Supporting Information

River Transport of Mercury from Artisanal and Small-Scale Gold Mining and Risks for Dietary Mercury Exposure in Madre de Dios, Peru

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Statistical Analyses

Table S1: Regression model of total suspended solids (TSS) in mg of solid per L and suspended particulate Hg (Hg_P) in ng Hg per Liter on river section within season 1 and season 2. Estimates on river section indicate regression slope and confidence interval in mg L^{-1} km^{-1}. Differences between section are represented in mg L^{-1} km^{-1}.

	Regression model of Hg_P in ng/L on river section				Regression model of TSS in mg/L on river section			
	Season 1 (n = 21)		Season 2 (n = 25)		Season 1 (n = 25)		Season 2 (n = 26)	
Covariate	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value
River Section 1 (km 1 to 180)	0.05 (0.02, 0.08)	**0.003**	0.02 (-0.04, 0.07)	0.481	0.04 (-1.35, 1.43)	0.955	0.67 (-0.33, 1.67)	0.188
River Section 2 (km 181 to 400)	0.04 (0.01, 0.06)	**0.002**	0.04 (0.00, 0.07)	**0.034**	0.06 (-1.00, 1.12)	0.906	0.43 (-0.20, 1.05)	0.180
River Section 3 (km 401 to 560)	-0.04 (-0.09, 0.01)	0.134	0.01 (-0.05, 0.08)	0.743	-1.62 (-3.60, 0.36)	0.109	-0.91 (-2.06, 0.24)	0.122
Difference between Sections 1 and 2	-0.01 (-0.06, 0.04)	0.686	0.02 (-0.06, 0.1)	0.656	0.02 (-2.22, 2.27)	0.983	-0.24 (-1.70, 1.22)	0.746
Difference between Sections 2 and 3	0.08 (0.01, 0.15)	**0.032**	0.03 (-0.07, 0.12)	0.564	1.68 (-1.14, 4.50)	0.242	1.34 (-0.29, 2.96)	0.108
Difference between Sections 1 and 3	-0.09 (-0.14, -0.04)	**<.001**	-0.01 (-0.08, 0.06)	0.815	-1.66 (-3.69, 0.37)	0.109	-1.58 (-2.90, -0.26)	**0.019**

Table S2: Estimated difference between sections based on regressions of Hg_P and TSS with river section as a categorical variable, adjusted for season. Type 3 p-value is a measure of the overall significance of the section categorical variable, whereas the individual p-values are pairwise.

	Particulate Mercury, Hg_P (ng/L) regressed on River Section, Adjusted for Season		Total Suspended Solids, TSS (mg/L) regressed on River Section, Adjusted for Season	
Level	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value
River Section 1 (km 1 to km 180)	reference		reference	
River Section 2 (km 181 to km 400)	9.94 (6.11, 13.76)	**<.001**	-5.95 (-114.84, 102.94)	0.915
River Section 3 (km 401 to km 560)	13.47 (9.24, 17.69)	**<.001**	107.64 (10.42, 204.85)	**0.030**
River Section 3 vs. River Section 2	3.53 (-0.20, 7.26)	0.063	-113.59 (-206.00, -21.18)	**0.016**
Type III p-value		**<0.001**		**0.026**

Table S3: Estimated difference between sections based on regressions of total mercury, total methylmercury, and % methylmercury of the total mercury in sediment with river section as a categorical variable. Type 3 p-value is a measure of the overall significance of the section categorical variable, whereas the individual p-values are pairwise.

	Total Mercury (mg/kg)		Total Methylmercury (dry weight, pg/g)		Percent Methylmercury (dry weight, %)	
Level	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value	Estimate (95% CI)	p-value
River Section 1 (km 1 to km 180)	reference		reference		reference	
River Section 2 (km 181 to km 400)	13.52 (4.40, 22.64)	**0.004**	254.85 (22.80, 486.90)	**0.031**	0.70 (-0.52, 1.91)	0.261
River Section 3 (km 401 to km 560)	21.94 (11.47, 32.41)	**<.001**	79.58 (-192.67, 351.83)	0.567	-0.10 (-1.47, 1.27)	0.881
River Section 3 vs. River Section 2	8.42 (-0.49, 17.33)	0.064	-175.27 (-388.84, 38.30)	0.108	-0.80 (-1.80, 0.20)	0.117
Type III p-value		**<0.001**		0.075		0.242

Table S4. General surface water quality averaged for all 47 sites along the MDD River for two sampling events: March/April 2013 (wet season) and June/July 2013 (dry season).

Parameter	Mar/Apr 2013	June/July 2013
pH	7.2 ± 0.3	7.6 ± 0.2
DO (%)	86.9 ± 7.0	93.5± 5.7
DO (mg/L)	6.95 ± 0.73	7.41 ± 0.48
Specific Conductivity (µS/cm)	82.9 ± 29.9	97.6 ± 36.6
Temperature (°C)	25.3 ± 19.9	25.7 ± 1.4

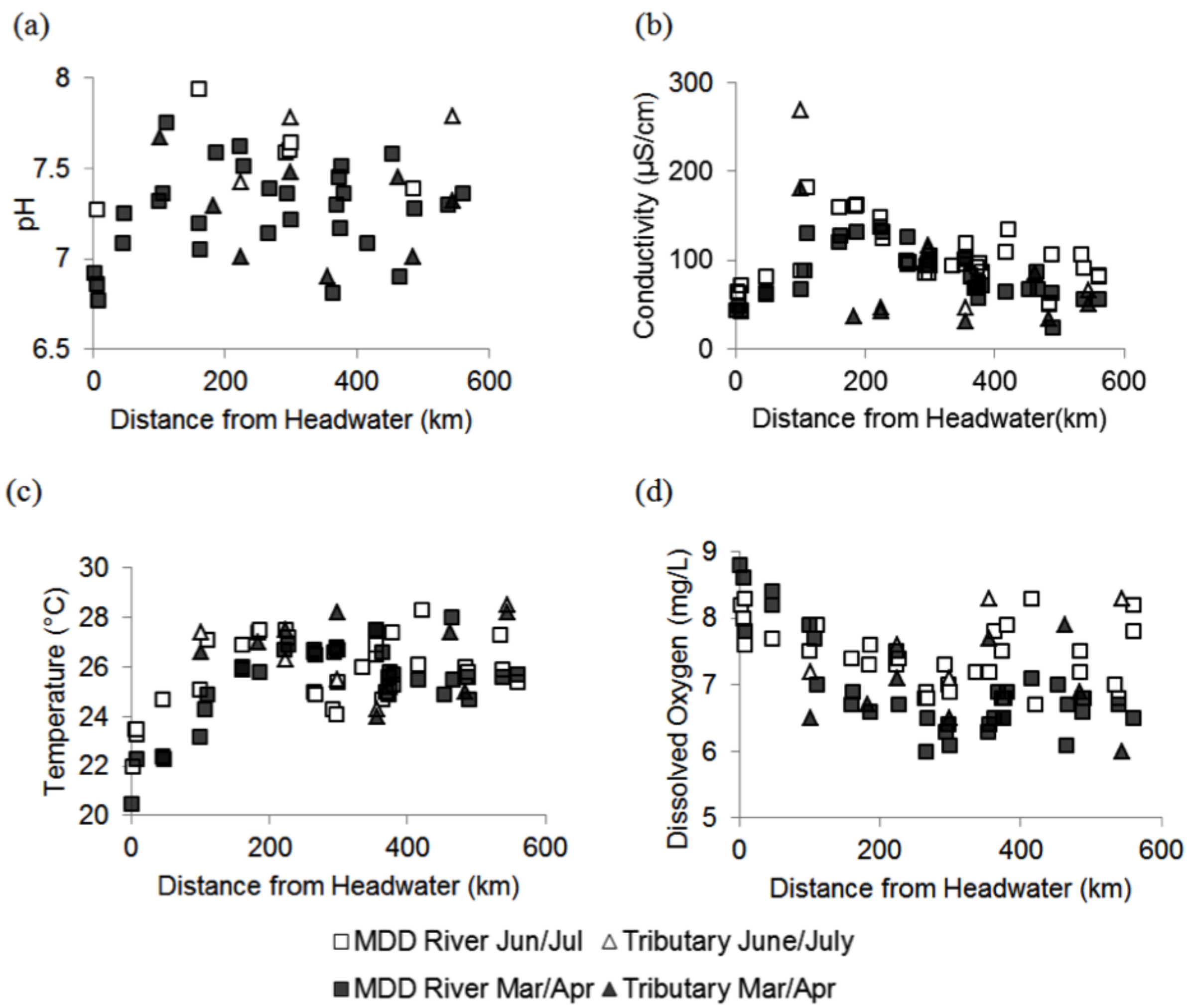

Figure S1. General surface water quality at sampling sites along MDD River and major tributaries for the wet season (March/April 2013) and dry season (June/July 2013). (a) pH; (b) Conductivity; (c) Temperature; and (d) Dissolved oxygen.

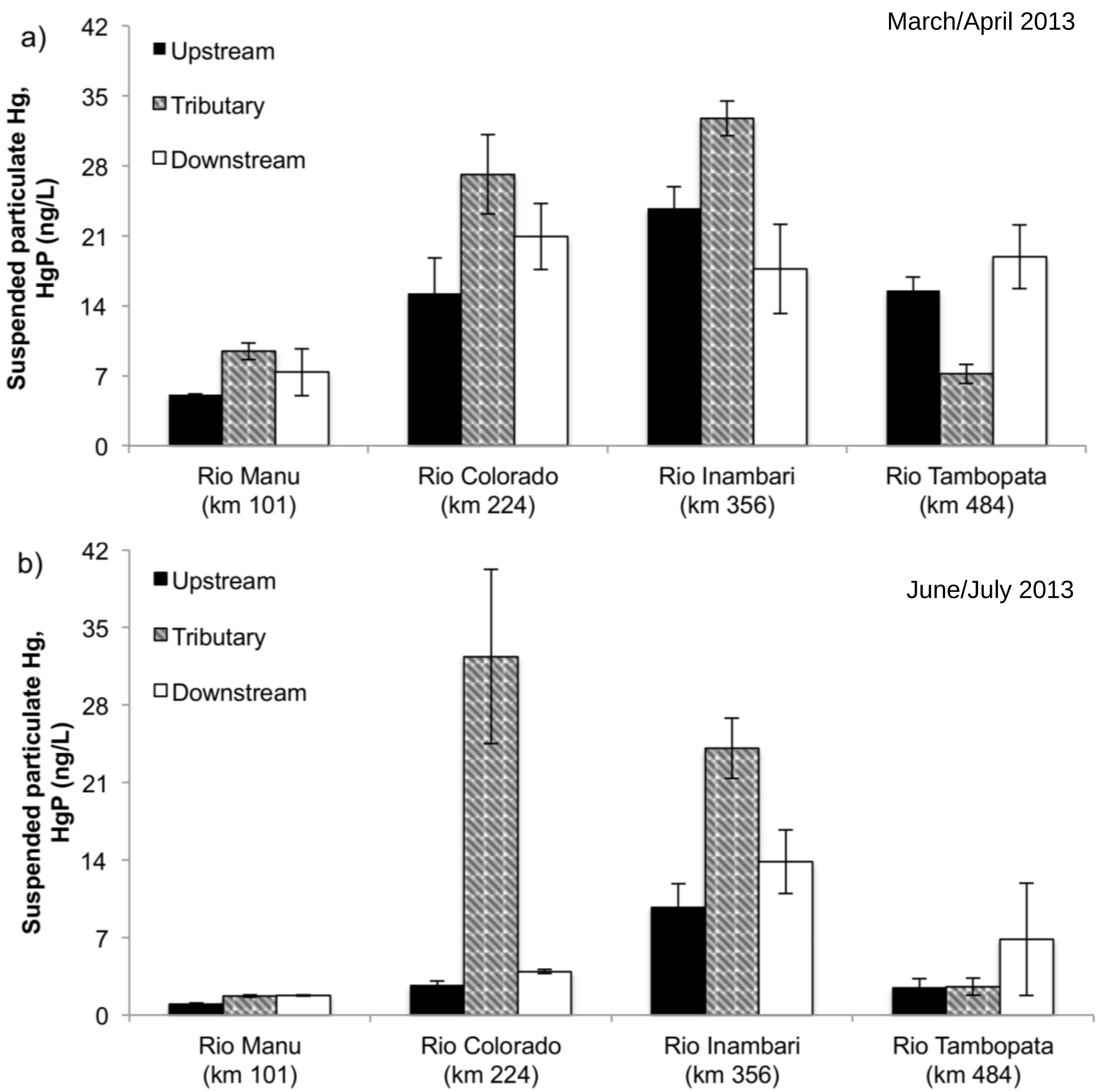

Figure S2. Suspended particulate mercury, Hg_P (ng/L), in surface water samples of the Madre de Dios River collected at sites within 40 km upstream and 40 km downstream of each tributary confluence and in surface water of the tributary upstream of the confluence. (a) March/April 2013 (wet season); (b) June/July 2013 (dry season). Error bars indicate standard deviation of duplicate or triplicate field samples.

(a)

(b)

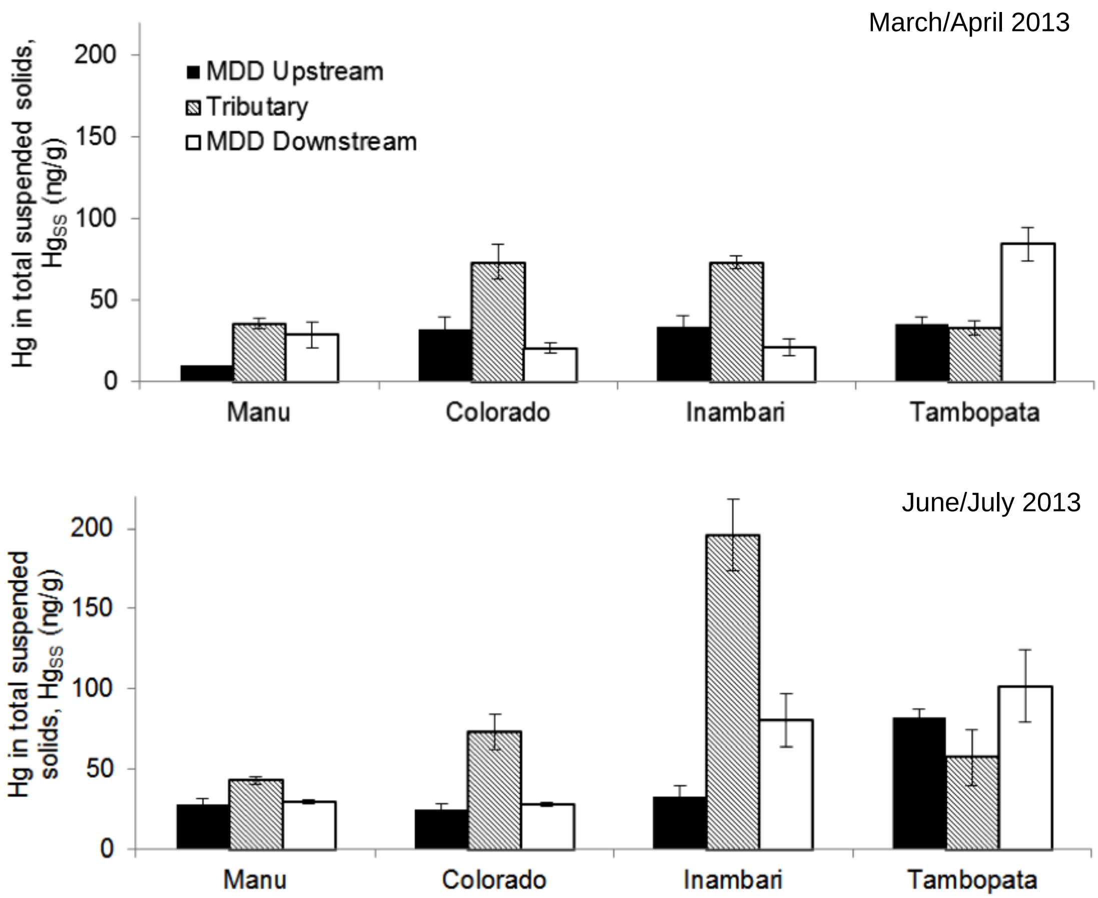

Figure S3. Concentrations of Hg in suspended solids (Hg_{SS}) in surface water samples of the Madre de Dios River collected up to 40 km upstream and 40 km downstream of each tributary confluence and in surface water of the tributary collected upstream of the confluence with the MDD River. (a) March/April 2013 (wet season); (b) June/July 2013 (dry season). Error bars indicate standard deviation of duplicate or triplicate field samples.

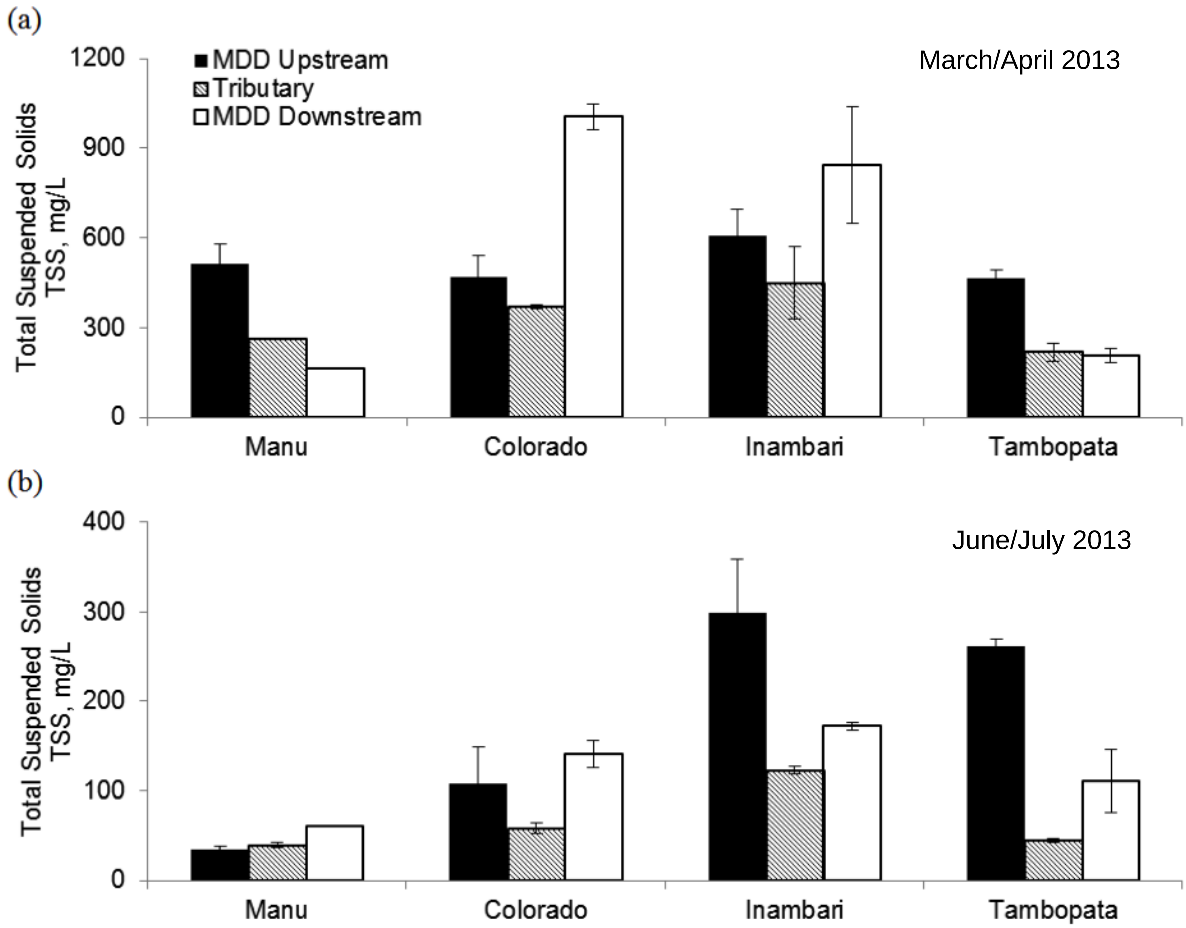

Figure S4. Total Suspended Solids (TSS) in surface water samples of the Madre de Dios River collected within 40 km upstream and 40 km downstream of each tributary confluence and in the surface water of the tributary upstream of the confluence. (a) March/April 2013 (wet season); (b) June/July 2013 (dry season). Error bars indicate standard deviation of duplicate or triplicate field samples.

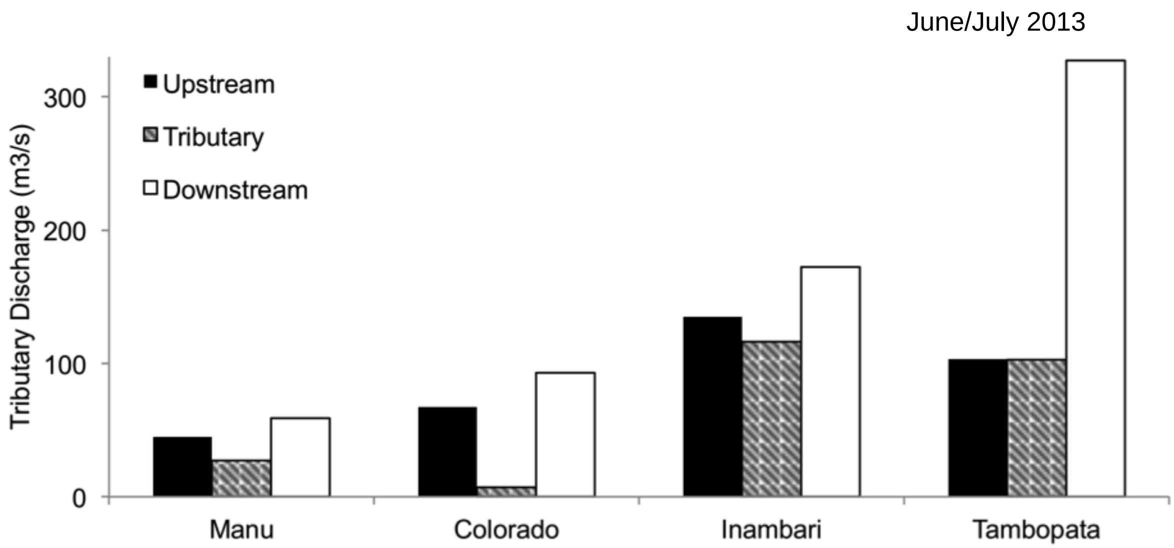

Figure S5. River flow rate from each tributary, upstream and downstream of the confluence on the MDD River during the June/July 2013 sampling event. Discharge rate was calculated by multiplying the surface water velocity (m/s) and cross sectional area (m^2).

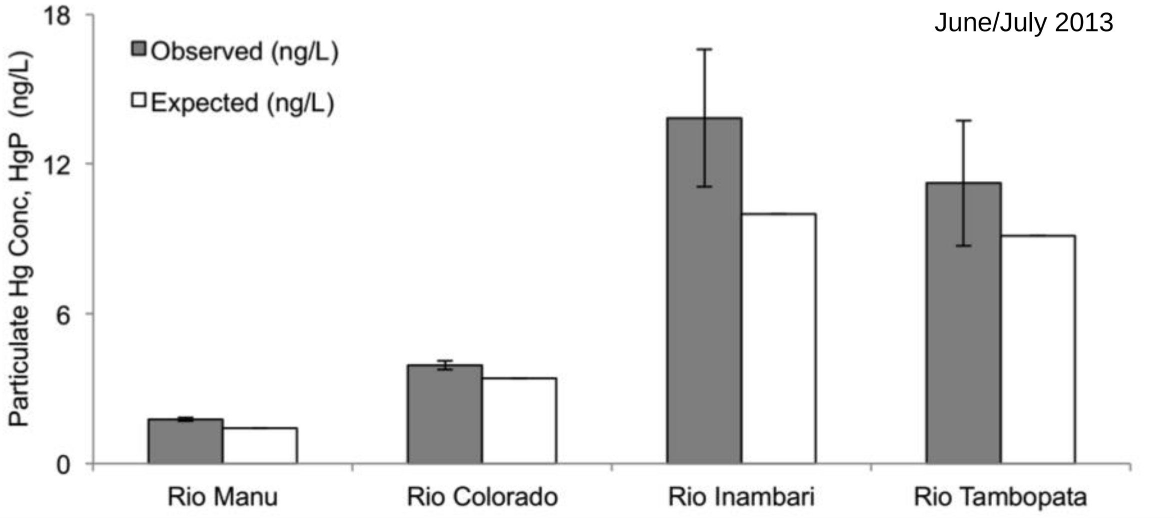

Figure S6. Observed and calculated particulate mercury concentrations Hg_P (ng/L) for locations directly downstream of major tributary confluences (June/July sampling event). Error bars represent standard deviation of replicate samples for the measured value (n = 3-6).

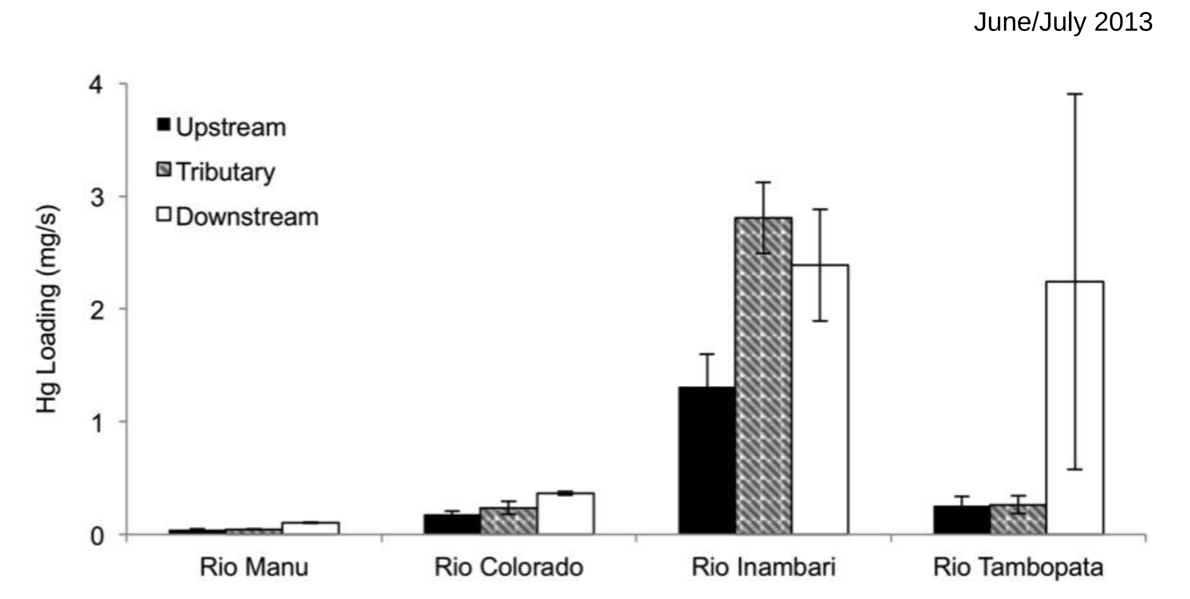

Figure S7. Particulate mercury loading rate at the confluence of each major tributary for the Madre Dios River (June/July sampling event). Loading rates were calculated by multiplying the river discharge rate with the suspended particulate mercury concentration (Hg_P). Error bars indicate standard deviation of the load rate estimated by propagating the standard deviation of replicate Hg_P measurements (n = 3-6).

Table S5. Fish samples caught during both Mar/Apr and Jun/Jul 2013 sampling events. Trophic level 1 is designated for non-carnivorous species, including detritovores, herbivores, and filter feeders. Trophic level 2 refers to omnivorores, while trophic level 3 refers to obligate carnivores. Four fish were not identified by common name or species. Migratory species are defined as species known to migrate more than 100 km while non-migratory species migrate less than 100 km throughout their known lifetime.

Local Name	Species Name	Samples (n=196)	Avg Hg in mg/kg (95% CI)	Standard Deviation	Avg Length, cm (Min, Max)	Avg Weight, g (Min, Max)	Trophic Level	Migratory?
Añashua or *Anhasua*	*Crenicichla semicincta*	2	0.17 (0.10, 0.24)	0.053	19 (15, 22)	75 (50, 100)	2	No
Ashara	*Leiarius marmoratus*	5	0.29 (0.01, 0.57)	0.31	50 (23, 63)	1600 (100, 2600)	3	No
Bacalao	*Pellona altamazonica*	2	0.38 (0.03, 0.73)	0.25	27 (26, 28)	150 (100, 200)	3	No
Bagre	*Megalonema platycephalum, Pimelodus sp.,*	21	0.15 (0.12, 0.18)	0.066	23 (15, 39)	111 (10, 300)	3	Yes
Bocachico	*Prochilodus nigricans*	35	0.22 (0.16, 0.28)	0.19	31 (21, 56)	407 (100, 2230)	1	Yes
Bocón/Toa	*Ageneiosus inermis*	4	0.63 (0.02, 1.24)	0.61	35 (23, 48)	494 (250, 850)	3	No
Canero	*Hemicetopsis sp*	1	1.4	--	27	100	3	No
Carachama	*Liposarcus sp/ Pterigoplictis disjuntivus/Hypostomus sp/Squaliforma phrixosoma*	6	0.06 (0, 0.12)	0.074	29 (22, 39)	311(10, 800)	1	No
Chambira	*Hydrolycus pectoralis/ Raphiodon vulpinus*	21	1.2 (0.96, 1.44)	0.56	46 (30, 63)	668 (200, 1600)	3	Yes
Chio chio	*Psectrogaster rutiloides*	3	0.12 (0.08, 0.16)	0.032	20 (18, 21)	167 (150, 200)	1	Yes
Corvina	*Plagioscion squamosissimus*	6	0.51 (0.27, 0.75)	0.30	35 (15, 51)	703 (25, 1500)	3	No
Denton	*Cynopotamus amazonus*	4	0.39 (0.39, 0.39)	0.087	18 (17, 19)	28 (15, 50)	2	Unknown
Doncella	*Pseudoplatystoma punctifer*	3	0.79 (0.75, 0.83)	0.49	56 (49, 60)	1400 (800, 1800)	3	Yes
Dorado	*Brachyplatystoma rousseauxii*	2	0.82 (0.81, 0.84)	0.26	67 (21, 112)	5945 (90, 11800)	3	Yes
Huasaco	*Hoplias malabaricus*	5	0.31 (0, 0.62)	0.37	36 (34, 38)	459 (275, 620)	3	No
Leguia	Ageneiosus sp.	1	0.33	--	21	50	3	Yes
Lisa	*Leporinus sp*	4	0.16 (0, 0.41)	0.14	27 (25, 30)	263 (200, 350)	2	Yes
Maparate	*Hypophthalmus edentatus*	1	0.33	--	38	400	1	Yes
Canero/ Pablucha	*Cetopsis plumbea*	2	0.06 (0.04, 0.07)	0.015	13(12, 13)	13 (10, 15)	2	No
Palometa	*Mylossoma aureum/*	11	0.04 (0.02, 0.05)	0.014	25 (16, 32)	423 (100, 900)	2	Yes

	Mylossoma duriventre							
Pico de Pato	*Sorubim lima/ Hemisorubim platyrhynchos/ Platystomatichthys sturio*	10	0.48 (0.33, 0.63)	0.15	33 (19, 47)	225 (25, 525)	3	No
Piraña or Piranha	*Serrasalmus sp*	3	0.27 (0.15, 0.39)	0.12	23 (19, 27)	250 (150, 400)	3	No
Piro	*Megalodoras irwini*	1	0.01	--	80	7800	2	Unknown
Sabalo	*Salminus affinus*	5	1.2 (0.44, 2.0)	0.58	34 (17, 61)	1317 (10, 3500)	2	Yes
Sapamama	*Triportheus angulatus*	7	0.28 (0.02, 0.54)	0.26	23 (19, 32)	175 (100, 300)	2	Yes
Sardina	*Tetragonopterus argenteus*	2	0.22 (0.12, 0.32)	0.10	15 (14, 15)	95 (90, 100)	2	No
Yahuarachi	*Potamorhina altamazonica / Potamorhina latior*	21	0.23 (0.10, 0.36)	0.13	27 (19, 36)	265 (95, 400)	1	Yes
Yulilla	*Anodus elongatus*	7	0.36 (0.04, 0.68)	0.32	31 (29, 33)	298 (250, 400)	1	Yes
Zúngaro	*Zungaro zungaro*	1	0.25	--	52	1100	3	Yes

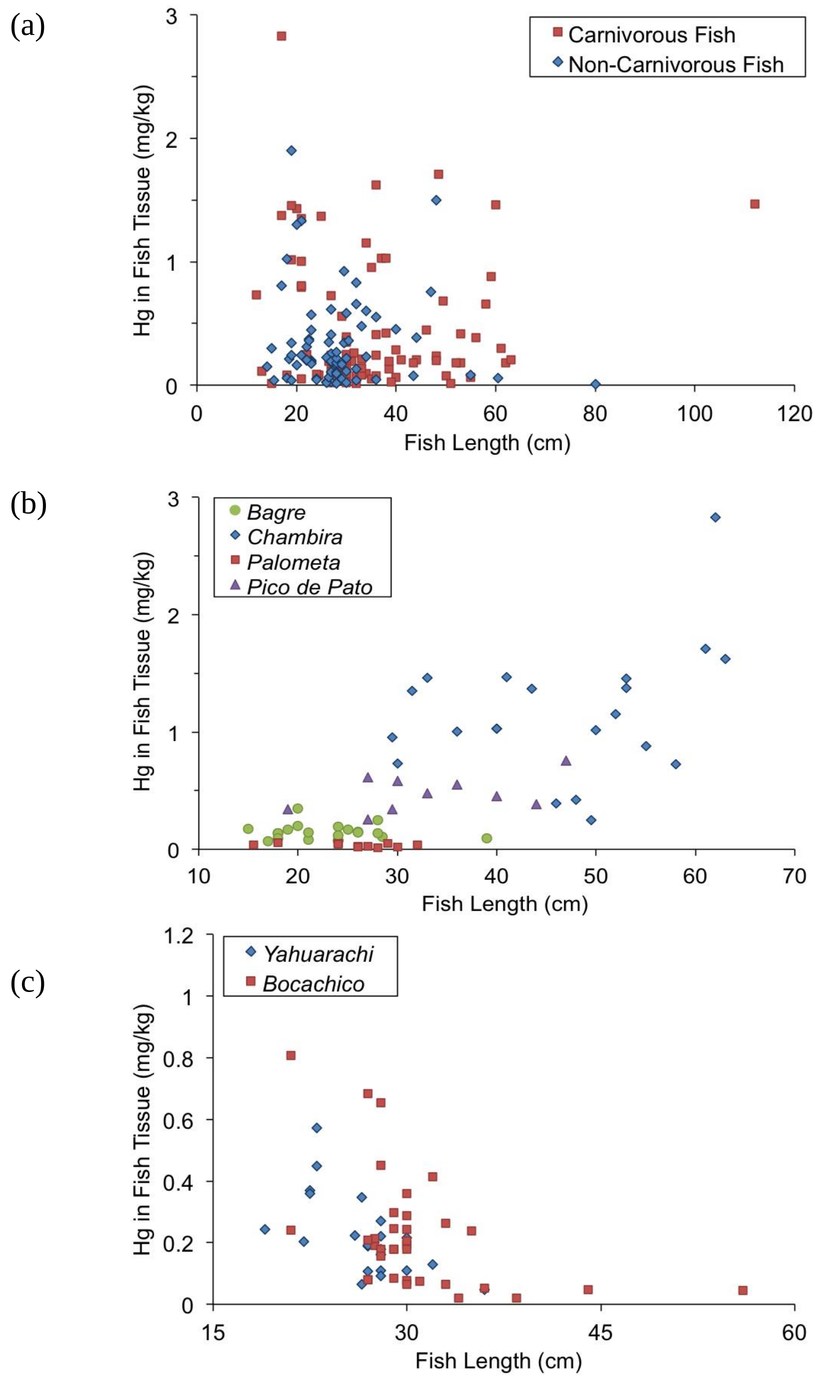

Figure S8. Correlations between fish length and fish tissue total Hg. Results indicate fish length and Hg concentration are not strongly correlated throughout the watershed, indicating fish size is not the only factor contributing to Hg concentration. (a) Correlation between Hg concentration in fish tissue with length for all carnivorous fish (n = 123, PC = 0.40) and all non-carnivorous fish (n = 74, PC = -0.25); (b) Correlation for individual carnivorous fish with 10 or greater samples: *Bagre* (n = 21, PC = -0.11), *Chambira* (n = 21, PC = 0.32), *Palometa* (n = 11, PC = -0.44), and *Pico de Pato* (n = 10, PC = 0.47). (c) Correlation for individual non-carnivorous fish with 10 or greater samples: *Bocachico* (n = 35, PC = -0.45), *Yahuarachi* (n = 21, PC = -0.62).

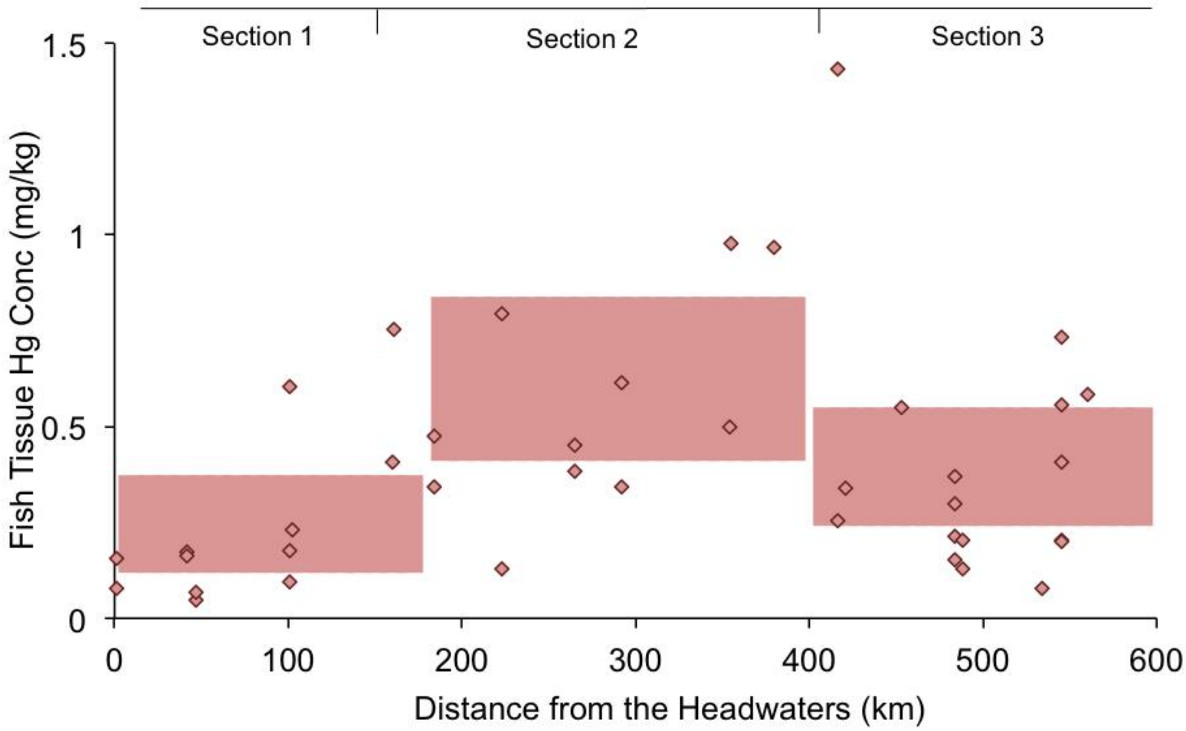

Figure S9. Examination of Hg concentrations within non-migratory, carnivorous fish (n = 41; *Anashua, Ashara, Bacalao, Canero, Corvina, Huasaco, Pablucha, Pico de Pato, Pirhana*, and *Sardina*). 95% confidence intervals around the mean fish concentration within each section are represented by red, solid boxes.

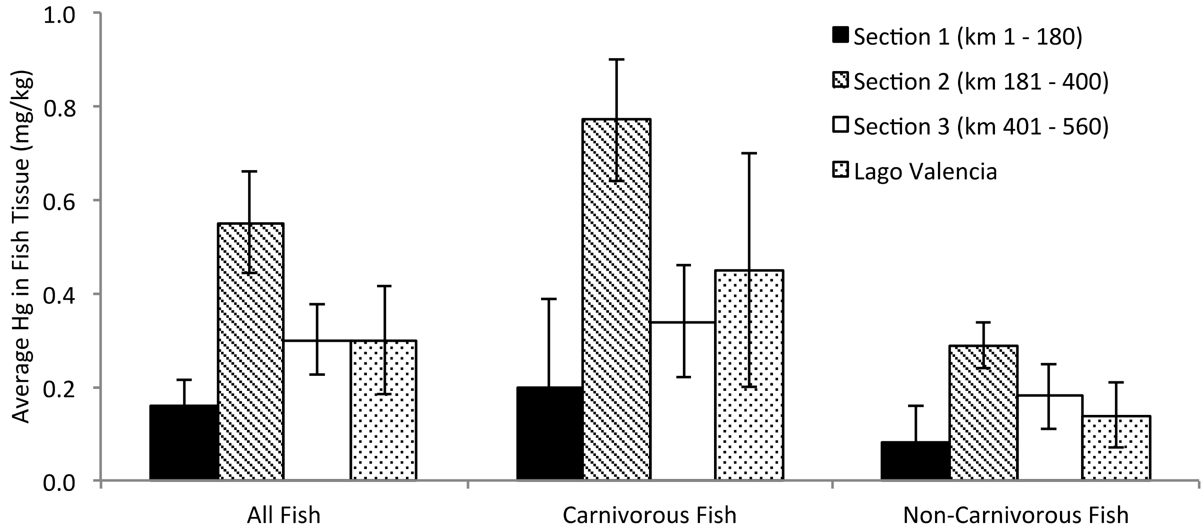

Figure S10. Average fish tissue concentration of Hg in each region for all fish, carnivorous fish, and non-carnivorous fish in both seasons. Error bars represent 95% confidence intervals.

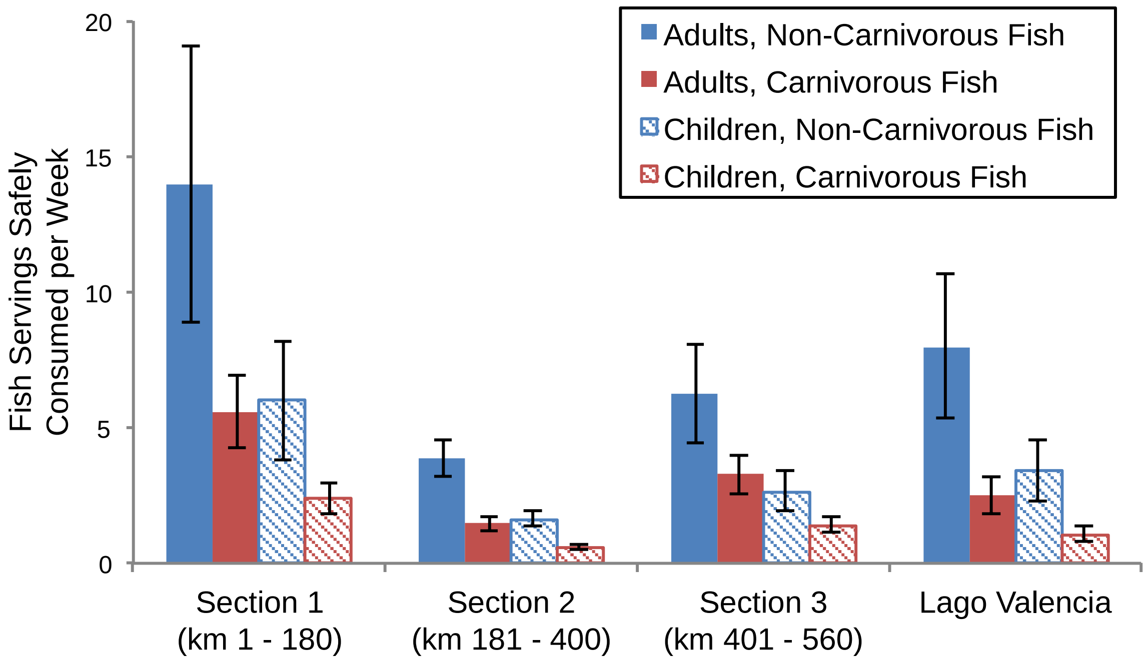

Figure S11. Number of fish safely consumed by adults (70 kg) and children (30 kg) of both carnivorous and non-carnivorous fish based on UNEP provisional tolerable weekly intake value of 3.2 μg Hg/kg bw for children and women of child bearing age. Note that pregnant women should consume up to half the recommended quantity of fish as recommended for children to avoid exceeding PTWI for fetal development (1.6 μg Hg/kg bw). Error bars represent 95% confidence intervals around fish tissue concentrations in section.