Co-production of contaminated landscapes: anthropogenic loading and food web structure drive mercury bioaccumulation in abandoned gold mines

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Title Page

Co-production of contaminated landscapes: anthropogenic loading and food web structure drive mercury bioaccumulation in abandoned gold mines

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Abstract

Artisanal and small-scale mining is a significant and growing livelihood across the global South, which all too often leaves a legacy of contaminated landscapes. Given the increasing reliance of economies on metals and minerals, it is critical to understand what controls contamination outcomes in this rapidly developing extractive practice. Here, we demonstrate that the emerging concept of co-production offers a novel way to elucidate the joint contributions of natural and societal factors in shaping contaminant exposure from artisanal and small-scale mining. Specifically, understanding the co-production of contaminated landscapes requires attention to both the political economy of mining, including how labor and extraction methods differ across mines, as well as the sources and pathways of mercury exposure. In Madre de Dios, Peru, we measured mercury levels in wildlife inhabiting abandoned gold mining sites worked with different extraction technologies. We found that the type of technology used, whether heavy machinery or suction-pump based, influenced mercury loading into mines, and together with differences in food-web structure, mediated mercury biomagnification rates. Mercury concentration increased 2.1 to 3.7-fold per trophic level, and bioaccumulation levels were high in both mined and unmined sites—indicating elevated background levels in the region. We also found evidence of lateral transfer of mercury from abandoned mining pits to terrestrial food webs. This observation indicates that the footprint of mercury contamination extends well beyond individual mines, affecting the larger landscape. Our findings underscore the necessity of understanding the entangled ways in which social and ecological factors contribute to the production of toxic landscapes.

Keywords

Artisanal and small-scale gold mining, bioaccumulation, biomagnification, food webs, co-production

Main Text

Introduction

Artisanal and small-scale mining (ASM) is a complex, entangled social-natural system. Across the global South, ASM currently underpins the well-being of over 200 million people (1, 2), many of whom work under dangerous conditions to extract precious metals, gems, and other non-renewables (3). However, like most mining, ASM leaves a legacy of contaminated landscapes and sick people (4). Mining often causes the degradation of land, rendering it unusable for subsistence activities; all the while toxic elements used in extraction can cause serious health problems for individuals that consume contaminated food (5-9). We contend that understanding and addressing the environmental and social problems caused by ASM requires acknowledging that toxicity is co-produced by social and natural processes. This approach needs to integrate environmental sources and pathways of contaminant exposure, differences in labor relations and technologies used in ASM, and the ties between global resource politics and local resource extraction.

The case of artisanal and small-scale gold mining (AGSM) is particularly noteworthy because of the pronounced social and ecological changes brought on by the rapid expansion of ASGM throughout the global South. ASGM is a form of gold extraction characterized by the use of simple technologies such as repurposed car motors and suction pumps. ASGM largely occurs outside of formal economies and represents the principal livelihood for approximately 40 million workers and 150 million indirect beneficiaries—supplying more than 20% of global annual gold production (1, 2). While the livelihood benefits of ASGM are extensive, especially in underdeveloped rural communities (10), we still have a limited understanding of its full range of social and ecological impacts.

Miners commonly use mercury to extract gold (11, 12). Processing gold releases inorganic mercury into the atmosphere, which can be redeposited in nearby waterbodies. In the bottom sediments of these waterbodies, under certain abiotic conditions, inorganic mercury can undergo a microbially-mediated transformation into methylmercury, the most toxic and biologically available form of mercury (13-15). Many studies have documented bioaccumulation of mercury in the vicinity of active mines (16-19), in sediments and tailings downstream of mining (20-22), and even legacy contamination from mines that have long been abandoned (23, 24). Bioaccumulation of mercury can cause a wide range of detrimental impacts to wildlife such as a reduction in growth (25-27), juvenile survivorship (28), reproductive success (29, 30), and even mortality (31). Contaminant exposure extends beyond wildlife, as humans are also exposed to mercury through the consumption of fish and other top predators (6). Because ASGM is concentrated in culturally and biologically diverse areas of the globe (7), currently occurring in more than 80 countries in the global South (1,2), understanding risk of contaminant exposure in these unique ecosystems is critical.

The emerging concept of co-production (32, 33) represents a novel way to conceptualize the joint contributions of natural and societal factors in determining the risk from exposure to contaminants from ASGM. Co-production of ecosystem services (or disservices) is the process by which societies leverage labor, artefacts, and technology to use material and non-material flows of nature to produce goods and services that benefit (or harm) humanity (34). For example, hydropower is an ecosystem service co-produced by using built infrastructure (i.e., dams) to harness river flow regimes - but dam-induced habitat fragmentation and altered flows represent disservices that exact a high environmental cost (35, 36). Another example of co-production is gold mining, wherein humans transform the natural wealth of geologic deposits into commodities by using extractive technologies to remove, process, and refine auriferous rock. Just as the

number and spatial arrangement of dams in a river network mediates the amount of harm done to salmonid fisheries due to habitat fragmentation, the intensity and scale of mining impact the conditions of a landscape and its ability to support bacterial production of toxic methylmercury. Such place-specific nuances of ASGM underscore the need to improve our mechanistic understanding of how co-production of contaminated landscapes is mediated by differences in the political economy of gold production *and* the local ecological context.

The case of the bioaccumulation and biomagnification of mercury in wildlife inhabiting abandoned gold mines in Madre de Dios, Peru illustrates the potential of using a co-production approach. In Madre de Dios, a diversity of gold extraction methods co-exist with differing levels of mechanization ranging from non-mechanized artisanal operations to heavily mechanized operations using front loaders and excavators. Mechanization drives variation in the degree of environmental impact (37) and in daily production volume which is often directly correlated with mercury usage. A legacy of the increasing mechanization of ASGM is the creation of networks of abandoned mining pits that cover approximately 28% of the land area deforested due to mining, or an area equivalent to ~20,000 ha (38). Over time, these ponds are colonized by invertebrates, fish, and other wildlife forming new networks of aquatic habitat. Many of these abandoned mines also become sources of wild fish for people inhabiting nearby areas. However, these ponds are also potential hotspots where inorganic mercury can be transformed into methylmercury, and bioaccumulate in wildlife (24, 39). In Madre de Dios, there is growing evidence that mercury contamination from ASGM extends beyond the boundaries of mining areas as high levels of total inorganic mercury (THg) have been documented in downstream river sediments (21, 22), fish (16), indigenous populations upstream of mining (40), and in wildlife found far from mining areas (19).

Here we examined how co-production of contaminated landscapes mediated exposure risk to wildlife in three distinct ways. First, we evaluated whether mercury bioaccumulation in wildlife inhabiting abandoned gold mines differed when compared to mercury concentrations in wildlife found in unmined sites. We found that bioaccumulation was high in all sites, even unmined sites, but that bioaccumulation in wildlife was highest in those sites where ASGM occurred.

Second, we evaluated whether extraction technologies influenced mercury biomagnification rates by comparing natural lakes in an unmined watershed to areas worked with heavy machinery (HM) and those worked with suction-pump based techniques (SP). We estimated the trophic magnification slope (TMS) by regressing THg concentrations against stable nitrogen isotopes ($\delta 15N$) of multiple consumers common to all mining pits. Our estimated TMS of 0.46 ± 0.03 was on the high end of reported values for tropical freshwater, lentic environments. Further, we found that the type of technology used in gold production influenced the degree of mercury loading (as measured by sediment THg concentration), and together with differences in food-web structure across abandoned mines, these factors controlled the rate of mercury biomagnification at each site.

Finally, we determined whether mining pits subsidize mercury for terrestrial ecosystems via export of contaminated prey that are then consumed by riparian predators. We collected a riparian predator common to all sites, long-jawed orb-weaving spiders (Family: Tetragnathidae) and compared THg concentration and trophic position of these spiders to those of aquatic consumers. We confirmed that cross-ecosystem subsidies of aquatic prey transfer not only energy but also contaminants to riparian and terrestrial food webs. This result confirms that the footprint of mercury contamination extends beyond boundaries of individual mining pits, further exposing humans and wildlife.

Results

We found high levels of mercury bioaccumulation and biomagnification in taxa sampled across all mined and unmined sites (Fig. 2b, and *SI Appendix*, Table S1). However, the highest mercury concentrations were found in wildlife collected in abandoned gold mines as compared to unmined sites (Table S1). Taxonomic groups showed relatively consistent trophic positions across sites (as measured by δ15N), but spanned >2 trophic levels from snails (caenogastropoda), lowest in the food chain, to piranha (*Serrasalmus spp.*) and wolf fish (*Hoplias malabaricus*), apical predators (Fig. 2a). Accordingly, predatory fishes contained the highest average concentrations of THg (Fig. 2b). We also found strong evidence of biomagnification. Total inorganic mercury concentrations (THg) were significantly and positively correlated with estimated trophic position (δ15N) across all sites ($R^2 = 0.61$, $p < 0.0001$). The rate of mercury biomagnification represented by the average trophic magnification slope (TMS) calculated from the linear regression of $\log_{10}$-transformed total mercury concentrations against trophic position (δ15N ‰) was 0.46 ± 0.03 ($p < 0.0001$, n = 115, Fig. 3). Back-transformed, this TMS slope is equivalent to a 2.7 to 3.1-fold increase in mercury concentration with each trophic transfer. Given that TMS > 0, these data indicate that THg is being biomagnified through aquatic food chains in mining pits and unmined, natural oxbow lakes.

Beyond differences in bioaccumulation and biomagnification between unmined and mined sites, we found evidence of co-production of contamination: THg loading was influenced by both social and natural factors (Table 1). To fully parse the relative importance of trophic position, THg loading in sediments, and the effect of extraction technology on bioaccumulation of mercury in biota, we compared fits of different explanatory model structures. Specifically, we compared support across four linear mixed-effect models using an information-theoretic approach via BIC (Table 1). We found that including an interaction effect between organism trophic

position and extraction technology (i.e., whether the site was worked with HM, SP, or was an unmined site) greatly improved model fit over a restricted model that included the effects of technology and trophic position separately. We also evaluated the two best fit models using log-likelihood tests (Table 1, models 3 & 4). We found that model 4, which included the interaction effect as well as the fixed effect of mercury loading (THgSed), was significantly different from model 3 (χ^2 (1) = 5.806, p = 0.016). This finding suggests that mercury loading, influenced by the type of extraction technology used, drives patterns in mercury bioaccumulation across a diversity of taxonomic groups.

In addition, we found that sediment total mercury concentration (THgSed), a proxy for mercury loading to each mining pit or reference lake system, varied across sites worked with different technologies (Fig. 3, *SI Appendix*, Fig. S1) with a significant difference in mean total mercury concentration across site types ($F_{2,36}$ = 3.438, p = 0.043). The highest levels of loadings were recorded in pits worked by suction pumps, followed by pits worked by HM, and then unmined oxbow lakes. A post-hoc Tukey HSD analysis indicates that sites worked with suction pump machinery were significantly different in THgSed concentrations than those in unmined sites. There was no difference in THgSed between unmined sites and heavy machinery sites. However, we found that one of the oxbow lakes in our control sites was elevated in THgSed possibly due to the fact that this lake is a palm swamp with low concentrations of dissolved oxygen (*SI Appendix*, Table S2). If this lake were excluded from the analysis, then the mean THgSed would be significantly different between all paired comparisons of sites.

Finally, we found evidence of lateral transfer of mercury from abandoned mining pits to terrestrial, riparian consumers (Fig. 4). Stable isotopes of δ15N as well as field observations of feeding behavior, confirm that a riparian predator, long-jawed orb weaving spiders, consume

aquatic prey, acting as recipients of mercury subsidies from abandoned mining pits. These riparian consumers bioaccumulated mercury at concentrations that fell within those predicted by the relationship between THg concentrations and trophic position of our aquatic taxa.

Discussion

Co-production drives mercury accumulation and biomagnification in AGSM sites

We found strong novel evidence that mercury contamination in Madre de Dios, Peru, is co-produced by social and natural processes. Both mercury bioaccumulation levels and biomagnification rates were driven by variation in mining practices and differences in food-web structure. Participant observation and interviews with miners by JDL confirm that different methods of gold extraction influenced the degree of mercury loading into mining sites. Where heavy machinery-based mining (HM) occurs, there is never direct amalgamation in mining pits and thus little or no input of elemental mercury from tailings [Diaz Leiva, 2020, *in prep*]. Instead, these HM pits are constructed as water storage ponds and are filled-in by groundwater infiltration. Water is pumped out of the ponds to constructed sluice boxes sometimes more than 200 meters away to wash auriferous material that is brought from elsewhere on-site. Therefore, the method of gold extraction, whether using HM or SP technologies plays an important role in directly mediating the quantity and location of mercury discharges on the landscape. Our finding adds evidence to the argument that local or regional-level differences in ASGM practices may affect mercury loading into aquatic ecosystems (41).

Second, we found that the trophic position of taxa and average food chain length (FCL; as estimated by subtracting trophic position of the top consumer from the primary consumers), which is known to affect biomagnification rates, varied by sites worked with different extraction technologies. In pits worked with heavy machinery, the average FCL was 2.70, for SP pits it was

200 2.30, and for reference lakes it was 2.05. While we do not know the mechanism driving
201 differences in FCL across sites worked with different technologies, we postulate that differences
202 in feeding behavior of the same organism across sites (i.e., omnivory in predatory fish) or food
203 web complexity (i.e., addition or insertion of top consumers lengthening the food chain) may be
204 responsible for this variation (42). Understanding the exact mechanism is particularly important
205 given that wildlife in these landlocked ponds will readily bioaccumulate contaminants from
206 dietary exposure and differences in FCL will mediate the accumulation of contaminants at the top
207 of the food chain. However, trophic position alone did not explain variation in bioaccumulation of
208 THg in wildlife across sites, as taxa with the same trophic position (e.g., *Hypostomus spp.*,
209 *Serrasalmus spp*.) were consistently higher in THg concentration in SP pits relative to HM pits.
210 Only when we took the full suite of co-production factors - trophic position, extraction
211 technology, and mercury loading - into account did our model (Table 1) best explain the patterns
212 in bioaccumulation and biomagnification we observed across sites.
213
214 **Diffuse Mercury Contamination in Madre de Dios**
215 We found extremely high mercury biomagnification rates in our study sites. Our
216 estimated TMS (0.46 ± 0.03) is more than three times that of the mean global value for freshwater
217 tropical sites, 0.12 ± 0.12 (43), and at the high end for studies restricted to Amazonia, which
218 ranged from 0.21 – 0.43 (44 - 49). These magnification rates have led to levels of top-predator
219 mercury bioaccumulation that represent a significant public and environmental health risk.
220 Predatory fish species (*H. malabaricus* and *Serrasalmus spp.*) caught in both unmined natural
221 lakes and abandoned mines contained concentrations of mercury that exceeded international
222 consumption limits by two to five times on average. Our findings are consistent with previous
223 measurements of bioaccumulation in *H. malabaricus* in oxbow lakes in the Bolivian Amazon
224 (46). The highest levels of bioaccumulation were recorded in predatory fish caught in abandoned

225 mines worked with suction-pump based technologies. The concentration of THg in two piranha
226 (*Serrasalmus spp.*) was 26.7 and 26.1 mg/kg respectively, more than 50 times the European
227 Union's recommended consumption limit of 0.5 mg/kg. These values are similar to those reported
228 in highly contaminated spill sites (50).
229
230 Perhaps most concerning, we found consistently high concentrations of THg in higher
231 trophic-level organisms—irrespective of presumed differences in mercury loading across
232 abandoned mining pits and unimpacted oxbow lakes. This result suggests that the
233 bioaccumulation potential of organisms inhabiting lentic environments in this region is high.
234 Importantly, our reference sites were located in a densely forested, protected watershed far from
235 direct elemental mercury inputs. Future research should focus on understanding the sources of
236 this mercury and quantifying the methylation potential of ponds and oxbow lakes to parse apart
237 why there are such elevated background levels in Madre de Dios. Researchers should leverage
238 environmental tracers such as mercury stable isotopes to determine whether these high
239 background levels bear the signature of mercury used in ASGM or whether these high levels are
240 due to the release of mercury from natural sources such as soil erosion (51). This question
241 remains unresolved in the literature as some studies have found that the signature of mercury in
242 sediments collected downstream of ASGM does not belong to mercury used in these operations
243 but is instead from mercury released from soils and trees due to deforestation from mining and
244 other land uses (41-43).
245
246 **Cross-Ecosystem Transfer of Mercury Extends the Footprint of Contamination**
247 Beyond high background levels, our findings indicate that networks of abandoned gold
248 mining pits may act as hotspots of biomagnification in landscapes impacted by informal gold
249 mining. Given that long jawed orb weaving spiders are consumed by mobile predators such as

bats and are not the only terrestrial consumers of emergent aquatic insects, we expect this contaminant subsidy to propagate through food webs far beyond the riparian zone of abandoned mining pits. Previous studies in the Amazon have found elevated levels of mercury in alluvial sediments downstream of mining sites and soils taken from areas proximate to active mining, but this study is the first to demonstrate that abandoned mining pits are an important source of mercury to the terrestrial biota inhabiting these highly impacted systems. More work is needed to fully understand the risk of contaminant exposure to higher trophic levels organisms. Previous work suggests that the degree to which these mining pits export mercury via emergent aquatic insects depends on the life history of these insects, as metamorphosis of aquatic larvae into adults reduces heavy metal burden in aquatic predators such as dragonflies and consequent accumulation in cross-system predators (52). In addition, body size of emergent aquatic insects, mediated by top-down interactions, can also alter contaminant flux into terrestrial ecosystems (53). In addition, these mosaics of abandoned mining pits form new habitat for migratory birds and other higher trophic levels organisms whose mobility allows them to further extend the footprint of mercury contamination from ASGM.

While these pits can act as sources of methylmercury to wildlife, it should be noted that these results also indicate that there is high heterogeneity in mercury bioaccumulation and biomagnification potential within abandoned gold mining landscapes driven largely by the differences in the political economy of ASGM. The diversity in mining operations drive variation in quantity and location of mercury discharges such that not everywhere in a mined landscape will there be evidence of high levels of mercury in biota. In turn, areas far removed from gold mining could have high levels of mercury in wildlife due to cross-ecosystem subsidies and mobility of consumers. Thus, sampling only one compartment such as the sediment of mining pits or the soils around abandoned mines can lead to inconclusive results, as differences in mercury

bioaccumulation higher up in the food chain are not apparent from the relatively small differences in mercury loading in sediment (on the order of 10 ng). Further, proposals to utilize these contaminated landscapes for production of fish through aquaculture or for farming should take into account the type of technology that was used in gold production (38). Sites worked with HM may be safer candidates for remediation for aquaculture than sites worked SP. Additionally, top consumers like predatory fish, fish-eating birds, and mammals should be closely monitored due to their susceptibility to accumulate mercury especially from sites where SP technologies were used.

Conclusion

Environmental contamination is not an isolated or unique phenomenon, but instead it is part of a ubiquitous pattern that implicates human's continued consumption of resources in the degradation of the natural world (54). Our globalized economy's reliance on non-renewable resources is unlikely to diminish in the near future, especially with the use of precious metals in climate-change mitigating technologies like solar panels and electric car batteries (55-57). To have any hope of mitigating the worst effects of contaminant exposure or remediating already contaminated landscapes, we must understand the entangled ways in which social and ecological factors contribute to the production of toxic landscapes. A co-production approach, that explicitly incorporates the social and natural, can help guide the kind of studies that are urgently needed to ensure that our planet continues to provide the life supporting services that we depend on while also ensuring just outcomes for those people who depend on resource extraction for their livelihoods.

Materials and Methods

Study Site & Field Sampling. We conducted our study in the Department of Madre de Dios, Peru, a region located at the western edge of the Amazon basin (Fig. 1). We sampled across four

sites, with each site characterized by the use of either suction-pump (SP) or heavy machinery (HM) extraction technologies as well as one unmined site. On the surface, the mining pits created by these two different mining technologies appear similar, however they differ in key aspects. Suction pump-based technologies create mining pits that are more heterogeneous in their bathymetric profile and on average much deeper than their counterparts created with heavy machinery. The volume of gold-bearing alluvium processed daily by these two types of production also differs. Daily production volume is approximately proportional to the level of mechanization and subsequently influences the amount of mercury used in amalgamation of gold at the end of a day's shift.

To test whether production practices mediate mercury biomagnification, we sampled multiple abandoned gold mining pits (≥2) at each site. A total of 10 mining pits were sampled including seven mining pits where SP technologies were used and three mining pits where HM was used. We were unable to access more sites worked with HM due to the rapidly changing security threats posed by a state of emergency declared in the region. At the unmined site, we assessed background biomagnification rates in four natural oxbow lakes located along a river in a protected watershed unimpacted by gold mining. Mining pits resembled artificial ponds and many contained floating and emergent macrophytes including *Paspalum repens, Hymenache spp*., as well as a diversity of fauna. Pits ranged in size from 0.3 ha to ~ 4 ha and varied in bathymetric profile based on the type of technology used in gold production. The average depth of pits worked with SP was 2.48 m and 1.75 m for pits worked with HM.

To estimate the rate of biomagnification across sites, we sampled for the same taxa across all pits and oxbow lakes including benthic macroinvertebrates and algivorous and piscivorous fish. Benthic macroinvertebrates common across all pits and oxbow lakes included predatory dragonflies (Families: Gomphidae, Libellulidae), giant water bugs (Family: Belostomatidae), and

water scorpions (Family: Nepidae). Primary consumer invertebrates (herbivorous/detritivorous strategies) included (Family: Caenogastropoda) and burrowing mayflies (Family: Polymitarcyidae). Benthic macroinvertebrates were live sorted to family level and then kept in water for 12-24 hours to clear gut contents. In the lab, the invertebrates were counted, measured and pooled by family to produce a composite sample for each pit. The invertebrates were kept frozen until they were freeze-dried at -56°C for 72 hours. Gastropods were used for estimation of baseline $\delta^{15}N$ following Post (2002, 58). In addition, a maximum of three individuals of the following fish species were collected at each mining pit or oxbow lake; piranha (*Serrasalmus spp*.), wolf fish (*Hoplias malabaricus*), and armored catfish (*Hypostomus spp*.). Fish were collected using a gill net deployed on the same day as invertebrate collection; length and weight were measured, and dorsal muscle tissues were removed, placed on ice until they could be frozen, and later freeze-dried.

To determine whether riparian consumers of emergent aquatic insects bioaccumulate mercury from aquatic resource subsidies, we collected long jawed orb-weaving spiders (Family: Tetragnathidae) from the vegetation at the margins of each pit and lake. Many studies have shown that in temperate zones, long-jawed orb-weaving spiders (Family: Tetragnathidae) and other riparian consumers feed on emergent insects that accumulate mercury and other contaminants during their aquatic larval phase (59-62). Hand collection of spiders occurred on the same day as collection of fish and benthic macroinvertebrates. Spiders were counted, frozen on ice and later freeze-dried at -56°C for 72 hours.

To estimate mercury loading in each pit or lake, surface sediment samples were collected in triplicate using an Ekman dredge and analyzed for total mercury content. Samples for water

350 chemistry were also collected *in situ* at each point where a sediment sample was taken using a
351 multiparameter Hanna HI91894. All sampling was conducted during the dry season from April-
352 August 2019.
353
354 Research protocol and methodologies for handling fish specimens were approved by UC
355 Berkeley's Animal Care and Use Committee (#AUP-2018-06-11147).
356
357 **Laboratory Analyses.** Laboratory processing and analysis followed similar procedures to Wyn et
358 al. (2014, 63). Individual fish muscle samples and invertebrate samples (pooled with greater than
359 5 individuals per taxon) were freeze-dried, homogenized, and subsampled once for each analysis.
360 Approximately 20 mg (±10 mg) of homogenized fish muscle or whole macroinvertebrate
361 composite was analyzed on a Milestone DMA-80 direct mercury analyzer (Milestone Inc., Shelton,
362 CT, USA) at the Carnegie Institute for Global Ecology at Stanford University (Stanford, California,
363 USA). Samples of certified reference material (DORM4 or TILL3) were within acceptable recovery
364 limits. All Hg data are expressed on a dry weight basis. Duplicate analytic measurements were
365 analyzed for each fish, invertebrate, and sediment sample analyzed for THg, and the two results
366 were averaged for the reported result. The average relative percent difference in THg measurements
367 of duplicate samples was 1.1%. Quality control measures included testing of standard reference
368 materials and internal laboratory standards. Continuing calibration verification and continuing
369 calibration blank measurements were determined on every tenth sample analyzed in accordance
370 with US EPA Method 7473 (64). The Method Detection Limit for total Hg analysis is 2 ng/g. No
371 fish, invertebrate, or sediment samples were rejected based on quality control results or duplicate
372 relative percent differences. Additional information on QA/QC of samples can be found in
373 supplemental materials (*SI Appendix).*
374

375 Samples were analyzed for $\delta^{15}N$ and $\delta^{13}C$ at the Center for Stable Isotope Biogeochemistry at the
376 University of California, Berkeley (Berkeley, California, USA). Stable isotope composition is
377 expressed in parts per thousand as a deviation from a standard reference material. Nitrogen
378 isotopic values were standardized against N_2 gas in air as follows:

379 $$\delta 15N = \left(\left(R_{sample} \div R_{standard}\right) - 1\right) * 1000 \quad (1)$$

380 Stable nitrogen isotopes were used to approximate the relative trophic position of an organism
381 because the relative abundance of ^{15}N in proportion to ^{14}N in a consumer is enriched by an
382 estimated 3-4‰ per trophic level (58, 65-69). The trophic level of an organism can then be
383 adjusted by measuring $\delta^{15}N$ in a primary consumer such as algivorous freshwater snails and
384 correcting $\delta^{15}N$ of higher-level consumers as in the following equation:

385

386 $$TL_{consumer} = \frac{\delta 15N_{consumer} - \delta 15_{primary\ consumer}}{\Delta 15N} + \lambda \quad (2)$$

387

388 Where $\Delta^{15}N$ is one of the most common enrichment factors used in studies of aquatic food webs,
389 3.4‰ (this however is not the only enrichment factor used in studies) and λ is the trophic level of
390 the baseline primary consumer (TL = 2). Additional information on QA/QC of samples can be
391 found in supplemental materials (*SI Appendix).*

392

393 **Data Analyses.** To test for differences in the biomagnification rate across sites worked with
394 different extraction technologies, we used tests of analysis of covariance (ANCOVA; R Version
395 3.6.1) (70). Trophic position (continuous independent variable) and technology (categorical
396 independent variable) on total mercury concentration in biota (THg; continuous dependent
397 variable). We tested for an interaction effect between technology and trophic position, which
398 would indicate that the slope estimate for the relationship between trophic position and mercury
399 concentration was dependent on the type of technology used. If the interaction effect was not
400 significant, we removed the term and assessed the main effects of trophic position and technology

independently. We also tested for differences in mercury loading across pits worked with the same technology using ANOVA and a post-hoc Tukey HSD test to evaluate whether there was a difference in sediment mercury loading across sites. Finally, we used a generalized linear mixed effects model, lme4 (lmer package, 71) to evaluate the proportion of variance in total mercury concentration of biota that was explained by a model that accounted for the random effect of pits nested within sites worked with different extraction technologies, as well as the fixed effects of mercury loading as estimated by the average pit or lake sediment total mercury concentration (THgSed), trophic position, and the interaction between trophic position and extraction technology. We compared our models using Bayesian information criterion (BIC) scores and chi-square log-likelihood ratio tests.

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Author Contributions

J.D.L., A.R., and M.D.P designed research; J.D.L performed research; J.D.L analyzed data; and J.D.L, A.R., and M.D.P wrote the paper.

Figures and Tables

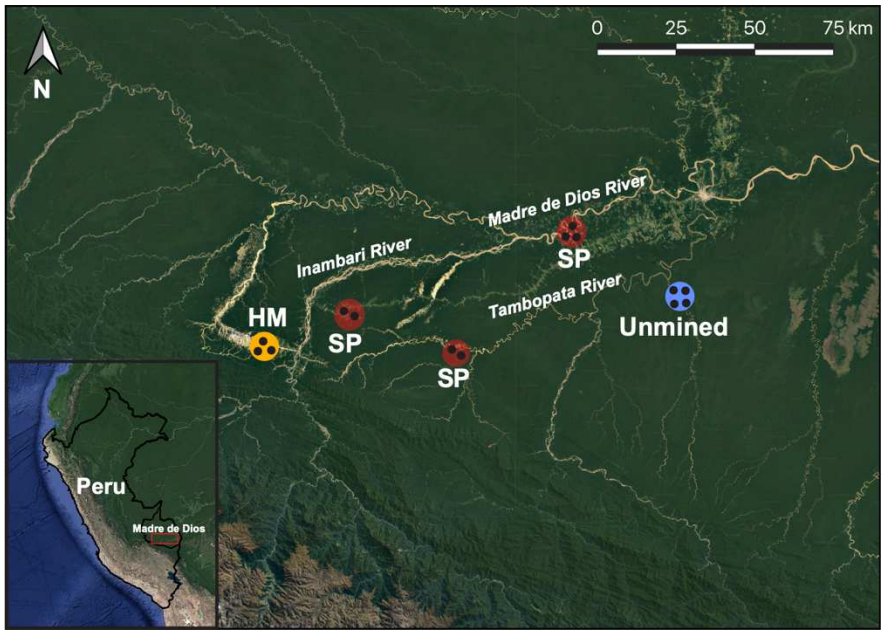


Figure 1. Map of study region showing approximate sampling locations and number of mines sampled (black dots) by type of machinery used in gold production: suction-pump based (SP) or heavy machinery (HM), and one unmined site.

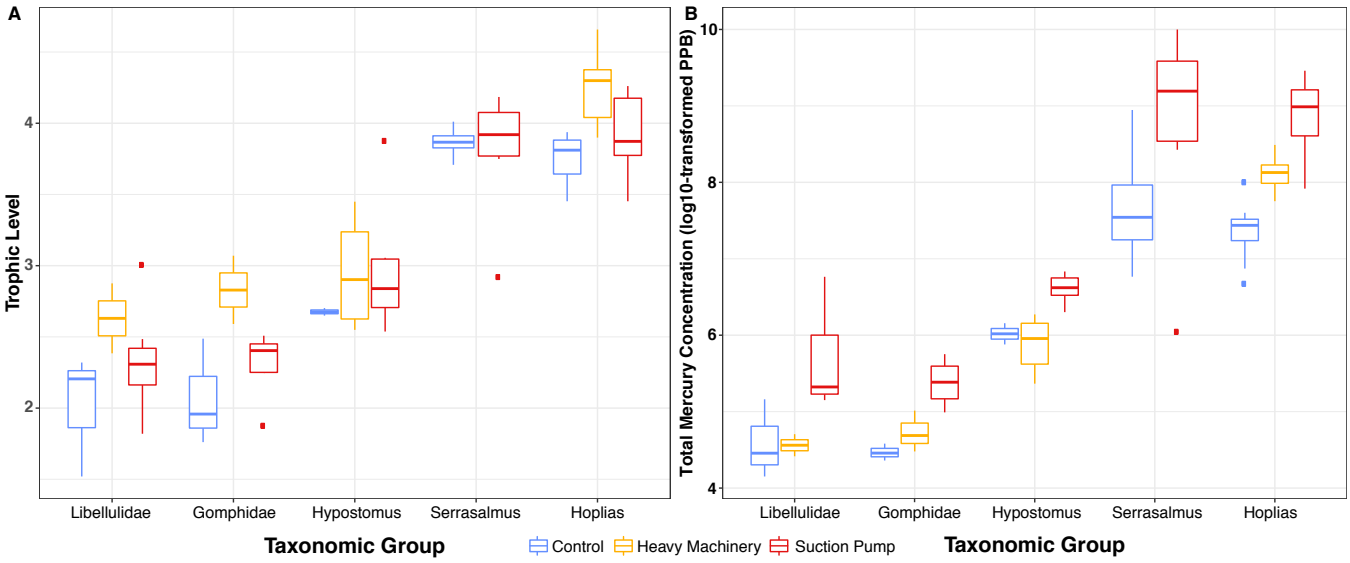


Figure 2. A. Differences in trophic level (calculated from δ15N, see methods for more details) of taxa varied consistently across sites worked with different extraction technologies and reference oxbow lakes. **B.** Bioaccumulation of total inorganic mercury across sites worked with different extraction technologies. Taxonomic groups organized by increasing trophic level.

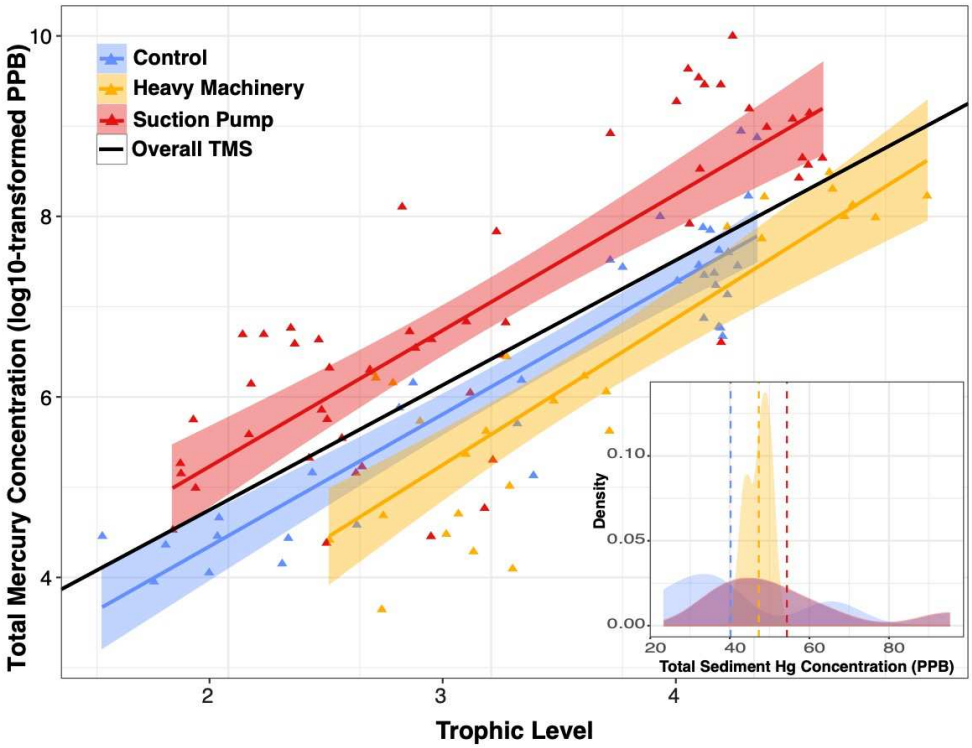


Figure 3. Trophic magnification slope (TMS) estimates overall (black line), and by type of extraction technology used (colored lines). Lines represent least squares estimate of the linear regression of total mercury concentration of biota against trophic level of taxa (calculated using δ15N estimates corrected using δ15N of a baseline consumer – see methods), with one standard error in slope estimate represented by shading. Inset – density plot of total sediment mercury concentration (PPB) by type of extraction technology. Dashed lines represent the mean total sediment mercury concentration across pits that differ in extraction technology.

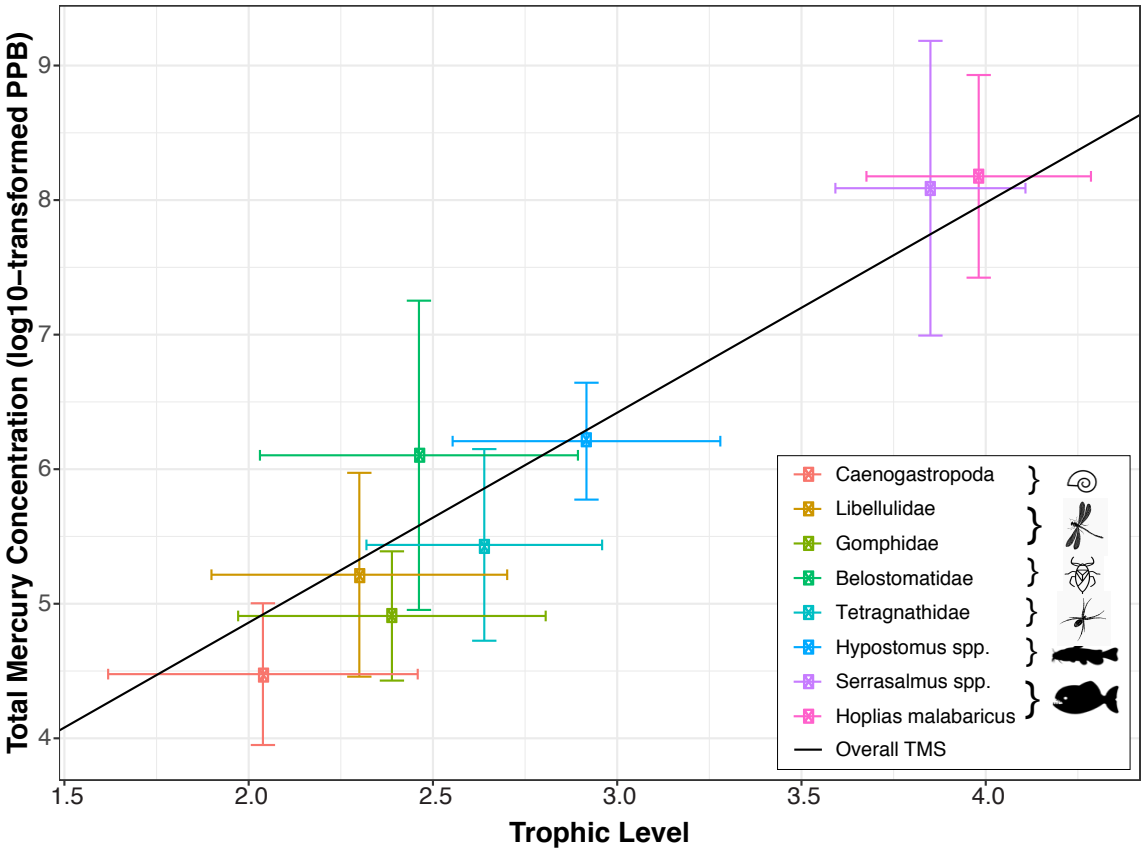
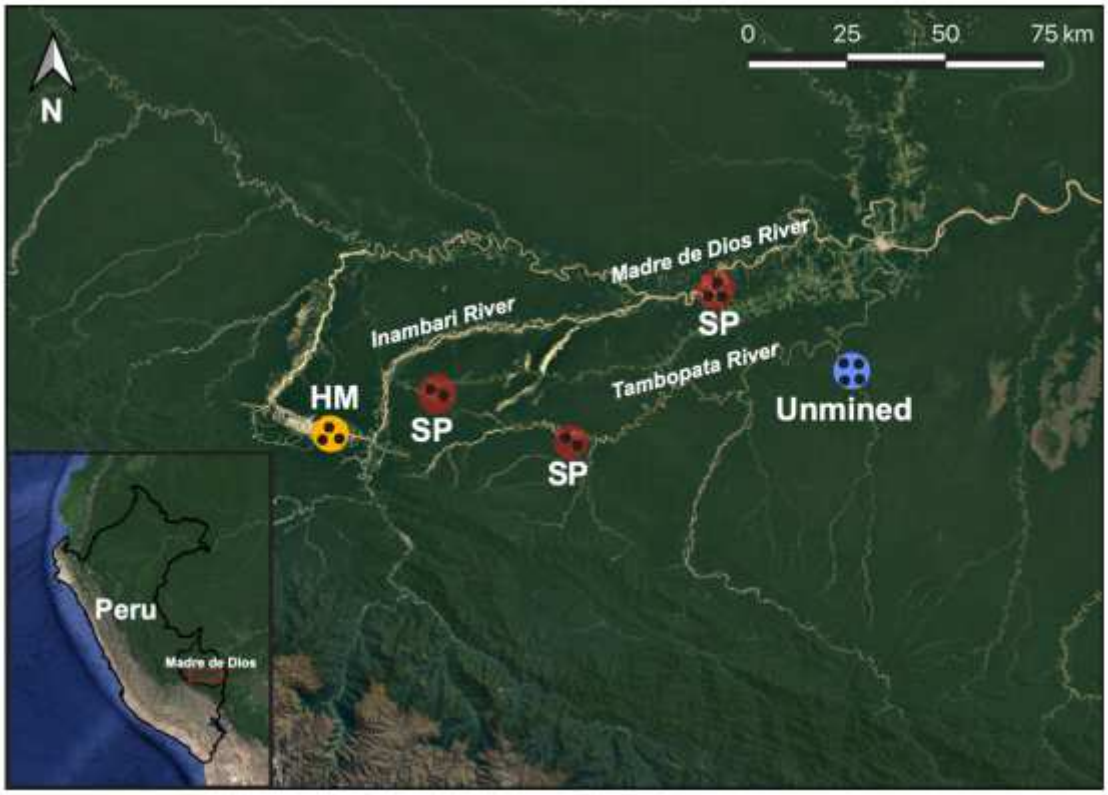


Figure 4. Mercury biomagnification by aquatic and terrestrial taxa sampled. Data points represent the mean trophic level and $\log_{10}$ transformed THg concentration $\pm$ 1 S.E. for each taxonomic group sampled across all sites. Black line represents the least squares estimate of the trophic magnification slope calculated from linear regression of total mercury concentration in biota against estimated trophic level of taxa (calculated from δ15N concentrations corrected by baseline consumer δ15N – see methods).

Table 1. Evaluation and comparison of linear mixed effects models: 1) restricted model including fixed effects accounting for taxonomic groups (Taxa), extraction technology (Tech), trophic position (Trophic Position), and a random effect of pit identity (1|Pit.ID), 2) model 1 plus a fixed effect of mercury loading (THgSed) in pits, 3) model 1 plus interaction between trophic position and extraction technology (1|Trophic Position:Tech), 4) model 3 plus the addition of mercury loading fixed effect.

No.	Model	Residual Deviance	Residual df	BIC	Log Likelihood
1	Taxa + Tech + Trophic Position + (1|Pit.ID)	473	15	559	-236
2	Taxa + Tech + Trophic Position + THgSed + (1|Pit.ID)	469	16	561	-234
3	Taxa + Tech + Trophic Position + (1|Pit.ID) + (1|Trophic Position:Tech)	-210	16	-119	105
4	Taxa + Tech + Trophic Position + THgSed + (1|Pit.ID) + (1|Trophic Position:Tech)	-216	17	-119	108

Figures



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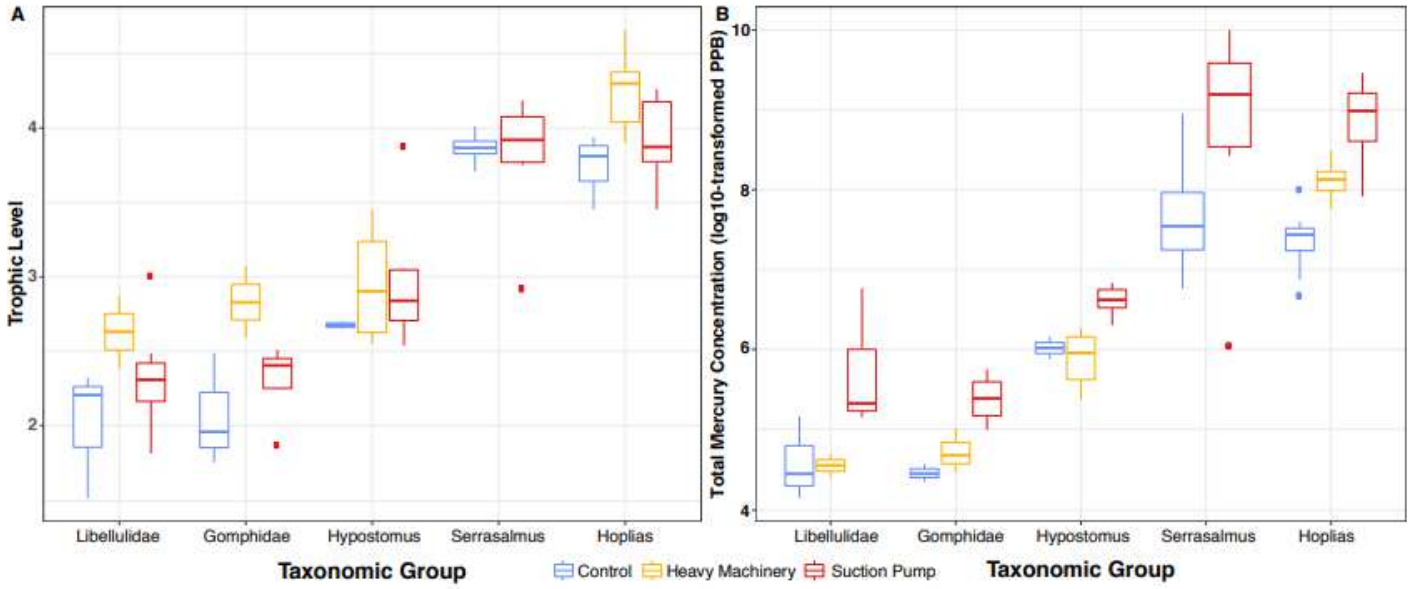

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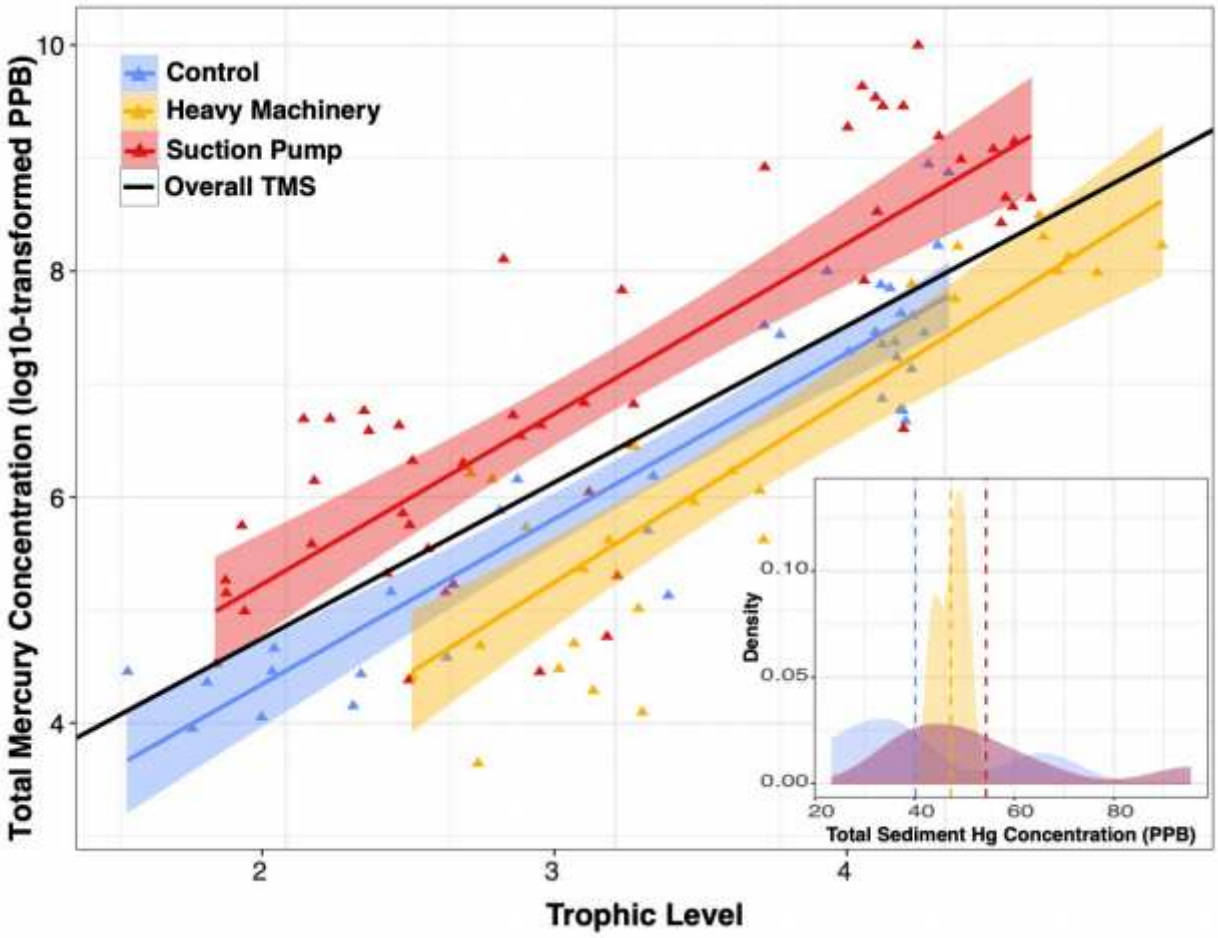

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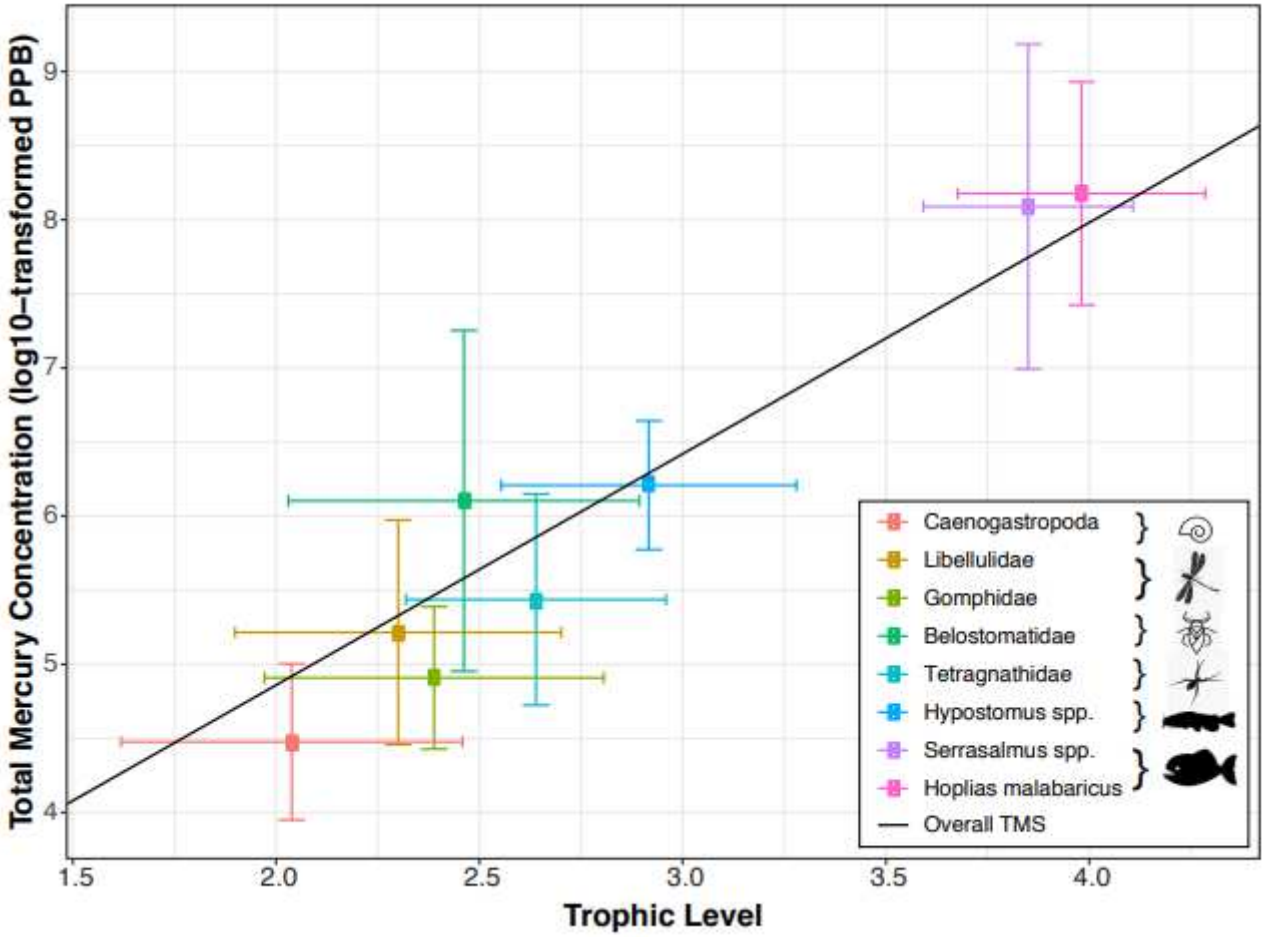

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Mercury biomagnification by aquatic and terrestrial taxa sampled. Data points represent the mean trophic level and log10 transformed THg concentration ± 1 S.E. for each taxonomic group sampled across all sites. Black line represents the least squares estimate of the trophic magnification slope calculated from linear regression of total mercury concentration in biota against estimated trophic level of taxa (calculated from δ15N concentrations corrected by baseline consumer δ15N – see methods).

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