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Population-based dietary exposure to mercury through fish consumption in the Southern Peruvian Amazon.

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Abstract

Background: Mercury exposure related to artisanal and small gold mining (ASGM) has raised environmental and public health concerns globally. Exposure to mercury, a potent neurotoxin that bioaccumulates in fish, is especially of concern to women of childbearing age (WCBA) and children in high-fish consuming populations. In Madre de Dios (MDD), Peru, an Amazon region with naturally occurring mercury and high ASGM activity, there is significant exposure concern among the mainly riverine, fish-consuming communities. The objective of this study was to conduct the first assessment of mercury exposure in a population-based sample of MDD, identify factors associated with elevated levels and compare the relationship between fish consumption and hair total mercury (H-THg) among persons living in ASGM affected and non-ASGM affected watersheds.

Methods: Hair samples and household demographic surveys, including a module on fish consumption, were collected from 723 participants across 46 communities within 10 km of the Interoceanic Highway in MDD, who were previously enrolled in the first population-based study in MDD spanning areas affected and unaffected by ASGM. H-THg concentration (natural log transformed) was evaluated for association with independent demographic variables through multilevel multivariate regression models accounting for clustering among households and communities. Samples from canned fish available at local stores were also tested for total mercury.

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Results: Fish consumption (diversity and total consumed) varied spatially along the highway. 269 participants (37.2%) had elevated H-THg (>2.2 μg/g; median 1.60 μg/g; mean 2.24 μg/g), including 42.7% of WCBA and 20.0% of children under 5. Overall, H-THg was higher among people living in ASGM-affected areas. H-THg concentrations were strongly associated with fish consumption; however, in the multivariate models, household consumption of high trophic level fish was associated with elevated H-THg only in communities located in the ASGM-impacted watersheds. Similarly, the relationship between living in a household engaged in economic activities of fishing or Brazil nut harvesting was associated with higher H-THg, but only among households in the ASGM-affected area. In the non-ASGM affected areas, we observed a positive relationship between household daily fruit consumption and H-THg that was not observed in ASGM-affected areas.

Conclusion: Diet, residential location, and occupation are strong predictors of mercury exposure in Madre de Dios, Peru. Canned fish may represent a previously overlooked source of dietary Hg exposure in the region. In accordance with the Minamata Convention, the significant environmental health concern of mercury exposure in ASGM areas demands policy and programmatic attention.

Keywords

mercury; hair; ASGM; fish; Amazonia

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1. Introduction

The Amazon Basin is a biodiversity hot spot spanning eight South American countries. Deforestation driven by agricultural expansion, urbanization, road construction, and natural resource extraction – including oil, gas and mining – threaten the health of the environment as well as the indigenous and non-indigenous populations living there (Asner and Tupayachi, 2017; Gregory P, 2016; Rudel et al., 2009). While such land use / land cover changes (LULCC) are often associated with emerging infectious disease risks, they are also associated with increased exposures to environmental contaminants.

For example, exposure to the toxic trace element mercury is prevalent across the Amazon Basin (de Castro and de Oliveira Lima, 2014; Dorea and Marques, 2016). Previous studies have established that the main sources of exposure to mercury in the Amazon are from: a) naturally occurring mercury in soils liberated via anthropogenic activities such as deforestation (Dorea and Marques, 2016; Fadini and Jardim, 2001; Lacerda et al., 2004; Miserendino et al., 2018); and b) the use of elemental mercury in artisanal and small scale gold mining (ASGM) and processing (Malm, 1998; Marshall et al., 2018). Following its release into the environment, it is methylated by anaerobic microorganisms to monomethyl mercury (MeHg) (Dorea and Marques, 2016). The main human exposure routes to mercury are occupational-via direct inhalation of inorganic mercury vapor from the burning of gold-mercury amalgams, and dietary-via the consumption of riverine fish that bioaccumulate MeHg. While those occupationally exposed do face neurological, immunological and other health risks from the inhalation of the Hg vapor (Barbieri and Gardon, 2009; Gibb and O'Leary, 2014), in the Amazon, chronic population exposure is largely via diet as fish is a food staple of the local culture, and fishing remains a dominant economic activity, even downstream of ASGM and in other heavily deforested areas. Dietary exposure to MeHg is considered a pressing public health threat around the world owing both to higher trophic level fish bioaccumulating and biomagnifying this organomercury compound (Grandjean et al., 2010; World Health Organization (WHO), 1990) and the ability of MeHg to cross both the blood-brain barrier and the placenta, eliciting deleterious effects on fetal, infant and child development (Basu et al., 2018; Ha et al., 2017).

Exposure to mercury can be measured in human tissue. In the Amazon, and around the world, hair-total mercury (H-THg) is a commonly used biomarker of recent dietary exposure when measured in scalp hair, (reflecting MeHg exposure from blood Hg concentrations at the time of hair growth) (Berglund et al., 2005; Sheehan et al., 2014). MeHg preferentially deposits in hair, and thus the measure of total mercury in hair is considered an appropriate biomarker to measure dietary exposure to MeHg. Collecting hair for analysis is non-invasive, does not require a cold chain, and the measures are reliable. The first centimeter of scalp hair reflects integrated exposures ~20 days to 1 month prior to the date of sampling.

In 2006, the Joint WHO/FAO Expert Committee on Food Additives (JECFA) adopted the Provisional Tolerable Weekly Intake (PTWI) of 1.6 μg/kg bodyweight per day for methylmercury to protect the most sensitive population of pregnant women and infants (Joint FAO/WHO Expert Committee on Food Additives, 2006). Accounting for adjustments for inter-individual variability, this PTWI is associated with 2.2 μg total mercury / g hair. It should be noted that Peru does not have any specific standards related to chronic dietary exposure to mercury, as their guidelines related to mercury exposure are directed more towards acute occupational exposures, and thus rely on measuring mercury in urine (Ministerio de Salud, 2013). In addition to this reference value, there are other reference values often referred to including the US National Research Council’s reference value for non-fish eating populations of 1.2 μg/g corresponding to a reference dose (RfD) for methylmercury of 0.1 μg/kg bodyweight/day (NRC (National Research Council), 2000). WHO previously established 10 μg/g as the hair mercury level of concern on the developing fetus of pregnant mothers (World Health Organization (WHO), 1990), which has been used as a reference level in many studies conducted in the Amazon (e.g., Barbieri and Gardon, 2009).

Madre de Dios is located in the southern Peruvian Amazon, bordering the Brazilian state of Acre and the Bolivian state of Pando (Figure 1). This area has been undergoing rapid land use and land cover change in recent decades, particularly related to the expansion of ASGM, the development of the Interoceanic Highway, and the intensification of agriculture, all of which have resulted in rapid urban growth in the capital city of Puerto Maldonado (Mastel et al., 2018; Southworth et al., 2011; Swenson et al., 2011). In Madre de Dios, studies have revealed mercury contamination in both environmental (fish, sediment, water, wildlife) (Diringer et al., 2015; Martinez et al., 2018; Moreno-Brush et al., 2018; Moreno-Brush et al., 2016) and human populations (Ashe, 2012; Berky et al., 2018; Weinhouse et al., 2017; Wyatt et al., 2017; Yard et al., 2012). In May 2016, then Peruvian President Ollanta Humala issued a Supreme Decree (No. 034-2016-PCM) declaring a State of Emergency in 11 districts of Madre de Dios in response to the elevated mercury exposures observed in the region (Humala Tasso, 2016; Tarabochia, 2016). Although exposure studies and the declaration brought attention to environmental and human health issues related to mercury exposure and ASGM, human exposure studies in Madre de Dios have been limited largely to occupationally exposed populations, with no studies to date designed to provide population level mercury exposure assessment.

The objective of this analysis was to assess mercury exposure at the population level in Madre de Dios using data obtained from a 2-stage cluster sample of communities and households located within 10 Kmof the Interoceanic Highway and identify factors associated with exposure. Specifically, we sought to test whether people living in *ASGM-affected* watersheds (that is, in localities within and downstream of ASGM operations) have higher mercury exposure compared to those living in watersheds without known ASGM activities (here called *non ASGM-affected communities* or *areas)*. Further, building off our previous research that demonstrated: (1) higher mercury concentrations in carnivorous fish located in ASGM-vs. non-ASGM-affected areas (Diringer et al., 2015); and (2) more frequent fish consumption was associated with higher mercury exposure (Wyatt et al., 2017); we hypothesized that more frequent consumption of higher trophic level fish would have a greater association with mercury exposure among people in ASGM-affected areas, where mercury is both input to the system exogenously and released from sediment in areas deforested for ASGM, compared to those in non-ASGM affected areas. Lastly, we evaluated methodology to improve reported household fish consumption frequency.

2. Materials and Methods

2.1 Study Design and Participants

Data are from follow-up visits (2014) of the *Investigacion de Migracion, Ambiente, y Salud* (IMAS Study) (Investigation of Migration, Environment, and Health). We enrolled IMAS study participants between August 2011 and April 2012, with follow-up visits between August and November 2014. We used a two-stage probability sample, stratified by urban/rural status. At baseline, ten (10) urban and 38 rural communities within 10 km of the Interoceanic Highway were selected via probability proportional to estimated size at baseline (Figure 1, excluding the capital city of Puerto Maldonado). We used the Peruvian Institute for Statistics (INEI) definition to classify urban communities, which is a population center with at least 100 households. In the second stage, we randomly selected households within each of the selected communities based on community maps and a random number generator. In urban communities, we procured maps from Peru’s Land Titling Agency (COFOPRI), randomly selected blocks within each locality, and then used a random number generator to select households, with replacement. In rural communities, we drew a map of the town (including all households) with town leaders and used a random number generator to select households. We selected eighteen and twelve households per urban and rural community, respectively. In some cases, less than 12 households existed in a community. Of the 1,664 participants in 486 households interviewed at baseline, 64% of the households were re-interviewed in 2014 (n=1,021 participants in 310 households in the follow-up). Loss at follow-up was primarily due to migration as no households refused follow-up enrollment.

After obtaining informed consent from all adults (and assent for all children under 12), a pair of trained local interviewers surveyed the head of the household or the spouse of the head of the household. Surveys consisted of modules on migration, mobility, economic activities, household dietary patterns and health for all household members, including measuring anthropometrics (height, weight, hip/waist circumference). Specific variables for this analysis included age, sex, occupation, geographic location, household consumption of fish, and household consumption of fruits and vegetables. Following procedures described by the WHO (World Health Organization, 2008) for the collection hair specimens, three samples of approximately the width of a pencil eraser (6 mm) were clipped using stainless steel scissors from the occipital region of the scalp by trained fieldworkers. Samples were identified only by sample number and date, affixed onto folded sticky notes with the scalp end noted, and stored in plastic bags in an air-conditioned laboratory until transport and analysis.

One section of the household survey was devoted to ascertaining both riverine and canned fish consumption patterns at the household level. There were three stages to this section: First, a question asked about *total fish consumption* in the household (daily, weekly, monthly, never). Second, the respondent was asked “In your household, which species of fish do you consume?” but was unprompted with potential fish types as answers. Third, after completion of this unprompted question, the interviewer *prompted* the respondent about consumption frequencies of each of the non-mentioned fish types that were identified in the pilot-tested survey list of commonly consumed fish.

2.2 Laboratory Mercury Analysis

At Duke University, we analyzed hair total mercury (H-THg) in the 2 cm subsection of the hair closest to the scalp by thermal decomposition, amalgamation, and atomic absorption spectrometry (Milestone DMA-80, Milestone SRL, Italy) (U.S. EPA, 2007). We calibrated the instrument with a certified standard solution of dissolved $Hg(NO_3)_2$ in 1% HNO_3. The lower limit of quantification (LOQ) was 0.5 ng, calculated as 10 times the instrument detection limit. This LOQ corresponds to 0.05 μg/g for hair and 0.002 μg/g for fish (based on the typical analyzed mass of 0.01 g for hair and 0.25 g for fish, respectively). One fish sample was below the LOQ. We assigned it a value of 0.001 μg/g. The analytical quality was ensured by the analysis of a human hair standard reference material (ERM-DB001, European Reference Materials) or a fish muscle tissue reference material (DORM-4, National Research Council Canada). Measurement of the reference material within the certified range (0.365 ± 0.028 μg/g for hair; 0.412± 0.036 μg/g for fish reference) was required prior to further analyses of samples. We repeated the analyses of an analytical blank and standard reference material every 10 samples, and sometimes more frequently, during a batch run.

Because “canned tuna” was frequently reported as part of the diet, and because “canned tuna” can refer to other canned chunk fish like mackerel and not just tuna, we obtained canned fish (tuna and mackerel) of five different Peruvian brands at markets located in sampled communities in December 2015. Samples were tested in triplicate: one was dried by freeze-drying and two by oven drying at 110°C; samples were then analyzed for total mercury (THg) content by DMA-80. The drying method did not appear to influence THg measurements. Thus, we pooled the Hg measurements of dried samples and, after conversion with the measured wet:dry mass ratio, we calculated the average THg content (wet mass basis) for each brand (raw data in Supplemental Table 2; summary data in Supplemental Table 3).

2.3 Data management and statistical analysis

A trained interviewer entered survey data into a CSPro database. We exported the data to Stata and R for cleaning. All analyses for this study were conducted in Stata version 14 (StataCorp, College Station, TX) and one figure was created in each of Sas version 9.4 and ArcGIS version 10.6 (Esri Corp, Redlands, CA)

We assigned the appropriate trophic status to each fish, according to our previously published fish study (detritovores, herbivores, and filter feeders [trophic level 1]; omnivores [trophic level 2]; and obligate carnivores [trophic level 3]) (Diringer et al., 2015). We created a trophic-level adjusted fish consumption index, adjusting for the potential over-reporting of consumption of individual fish types (Equation 1). This was calculated by multiplying the indicator of frequency of consumption of each reported fish type (daily=3, weekly=2, monthly=1, or never=0) by its trophic level (1, 2, or 3), summing the products across all individual fish types reported, and then dividing this value by the amount of total fish reported in the household overall in the first question:

$$\textit{Fish consumption index} = \frac{\sum_{x=1}^{39}\left(\textit{Trophic Level}_{\textit{fish } x} * \textit{Frequency of consumption}_{\textit{fish } x}\right)}{\textit{Reported frequency of total fish consumption}} \qquad \text{Equation 1}$$

where reported frequency values were indicated as daily=1, weekly=2 and monthly=3 in the denominator. We then grouped the index into tertiles for statistical analysis: low (tertile 1), medium (tertile 2) and high (tertile 3), accordingly. While tertiles do not specifically correspond to a given intake of fish, households that primarily consume fish from lower trophic levels comprise tertile 1, while households that primarily consume fish from the higher trophic levels comprise tertile 3.

Community locations were grouped into bins of distance (>150km, 50–150km, 0–50km) and direction (north / west) along the highway measured from the central location of the capital city of Puerto Maldonado (green star in Figure 1) to approximate access to markets and decipher between areas of different dominant economic activities. The category furthest to the west (>150km west) is historically heavily deforested from ASGM. The 50–150km west area comprises most of the region's mining concessions, including the area known as "La Pampa" where ASGM activity is concentrated in established and transient mining towns along the Interoceanic Highway, while there is also still agricultural activity present. The areas proximal (<50 km) to Puerto Maldonado to the west and the north are largely rural and agricultural. The area 50–150km north of Puerto Maldonado is agricultural, primarily consisting of Brazil nut concessions. The region furthest to the north (>150km N) includes forest management areas for logging, agriculture, and is the border region with Brazil, where the highway continues through to the Atlantic Ocean. In order to compare participants from ASGM affected and non-ASGM affected areas, we defined a locality to be ASGM *affected* if active ASGM takes place within it or upstream in the same watershed, which corresponds largely with the mining corridor west of Puerto Maldonado (outlined in white oval in Figure 1).

We evaluated independent associations between H-THg and demographic, dietary, and household variables. Because H-THg data were non-normally distributed, we used Kruskal-Wallis H to determine whether H-THg concentrations differed among groups. Since the distribution of H-THg was right skewed, it was natural log transformed (ln) for regression analyses. To test the hypothesis that there were differences between ASGM and non-ASGM areas, intercept-only models of the natural log of mercury (ln(H-THg)) that partitioned variance among individuals, households, and communities in ASGM and non-ASGM communities was used. We used likelihood ratio tests to assess statistical significance of the addition of random intercepts. To assess the effects of independent variables with the natural log of H-THg (ln(H-THg), we created separate ASGM and non-ASGM three-level mixed effects linear regression using maximum likelihood estimation (MLE) with random intercepts for household and community to account for clustering. For all variables with $p<0.2$ in the separate ASGM and non-ASGM univariate models, we conducted subsequent separate multivariate analyses using three-level mixed effects linear regression models using MLE with random intercepts for household and community. Statistical significance was defined as $p<0.05$ for a two-tailed test. To estimate the predicted probability of exceeding the WHO JECFA reference level of 2.2 μg/g for demographic groups we created separate three-level mixed effects logistic regression models using MLE with random intercepts for household and community to account for clustering for ASGM-and non ASGM-affected areas. This research was approved by the institutional review board at United States Navy Medical Research Unit-6, Lima, Peru (#NAMRU6.2011.0004).

3. Results

3.1. Study Participants

The dataset for this analysis is comprised of the study participants who provided hair samples and had complete survey data (723 of 1,021 study participants, 71%, Table 1). Mean age was 33 years (range 0–94), and 51% of the participants were female. Household income was reported from a variety of locally relevant economic activities, with agriculture as the most common source (48% of households) and fishing as the least common. The “other” income category, which includes commercial, professional, transportation, employment in local businesses, and merchant economic activities, was also common. Most households earned less than 1,000 Peruvian New Soles per month (2014 equivalent to USD: $336). 33% of the sample lived in urban areas. 97% of households consumed rice daily, while 87% of households consumed fruit at least weekly. Communities were located across the entire span of the Interoceanic Highway (Figure 1, Figure 3a). 58% of the sample live in ASGM-affected communities, i.e. those located in the Colorado, Inambari, and main stem of the Madre de Dios, Malinowski, and Tambopata River watersheds, which included all communities west of Puerto Maldonado and a few communities surrounding the city (Figure 1, Figure 2a). Communities situated further north of Puerto Maldonado were non-ASGM affected.

There were no statistically significant differences between participants in ASGM affected areas versus non-ASGM affected areas in terms of gender or age category (Supplementary Table 1). While fish consumption frequency varied between the two groups, the tertiles of trophic level adjusted index of fish consumption were comparable between the two groups (Supplementary table 1). The two groups also differed in terms of self-reported household economic activities, income categories, fruit consumption, and the main source of fish for the households (river, fish farms, markets, or other sources). In the ASGM-affected areas, there is more purchasing of fish at markets compared to non-ASGM affected areas, where fish is more often from the river or fish farms.

3.2 Household Fish Consumption

Among the 298 households that answered the questions about fish consumption, only three households indicated that they did not eat fish because of a preoccupation with contamination. 281 indicated that they believed fish consumption to be important for their family’s health (94.6%). 96.7% of households reported consuming fish (9.5% reported daily, 41.3% reported weekly, and 45.8 % reported monthly consumption). There were significant differences in recall of fish consumption when comparing unprompted vs. prompted consumption (Supplemental Figure 1). As is often the case in self-reported fish consumption, most fish consumption was underreported without prompting, and in this case, specifically canned fish, which would be considered fish purchased from a market, was grossly underreported. Averaged across all fish in a given trophic level, there was a 45%, 51% and 59% increase in reporting in trophic levels 1, 2, and 3, respectively, when interviewees were prompted about specific fish consumption in the home (supplemental Figure 1)

Trophic level adjusted fish consumption index values ranged from zero (no fish consumption) to 113 (highest consumption of highest trophic level fish) among the households living in ASGM-affected areas, and ranged from 0–67 for households living in non-ASGM affected areas in this study. The fish consumption index varied across community locations and generally was higher among households in communities from 50km west of Puerto Maldonado through to the northern end of the highway compared to the more western communities (Figure 2c). Household fish consumption was more frequent in fishing households compared to non-fishing households, and was less frequent in mining households compared to non-mining households (data not shown).

3.3 Mercury exposure

The geometric mean for H-THg was 1.54 μg/g (95% CI: 1.44–1.65), with values ranging from below the detection limit to 16.65 μg/g (Table 1, section A). There was no statistically significant difference between males and females, but exposure varied among age groups. In general, children had lower H-THg than adults, though even 20% of the youngest children below 5 years of age had levels exceeding the 2.2 μg/g. 43% of women of childbearing age (ages 15–49) also exceeded this exposure level.

Households engaged in fishing as an economic activity had the highest H-THg concentrations compared to households engaged in other types of economic activities (Table

1, section B). No relationship was detected between income and H-THg. Consumption of rice and fruits was frequent among households in this study, though overall no statistically significant trends were observed with H-THg. Fish consumption, reported as both frequency of consumption and the Fish Consumption Index, was positively associated with H-THg exposure. Participants living in households that mainly obtained fish from the river had statistically significantly higher exposure than those living in households who procured their fish from non-river sources.

While there was no difference between the distribution of the fish consumption index itself between ASGM-affected and non-affected areas ($p<0.10$, Supplementary Table 1), the participants in the ASGM-and non ASGM-affected areas did differ in the relationship observed between the household fish consumption index and individual level exposure to mercury. Figure 3 illustrates that the distribution of H-THg by location and tertile of fish consumption differs in ASGM-affected vs. non-ASGM affected areas (Figure 3, $p=0.002$, Fishers exact test) by indicating the percentage of participants that exceed the frequently used H-THg cut-off values. Notably, 40.4% of participants in the ASGM-affected watersheds exceed 2.2 μg/g H-THg compared to 32.4% in non-ASGM affected watersheds. Six (1.4%) participants have H-THg values above 10 μg/g in the ASGM-affected areas compared to 2 (0.7%) in non-ASGM affected areas. Focused only on participants in the high tertile of the fish consumption index, 61.7% of them in the ASGM-affected areas exceed 2.2 μg/g H-THg while only 27.8% of the participants in the non-ASGM affected areas exceed this threshold (Figure 3 – third column of each of the two groups).

Living in rural (vs. urban; $p<0.0224$) and ASGM-impacted watersheds (vs. non-impacted, $p<0.0015$) both had statistically significant positive associations with H-THg (Table 1, Section C). When community locations were binned into distance categories from Puerto Maldonado, the group with the highest exposure was 0–50 km west of Puerto Maldonado, although higher mean H-THg values were apparent graphically in these communities (Figure 2b). This was consistent with fish consumption data reported above, wherein the highest percentage of households in the highest tertile of the Fish Consumption Index were also living in this area.

Mercury exposure increased as a function of distance from the Western MDD border with the region of Cusco to Puerto Maldonado (Figure 2b), and overall, H-THg levels were lower in non-ASGM affected communities compared to the ASGM-affected ones. Variance in mean values also varied by community location. The intra-class correlation (ICC) from an intercept-only model of the natural log of mercury (ln(H-THg)) that partitioned variance among individuals, households, and communities show significant differences between ASGM vs. non-ASGM affected areas. Individual level ICC values were high in ASGM-and non-ASGM affected areas (ICC=69% and 55%, respectively), while household level ICC values were more than two times higher in non-ASGM affected areas (42%) compared to ASGM affected areas (18%). Plots of the random community variance demonstrate significantly more variation in ASGM (12.7%) vs non-ASGM affected areas (3.8%, Figure 2d).

3.4 Multivariate Analysis

Trophic level of fish consumption and occupation were significantly associated with ln(H-THg) in the multivariate model (Figure 4, Supplemental Table 1). Compared to the lowest tertile of fish consumption, participants living in households in non-ASGM affected watersheds in the 2nd tertile had a nearly statistically significant increase (p=0.079) in ln(H-THg), but with no additional increase for those in tertile 3. In the ASGM-affected areas, a statistically significant trend was observed, and tertile 3 was statistically significantly different than tertile 2 (p=0.034). We tested whether there was a difference in ln(H-THg) in being in tertile 2 (medium) or tertile 3 (high) of the fish consumption index compared to tertile 1 (low) in both ASGM-affected and non ASGM affected communities. There was no statistically significant increase in ln(H-THg) associated with the grouped categories of medium or high fish consumers compared to low fish consumers in the non ASGM-affected group (beta = 0.18; p=0.201), but there was a statistically significant relationship in the ASGM-affected areas (beta = 0.25; p=0.028).

Living in a household engaged in fishing (p=0.006) and the harvesting or processing of Brazil nuts (p=0.036) as an economic activity were associated with higher ln(H-THg) compared to living in non-fishing households in the ASGM-affected areas only (Figure 4, Supplementary Table 2). A similar relationship was seen for households whose primary fish source was from sources other than the river (including lakes, markets and fish farms) compared to the river (Figure 4, Supplementary Table 2). It should be noted that fish sold locally in markets is not necessarily from Madre de Dios but may be non-local fish from Brazil or other regions of Peru, such as the coast. ln(H-THg) did not differ between males and females, income groups, nor between mining and non-mining households. Among households in ASGM affected and unaffected areas, adults had elevated exposure compared to children. Notably, ln(H-THg) among older adults (≥65 years of age) only decreased in the non-mining affected areas. The only other variable that was statistically significantly associated with elevated mercury exposure among participants in the non-ASGM affected areas was daily (compared to non-daily) fruit consumption, though this relationship was not seen among the ASGM-affected households.

The same independent variables as specified in the continuous outcome models were included in a multivariate logistic regression model to ascertain the predicted probabilities of exceeding the WHO/JEFCA reference level of 2.2 μg/g H-THg. Overall, participants living in the ASGM affected area have a 39% probability of exceeding this threshold while those in the non-ASGM affected area have a 30% probability. Predicted probabilities of exceeding this threshold for children (0–14 years) and adults (15+ years) varied, and were the highest among participants in ASGM-affected areas of at least 15 years of age in households with this highest tertile of the trophic-leveld adjusted fish consumption index. (Table 2).

3.5 Canned fish mercury analysis

Canned fish are a local dietary staple. Our measurements of a limited sampling of store-bought canned fish had THg contents ranging from 0.01 to 0.22 μg/g (wet mass basis). Supplementary Table 3 reports the raw data and Supplementary Table 4 reports the summary data of the canned fish mercury analysis.

4. Discussion

This study is the first population-representative sample in Madre de Dios, providing greater inference and generalization of exposure compared to any previous studies published on human THg exposure in the region. In this study, we found that 37.7% of the people living in Madre de Dios exceed the WHO/FAO recommended H-THg threshold (>2.2 μg/g), including 42.7% of women of childbearing age and 20.0% of children under 5 years of age. These results confirm that mercury exposure is high enough to pose a risk to human health across all age groups. Fish consumption was, as hypothesized, an important factor related to elevated H-THg; however, both mercury exposure and fish consumption varied across the region. Participants living within and downstream of ASGM activity had higher average H-THg than those in non-ASGM-affected watersheds. Within the ASGM-affected group, H-THg was higher among participants living downstream compared to those living within ASGM communities, likely owing to the comparatively lower fish consumption among households in the ASGM communities.

Measuring fish consumption by trophic level was of key interest. A pilot-tested module accurately measured household level fish consumption reported by the head or spouse of the head of household, which allowed us to evaluate consumption recall and develop a tropic-level adjusted fish consumption index. Both these measures were strong predictors of H-THg levels, particularly the fish consumption index, which captures variability of dietary Hg exposure risk across major trophic levels. However, this method of measuring consumption was also limited due to the lack of individual level measures of consumption. We address this limitation by specifying random intercepts that can adjust for correlated behaviors and latent constructs at the household level, though future work should refine this by ascertaining consumption patterns of individuals.

In addition to fish consumption, our multivariate model for ASGM affected areas identifies elevated H-THg in households whose economic activities include fishing and working with the harvesting or processing of Brazil nuts, but not in those engaged in mining. None of these relationships are statistically significant in the non-ASGM areas. It is possible that this lack of association with mining is due to few mining households in the study (n=12), lower fish consumption among mining households, or that other biomarkers, specifically urine, would be more appropriate for measuring Hg inhalation exposure compared to hair, which primarily reflects dietary exposure to MeHg. Since exposure to Hg vapor among amalgam burners and neighbors of such establishments has been documented in previous research (Yard et al., 2012), this route of exposure should not be ignored. Anecdotally, in Madre de Dios,we have observed that gold shops are largely located in the capital city of Puerto Maldonado, and in urban gold-mining towns (the only two of these in the study are Laberinto and Mazuko). In total, we had only 27 participants from these areas combined, and ascertaining locations of gold shops in these towns was outside of the scope of the present study. Future studies should investigate all possible routes of exposure, as well as estimate the relative amounts of amalgam burned in these shops versus in the field.

Although our design excluded communities located beyond 10 km from the Interoceanic Highway and also excluded the capital city of Puerto Maldonado, it remains the only study specifically designed to draw inference about human, environment and health dynamics in Madre de Dios. Note also that while the sample frame and selection was in 2011 and the trace metals assessment in 2014, dropout between baseline and follow-up was non-differential with respect to age, sex, and urban/rural status. We do note that households not participating in 2014 were more likely to be from the district of Laberinto (an ASGM-affected region) compared to the other districts along the road (Pettigrew et al., 2019).

This study is also the largest assessment of Hg ever conducted in Madre de Dios (Supplementary Table 5). Hg exposure in this study is comparable to exposure levels measured in other recent studies in Madre de Dios,. However, H-THg levels were lower in this study compared to a non-population-based study obtaining data from individuals living along the Madre de Dios River (Wyatt et al., 2017). Notably, the considerably higher exposure to Hg reported in Yard (2012) is likely due to that author's non-random selection of participants and the different biomarkers of exposure employed in that study. In addition, Ashe (2012) found higher exposures among households living in ASGM communities compared to households near Puerto Maldonado which contrasts our results (Ashe, 2012). Some of these discrepancies observed between the present study and the previous studies may be due to the more robust randomized sampling of the current study, the biomarkers used (hair versus urine versus blood), methods of analysis and year of sampling. The levels observed in the current study are in the low end of the range of those observed in other ASGM communities, including those in other parts of the Amazon Basin and (Barbieri and Gardon, 2009; Gibb and O'Leary, 2014). As the authors of these two previous reviews note, a wide variation of exposure exists in the Amazon, which may be due to varying current and historical land uses, differences in the local use of elemental Hg in mining, differences in fish consumption patterns, differences in populations surveyed, or potentially due to differing laboratory Hg analysis methods. For instance, many of the studies assessing ASGM-related Hg exposure in Brazil have been conducted among small, isolated, traditional riverine communities Madeira and Tapajos River Basins, which are more remote than this current study area, and whose populations have a higher dependency on fish as the main source of protein than the highway-adjacent localities of Madre de Dios (Barbieri and Gardon, 2009).

The most important questions in our research is who is at risk and why. We find Hg exposure higher in adults than children, likely due to their higher level of fish consumption and longer time period consuming fish as has been noted previously in the Amazon (Dorea and Marques, 2016). In older adults, exposures fall in non ASGM-affected areas, but stay elevated in ASGM-affected areas. This is likely tied to fish consumption as 65% of participants in this age group (65+ years) that live in non-ASGM affected areas are in the low category of the Fish consumption index (Tertile 1) while this is the case for only 35% of older adults in the ASGM-affected areas. In the ASGM-affected areas, there are more children (and adults) with critical levels of exposure (>6 µg/g) that may impact cognitive development (Betancourt et al., 2015; Cordier et al., 2002; Reuben A et al., in submission).

While underreporting of food consumption is often the case in food frequency questionnaires, in the case of this study, the significant underreporting of canned fish may point towards the lack of knowledge or consideration of the potential dietary Hg exposure risk via canned tuna. As our analysis revealed a wide range of Hg content in canned fish, some of which exceeded United States EPA-FDA fish consumption guidance which recommends limiting weekly servings for fish with Hg contents >0.15 μg/g (United States Environmental Protection Agency, 2016), more detailed evaluations of Hg contents in household food sources, including fish, are warranted. Nevertheless, this work highlights an important consideration when discussing options for dietary behavior change and government intervention strategies to reduce dietary Hg exposure in the region.

Future research quantifying the relationship between fish consumption and measured H-THg should consider direct measures of mercury in food (E.g.., fish) consumed. Fish consumption (frequency and species availability) can be seasonal, influencing the relationship identified (Wyatt et al, 2017). In the Amazon, seasonality may cause another unintended problem as methylation rates of mercury in sediment can vary under different environmental conditions, resulting in differential bioaccumulation by season and over time (Azevedo et al., 2019; Guimaraes et al., 2000) In this study, all surveys and hair were collected at the end of the dry season, making seasonal effects on exposure unlikely in our study.

This study was limited to the measurement of THg in hair rather than MeHg. Since MeHg corresponds to up to 90% of mercury in H-THg in highly exposed populations, H-THg is considered a valid marker of dietary exposure (Barbieri and Gardon, 2009; Berglund et al., 2005). Notwithstanding, the measured H-THg in this study might reflect a combination of MeHg from fish consumption and exogenous inorganic Hg adsorbed to the hair (Sherman et al., 2015). However, our conclusions are likely not affected by this limitation as very few of the households were engaged directly in mining activities (only 5% of the study population), and mining as a household economic activity was not found to be statistically significant in the final model. Considering the potential for a small subset of this population to have mixed Hg exposure, further work might attempt to evaluate multiple biomarkers, such as blood, urine or toenails, and the Hg form in these biomarkers as a means to delineate the extent of acute versus chronic, and MeHg vs inorganic Hg exposures.

The origins of the mercury to which the MDD communities are exposed could include naturally occurring Hg, exogenous elemental Hg (Hg^0), and other sources. It is important to underscore that the source of Hg exposure observed here has not yet been determined. Source tracking with stable isotopic signatures of Hg might be a useful tool, as documented in other environmental mercury exposure cases (Laffont et al., 2011; Marshall et al., 2018; Miserendino et al., 2018; Schudel et al., 2018; Sherman et al., 2015); however, such studies have not yet been undertaken in Madre de Dios. Lastly, other future directions of research in this area should also focus on whether in utero exposure is an issue of concern, and ascertain information on relevant co-exposures including omega-3-fatty acids, selenium, stress, and co-exposures to other trace elements, such as arsenic, that may interact with mercury.

5. Conclusion

Hg exposure is prevalent in Madre de Dios, Peru, and is especially of public health importance in areas downstream of ASGM and deforestation activities. Monitoring of children and women of childbearing age for elevated Hg levels is necessary to assess potential cognitive developmental impacts of chronic dietary exposure. Educational outreach regarding benefits and risks of consuming fish from varying trophic levels should be strengthened and implemented, especially in target areas identified in this study. To direct successful scientific-based policies and comply with the Minamata Convention goals to reduce use of and exposure to mercury (www.mercuryconvention.org), further multidisciplinary research engaged with the Ministries of Health and Environment is required to ascertain the provenance of the Hg exposure, be it from elemental mercury introduced in ASGM or inorganic mercury liberated via deforestation activities. As research moves forward in this area, it is important to engage with these ministries, local communities and other stakeholder groups to develop and support sustainable policies and programs that promote warranted behavioral changes that reduce mercury exposure while not adversely affecting the nutritional status or cultural norms of this vulnerable population.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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References

(CAMEP), C. A. M. E. P., Mercury concentrations in fish and humans in Puerto Maldonado: Research Brief 1. 2013.

Ashe K, 2012. Elevated mercury concentrations in humans of Madre de Dios, Peru. PloS one. 7, e33305. [PubMed: 22438911]

Asner GP, Tupayachi R, 2017. Accelerated losses of protected forests from gold mining in the Peruvian Amazon. Environmental Research Letters. 12.

Azevedo LS, et al., 2019. Influence of the flood pulse on mercury accumulation in detritivorous, herbivorous and omnivorous fish in Brazilian Amazonia. Ecotoxicology. 28, 478–485. [PubMed: 31025154]

Barbieri FL, Gardon J, 2009. Hair mercury levels in Amazonian populations: spatial distribution and trends. Int J Health Geogr. 8, 71. [PubMed: 20025776]

Basu N, et al., 2018. A State-of-the-Science Review of Mercury Biomarkers in Human Populations Worldwide between 2000 and 2018. Environ Health Perspect. 126, 106001. [PubMed: 30407086]

Berglund M, et al., 2005. Inter-individual variations of human mercury exposure biomarkers: a cross-sectional assessment. Environ Health. 4, 20. [PubMed: 16202128]

Berky AJ, et al., 2018. Predictors of mitochondrial DNA copy number and damage in a mercury-exposed rural Peruvian population near artisanal and small-scale gold mining: An exploratory study. Environ Mol Mutagen.

Betancourt Ó, et al., 2015. Decline of General Intelligence in Children Exposed to Manganese from Mining Contamination in Puyango River Basin, Southern Ecuador. 12, 453–460.

Cordier S, et al., 2002. Neurodevelopmental investigations among methylmercury-exposed children in French Guiana. Environ Res. 89, 1–11. [PubMed: 12051779]

de Castro NS, de Oliveira Lima M, 2014. Biomarkers of mercury exposure in the Amazon. Biomed Res Int. 2014, 867069. [PubMed: 24895619]

Diringer SE, et al., 2015. River transport of mercury from artisanal and small-scale gold mining and risks for dietary mercury exposure in Madre de Dios, Peru. Environmental Science: Processes & Impacts. 17, 478–487. [PubMed: 25573610]

Dorea JG, Marques RC, 2016. Mercury levels and human health in the Amazon Basin. Ann Hum Biol. 43, 349–59. [PubMed: 27230737]

Fadini PS, Jardim WF, 2001. Is the Negro River Basin (Amazon) impacted by naturally occurring mercury? The Science of the total environment. 275, 71–82. [PubMed: 11482405]

Gibb H, O'Leary KG, 2014. Mercury Exposure and Health Impacts among Individuals in the Artisanal and Small-Scale Gold Mining Community: A Comprehensive Review. 122, 667–672.

Gonzalez D, 2015. Mercury exposure and risk among women of childbearing age in Madre de Dios. Tropical Resources Bulletin: The Bulletin of the Yale Tropical Resources Institute. 34, 16–24.

Grandez-Urbina JA, et al., 2014. [Mercury concentrations in women's hair in a native community of Peru]. Gac Sanit. 28, 177. [PubMed: 24300383]

Grandjean P, et al., 2010. Adverse effects of methylmercury: environmental health research implications. Environ Health Perspect. 118, 1137–45. [PubMed: 20529764]

Gregory P A. a. R. T., 2016. Accelerated Losses of Protected Forests From Gold Mining in the Peruvian Amazon. Environmental Research Letters. 12.

Guimaraes JRD, et al., 2000. Mercury methylation along a lake-forest transect in the Tapajos river floodplain, Brazilian Amazon: seasonal and vertical variations. Science of the Total Environment. 261, 91–98.

Ha E, et al., 2017. Current progress on understanding the impact of mercury on human health. Environ Res. 152, 419–433. [PubMed: 27444821]

Humala Tasso O, DECRETO SUPREMO : Declaran el Estado de Emergencia en once distritos de las provincias de Tambopata, Manu y Tahuamanu del departamento de Madre de Dios, por contaminación por mercurio en el marco de la Ley 29664 sobre Gestión de Riesgo de Desastres. 2016.

Joint FAO/WHO Expert Committee on Food Additives, Evaluation of certain food additives and contaminants : sixty-seventh report of the Joint FAO/WHO Expert Committee on Food Additives. World Health Organization, Rome, Italy 2006.

Lacerda LD, et al., 2004. The effects of land use change on mercury distribution in soils of Alta Floresta, Southern Amazon. Environmental Pollution. 129, 247–255. [PubMed: 14987810]

Laffont L, et al., 2011. Hg Speciation and Stable Isotope Signatures in Human Hair As a Tracer for Dietary and Occupational Exposure to Mercury. Environmental Science & Technology. 45, 9910–9916. [PubMed: 22003970]

Langeland AL, et al., 2017. Mercury Levels in Human Hair and Farmed Fish near Artisanal and Small-Scale Gold Mining Communities in the Madre de Dios River Basin, Peru. Int J Environ Res Public Health. 14.

Malm O, 1998. Gold mining as a source of mercury exposure in the Brazilian Amazon. Environmental Research. 77, 73–78. [PubMed: 9600798]

Marshall BG, et al., 2018. Evidence of transboundary mercury and other pollutants in the Puyango-Tumbes River basin, Ecuador-Peru. Environmental Science-Processes & Impacts. 20, 632–641. [PubMed: 29492485]

Martinez G, et al., 2018. Mercury Contamination in Riverine Sediments and Fish Associated with Artisanal and Small-Scale Gold Mining in Madre de Dios, Peru. Int J Environ Res Public Health. 15.

Mastel M, et al., 2018. Critical linkages between land use change and human health in the Amazon region: A scoping review. Plos One. 13.

Ministerio de Salud, Guia de practica clinica para el diagnostico y tratamiento de la intoxicacion por mercurio. In: D. G. d. S. d. l. P. E. S. N. d. V. y. C. d. R. p. C. c. M. P. y. O. s. Químicas, (Ed.), Lima, Peru, 2013.

Miserendino RA, et al., 2018. Mercury Pollution in Amapa, Brazil: Mercury Amalgamation in Artisanal and Small-Scale Gold Mining or Land-Cover and Land-Use Changes? Acs Earth and Space Chemistry. 2, 441–450.

Moreno-Brush M, et al., 2018. Mercury concentrations in bats (Chiroptera) from a gold mining area in the Peruvian Amazon. Ecotoxicology. 27, 45–54. [PubMed: 29101638]

Moreno-Brush M, et al., 2016. Is mercury from small-scale gold mining prevalent in the southeastern Peruvian Amazon? Environ Pollut. 218, 150–159. [PubMed: 27552048]

NRC (National Research Council), Toxicological Effects of Methylmercury. Washington, D.C., 2000.

Pettigrew SM, et al., 2019. In urban, but not rural, areas of Madre de Dios, Peru, adoption of a Western diet is inversely associated with selenium intake. Science of The Total Environment. 687, 1046–1054.

Reuben A, et al., in submission. Elevated hair mercury levels are associated with neurodevelopmental deficits in children living near artisanal and small-scale gold mining in Peru. Environmental Health Perspectives.

Rudel TK, et al., 2009. Changing Drivers of Deforestation and New Opportunities for Conservation. 23, 1396–1405.

Schudel G, et al., 2018. An investigation of mercury sources in the Puyango-Tumbes River: Using stable Hg isotopes to characterize transboundary Hg pollution. Chemosphere. 202, 777–787. [PubMed: 29609178]

Sheehan MC, et al., 2014. Global methylmercury exposure from seafood consumption and risk of developmental neurotoxicity: a systematic review. Bull World Health Organ. 92, 254–269F. [PubMed: 24700993]

Sherman LS, et al., 2015. Assessment of mercury exposure among small-scale gold miners using mercury stable isotopes. Environ Res. 137, 226–34. [PubMed: 25577187]

Southworth J, et al., 2011. Roads as Drivers of Change: Trajectories across the Tri-National Frontier in MAP, the Southwestern Amazon. Remote Sensing. 3, 1047–1066.

Swenson JJ, et al., 2011. Gold mining in the Peruvian Amazon: global prices, deforestation, and mercury imports. PloS one. 6, e18875. [PubMed: 21526143]

Tarabochia ML, PERÚ: Estado de emergencia en Madre de Dios se prolonga por 60 días más. Mongabay Latam, Vol. 2019, 2016.

U.S. EPA, Method 7473; Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophometry. US Environmental Protection Agency, Washington, DC, 2007.

United States Environmental Protection Agency, EPA-FDA Fish Advice: Technical Information. Vol. 2019 2016.

Weinhouse C, et al., 2017. Hair Mercury Level is Associated with Anemia and Micronutrient Status in Children Living Near Artisanal and Small-Scale Gold Mining in the Peruvian Amazon. Am J Trop Med Hyg. 97, 1886–1897. [PubMed: 29016304]

World Health Organization, Guidance for identifying populations at risk from mercury exposure. WHO; Inter-Organization Programme for the Sound Management of Chemicals, Geneva, 2008.

World Health Organization (WHO), Environmental Health Criteria 101: Methylmercury. World Health Organization, Geneva, 1990.

Wyatt L, et al., 2017. Spatial, Temporal, and Dietary Variables Associated with Elevated Mercury Exposure in Peruvian Riverine Communities Upstream and Downstream of Artisanal and Small-Scale Gold Mining. Int J Environ Res Public Health. 14.

Yard EE, et al., 2012. Mercury exposure among artisanal gold miners in Madre de Dios, Peru: a cross-sectional study. Journal of Medical Toxicology. 8, 441–448. [PubMed: 22926732]

Highlights:

- The arithmetic mean of Hair-total mercury concentration was 2.24 µg/g, (median: 1.60 µg/g; geometric mean: 1.54 µg/g).
- 37.2% of the population of Madre de Dios, Peru, including 42.7% of women of child-bearing age and 20.0% of children under 5, has hair-total mercury levels exceeding WHO guidelines.
- Fishing as a household economic activity and Higher consumption of higher trophic level fish is only associated with higher hair total mercury in persons residing in ASGM-affected areas, not in unaffected areas
- The predicted probabilities of exceeding the WHO guideline of 2.2 µg Hg/g hair was 0.730 for adults ≥ 15 years in the highest fish-eating tertile living in ASGM affected regions, compared to 0.412 and in the non-ASGM affected areas.
- Mean mercury content of two brands of canned tuna and three brands of canned mackerel varied from 0.012–0.223 µg/g wet weight.

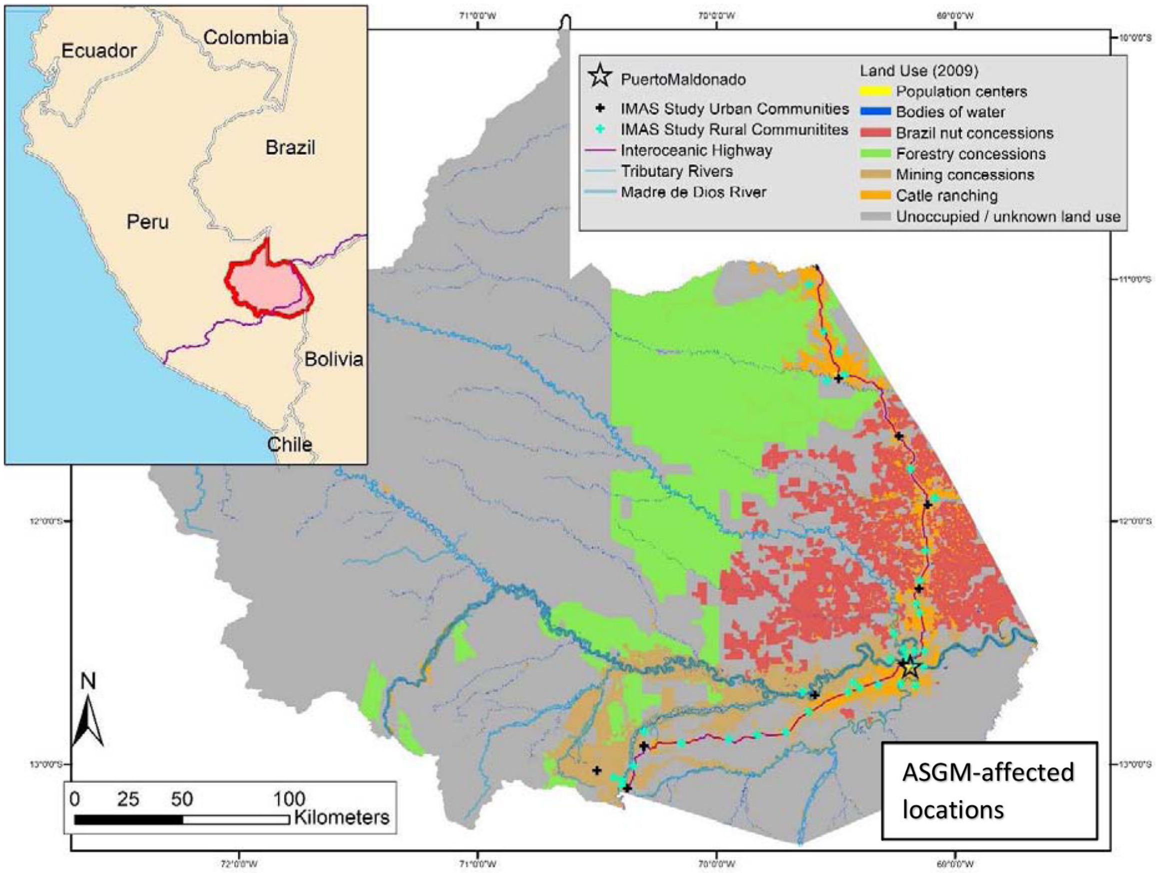

Figure 1.

Geographic Setting of Study Area. Source of Land Use Data Layer: Amazon Conservation Association. Localities designated as *ASGM-affected* are within the annotated white oval on the map. Source of 2009 Land Use data: Amazon Conservation Association

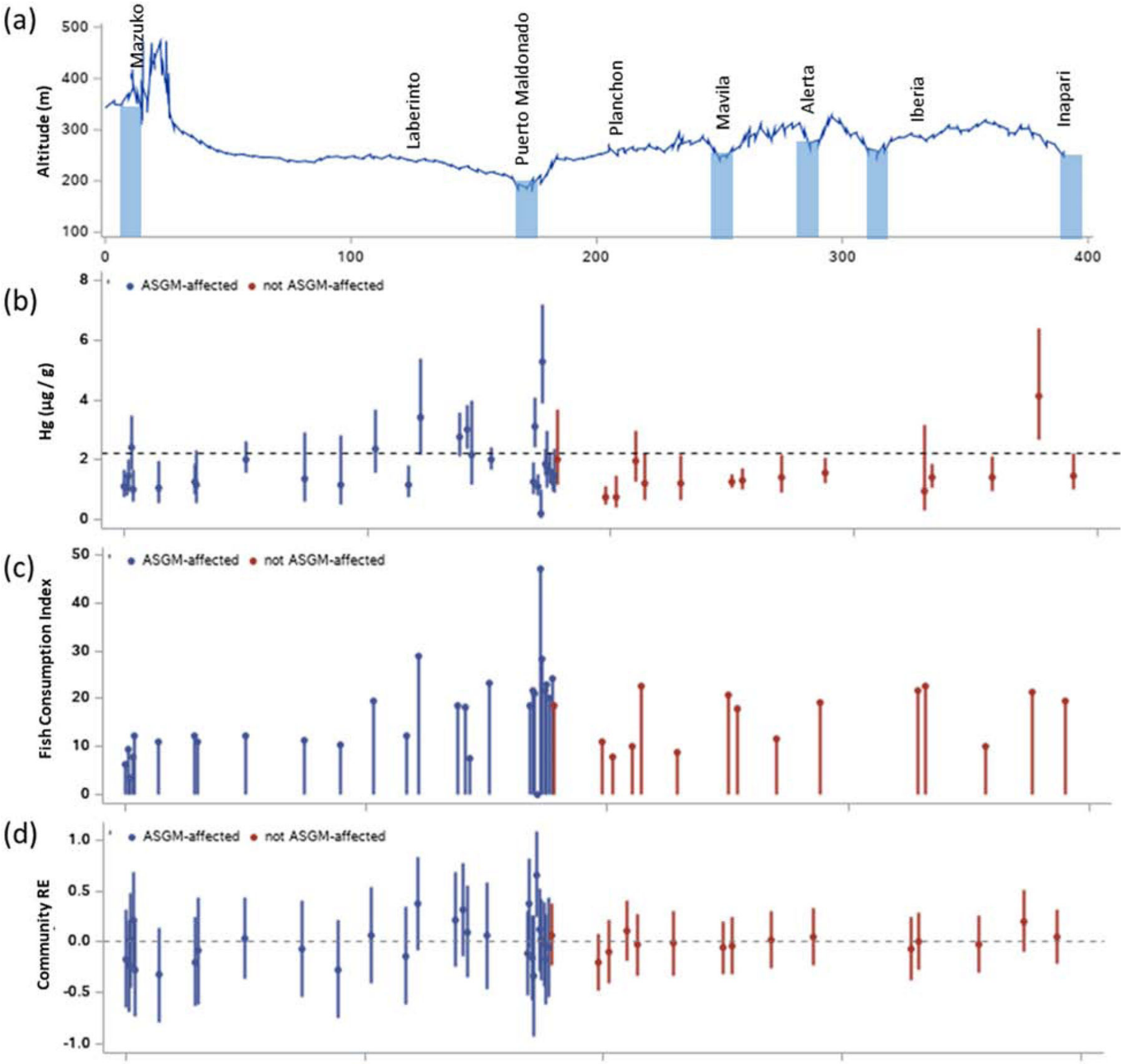

Figure 2.

Distribution of community location along the highway by hair total mercury (μg/g), fish consumption index, and community variation. The x-axis for all graphs is the relative distance (in km) of each community along the highway, starting with the most western community in the study. Figure 2a shows the road altitude, location of select communities and location of all rivers that cross the highway (in blue vertical bars); Figure 2b shows the average H-THg and 95% confidence interval (CI) by community along the highway; Figure 2c is average fish consumption index level by community along the highway; and Figure 2d is the community random intercept and 95% CI from an intercept-only model that partitions variance among individuals, households and communities. Communities located in ASGM affected watershed are denoted in blue while those in non-ASGM-affected watershed are denoted in brown.

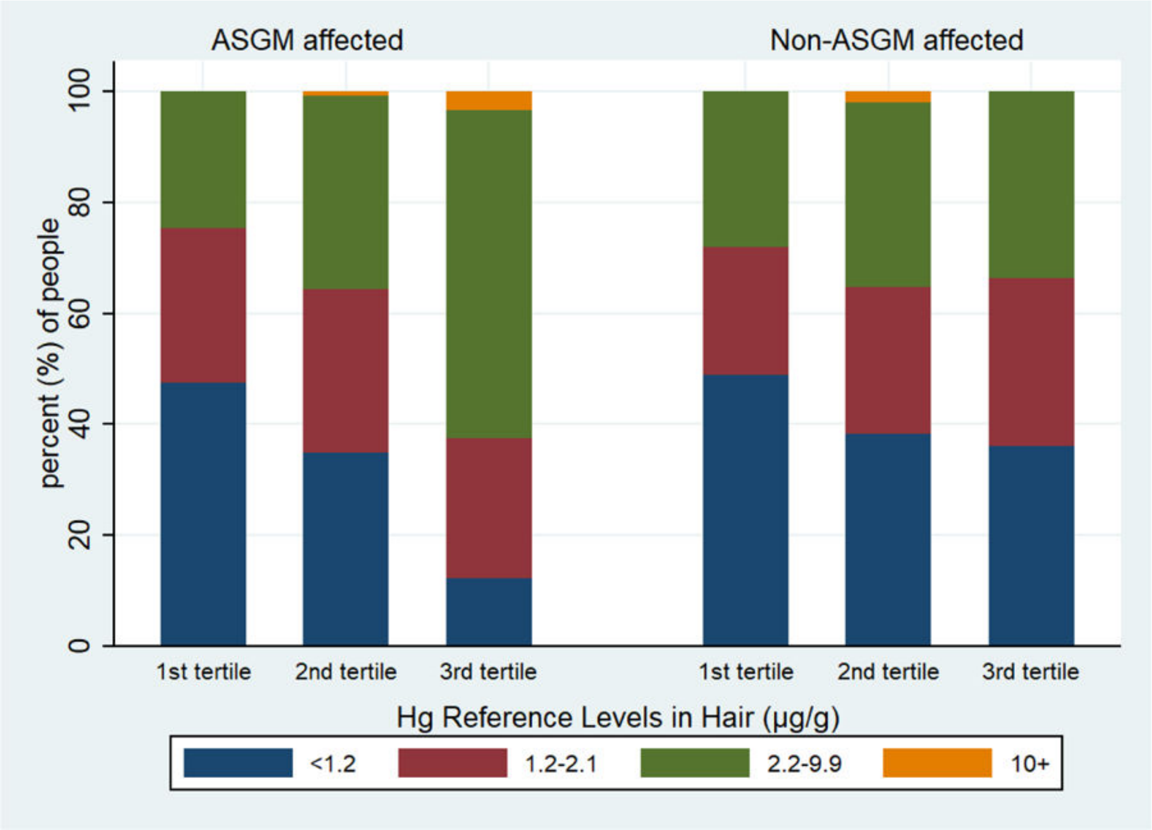

Figure 3.

Percentage of participants within each tertile of fish consumption that exceed established H-THg reference levels in µg/g in ASGM affected (left three bars) and Non-ASGM affected (right three bars) areas. Reference values indicated include the US National Research Council's reference value for non-fish eating populations of 1.2 µg/g corresponding to a reference dose (RfD) for methylmercury of 0.1 µg/kg bodyweight/day (NRC (National Research Council), 2000); the WHO JEFCA PTWI 1.6 µg/ kg bodyweight established in 2003 and confirmed in 2006 which Peru adheres to, corresponding to 2.2 µg/g in hair (Joint FAO/WHO Expert Committee on Food Additives, 2006), and 10 µg/g which WHO previously established as the level of concern for impact of methylmercury on the developing fetus of pregnant mothers (World Health Organization (WHO), 1990), and which is commonly used as a reference level in the Amazon (Barbieri and Gardon, 2009).

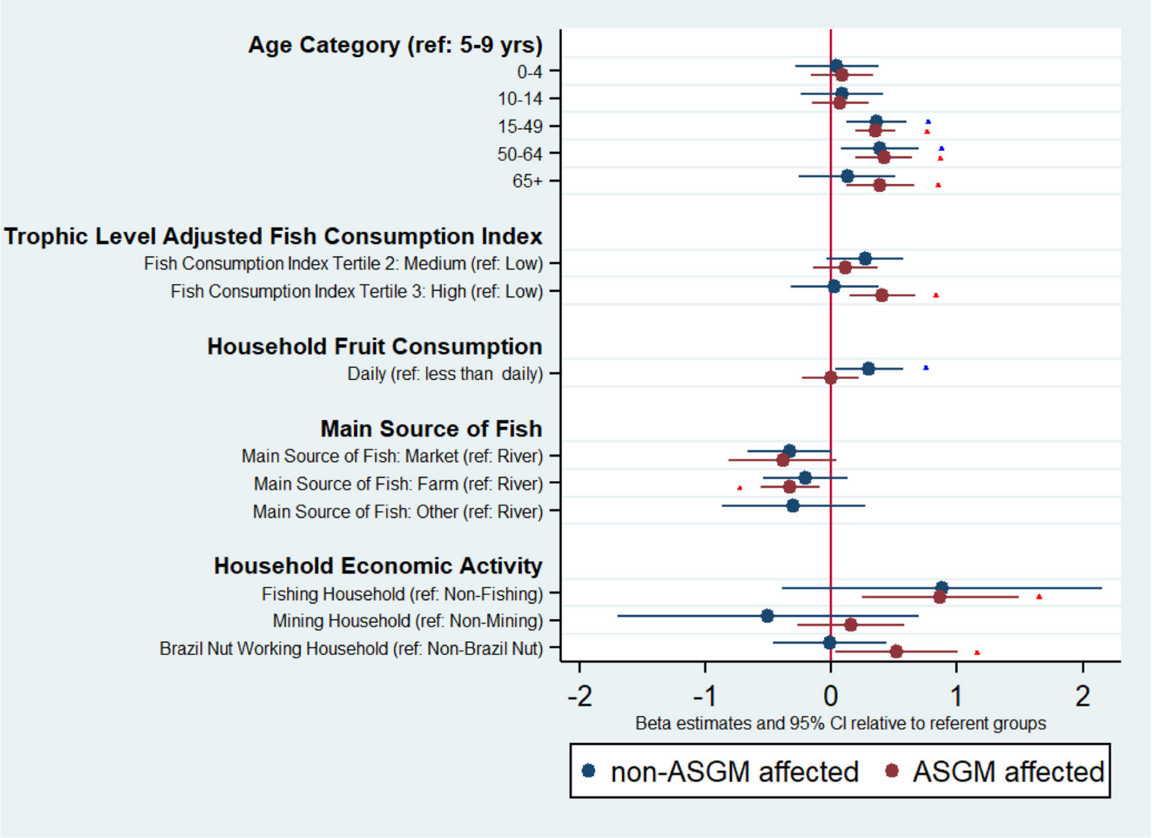

Figure 4.

Association between ln(Hair-THg) and individual and household variables in IMAS study households, 2014 (n=676) from separate multilevel multivariate regression models for non-ASGM affected and ASGM affected households with random intercepts at the household and community levels. Dots represent beta estimates and whiskers the 95% confidence intervals of the beta estimates. The small triangles indicate statistical significance for alpha<0.05 with blue corresponding to non-ASGM affected model and red corresponding to ASGM-affected model. Models are mutually adjusted for all listed variables, and also for sex, urban location, and household monthly income category (see Supplementary Table 2).

Table 1.

H-THg exposure by Individual, Household and Community Characteristics, Madre de Dios, Peru in 2014 (n=723). The lower limit of quantification (LOQ) for H-THg was 0.05 mg/g.

	N (%)	Range (min-max), µg/g	n(%) exceeding 2.2 µg/g H-THg	H-THg Geometric mean (95% CI), µg/g	H-THg Arithmetic mean (95% CI), µg/g	H-THg Median (IQR), µg/g	Kruskal-Wallis H test
All participants	**723 (100)**	**<LOQ-16.65**	**37.21**	**1.54 (1.44–1.65)**	**2.24 (2.09–2.38)**	**1.60 (0.86–2.99)**	
A. Individual Characteristics							
Male	352 (48.6)	<LOQ-13.22	134 (38.07)	1.54 (1.39–1.71)	2.29 (2.07–2.51)	1.60 (0.82–3.25)	p=0.8222
Female	371 (51.4)	0.08–16.65	135 (36.39)	1.54 (1.41–1.69)	2.18 (1.98–2.38)	1.60 (0.90–2.85)	
Age in years							
0–4	55 (7.61)	0.29–6.61	11(20.00)	1.19 (0.99–1.45)	1.54 (1.21–1.87)	1.22 (0.66–1.83)	p=0.0001
5–9	95 (13.14)	0.18–12.94	23 (24.21)	1.25 (1.05–1.48)	1.80 1.42–2.19)	1.15 (0.74–2.19)	
10–14	63 (8.71)	<LOQ-6.73	15 (23.81)	1.11 (0.82–1.50)	1.64 (1.30–1.99)	1.31 (0.69–2.18)	
15–49	321 (44.40)	0.11–13.22	138 (42.99)	1.81 (1.64–1.99)	2.52 (2.29–2.76)	1.86 (1.08–3.40)	
50–64	112 (15.49)	0.08–16.65	52 (46.43)	1.75 (1.46–2.10)	2.58 (2.14–3.02)	1.82 (1.06–3.44)	
65+	77 (10.65)	0.14–11.26	30 (38.96)	1.35 (1.08–1.69)	2.05 (1.63–2.47)	1.56 (0.65–3.06)	
Women of Childbearing Age (15–49 years)	185 (25.59)	0.11–12.16	79 (42.7)	1.79 (1.58–2.02)	2.45 (2.17–2.74)	1.84 (1.10–3.29)	--

B. Household Characteristics

Household Economics	Participants n(%)	Households n(%)	N (%) exceeding 2.2 µg/g Hair THg	H-THg Geometric mean (95% CI), µg/g	H-THg Arithmetic mean (95% CI), µg/g	H-THg Median (IQR), µg/g	Kruskal-Wallis H test
Household Economic Activity [a]							
Agriculture	327 (45.23)	142 (48.14)	126 (38.53)	1.48 (1.32–1.65)	2.20 (1.98–2.42)	1.60 (0.80–2.96)	p<0.5846 [b]
Mining	36 (4.98)	12 (4.07)	16 (44.44)	2.03 (1.57–2.65)	2.64 (2.02–3.27)	1.95 (1.17–4.12)	p<0.0581 [b]
Fishing	21 (2.90)	6 (2.03)	20 (95.24)	6.53 (5.06–8.43)	7.40 (5.88–9.19)	7.25 (4.81–9.32)	p<0.0001 [b]

B. Household Characteristics

Household Economics	Participants n(%)	Households n(%)	N (%) exceeding 2.2 µg/g Hair THg	H-THg Geometric mean (95% CI), µg/g	H-THg Arithmetic mean (95% CI), µg/g	H-THg Median (IQR), µg/g	Kruskal-Wallis H test
Logging	93 (12.86)	31 (10.51)	23 (24.73)	1.44 (1.24–1.66)	1.83 (1.54–2.11)	1.42 (0.98–2.18)	p<0.1864 [b]
Brazil Nuts	48 (6.64)	19 (6.44)	24 (50.00)	2.05 (1.64–2.58)	2.65 (2.14–3.16)	2.22 (1.28–3.92)	p<0.0153 [b]
Other	347 (47.99)	131 (44.41)	116 (33.43)	1.50 (1.37, 1.64)	2.12 (1.90–2.33)	1.52 (0.82–2.73)	p<0.0957 [b]
None	20 (2.77)	13 (4.41)	7 (35.00)	1.23 (0.71–2.11)	2.09 (1.17–3.00)	1.47 (0.55–3.15)	p<0.4544 [b]
Monthly Household Income (Peruvian New Soles)							
<600	129 (17.92)	63 (21.43)	47 (36.43)	1.36 (1.11–1.67)	2.14 (1.78–2.50)	1.54 (0.86–3.04)	p<0.1981
600–999	302 (41.94)	121 (41.16)	109 (36.09)	1.53 (1.39–1.68)	2.09 (1.90–2.27)	1.55 (0.90–2.86)	
1000–1999	196 (27.22)	68 (23.13)	82 (41.84)	1.76 (1.57–1.98)	2.42 (2.13–2.72)	1.83 (1.01–3.27)	
2000+	93 (12.92)	42 (14.29)	31 (33.33)	1.47 (1.19–1.83)	2.50 (1.90–3.09)	1.52 (0.67–2.73)	

Household Dietary Factors	Participants n(%)	Households n(%)	n(%) exceeding 2.2 µg/g H-THg	H-THg Geometric mean (95% CI), µg/g	H-THg Arithmetic mean (95% CI), µg/g	H-THg Median (IQR), µg/g	Kruskal-Wallis H test
Rice Consumption							
Never	6 (0.83)	2 (0.68)	5 (83.33)	2.70 (1.02, 7.15)	3.38 (1.70–5.07)	3.60 (3.14–4.59)	p<0.1019
Monthly	6 (0.83)	2 (0.68)	4 (66.67)	2.70 (1.14–6.34)	3.35 (1.39–5.32)	4.29 (1.15–4.52)	
Weekly	9 (1.25)	7 (2.38)	2 (22.22)	1.44 (0.90–2.30)	1.68 (0.95–2.41)	1.38 (1.29–3.07)	
Daily	699 (97.08)	283 (96.26)	258 (36.91)	1.54 (1.43–1.65)	2.23 (2.08–2.38)	1.60 (0.86–2.94)	
Fruit Consumption							
Never	14 (1.94)	5 (1.70)	5 (35.71)	1.51 (0.88–2.61)	2.20 (1.13–3.27)	1.57 (0.63–3.78)	p<0.0786
Monthly	81 (11.25)	31 (10.54)	28 (34.57)	1.56 (1.30–1.85)	2.03 (1.69–2.38)	1.70 (1.19–2.59)	
Weekly	334 (46.39)	136 (46.26)	117 (35.03)	1.43 (1.30–1.58)	2.10 (1.89–2.31)	1.45 (0.75–2.83)	
Daily	291 (40.42)	122 (41.50)	119 (40.89)	1.70 (1.52–1.91)	2.46 (2.21–2.72)	1.80 (1.00–3.25)	
Fish Consumption							

Household Dietary Factors	Participants n(%)	Households n(%)	n(%) exceeding 2.2 μg/g H-THg	H-THg Geometric mean (95% CI), μg/g	H-THg Arithmetic mean (95% CI), μg/g	H-THg Median (IQR), μg/g	Kruskal-Wallis H test
Never	22 (3.06)	10 (3.40)	1 (4.55)	0.32 (0.16–0.64)	0.57 (0.33–0.81)	0.38 (0.24–0.81)	p<0.0001
Monthly	322 (44.72)	135 (45.92)	99 (30.75)	1.37 (1.25–1.51)	1.94 (1.75–2.12)	1.46 (0.77–2.42)	
Weekly	301 (41.81)	121 (41.16)	126 (41.86)	1.75 (1.60–1.91)	2.31 (2.10–2.52)	1.80 (1.06–3.12)	
Daily	75 (10.42)	28 (9.52)	43 (57.33)	2.53 (2.03–3.16)	3.76 (3.02–4.50)	2.76 (1.25–4.92)	
Most common source of fish eaten in the house: (n=676)							
River	318 (47.04)	120 (43.96)	147 (46.23)	1.95 (1.77–2.14)	2.73 (2.47–3.00)	1.98 (1.08–3.66)	p<0.0001
Fish farm	85 (12.57)	35 (12.82)	30 (35.29)	1.26 (1.03–1.54)	1.82 (1.50–2.14)	1.44 (0.71–2.48)	
Market	260 (38.46)	112 (41.03)	82 (31.54)	1.47 (1.34–1.62)	1.97 (1.77–2.16)	1.53 (0.87–2.62)	
Other	13 (1.92)	6 (2.20)	3 (23.08)	1.54 (1.06–2.24)	1.83 (1.08–2.59)	1.59 (1.18–2.00)	
Trophic Level Adjusted Fish Consumption Index							
Tertile 1	262 (36.39)	115 (39.12)	68 (25.95)	1.13 (1.00–1.27)	1.61 (1.44–1.77)	1.28 (0.67–2.22)	p<0.0001
Tertile 2	228 (31.67)	90 (30.61)	81 (35.53)	1.59 (1.42–1.78)	2.31 (2.02–2.60)	1.60 (0.98–3.04)	
Tertile 3	230 (31.94)	89 (30.27)	120 (52.17)	2.17 (1.95–2.41)	2.90 (2.61–3.20)	2.39 (1.31–3.93)	

C. Community Characteristics

Community Location	Participants n(%)	Communities n(%)	N (%) exceeding 2.2 μg/g Hair THg	Geometric mean (95% CI)	Arithmetic mean (95% CI)	Median (IQR)	Kruskal-Wallis H test
Urban							
Rural	482 (66.67)	35 (76.09)	188 (39.00)	1.64 (1.51–1.77)	2.35 (2.16–2.54)	1.69 (0.98–3.17)	p<0.0224
Urban	241 (33.33)	11 (23.91)	81 (33.61)	1.37 (1.21–1.55)	2.00 (1.77–2.25)	1.49 (0.76–2.73)	
Mining-affected Area							
No	281 (39.13)	18 (39.13)	91 (32.38)	1.37 (1.24–1.52)	1.93 (1.72–2.14)	1.40 (0.80–2.61)	p<0.0015
Yes	442 (60.87)	28 (60.83)	178 (40.27)	1.66 (1.52–1.82)	2.43 (2.23–2.64)	1.74 (1.01–3.39)	
Community Location relative to Puerto Maldonado (PEM)							

C. Community Characteristics

Community Location	Participants n(%)	Communities n(%)	N (%) exceeding 2.2 µg/g Hair THg	Geometric mean (95% CI)	Arithmetic mean (95% CI)	Median (IQR)	Kruskal-Wallis H test
>150 km west of PEM	78 (10.79)	6 (13)	18 (23.08)	1.22 (1.03–1.44)	1.55 (1.32–1.78)	1.27 (0.77–2.04)	p<0.0002
51–150 km west of PEM	123 (17.01)	8 (17.39)	50 (40.65)	1.61 (1.31–1.97)	2.45 (2.08–2.84)	1.71 (1.01–3.53)	
≥0–50 km west of PEM	158 (21.85)	11 (23.91)	80 (50.63)	1.94 (1.67–2.24)	2.82 (2.44–3.21)	2.21 (1.04–2.78)	
>0–50 km north of PEM	156 (21.58)	9 (19.57)	54 (34.62)	1.45 (1.24–1.69)	2.19 (1.84–2.54)	1.57 (0.78–2.80)	
51–150 km north PEM	119 (16.46)	5 (10.87)	29 (24.37)	1.35 (1.20–1.53)	1.71 (1.47–1.95)	1.25 (0.86–2.15)	
>150 km north of PEM	89 (12.31)	7 (15.22)	38 (42.70)	1.58 (1.31–1.92)	2.27 (1.86–2.68)	1.66 (0.89–3.10)	

[a] Compared to households not engaged in the economic activity

[b] households may have more than one economic activity.

Table 2.

Predicted Probabilities of exceeding 2.2 μg/g hair THg in areas affected and not affected by ASGM activities, IMAS study 2014

	ASGM-affected areas	**Non ASGM-affected areas**
	mean (sd) by group	
All Participants (n=723)	0.391 (0.280)	0.301 (0.25)
Children <15 years, fish index tertile 1	0.063 (0.041)	0.125 (0.147)
Children <15 years, fish index tertile 2	0.168 (0.157)	0.145 (0.171)
Children <15 years, fish index tertile 3	0.392 (0.209)	0.099 (0.120)
Adults ≥ 15 years, fish index tertile 1	0.269 (0.164)	0.321 (0.249)
Adults ≥ 15 years, fish index tertile 2	0.400 (0.189)	0.407 (0.260)
Adults ≥ 15 years, fish index tertile 3	0.730 (0.190)	0.412 (0.230)