Table 16 in terms of local population factors or preferences could not be accounted for in this study. For the Samoan population of Tutuila, assumptions for body weight and serving size used to prepare Table 16 may not accurately reflect population parameters or typical consumption practices. Uncertainty cannot be avoided when estimating levels of safe exposure for environmental contaminants (ATSDR, 1999; US EPA, 2000; Tran et al., 2004), and there is no reliable population data for American Samoa to use as a basis to modify table values (Peshut and Brooks, 2005).

INSERT TABLE 16 ABOUT HERE

In the case of the Tutuila population, an under-estimation of average body weight would lead to an over-estimation of risk, and the conservative aspect of consumption limits would be enhanced, that is, overly conservative. Conversely, an under-estimation of serving size per meal for the local population would tend to under-estimate risk, and the conservative aspect of table values would be tend to be relaxed, that is, less conservative. With these considerations acknowledged, consumption limits were prepared in accordance with peer-reviewed standard practice (US EPA, 2000), and used as the basis for consumption rates.

An advantage of this research was that consumption rate tables (Tables 17 and 18) were based on actual MeHg concentration in tissue, whereas the consumption limit values (Table 16) are based on using THg concentrations in tissue, with the assumption that most or all Hg is present as MeHg (US EPA, 2000). The practice of using THg and then applying assumptions for MeHg is normally applied in the interests of expediency and costs. Using experimentally-derived MeHg values would tend to improve the usefulness of the standard table values, by mitigating at least one potential source of uncertainty. It is notable that if speciation analysis had not been used for Hg in muscle tissue for this study, consumption limits based on US EPA (2000) guidance would considerably over-estimate risk for *A. lineatus* from un-impacted reefs, where MeHg in muscle tissue occurred only as ~65% of THg. A marginal over-estimation of risk would have been calculated for the Mullids from the un-impacted sites (MeHg ~90-95% of Hg). Assumptions for 100% MeHg based

consumption limits did not affect the top-trophic *S. qenie*, or the Mullids from Pago Pago Harbour, where MeHg occurred as ~100% of THg.

Implications from these findings are that consumption limit tables based on THg residues in tissues can be overly conservative for lower-trophic fish, especially for remote locations. Speciation analysis for chemical contaminants that have varying toxicity depending on chemical form has been shown to mitigate overly conservative human health determinations (Liu et al., 2006; Yokel et al., 2006; Peshut et al., 2008). The assessment provided here is intended to be interpreted only as an indication of the potential for impacts from Hg on human health in the remote oceanic location of Tutuila, based on limited data, and is not a fully developed risk assessment. This assessment was based on a subjectively selected consumption rate threshold of ≤ 8 fish meals per month, consistent with the previous risk assessment for American Samoa (Peshut and Brooks, 2005).

INSERT TABLES 17 AND 18 ABOUT HERE

At the un-impacted sites of Aasu and Vatia, there was no basis to advise the public to limit consumption of surgeonfish or goatfish. Surprisingly, there was also no basis for an advisory for *A. lineatus* in Pago Pago Harbour (Table 17). Based on Hg residues in muscle tissue for *S. qenie*, consumption for the two largest fish was limited to 1 meal per month. Extrapolating from results for *S. qenie* suggest that fish >15 kg would have a recommendation of "no consumption". Results for the top-predator *S. qenie* clearly indicated that public health concerns for consumption of top-trophic reef-fish may be warranted, but this evidence must remain indicative at this time. The roving carnivorous goatfish from Pago Pago Harbour were assigned limited consumption status, consistent with previous findings for the Harbour (Peshut and Brooks, 2005).

Overall, results for reef fish from coastal reefs of Aasu and Vatia, and the Harbour reef at Loa, are consistent with conclusions from Peshut and Brooks (2005), that open coastal reefs of Tutuila do not warrant fish advisories, and that the fish advisory currently in place for Pago Pago Harbour is supported. These are encouraging results for the artisanal reef fish fisheries for Tutuila, because consumption of economically and culturally important species does not appear to be of concern. In contrast, the high levels of Hg accumulated in the relatively small barracuda from un-impacted reefs, supports concerns for the persistence and

bio-accumulation of Hg in un-impacted environments of remote locations, as a result of global proliferation of Hg pollution.

4. CONCLUSIONS

Total Hg and methyl Hg concentrations in marine waters around Tutuila are low and in general agreement with historical and other recent data. Reef sediment Hg concentrations are also low, with slightly higher concentrations in the Loa (harbour impacted site). The first reported data for Hg in turf algae (THg 10-14, MeHg <0.2-1 ng g^{-1} w/w) show evidence of significant biological accumulation (BAFs of ~$10^{4.5}$). Elevated Hg in muscle tissue of the top-trophic reef predator *Sphyraena qenie* is an important indicator of the ubiquity of Hg in remote global regions, and shows that Hg bio-accumulates to levels of concern for human health even in remote un-impacted environments. Health risks from the consumption of surgeonfish and goatfish from the artisanal reef fish fishery in American Samoa appear negligible for coastal reefs according to risk assessment protocols.

Tutuila Island presents an advantageous opportunity to establish a long-term atmospheric Hg monitoring station in a remote oceanic location of the southern hemisphere. Monitoring facilities for atmospheric monitoring are currently in place at Cape Matatula on Tutuila's easternmost point (Global Monitoring Division (GMD) of the US National Oceanic and Atmospheric Administration). Complementing such atmospheric monitoring, a long-term fish monitoring programme with a 3-5 year cycle, would be an efficient means of providing important data on how the un-impacted aquatic biological system on Tutuila responds to atmospheric Hg over time.

ACKNOWLEDGEMENTS

The authors acknowledge the assistance of Carl Goldstein of the United States Environmental Protection Agency (San Francisco, USA) who provided administrative support, funding, and sustained enthusiasm for this work (USEPA played no role in design, execution or reporting on this project). In the field, Howard Dunham and crew of Moon Divers (Tutuila Island) provided outstanding support throughout two years of field initiatives, that involved driving small boats, diving, and technical assistance for collections, by day and by night, on dozens of forays of up to 90 km, in all sea conditions and weathers. Edna Buchan, Water Program

Manager, and her staff of the American Samoa Environmental Protection Agency (Tutuila Island) provided technical assistance and logistical support, and endless good cheer, frequently outside regular duty hours.

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Figure 1 Location of Study Sites - Tutuila Island

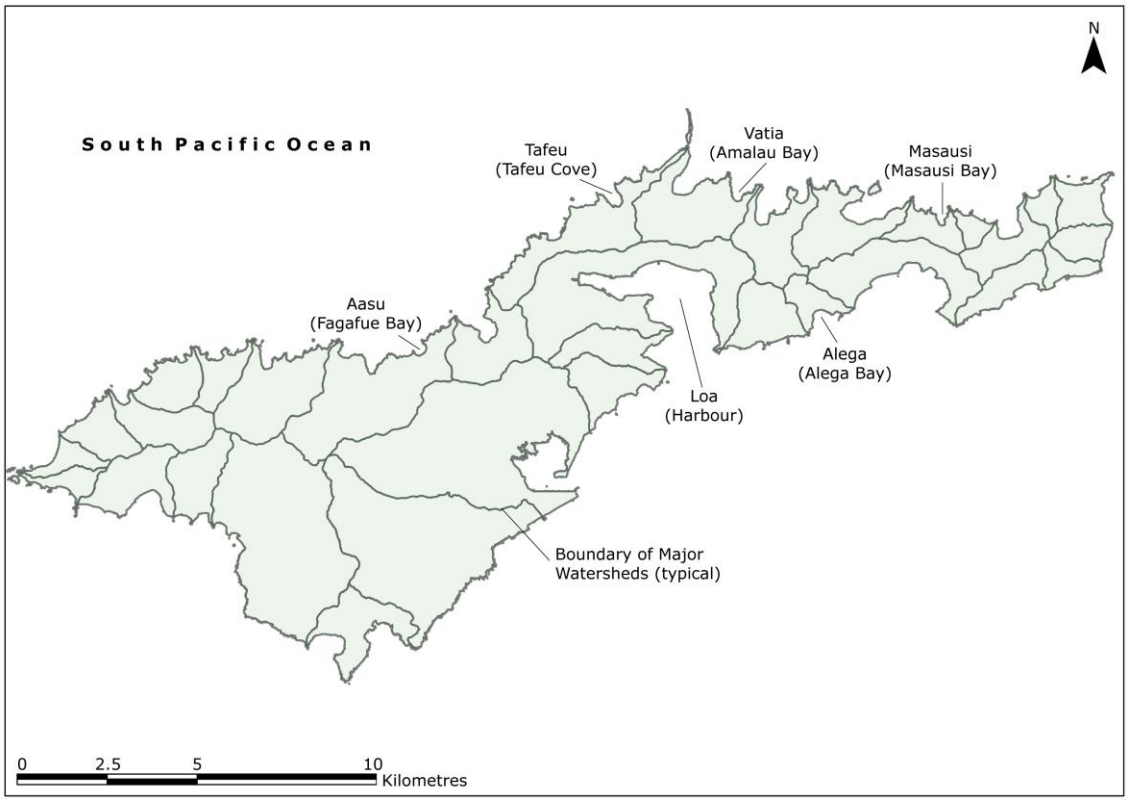

Figure 2. Total Hg in muscle tissue *vs* weight - *Parupeneus cyclostomus*

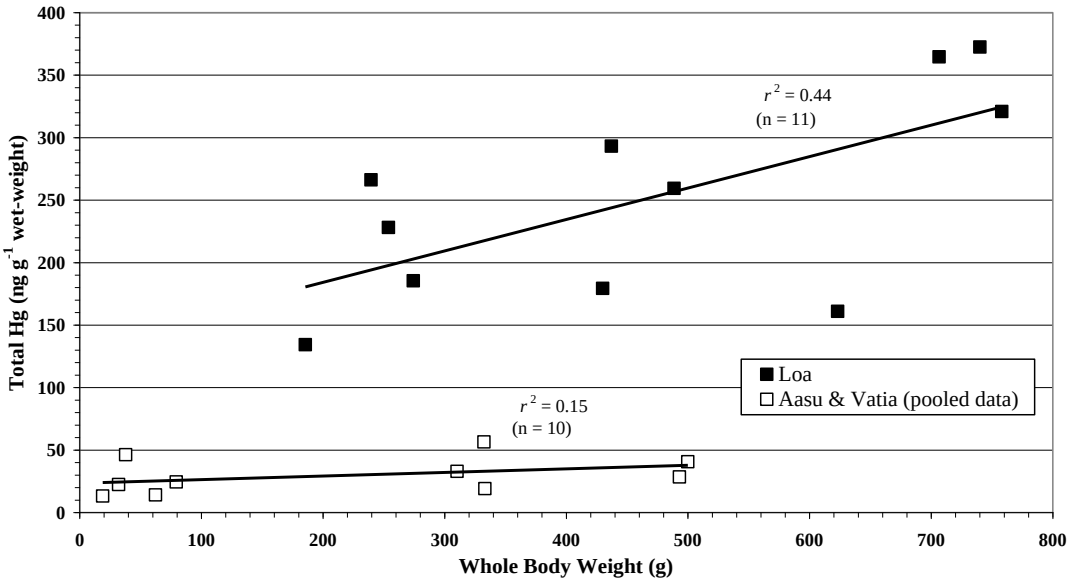

Figure 3. Methyl Hg in muscle tissue *vs* weight - *Parupeneus cyclostomus*

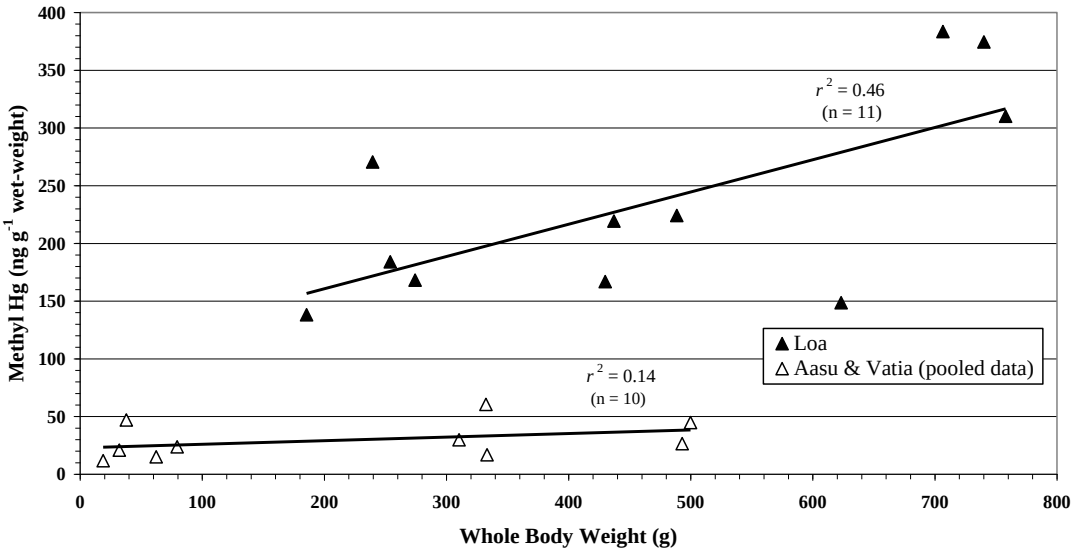

Table 1. Features of the study sites

Site	Catchment Area (km^2)	Catchment Maximum Elevation (m)	Population in Catchment
Aasu (Fagafue Bay)	8.441	~425	~1150
Tafeu	5.116	~490	<10
Vatia (AmalauBay)	4.884	~325	~300
Masausi	1.565	~300	<200
Alega	1.313	~350	<50
Loa (Pago Pago)	10.437	653	~2500[1]

1For Loa. Population in the vicinity of the study site ~2500; 0verall population of Pago Pago harbour catchment ~10,500

Table 2. Methods and detection limits for Hg in sediments and coral reef biota

Sample	Analyte	Instrument	Method[1]	Achieved DL
Sediment	Total Hg	CVAF	EPA 1631E	0.302 - 0.320 ng g^{-1} dry-weight
	Methyl Hg	CVAF	EPA 1630	0.0124 - 0.0164 ng g^{-1} dry-weight
	TOC		ASTMD4129-82	0.05%
Turf Algae	Total Hg	CVAF	EPA 1631E	0.182 ng g^{-1} wet-weight
	Methyl Hg	CVAF	EPA 1630	0.435 ng g^{-1} wet-weight
Fish	Total Hg	CVAF	EPA 1631E	0.105 - 0.108 ng g^{-1} wet-weight
	Methyl Hg	CVAF	EPA 1630	0.251 - 0.257 ng g^{-1} wet-weight

[1](ASTM, 1982; US EPA, 1994a, 1994b, 2001a, 2001b, 2002; Bligh and Dyer, 1959)

Table 3. Total Hg in Tutuila marine water (ng L^{-1})

	Study site		
Sample ID	Aasu	Vatia	Loa
MW1-1	0.378	0.427	0.524
MW1-2	0.374	0.485	0.634
MW1-3	0.389	0.419	0.483
MW1-4	0.484	0.359	0.512
MW2-1	0.278	0.290	0.322
MW2-2	0.282	0.245	0.349
MW2-3	0.286	0.324	0.387
MW2-4	0.254	0.288	0.404
MW3-1	0.295	0.259	0.348
MW3-2	0.271	0.248	0.383
MW3-3	0.391	0.264	0.352
MW3-4	0.232	0.206	0.320
Mean:	0.326 ±0.021	0.318 ±0.025	0.418 ±0.028

Table 4. Methyl Hg in Tutuila marine water (ng L^{-1})

	Study site		
Sample ID	Aasu[1]	Vatia[1]	Loa
MW1-1	**0.0094**	0.0256	0.0354
MW1-2	**0.0094**	**0.0094**	0.0365
MW1-3	0.0239	**0.0094**	0.0219
MW1-4	0.0265	**0.0094**	0.0486
MW2-1	0.0359	0.0261	0.0906
MW2-2	0.0347	0.0412	0.0454
MW2-3	0.0414	0.0417	0.0767
MW2-4	0.0346	0.0367	0.0996
MW3-1	**0.0094**	**0.0094**	0.0484
MW3-2	0.0262	0.0561	0.0338
MW3-3	0.0244	0.0651	0.0387
MW3-4	0.0232	0.0630	0.0439
Mean:	0.0249 ±0.0032	0.0328 ±0.0061	0.0516 ±0.0070

[1]Bold indicates analytical result < DL; value is ½ DL for calculation purposes

Table 5. TSS in Tutuila marine water (mg L^{-1})

	Study site		
Sample ID	Aasu	Vatia	Loa
MW1-1a	2.47	0.899	0.700
MW1-3a	2.45	0.816	0.812
MW2-1a	1.57	1.48	0.815
MW2-3a	1.21	0.731	1.74
MW3-1a	0.975	0.885	0.983
MW3-3a	0.802	1.18	1.10
Mean:	1.58 ±0.30	1.00 ±0.11	1.03 ±0.15
	Overall mean:	1.20 ±0.13	

Table 6. Total Hg in Tutuila marine sediments (ng g^{-1} dry-weight)

	Study site					
Sample ID	Aasu	Tafeu	Vatia	Loa	Alega	Masausi
1	13.5	3.74	2.12	21.9	1.84	6.01
2	15.6	3.27	1.58	28.4	3.43	2.59
3	4.93	3.98	2.80	29.9	2.64	2.40
4	15.8	4.31	4.09	40.6	2.03	3.63
5	7.10	3.98	1.86	19.4	4.75	4.25
6	12.9	5.48	1.68	15.1	3.21	3.43
7	7.56	4.54	3.81	29.9	3.12	3.34
8	6.79	5.82	4.34	31.9	3.56	3.03
9	12.2	3.74	3.39	16.0	2.19	2.45
10	3.41	4.95	3.63	21.9	2.00	2.00
11	4.64	5.56	3.21	27.1	5.39	2.58
12	8.46	4.24	3.48	20.7	4.21	2.60
13	10.0	4.00	3.73	25.4	3.37	2.80
14	7.22	3.52	1.82	29.5	2.03	2.76
15	9.29	3.78	1.91	37.7	2.17	4.62
16	7.99	3.37	2.25	39.0	1.54	2.64
17	5.33	5.03	4.70	---	3.84	3.40
18	7.77	4.98	4.20	---	3.99	3.97
19	5.99	3.95	5.05	---	3.34	3.19
20	8.83	3.49	2.54	---	3.25	3.36
21	5.39	3.36	3.26	---	5.45	2.52
22	11.4	5.81	3.54	---	5.17	7.45
23	6.33	3.81	3.16	---	2.07	4.45
24	7.08	5.48	2.58	---	4.76	2.61
25	9.44	5.80	2.57	---	4.80	3.21
26	11.2	5.22	1.32	---	3.26	2.96
27	6.37	4.20	2.04	---	4.29	3.51
28	3.93	4.55	2.00	---	5.30	3.19
29	6.58	5.19	1.90	---	2.37	4.60
30	9.15	5.18	2.79	---	5.77	4.01
31	8.00	5.30	3.66	---	3.60	3.86
32	11.6	4.68	3.46	---	2.39	5.12
Mean:	8.49 ±0.56	4.51 ±0.14	2.95 ±0.17	27.2 ±1.95	3.47 ±0.22	3.52 ±0.20
Range:	(3.41-15.8)	(3.27-5.82)	(1.32-5.05)	(15.1-40.6)	(1.54-5.77)	(2.00-7.45)

Table 7. Methyl Hg in Tutuila marine sediments (ng g^{-1} dry-weight)

	Study site					
Sample ID	Aasu	Tafeu	Vatia	Loa	Alega	Masausi
1	0.1822	0.0855	0.0283	0.1790	0.0635	0.0918
2	0.2074	0.0546	0.0265	0.1210	0.1587	0.0294
3	0.0693	0.1327	0.0208	0.2013	0.0795	0.0207
4	0.3725	0.1297	0.0917	0.1702	0.0734	0.1524
5	0.0883	0.0802	0.0491	0.2342	0.1739	0.1184
6	0.2755	0.0846	0.0188	0.1426	0.0646	0.0515
7	0.0815	0.0503	0.0282	0.2685	0.0554	0.0767
8	0.1204	0.1597	0.0491	0.2347	0.1081	0.0331
9	0.3207	0.0658	0.0240	0.1104	0.0536	0.0295
10	0.0321	0.1354	0.1313	0.2326	0.0219	0.0327
11	0.0548	0.0831	0.0462	0.2116	0.1141	0.0467
12	0.1593	0.0840	0.0916	0.1701	0.1264	0.0521
13	0.1664	0.1070	0.0721	0.1371	0.0808	0.1021
14	0.1355	0.0356	0.0191	0.1359	0.0690	0.0270
15	0.1721	0.0723	0.0123	0.1905	0.0407	0.1178
16	0.1446	0.0360	0.0207	0.1712	0.0555	0.0285
17	0.0856	0.1330	0.0635	---	0.0542	0.0501
18	0.1213	0.1240	0.0568	---	0.1267	0.1061
19	0.0724	0.0397	0.0285	---	0.0821	0.0717
20	0.1962	0.0822	0.0586	---	0.0974	0.0879
21	0.0603	0.0895	0.0710	---	0.1616	0.0885
22	0.1242	0.2166	0.0417	---	0.1341	0.1507
23	0.0568	0.0911	0.0475	---	0.0417	0.1289
24	0.0518	0.1565	0.0684	---	0.1519	0.0331
25	0.1298	0.1399	0.0572	---	0.1649	0.0456
26	0.3056	0.0930	0.0534	---	0.1236	0.0421
27	0.0926	0.0429	0.0315	---	0.1427	0.0494
28	0.0315	0.0860	0.0534	---	0.1924	0.1148
29	0.1240	0.1510	0.0586	---	0.0346	0.0853
30	0.2632	0.1685	0.0552	---	0.0782	0.0684
31	0.1738	0.2149	0.0826	---	0.1045	0.0958
32	0.3412	0.1422	0.0866	---	0.0384	0.1211
Mean:	0.1504	0.1052	0.0514	0.1819	0.0959	0.0734
	±0.0165	±0.0085	±0.0047	±0.0115	±0.0083	±0.0069
Range:	(0.0315-0.3725)	(0.0356-0.2166)	(0.0123-0.1313)	(0.1104-0.2685)	(0.0219-0.1924)	(0.0207-0.1524)

Table 8. Total organic carbon in Tutuila marine sediment (% dry-weight)

	Study site					
Sample ID	Aasu	Tafeu	Vatia	Loa	Masausi	Alega
1	0.96	0.29	0.20	0.41	0.32	0.31
2	2.26	0.32	0.25	0.74	0.32	0.51
3	0.55	0.62	0.30	0.43	0.25	0.26
4	3.46	0.47	0.29	1.21	0.49	0.40
5	0.79	0.51	0.34	0.35	0.53	0.40
6	0.87	0.53	0.39	0.28	0.53	0.42
7	1.04	0.32	0.79	0.50	0.57	0.34
8	0.71	0.24	0.30	0.84	0.40	0.43
9	1.65	0.30	0.25	0.33	0.29	0.34
10	0.56	0.30	0.28	0.27	0.34	0.21
11	1.07	0.39	0.26	0.81	0.38	0.42
12	1.27	0.51	0.29	0.44	0.38	0.31
13	0.86	0.30	0.22	0.43	0.25	0.24
14	2.18	0.31	0.25	0.48	0.33	0.34
15	0.97	0.31	0.18	0.96	0.43	0.38
16	0.85	0.37	0.16	0.96	0.34	0.33
17	0.43	0.44	0.25	---	0.29	0.35
18	0.67	0.35	0.27	---	0.43	0.45
19	0.83	0.30	0.35	---	0.57	0.38
20	0.84	0.24	0.22	---	0.44	0.33
21	0.57	0.56	0.26	---	0.42	0.41
22	0.96	0.63	0.23	---	0.49	0.77
23	0.52	0.28	0.31	---	0.51	0.46
24	0.77	0.60	0.29	---	0.28	0.48
25	1.16	0.50	0.33	---	0.40	0.49
26	0.78	0.41	0.56	---	0.35	0.45
27	0.64	0.27	0.31	---	0.38	0.66
28	1.17	0.31	0.28	---	0.36	0.28
29	2.06	0.46	0.24	---	0.43	0.38
30	1.14	0.37	0.33	---	0.31	0.42
31	0.93	0.38	0.42	---	0.21	0.50
32	1.16	0.56	0.32	---	0.53	0.34
Mean:	1.08	0.40	0.30	0.59	0.39	0.40

Table 9. Total Hg in Tutuila turf algae (ng g^{-1} wet-weight)

	Study Site		
Sample ID	Aasu	Vatia	Loa
TA-1	9.05	8.09	20.3
TA-2	12.1	10.2	11.7
TA-3	13.4	11.9	19.4
TA-4	12.7	15.5	11.0
TA-5	12.0	6.72	16.1
TA-6	12.5	9.06	13.3
TA-7	16.4	7.76	8.50
TA-8	20.1	12.0	14.3
TA-9	19.5	8.27	9.37
TA-10	12.1	14.8	6.47
Mean:	14.0 ±1.12	10.4 ±0.96	13.0 ±1.44

Table 10. Methyl Hg in Tutuila turf algae (ng g^{-1} wet-weight)

Sample ID	Study Site		
	Aasu	Vatia[1]	Loa[1]
TA-1	0.801	**0.217**	0.656
TA-2	0.972	**0.217**	0.461
TA-3	0.893	**0.217**	1.20
TA-4	0.870	**0.217**	**0.217**
TA-5	0.971	**0.217**	**0.217**
TA-6	1.34	**0.217**	0.464
TA-7	1.14	**0.217**	**0.217**
TA-8	1.48	**0.217**	0.590
TA-9	0.792	**0.217**	0.440
TA-10	0.923	**0.217**	0.471
Mean:	1.02 ±0.07	0.22	0.49 ±0.09

[1]Bold indicates analytical result < DL; value is ½ DL for calculation purposes

Table 11. Hg in *Acanthurus lineatus* muscle tissue (ng g^{-1} wet-weight)

Sample ID			Weight (g)	Total Hg	Methyl Hg	Ratio MeHg:THg
Loa	ALOGO	1-1-1	203.1	3.402	3.662	108%
Loa	ALOGO	1-1-2	165.4	4.218	3.468	82%
Loa	ALOGO	1-1-3	119.8	4.320	3.903	90%
Loa	ALOGO	1-1-4	130.8	6.025	4.992	83%
Loa	ALOGO	1-1-5	146.8	4.246	4.259	100%
Loa	ALOGO	1-2-1	135.1	5.977	4.985	83%
Loa	ALOGO	1-2-2	163.0	4.444	4.202	95%
Loa	ALOGO	1-2-3	130.3	3.683	3.965	108%
Loa	ALOGO	1-2-4	150.3	4.008	3.632	91%
Loa	ALOGO	1-2-5	138.7	13.49	14.50	107%
Loa	ALOGO	2-1-1	134.1	6.285	5.148	82%
Loa	ALOGO	2-1-2	165.1	5.871	5.058	86%
Loa	ALOGO	2-1-3	186.5	7.596	6.738	89%
Loa	ALOGO	2-1-4	175.2	6.170	6.406	104%
Loa	ALOGO	2-1-5	181.2	9.772	9.936	102%
Loa	ALOGO	2-2-1	183.5	4.528	4.913	109%
Loa	ALOGO	2-2-2	186.4	7.352	6.695	91%
Loa	ALOGO	2-2-3	202.7	5.946	5.629	95%
Loa	ALOGO	2-2-4	187.7	5.086	4.539	89%
Loa	ALOGO	2-2-5	174.5	5.911	5.295	90%
		Mean:	163.0 ±5.7	5.92 ±0.53	5.60 ±0.57	94.1%
		Range:	(119.8-203.1)	(3.40-13.5)	(3.47-14.5)	---
Vatia	ALOGO	1-1-1	329.5	0.849	0.473	56%
Vatia	ALOGO	1-1-2	223.5	1.028	0.502	49%
Vatia	ALOGO	1-1-3	273.2	0.845	0.309	37%
Vatia	ALOGO	1-1-4	244.9	0.752	0.436	58%
Vatia	ALOGO	1-1-5	228.6	0.841	0.466	55%
Vatia	ALOGO	1-2-1	223.8	0.830	0.552	66%
Vatia	ALOGO	1-2-2	239.3	1.300	0.635	49%
Vatia	ALOGO	1-2-3	233.7	1.163	0.635	55%
Vatia	ALOGO	1-2-4	220.1	1.410	0.440	31%
Vatia	ALOGO	1-2-5	278.1	1.479	1.069	72%
Vatia	ALOGO	2-1-1	230.2	0.735	0.737	100%
Vatia	ALOGO	2-1-2	216.6	0.840	0.766	91%
Vatia	ALOGO	2-1-3	226.7	1.183	0.928	78%
Vatia	ALOGO	2-1-4	216.0	1.189	0.951	80%
Vatia	ALOGO	2-1-5	249.7	0.828	0.535	65%
Vatia	ALOGO	2-2-1	249.8	1.217	0.901	74%
Vatia	ALOGO	2-2-2	252.5	1.012	0.775	77%
Vatia	ALOGO	2-2-3	295.8	1.273	0.690	54%
Vatia	ALOGO	2-2-4	277.9	1.229	1.006	82%
Vatia	ALOGO	2-2-5	246.3	1.075	0.830	77%
		Mean:	247.8 ±6.6	1.05 ±0.05	0.682 ±0.049	65.3%
		Range:	(216.0-329.5)	(0.735-1.48)	(0.309-1.07)	---
Aasu	ALOGO	1-1-1	248.4	0.925	0.967	104%
Aasu	ALOGO	1-1-2	231.1	0.605	0.415	69%
Aasu	ALOGO	1-1-3	207.7	0.741	0.450	61%
Aasu	ALOGO	1-1-4	259.9	1.112	0.584	53%
Aasu	ALOGO	1-1-5	227.8	0.794	0.427	54%

Aasu	ALOGO	1-2-1	286.6	1.009	0.416	41%
Aasu	ALOGO	1-2-2	168.3	0.835	0.384	46%
Aasu	ALOGO	1-2-3	223.3	0.833	0.447	54%
Aasu	ALOGO	1-2-4	234.8	0.932	0.376	40%
Aasu	ALOGO	1-2-5	165.5	1.059	0.351	33%
Aasu	ALOGO	2-1-1	224.1	0.909	0.683	75%
Aasu	ALOGO	2-1-2	190.8	1.271	1.073	84%
Aasu	ALOGO	2-1-3	214.1	1.607	0.947	59%
Aasu	ALOGO	2-1-4	236.0	1.272	0.772	61%
Aasu	ALOGO	2-1-5	243.9	1.247	0.853	68%
Aasu	ALOGO	2-2-1	227.4	1.220	0.857	70%
Aasu	ALOGO	2-2-2	216.1	0.986	0.672	68%
Aasu	ALOGO	2-2-3	207.3	1.056	0.746	71%
Aasu	ALOGO	2-2-4	226.4	1.282	0.866	68%
Aasu	ALOGO	2-2-5	229.5	1.087	0.622	57%
		Mean:	223.5 ±6.3	1.04 ±0.05	0.645 ±0.051	61.8%
		Range:	(165.5-286.6)	(0.605-1.61)	(0.351-1.07)	---

Table 12. Hg in *Parupeneus cyclostomus* muscle tissue (ng g^{-1} wet-weight)

Sample ID			Weight (g)	Total Hg	Methyl Hg	Ratio Methyl:THg
Loa	IASINA	1-1-2	274.3	185.4	168.1	91%
Loa	IASINA	1-1-4	488.6	259.4	224.1	86%
Loa	IASINA	1-1-5	758.0	320.8	310.1	97%
Loa	IASINA	1-2-5	437.2	293.2	219.3	75%
Loa	IASINA	2-1-1	430.0	179.4	166.8	93%
Loa	IASINA	2-1-2	253.9	228.1	184.0	81%
Loa	IASINA	2-1-4	740.1	372.5	374.5	101%
Loa	IASINA	2-1-5	239.6	266.1	270.5	102%
Loa	IASINA	2-2-2	706.6	364.6	383.4	105%
Loa	IASINA	2-2-3	623.2	161.0	148.6	92%
Loa	IASINA	2-2-4	185.2	134.3	138.2	103%
		Mean:	467.0 ±64.5	251.3 ±24.6	235.2 ±26.5	93.2%
		Range:	(185.2-758.0)	(134.3-372.5)	(138.2-383.4)	---
Vatia	IASINA	1-1-2	18.9	13.14	11.66	89%
Vatia	IASINA	2-1-5	332.4	56.52	60.47	107%
Vatia	IASINA	2-2-1	493.1	28.44	26.42	93%
Vatia	IASINA	2-2-4	37.8	46.27	47.04	102%
Vatia	IASINA	2-2-5	333.2	19.15	16.79	88%
		Mean:	243.1 ±92.5	32.7 ±8.17	32.5 ±9.25	95.6%
		Range:	(18.9-493.1)	(13.1-56.5)	(11.7-60.5)	---
Aasu	IASINA	1-1-1	499.9	40.62	44.73	110%
Aasu	IASINA	1-1-3	62.3	14.14	15.13	107%
Aasu	IASINA	1-2-5	32.0	22.35	20.81	93%
Aasu	IASINA	2-2-2	79.5	24.42	23.79	97%
Aasu	IASINA	2-2-5	310.3	32.92	29.79	90%
		Mean:	196.8 ±90.5	26.9 ±4.55	26.8 ±5.06	99.6%
		Range:	(32.0-499.9)	(14.1-40.6)	(15.1-44.7)	---

Table 13. Hg in muscle tissue for *Sphyraena qenie* (ng g^{-1} wet-weight)

Sample ID	Weight (g)	Total Hg	Methyl Hg	Ratio MeHg:THg
Bar 01	9050	741	752	101%
Bar 02	7470	650	643	99%
Bar 03	3570	105	105	100%

Table 14. BAFs for Total Hg (referenced to marine water)

Location	Turf Algae	*A. lineatus*	*P. cyclostomus*	*S. qenie*[1]
Aasu	$10^{4.6}$	$10^{3.5}$	$10^{4.9}$	----
Vatia	$10^{4.5}$	$10^{3.5}$	$10^{5.0}$	----
Loa	$10^{4.5}$	$10^{4.6}$	$10^{5.8}$	----
Coastal	----	----	----	$10^{6.4}$

[1]Bar01; calculation based on THg in Aasu and Vatia marine water (mean)

Table 15. BAFs for Methyl Hg (referenced to marine water)

Location	Turf Algae	*A. lineatus*	*P. cyclostomus*	*S. qenie*[1]
Aasu	$10^{4.6}$	$10^{4.4}$	$10^{6.0}$	----
Vatia	$10^{3.8}$	$10^{4.3}$	$10^{6.0}$	----
Loa	$10^{4.0}$	$10^{5.0}$	$10^{6.7}$	----
Coastal	----	----	----	$10^{7.4}$

[1]Bar01; calculation based on MeHg in Aasu and Vatia marine water (mean)

Table 16. Consumption limits for MeHg in fish tissue

Fish meals per month	Muscle tissue MeHg (ng g^{-1} wet-weight)
un-restricted (>16)	≤ 29
16	>29 - 59
12	>59 - 78
8	>78 - 120
7	134
6	156
5	190
4	230
3	310
2	470
1	940
½	1900
none (< ½)	>1900

Adapted from Peshut and Brooks (2005); prepared in accordance with US EPA (2000) method

Table 17. Consumption rates for un-impacted artisanal fishery

Species	Location (specimens)	Muscle tissue MeHg (ng g^{-1} wet-weight)	Consumption (fish meals per month)
A. lineatus	Aasu (n=20)	0.645	un-restricted
	Vatia (n=20)	0.682	un-restricted
M. vanicolensis	Aasu (n=3)	20.5	un-restricted
	Vatia (n=3)	11.3	un-restricted
P. barberinus	Aasu (n=1)	53.6	16
P. crassilabris	Aasu (n=4)	33.4	16
	Vatia (n=1)	60.4	12
P. cyclostomus	Aasu (n=5)	26.8	un-restricted
	Vatia (n=5)	32.5	16
P. multifasciatus	Aasu (n=7)	22.2	un-restricted
	Vatia (n=11)	35.4	16
S. qenie	Coastal (n=1)	752	1
	Coastal (n=1)	643	1
	Coastal (n=1)	105	8

Table 18. Consumption rates for impacted artisanal fishery

Species	Location	Muscle tissue MeHg (ng g^{-1} wet-weight)	Consumption (fish meals per month)
A. lineatus	Loa (n=20)	5.60	un-restricted
M. vanicolensis	Loa (n=2)	169	5
P. barberinus	Loa (n=3)	72.6	12
P. crassilabris	Loa (n=3)	239	4
P. cyclostomus	Loa (n=11)	235	4
P. multifasciatus	Loa (n=1)	233	4