

Mercury in the gold mining district of San Martin de Loba, South of Bolivar (Colombia)

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Abstract Gold mining is responsible for most Hg pollution in developing countries. The aims of this study were to assess the levels of total Hg (T-Hg) in human hair, fish, water, macrophyte, and sediment samples in the gold mining district of San Martin de Loba, Colombia, as well as to determine fish consumption-based risks for T-Hg ingestion. T-Hg levels were measured by electrothermal atomization and atomic absorption spectroscopy. The overall mean T-Hg level in hair for humans in the mining district of San Martin de Loba was 2.12 µg/g, whereas for the reference site, Chimichagua, Cesar, it was 0.58 µg/g. Mean T-Hg levels were not different when considered within localities belonging to the mining district but differed when the comparison included Chimichagua. T-Hg levels in examined locations were weakly but significantly associated with age and height, as well as with fish consumption, except in San Martin de Loba. High T-Hg concentrations in fish were detected in *Pseudoplatystoma magdaleniatum*, *Caquetaia kraussii*, *Ageneiosus pardalis*, *Cyrtocharax magdalenae*, and *Triporthus magdalenae*, whereas the lowest appeared in *Prochilodus magdalenae* and *Hemiancistrus wilsoni*. In terms of Hg exposure due to fish consumption, only these last two species offer some guarantee of low risk for Hg-related health problems. Water, floating macrophytes, and sediments from effluents near mining sites also had high Hg values. In mines of San Martin de Loba and Hatillo de Loba, for instance, the geoaccumulation index (I_{geo}) for sediments reached values greater than 6, indicating extreme pollution. In short, these data support the presence of a high Hg-polluted

environment in this mining district, with direct risk for deleterious effects on the health of the mining communities.

Keywords Gold mining · Contamination · Aquatic ecosystems · Bioaccumulation · Heavy metal

Introduction

Heavy metals are a term used to describe metals and metalloids that have been associated with contamination and potential toxicity (Duffus 2002). These elements have received increasing attention due to the better understanding of their toxicological importance in ecosystems and human health. Within this group of chemicals, mercury (Hg) is considered to be one of the environmental pollutants with the greatest impact on the biosphere (Miller et al. 2011). This metal has been utilized by humans since Roman mining operations 3,500 years ago. Today, its uses are equally numerous, ranging from electronics to gold mining. The process by which many developing countries use elemental Hg to extract gold is becoming one of the primary sources of Hg pollution (Tomiyasu et al. 2013). During gold mining, Hg is employed in its elemental form to produce gold-Hg amalgams. Once it reaches the ecosystems, Hg is distributed into the air, soil, water, and sediments, forming the largest metal deposits in the environment (Martinez-Finley and Aschner 2014). Elemental Hg is highly volatile and easily dispersed at the high temperatures that often occur at sites of amalgamation. Once in the air, Hg undergoes global long-range atmospheric transport and deposition that allows its accumulation in biota, with subsequent human health risks (Falandyasz et al. 2014).

People living in close proximity to artisanal mining areas are vulnerable to Hg exposure. One of the major problems of Hg is its ability to cause neurotoxicity (Woods et al. 2013) and teratogenesis (Heinz et al. 2011), as well as lesions in organs

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such as the liver and kidneys (Sonne et al. 2013). Much of the neurotoxicity of Hg is associated with its ability to reach the brain by binding to cysteine, which uses the neutral amino acid transporter (Aschner and Aschner, 1990). Through the same process, the complex methylmercury (CH_3Hg^+)-cysteine also crosses the placenta and reaches the fetus where the developing nervous system is particularly sensitive to its deleterious effects (Kajiwara et al. 1996). Therefore, prenatal life is more susceptible to brain damage than adults (Cordier et al. 2002). Methylmercury also depletes cellular glutathione, resulting in the generation of oxidative stress (Lebel et al. 1998), and inhibits Se-dependent enzymes, key molecules in preventing neuronal oxidative damage. Therefore, Se-enriched diets are recommended to ameliorate organic Hg neurotoxicity symptoms (Ralston and Raymond 2010).

In Latin America, small-scale and artisanal gold extraction is one of the leading causes of Hg release to aquatic ecosystems (Veiga 2010). Although this activity contributes significantly to rural employment in Colombia, the apparent wealth resulting from gold mining contrasts with the loss of ecosystems, as well as both health and social impairment on communities. Despite the existing alternatives for avoiding Hg use in Colombian artisanal gold extraction, Hg pollution in mining areas has been almost exclusively associated with this activity (Olivero-Verbel et al. 2011; Marrugo-Negrete et al. 2008). The inappropriate use of Hg and the poor technical knowledge on handling and recovering of this metal have facilitated Hg contamination of water bodies and the atmosphere. Hg pollution possesses a daily threat to human health in towns surrounding these mines. The main objectives of this research were to determine the extent of Hg pollution derived from gold mining in the district of San Martin de Loba, one of the most prominent gold mining areas in Colombia, as well as assess sediment contamination and the health risk due to Hg intake via fish consumption.

Materials and methods

Study area

This study was conducted in the Mining District of San Martin de Loba, located in Southern Bolivar. This district registers all auriferous mining activities taking place in several municipalities, being San Martin de Loba, Barranco de Loba, and Hatillo de Loba, some of the most representative locations (Fig. 1). This area belongs to the Mompox Depression, a flood plain zone between the Magdalena and Cauca Rivers. Its main characteristic is the abundance of marshes and high temperatures throughout the year.

Hair collection

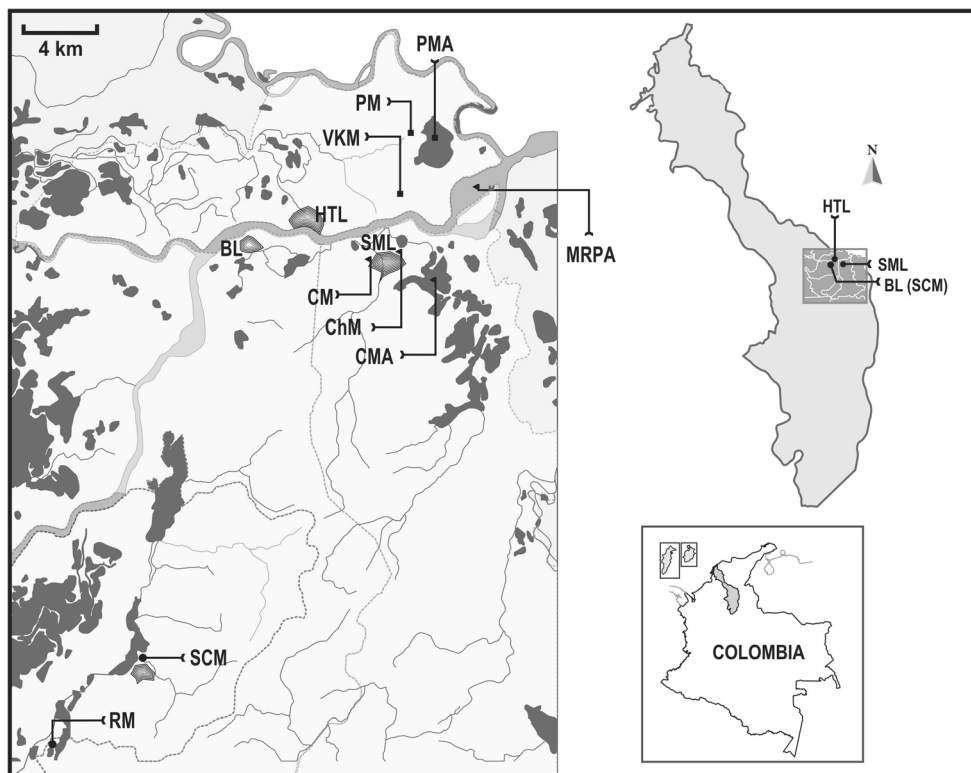
A total of 426 human hair samples were collected during October to December 2011 in the mining district: 200 from San Martin de Loba, 76 from Hatillo de Loba, and 167 from Barranco de Loba (Santa Cruz mine). In addition, 112 samples were obtained from Chimichagua, Cesar, a reference site located northeast of the mining area where fishing is a major economic activity, but no gold mining is present. A sample of approximately 100–200 mg of hair from the occipital scalp of each voluntary was collected using ethanol-cleaned scissors. After removal, samples were immediately deposited in white envelopes, labeled, and transported to the laboratory. Hair strands were cut with stainless steel scissors into 1–2-mm pieces and stored in a desiccator before analysis. The participants were interviewed by trained health professionals who carefully explained the objectives of the study. Gathered information included sociodemographic characteristics and several possible factors that may be associated with hair Hg concentration. A written informed consent was signed by each voluntary after receiving detailed explanations of the study and its potential consequences prior to enrollment.

Collection of fish, sediment, and macrophyte samples

A total of 195 fish were caught from October 2011 to January 2012 with the help of local fishermen: 90, 6, and 99 fish specimens were obtained from San Martin de Loba, Barranco de Loba, and Hatillo de Loba, respectively. Moreover, some specimens of *Pseudoplatystoma magdaleniatum* (Bagre tigre) were purchased at San Martin de Loba due to their large size and food habits (carnivorous). After collection, specimens were stored in ice and transported to the laboratory. The total length and weight of each specimen were recorded. A dorsal muscle subsample was removed using plastic knives and kept at $-20\text{ }^\circ\text{C}$ until chemical analyses.

Forty-seven sediment samples were obtained by lowering an Eckman grab from a boat. At each station, at least three to four subsamples were collected to make a composite sample of approximately 500 g. Each sample was placed in plastic bags, labeled and packed on ice, transported to the lab, and stored at $-20\text{ }^\circ\text{C}$ for their subsequent freeze-drying (Labconco Freezone 2.5) at $-50\text{ }^\circ\text{C}$ for 20 h and then homogenized. During sediment sampling, 43 macrophyte (*Eichhornia crassipes*) samples were manually collected from different sites. The plants were rinsed with marsh water and transported on ice to the lab. Upon arrival, plants were washed twice with Milli-Q water, stored at $-20\text{ }^\circ\text{C}$, freeze-dried as previously described, and homogenized (Jampeetong et al. 2012; Mechora et al. 2014). Additionally, 50 water samples (250 mL per station) were collected in several sites from the mining district, and then poured into clean, acid-washed

Fig. 1 Geographic location of the study area in the Bolivar Department. *HTL* Hatillo de Loba, *SML* San Martin de Loba, *BL* Barranco de Loba, *SCM* Santa Cruz Mine, *VKM* Villa Kelly Mine, *PM* Palenquillo Mine, *PMA* Palenquillo Marsh, *CM* Catanga Mine, *ChM* Charco Marsh, *CMA* Chimi Marsh, *MRPA* Magdalena River, La Peña Area, *RM* Redonda Marsh



polyethylene containers, acidified with HNO₃ to pH <2, and kept refrigerated until analysis within 1 week after collection (Marrugo-Negrete et al. 2008).

Mercury analysis in hair, fish, sediment, macrophytes, and water

The concentrations of total Hg (T-Hg) in human hair, fish muscle, sediments, and macrophytes were assessed employing 10, 140–160, 25, and 3–10 mg, respectively (Olivero-Verbel et al. 2009; Olivero-Verbel and Caballero-Gallardo 2013). Subsamples were pyrolyzed at 800 °C, and the vaporized Hg was detected by a RA-915+ Zeeman mercury spectrometer (Sholupov et al. 2004). T-Hg quantification was performed employing calibration curves constructed by measuring the absorbance for different weights of certified materials. Curves were considered optimal if the regression coefficient was ≥0.99. The accuracy of the method was assessed by analysis of blanks, and the use of certified reference materials such as IAEA-086 (human hair), IAEA-085 (human hair), IAEA-433 (marine sediment) from the International Atomic Energy Agency, NIES-13 (human hair) from National Institute for Environmental Studies, Japan, DORM-2 (dogfish muscle), and TORT-2 (lobster hepatopancreas) from National Research Council Canada. T-Hg in unfiltered water samples was measured using cold vapor atomic absorption spectroscopy (CVAAS) after digestion with diluted KMnO₄-K₂S₂O₈ solutions for 2 h at 95 °C (US-EPA 1994).

Detection limits for analyzed matrices are reported elsewhere (Marrugo-Negrete et al. 2008).

Assessment of sediment contamination

The geochemical accumulation index (*I_{geo}*) was employed as a quantitative tool to assess the level of Hg contamination in sediments. This index was calculated using Eq. 1, proposed by Muller (1969):

$$I_{geo} = \log_2 [Cn/ABn] \tag{1}$$

where Cn is the sediment metal concentration; A is the constant for modifying the fluctuation of the background value caused by lithological movement, usually 1.5 (Bhuiyan et al. 2010); and Bn is the geochemical background value of the metal. In this work, two reported background values were utilized: 0.06 (Lecce and Pavlowsky 2014) and 0.08 µg/g (Hortellani et al. 2013). Based on the results, Hg pollution in the sediment was classified into seven different categories: class 0 (unpolluted), *I_{geo}* ≤ 0; class 1 (unpolluted to moderately polluted), 0 ≤ *I_{geo}* ≤ 1; class 2 (moderately polluted), 1 ≤ *I_{geo}* ≤ 2; class 3 (moderately to strongly polluted), 2 ≤ *I_{geo}* ≤ 3; class 4 (strongly polluted), 3 ≤ *I_{geo}* ≤ 4; class 5 (strongly to extremely polluted), 4 ≤ *I_{geo}* ≤ 5; and class 6 (extremely contaminated), *I_{geo}* > 5.

Risk-based consumption limits

Risk factors were calculated according to the guidelines of the US Environmental Protection Agency (US-EPA 1989, 2000), previously reported by different authors (Marrugo-Negrete et al. 2008; Copat et al. 2013a, b). It was assumed that the ingestion dose was equal to the adsorbed T-Hg dose and that cooking had no effect on muscle T-Hg levels (Chien et al. 2002). Hg consumption limit calculations were based on the reference dose (RfDo) set by the US-EPA for methylmercury (MeHg). In summary, this risk was calculated using Eqs. 2 and 3 for the estimated daily intake per meal (EDIm) and for the target hazard quotient (THQ):

$$\text{EDIm} = (\text{MS} * \text{C})/\text{BW} \quad (2)$$

$$\text{THQ} = \text{EDIm}/\text{RfD} \quad (3)$$

where EDIm is the estimated daily intake of Hg per meal size; MS is the standard portion size of 230 g for adults (Hosseini et al. 2013); C is the MeHg mean concentration in fish ($\text{MS} = 0.90 \times \text{T-Hg}$) (Marrugo-Negrete et al. 2008); BW is the body weight of 70 kg for adults (Copat et al. 2013a); THQ is the target hazard quotient, the ratio between exposure and the reference dose, which indicates that systemic effects may occur when it results above 1. RfD for MeHg is 0.1 $\mu\text{g}/\text{kg}/\text{day}$ (US-EPA 1989).

The allowable number of fish meals of a specific meal size that may be consumed over a given period of time was also evaluated. For noncarcinogenic effects, we obtained the maximum allowable fish consumption rate in meals/week (CRmw) (US-EPA 2000) that would not be expected to cause any chronic systemic effects.

$$\text{CRmw} = 49/(\text{C} * \text{MS}) \quad (4)$$

Considering an average adult body weight of 70 kg (US-EPA 1994), the MeHg USEPA Acceptable Daily Intake (ADI) can be approximated as 7 $\mu\text{g}/\text{day}/\text{adult}$ (49 μg Hg/week) (Hosseini et al. 2013).

Data analysis

Data are presented as mean \pm standard error. ANOVA was used to evaluate mean differences for T-Hg concentrations between sampling locations, previously checking for normality and variance homogeneity, using Kolmogorov–Smirnov and Bartlett tests, respectively. When normality was not achieved, Kruskal–Wallis was used instead. Spearman correlation

analysis was conducted to determine associations between T-Hg and other variables. Also, Student's *t* test was done to compare between genders. The criterion of significance was set at $P < 0.05$.

Results

Sociodemographic and other characteristics of participants

The sociodemographic and other general characteristics of the communities evaluated in this study are summarized in Table 1. No significant differences were found between mining localities regarding age ($P = 0.343$). However, when considering the four groups, there were differences for fish intake habits, dental amalgam filling status, education, and gender ($P < 0.05$).

Hair mercury concentrations

The results of the analysis of T-Hg in human hair from the mining district of San Martin de Loba are presented in Fig. 2. The average concentration for all tested samples from the mining district was 2.12 ± 0.14 $\mu\text{g}/\text{g}$ (0.12–34.39 $\mu\text{g}/\text{g}$). This value is in average four-fold greater than that registered in the reference site (0.58 ± 0.05 $\mu\text{g}/\text{g}$). Hair T-Hg values within localities decreased in the order San Martin de Loba (2.50 ± 0.26 $\mu\text{g}/\text{g}$) > Barranco de Loba (1.85 ± 0.21 $\mu\text{g}/\text{g}$) > Hatillo de Loba (1.74 ± 0.15 $\mu\text{g}/\text{g}$). These values exceed the internationally accepted safety limits for T-Hg (1 $\mu\text{g}/\text{g}$) (Silbernagel et al. 2011).

The over-limit ratio values for T-Hg levels in hair from people living in evaluated localities are shown in Table 2. The risk threshold level for human health used in this study was 1.0 $\mu\text{g}/\text{g}$ in hair, as suggested by USEPA (Silbernagel et al. 2011). Data showed that 73 % of volunteers from San Martin de Loba, 69.7 % from Hatillo de Loba, and 76.7 % from Barranco de Loba had T-Hg concentrations greater than this level, with no significant differences detected between sites ($\chi^2 = 1.26$, $P = 0.532$). However, when the analysis included the reference site, significant differences were recorded between the proportions ($\chi^2 = 111.1$, $P < 0.001$), indicating that people from the mining district indeed had greater T-Hg concentrations than a municipality where gold mining is not a major activity.

Hair T-Hg levels according to gender are shown in Fig. 3. In all cases, male has significant greater average T-Hg levels. Outliers in the community of San Martin de Loba included three males, two miners (23.3 and 27.8 $\mu\text{g}/\text{g}$), and a fisherman (31.4 $\mu\text{g}/\text{g}$). However, in Barranco de Loba, a female, a daughter of a miner, had T-Hg level of 34.4 $\mu\text{g}/\text{g}$.

Table 1 Sociodemographic and other characteristics of volunteers

Municipality					Statistics	P value
Characteristic	SML	BL	HTL	CH		
Age <i>n</i> (year, mean±SEM)	197 (27.66±1.26)	167 (30.37±1.53)	76 (28.54±2.52)	111 (31.77±1.95)	KW=3.36	0.343
T-Hg (µg/g, mean±SEM)	200 (2.50±0.26)	167 (1.85±0.21)	76 (1.74±0.15)	112 (0.58±0.05)	KW=150.90	<0.001
Gender, <i>n</i> ^a (%) ^b						
Male	92 (46.00)	116 (69.46)	33 (43.42)	59 (52.68)	$\chi^2=16.24$	0.001
Female	108 (54.00)	51 (30.51)	43 (56.58)	53 (47.32)		
Education						
None	55 (27.50)	51 (30.54)	11 (14.47)	24 (21.43)	$\chi^2=23.77$	0.005
≤High school degree	132 (66.00)	111 (66.47)	54 (71.05)	72 (64.29)		
Some college ^c	8 (4.00)	2 (1.20)	2 (2.63)	9 (8.04)		
College degree	5 (2.50)	3 (1.80)	9 (11.84)	7 (6.25)		
Fish intake (times/week)						
0	9 (4.50)	14 (8.38)	2 (2.63)	2 (1.79)	$\chi^2=20.65$	0.014
1–4	89 (44.50)	102 (61.08)	46 (60.53)	79 (70.54)		
5–10	54 (27.00)	27 (16.17)	16 (21.05)	20 (17.89)		
>10	48 (24.00)	24 (14.37)	12 (15.79)	11 (9.82)		
Dental amalgam filling						
Yes	68 (34.00)	26 (15.57)	12 (15.79)	10 (8.93)	$\chi^2=22.50$	<0.001
No	132 (66.00)	141 (84.43)	64 (84.21)	102 (91.07)		

^a Number of data

^b Percentage frequency

^c Includes vocational/technical school

SML San Martin de Loba, BL Barranco de Loba, HTL Hatillo de Loba, CH Chimichagua (reference site), KW Kruskal–Wallis, χ^2 Chi-square, P significance level

According to Spearman correlation data (Table 3), age and height of volunteers correlated with T-Hg levels for all examined localities, including the reference site, being the maximum correlation found between T-Hg and age in

Chimichagua. In the case of fish intake and T-Hg, with the exception of San Martin de Loba ($\rho=0.108$, $P=0.136$), all localities showed significant correlations.

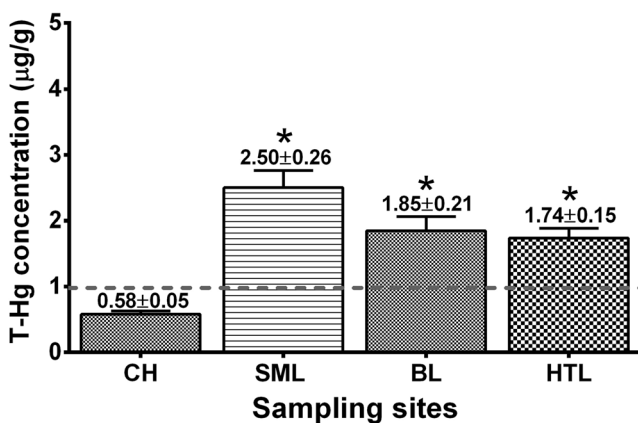


Fig. 2 Distribution of hair mercury levels from different sites. Significant differences were found between the groups (Kruskal–Wallis, post-test Dunnett, $P<0.01$). Dotted lines correspond to the maximum international permissible levels in hair (1 µg/g)

Fish mercury concentrations

The average muscle T-Hg concentrations in fish are depicted in Fig. 4. The highest T-Hg concentrations were observed in fish collected in marshes from San Martin de Loba, Chimi Marsh (*Caquetaia kraussii*, 0.45 ± 0.16 µg/g; *Cyrtcharax magdalenae*, 0.42 ± 0.05 µg/g; and *Triporthesus magdalenae*, 0.39 ± 0.09 µg/g) and Charco Marsh (*C. magdalenae*, 0.36 ± 0.09 µg/g). In Barranco de Loba and Hatillo de Loba, *C. magdalenae* presented lower T-Hg levels (0.29 ± 0.06 µg/g and 0.17 ± 0.02 , respectively). Interestingly, *Prochilodus magdalenae* (0.02 – 0.03 µg/g) always showed low T-Hg levels, even when captured from water bodies impacted by gold mining.

Large-size specimens of *P. magdaleniatum* obtained from San Martin de Loba area were tested for T-Hg in several edible parts along their bodies, and the results are presented in Fig. 5.

Table 2 Over-limit ratio of hair Hg concentration in the Mining District of San Martin de Loba

Parameter	SML	BL	HTL	CH	χ^2	<i>P</i>
<1 $\mu\text{g/g}$, <i>n</i> (%)	54 (27.00)	39 (23.35)	23 (30.26)	97 (86.61)	1.26 ^a , 111.1 ^b	0.532 ^a , <0.001 ^b
>1 $\mu\text{g/g}$, <i>n</i> (%)	146 (73.00)	128 (76.65)	53 (69.74)	15 (13.39)		

^a Chi-square without the reference site

^b Chi-square including the reference site

T-Hg concentrations were similar within specimens, although significant differences were found when compared to each other. As expected, T-Hg levels in these specimens increased with fish length and weight.

Risk-based consumption limits

Estimated EDIm, THQ, and CRmw values are presented in Table 4. Overall data indicate that for several species, the EDIm is greater than that suggested by the Joint FAO/WHO Expert Committee on Food Additive (JECFA) online database for total Hg (JECFA 2010) (0.004 mg/kg weekly, thus 0.571 $\mu\text{g/kg}$ daily). Of special concern are Arenca, Mojarra amarilla, and Chango collected from San Martin de Loba, Chango from Barranco de Loba, and Blanquillo from Hatillo de Loba. These results are in agreement with calculated THQ values. In fact, in addition to species mentioned in the previous paragraph, other fish species that represent a possible risk for chronic systemic effects derived from their Hg content are Doncella and Barbul, with THQ values greater than 2.0. Although unfortunately all the species could not be obtained in all sampling stations, it is of special interest to point out that from the data gathered for *C. magdalenae* (Chango), with

THQ values higher than 5 throughout the gold mining district, it is possible to suggest that for other fish species with similar T-Hg content, consumption should be avoided. This information, together with CRmw results, indicates that the species that can be eaten with low risk for human health are Bocachico, Coroncoro, and Comelon, being the first one of the most commercially available in the region.

Water mercury concentrations

T-Hg concentrations in water samples collected from different sites along the mining district are presented in Table 5. Although all collected samples were detected with measurable levels of T-Hg, greatest concentrations were registered in water samples collected from mills where ores are crushed at Catanga Mine, in San Martin de Loba ($39.18 \pm 7.50 \mu\text{g/L}$), whereas the lowest was observed at Villa Kelly Mine, Loma de Gallo Wetland ($0.17 \pm 0.01 \mu\text{g/L}$).

Sediment mercury concentrations and geoaccumulation analysis

Levels of T-Hg in sediment samples revealed high variability (Table 6). Sediments from Catanga Mine presented the highest metal concentrations ($63.46 \pm 25.88 \mu\text{g/g}$), followed by Palenquillo Marsh ($50.32 \pm 35.58 \mu\text{g/g}$), and Villa Kelly Mine ($31.75 \pm 11.85 \mu\text{g/g}$) in Hatillo de Loba. These samples were collected in nearby streams that conduct effluents from the mines to marshes or rivers. The lowest levels were observed in Charco Marsh ($0.40 \pm 0.09 \mu\text{g/g}$) at San Martin de Loba.

The I_{geo} data for the sampling sites are presented in Table 6. Based on these results, I_{geo} class 6, this is highly polluted sediments, which were found in San Martin de Loba (Caño and Catanga Mines) and Hatillo de Loba (Juana Sanchez, Villa Kelly Mine and Palenquillo Marsh). I_{geo} class 4, representing moderately to highly polluted sediments, was found only in Barranco de Loba. Moderately polluted sediments with I_{geo} classes 2 and 3 were found in two sites from San Martin de Loba.

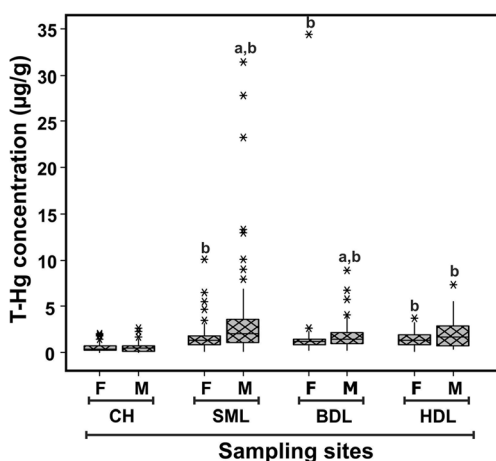


Fig. 3 Box plot average levels of T-Hg by gender. **a** Significant difference between female and male for each sampling site, using a *t* test ($P < 0.05$). **b** Significant difference for the same gender when comparing each sampling site with the reference group, using a *t* test ($P < 0.05$). *F* female and *M* male

Table 3 Spearman correlations between T-Hg in hair and some general characteristics of human population under study (significance level in parentheses)

Characteristic	Municipality			
	SML	BL	HTL	CH
Age (years)	0.282 (0.001)	0.313 (0.001)	0.383 (0.001)	0.508 (0.001)
Height (cm)	0.370 (0.001)	0.316 (0.001)	0.265 (0.022)	0.334 (0.001)
Fish intake ^a	0.108 (0.136)	0.418 (0.001)	0.411 (0.001)	0.433 (0.001)

^a Number of times (meals) a week that fish is part of the diet

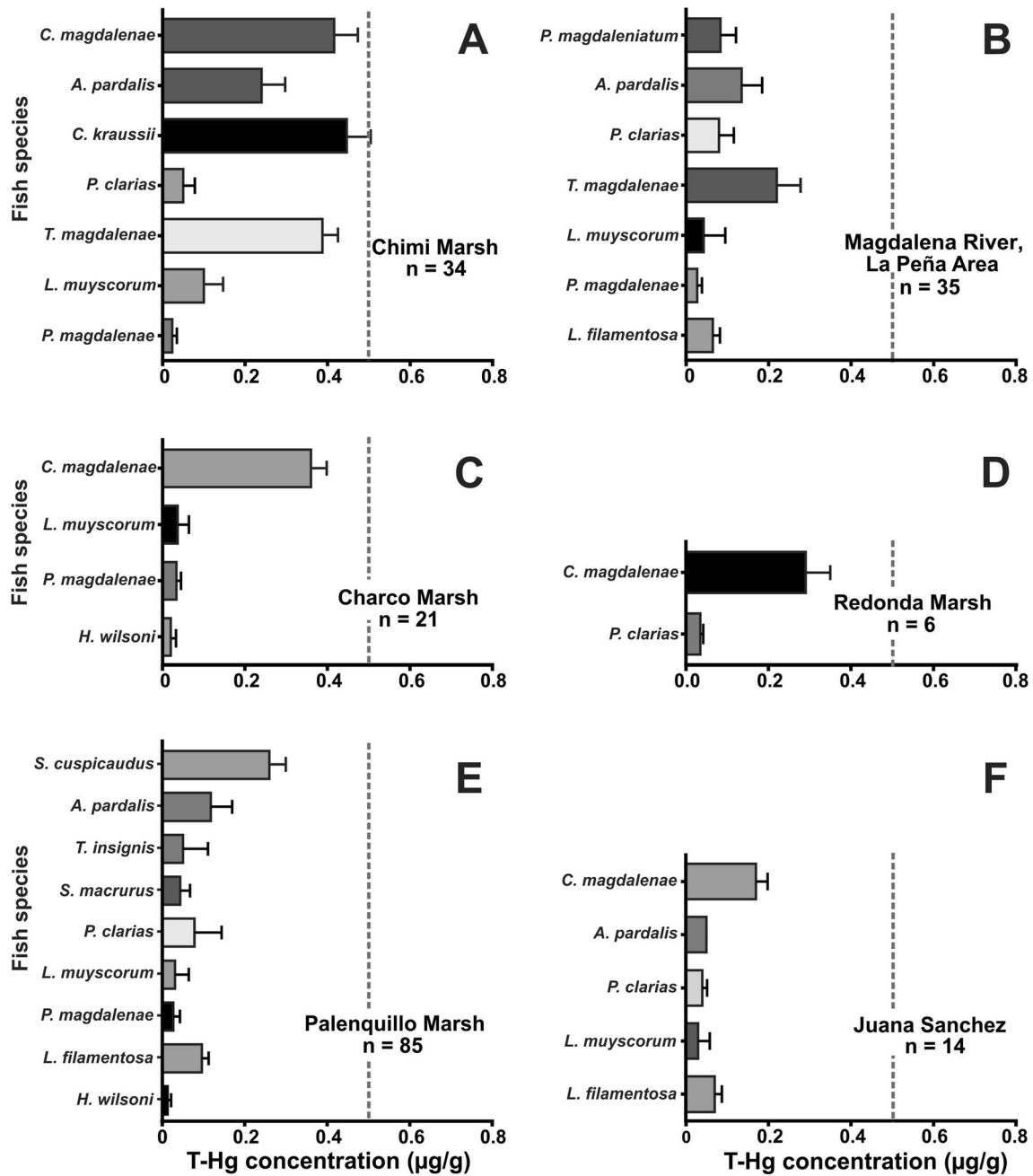
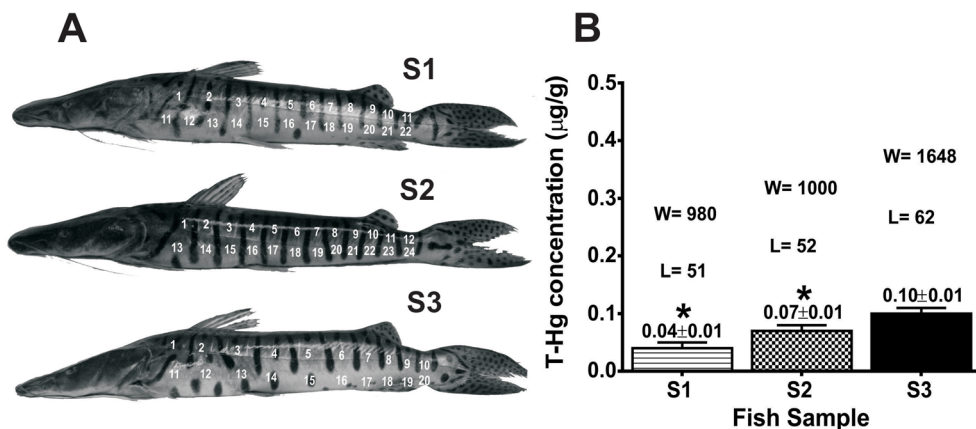


Fig. 4 T-Hg concentrations in muscle tissue of fish caught in the mining district of San Martin de Loba, Colombia. San Martin de Loba (a–c), Barranco de Loba (d), Hatillo de Loba (e, f). Data are presented as mean±

standard error for different sampling campaigns. Dotted lines correspond to the maximum international permissible levels in fish muscle (0.5 µg/g)

Fig. 5 T-Hg in muscle of *P. magdaleniatum*. Numbers inside represent the sample tissue used for results shown in B (a). Average T-Hg levels in different muscle sections of specimens (S1-S3) depicted on a (b). *W* weight (g) and *L* length (cm) measurements are shown for comparison. *Statistically different when compared to muscle samples from S3



Macrophyte mercury concentrations

Mercury content in root samples of the floating macrophyte *E. crassipes* collected in the mining district is shown in Fig. 6. Macrophyte roots from San Martin de Loba (Caño Mine) and Hatillo de Loba (Loma de Gallo Wetland) had the highest T-Hg concentrations (Fig. 6a; 0.48 ± 0.14 and 0.22 ± 0.06 µg/g, respectively). Mercury levels in different parts of *E. crassipes* (Fig. 6b) collected at the most polluted site (Caño Mine)

revealed that T-Hg levels decrease in the order roots>leaves>flowers>stem.

Discussion

Mercury is a persistent environmental pollutant considered a prominent toxicant among heavy metals (McAloon and Mason

Table 4 Estimated daily intake per meal (EDIm) (µg/kg daily), target hazard quotient (THQ), and maximum allowable fish consumption rate in meals/week (CRmw) in adults

Sampling site	Common name	Scientific name	T-Hg (µg/g) Mean±SE	EDIm	THQ	CRmw
San Martin de Loba	Coroncoro	<i>Hemiancistrus wilsoni</i>	0.02±0.01	0.06	0.59	11
	Alcalde	<i>Loricaria filamentosa</i>	0.07±0.01	0.21	2.07	3
	Bocachico	<i>Prochilodus magdalenae</i>	0.03±0.01	0.09	0.89	7
	Comelón	<i>Leporinus muyscorum</i>	0.05±0.01	0.15	1.48	4
	Arenca	<i>Triportheus magdalenae</i>	0.29±0.06	0.86	8.58	1
	Barbul	<i>Pimelodus clarias</i>	0.07±0.01	0.21	2.07	3
	Mojarra amarilla	<i>Caquetaia kraussii</i>	0.45±0.16	1.33	13.31	0
	Doncella	<i>Ageneiosus pardalis</i>	0.19±0.07	0.56	5.62	1
	Chango	<i>Cyrtocharax magdalenae</i>	0.40±0.04	1.18	11.83	1
	Bagre tigre	<i>Pseudoplatystoma magdaleniatum</i>	0.08±0.02	0.24	2.37	3
Santa Cruz Mine	Barbul	<i>Pimelodus clarias</i>	0.04±0.01	0.12	1.18	5
	Chango	<i>Cyrtocharax magdalenae</i>	0.29±0.06	0.86	8.58	1
Hatillo de Loba	Coroncoro	<i>Hemiancistrus wilsoni</i>	0.01±0.01	0.03	0.30	21
	Alcalde	<i>Loricaria filamentosa</i>	0.08±0.01	0.24	2.37	3
	Bocachico	<i>Prochilodus magdalenae</i>	0.03±0.01	0.09	0.89	7
	Comelón	<i>Leporinus muyscorum</i>	0.03±0.01	0.09	0.89	7
	Barbul	<i>Pimelodus clarias</i>	0.07±0.02	0.21	2.07	3
	Mayupa	<i>Sternopygus macrorus</i>	0.04±0.01	0.12	1.18	5
	Capitanejo	<i>Trachelyopterus insignis</i>	0.05±0.02	0.15	1.48	4
	Doncella	<i>Ageneiosus pardalis</i>	0.10±0.02	0.30	2.96	2
	Chango	<i>Cyrtocharax magdalenae</i>	0.17±0.02	0.50	5.03	1
	Blanquillo	<i>Sorubim cuspicaudus</i>	0.26±0.09	0.77	7y69	1

EDIm values higher than the PTWI and values of THQ greater than 1 are indicated in bold

Table 5 T-Hg concentrations in water (µg/L) collected in the Mining District of San Martín de Loba, Colombia

Municipality	Site	n	T-Hg (µg/L) mean±SE
Hatillo de Loba	Villa Kelly Mine, Loma de Gallo Wetland	5	0.17±0.01
	Palenquillo Marsh	3	1.96±0.79
San Martín de Loba	Caño Mine	7	2.24±1.60
	Charco Marsh	3	0.26±0.05
	Catanga Mine	11	39.18±7.50
	Chimi Marsh	10	0.18±0.02
Santa Cruz Mine	Redonda Marsh and Pacha Marsh	10	0.33±0.06

2003) due to its high toxicity and association with historic events of contamination (Kasper et al. 2007). One of the most prominent uses of Hg in Latin America is artisanal gold mining. In this work, T-Hg levels in several environmental matrices were evaluated at the Mining District of San Martín de Loba, Northern Colombia, aiming to establish human health risks based on Hg exposure. Mean T-Hg concentrations in human hair reported here were greater than those registered back in 2006 (Olivero-Verbel et al. 2011), as follows: San Martín de Loba (2.50±0.26 vs 0.92±0.14 µg/g), Barranco de Loba (1.85±0.21 vs 1.77±0.19 µg/g), and Hatillo de Loba (1.74±0.15 vs 1.09±0.11 µg/g). This information indicates that the increase in Hg concentration was sustained in San Martín de Loba, probably as a result of a more extensive mining process taking place in that area. However, all mining towns of this district had greater T-Hg values than those found in Chimichagua, a location where mining is not practiced and fishing is one of the major economic activities. Overall, human Hg exposure usually occurs through inhalation of elemental Hg vapor via occupational or dental amalgam exposure, or by ingestion of contaminated fish (organic mercury) (WHO 1991; Richardson 1996). As the district is surrounded by river and marshes, Hg release from gold mining may be reaching humans both as a result of direct breathing of Hg vapor or through consumption of Hg-containing fish.

In addition to be a great fishery resource, fish are considered excellent indicators for the assessment of Hg pollution (Kasper et al. 2007). In tropical ecosystems, several studies have reported that fish species with carnivorous habits accumulate more Hg

than those not carnivorous (Marrugo-Negrete et al. 2008; Olivero-Verbel et al. 2009; Zhu et al. 2012). The results presented here for different fish species suggest that based on their Hg content, only few species are suitable for human consumption, in particular *P. magdalenae* and *Hemiancistrus wilsoni*, which usually report T-Hg concentrations below 0.1 µg/g fresh weight, even when caught at wetlands with consistent heavy Hg discharges from artisanal gold mining, such as the Redonda Marsh, near Santa Cruz mine (Barranco de Loba). On the other hand, species such as *A. pardalis*, *P. magdaleniatum*, *C. kraussii*, *C. magdalenae*, and *T. magdalenae* possess T-Hg concentrations that effectively constitute a risk for human health. Several of these species collected in other freshwater ecosystems of Colombia have similar or greater Hg levels (Marrugo-Negrete et al. 2008). The species *C. magdalenae* presented high concentrations of T-Hg in all sampling stations along the Mining District of San Martín de Loba. The high Hg bioaccumulation capacity of this species had not been reported in the literature.

The identification of fish species with high Hg bioaccumulation potential is of great importance for populations whose diet is based on fish, as they are the most vulnerable and the least aware of their danger. Regardless of whether these species are obtained from marshes impacted by gold mining or not, these should be avoided for human consumption, especially by children and pregnant women, since they are the most susceptible to the deleterious effects of the heavy metal (Sadagoparamanujam et al. 2011). Prenatal exposure to Hg is of particular concern because the fetal central nervous system is highly vulnerable to

Table 6 T-Hg concentrations in sediment (µg/g) collected in the Mining District of San Martín de Loba, Colombia

Municipality	Site	n	T-Hg (µg/g) mean±SE	Range (µg/g)	I _{geo}	Sediment quality
Hatillo de Loba	Juana Sanchez,	8	31.8±11.9	0.1–83.8	8.0–8.5*	Extremely polluted
	Villa Kelly Mine	3	50.3±33.6	10.1–117.0	8.7–9.1	Extremely polluted
San Martín de Loba	Palenquillo Marsh					
	Caño Mine	7	14.6±12.2	0.6–87.8	6.9–7.3	Extremely polluted
	Charco Marsh	3	0.4±0.1	0.3–0.6	1.7–2.2	Moderately polluted
	Chimi Marsh	7	0.7±0.1	0.5–1.0	2.6–3.0	Moderately to strongly polluted
Barranco de Loba	Catanga Mine	9	63.5±25.9	5.9–232.8	9.0–9.5	Extremely polluted
	Santa Cruz Mine (Port and Marsh)	10	1.0±0.3	0.1–3.4	3.1–3.5	Strongly polluted

*Background levels: 0.08 (Hortellani et al. 2013) and 0.06 (Lecce and Pavlowsky 2014)

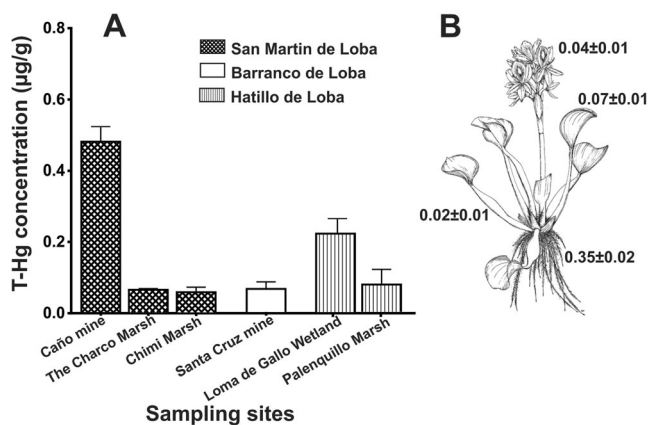


Fig. 6 T-Hg levels in the roots of *E. crassipes* collected from different sites at the mining district of San Martin de Loba (a) and in different parts of specimens from Caño Mine (b) at the primary effluent channel

its effects, affecting neurodevelopment, primarily measured through decrements in IQ (Dickenson et al. 2013), although other cognitive and behavioral deficits have also been observed, including attention-deficit/hyperactivity disorder-related behavior in children (Llop et al. 2012; Jain 2013).

P. magdaleniatum is one of the most desirable species in the national market for the quality of its meat (CCI-INCODER 2007). These species usually reach large sizes, with piscivorous habits, and are highly distributed in the Magdalena basin (Carvalho-Costa et al. 2011), where the mining district is located. *P. magdaleniatum* samples from the Magdalena River have been found with T-Hg levels of $0.17 \pm 0.06 \mu\text{g/g}$ (Alvarez et al. 2012)

for specimens larger than those captured in this study. At the Dique Channel, approximately 200 km downstream of the mining district, this species has reported T-Hg levels of $0.07 \pm 0.01 \mu\text{g/g}$, suggesting a clear reduction in Hg exposure with distance from the source (Olivero-Verbel and Caballero-Gallardo 2013).

Data on consumption limits (Table 4) indicate that although the T-Hg concentrations detected in examined fish are under recommended limits (less than $0.5 \mu\text{g/g}$), the intake per meal size in many cases exceeds the doses suggested by the JECFA (JECFA 2003, 2010). These data are the first to estimate the risk of eating fish from this mining area and confirm the potential public health risks based on Hg increases with trophic levels, the length of the food web, and body size (Gentès et al. 2013).

This research reports that most water samples have average T-Hg levels above the method detection limit ($0.1 \mu\text{g/L}$), which is a clear evidence of the extent of Hg pollution in the mining district of San Martin de Loba. The highest values of T-Hg in water were recorded in channels that collect effluents from Catanga Mine, San Martin de Loba (up to $73.2 \mu\text{g/L}$). This value exceeded several times the T-Hg levels found in an abandoned gold mine in China (up to $9.3 \mu\text{g/L}$) (Qiu et al. 2009). T-Hg concentrations found at Redonda Marsh and The Pacha Marsh were low ($0.33 \pm 0.06 \mu\text{g/L}$), consistent with levels found in sediments and macrophytes. Interestingly, similar values were reported by Marrugo-Negrete et al. (2008) in water samples from Grande Marsh in South Bolivar.

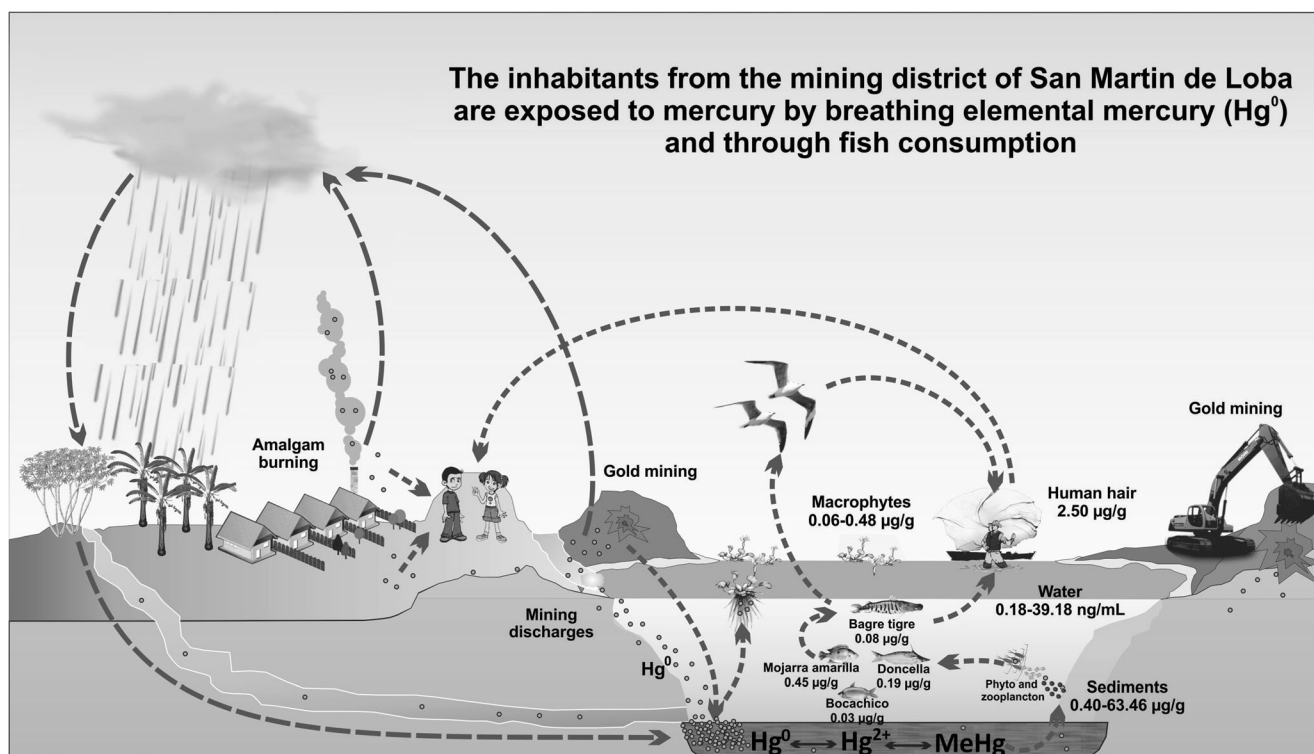


Fig. 7 Diagram for plausible mercury distribution pathways along the mining district of San Martin de Loba

T-Hg levels in sediments collected in the mines of the Mining District of San Martin de Loba (Table 6) reflect the high Hg contamination in liquid waste generated from gold mines. T-Hg values as high as 232.8 and 117.0 $\mu\text{g/g}$ in sediments from mines at San Martin de Loba and Hatillo de Loba, respectively, demonstrated the magnitude of the problem. According to the I_{geo} indexes, Hg pollution in these sediments classifies them as highly to strongly polluted (class 6), comparable to sediments found in an abandoned gold mining area in southern Minas Gerais State, Brazil (Cesar et al. 2011) and the Middle Odra River (Germany/Poland) (Boszke et al. 2004). These concentrations are far from 0.81 $\mu\text{g/g}$, which is the allowable limit suggested by the International Atomic Energy Agency (IAEA) (Kwaansa-Ansah et al. 2011). Due to its high density, the bulk Hg amounts deposited in nearby streams of the mining operations may move slowly to nearby marshes, polluting their sediments (Marrugo-Negrete et al. 2008).

One of the most predominant macrophyte species in the Magdalena River basin is *E. crassipes* (water hyacinth). This species has received considerable attention due to its ability to grow in polluted water bodies and its accumulation capacity of toxic metal ions and other contaminants (Lu et al. 2004; Mishra et al. 2007; Vesik et al. 1999; Zhu et al. 1999). Even under stress, this plant is usually able to grow without showing any symptoms of toxicity (Lage-Pinto et al. 2008). This plant is an effective Hg bioaccumulator, particularly if it is in direct contact with the sediment in shallow waters, as in water channels leading to nearby marshes (Caño mine, San Martin de Loba). In general, Hg concentrations in macrophytes from the marshes surrounding gold mines were usually fairly low, probably as a result of the short residence time experienced in these water bodies. Results presented here are similar to those reported by Marrugo-Negrete et al. (2010) in the Ayapel marsh from the Mojana Region, an ecosystem indirectly impacted by gold mining. Interestingly, T-Hg concentrations in the different organs of this macrophyte also have a differential distribution as observed in the Caño Mine, San Martin de Loba (Fig. 6), where maximum Hg values were recorded in roots, followed by leaves and flowers, and lower levels in the stems.

According to the results found in this study, environmental dynamics of Hg in the mining district of San Martin de Loba may occur in the following manner (Fig. 7). First, Hg used in gold mining is poured into the marshes and rivers where it is deposited in the sediments. Next, the Hg undergoes a process of methylation by bacteria present in it and is turned into methylmercury, which passes through planktonic organisms (phytoplankton and zooplankton) which are eaten by small fish, and these by larger fish like Bagre Tigre, Moncholo, Mojarra amarilla, and Pacora, or even bird species that are to be consumed by the population of the district. Similarly, the Hg reaches waterbodies by both wet and dry deposition, this

last associated with dust particles on which Hg atoms in the air have been adsorbed in the atmosphere. Furthermore, metallic Hg deposited in sediments may pass the water column and enter air where it is dispersed. Hg from water could also be captured by aquatic plants and transported to nearby streams of the same ecosystems or, in some cases, remote sites. These plants may also die and settle to the bottom of the swamps where Hg is released, starting its biogeochemical cycle again. One aspect to highlight in the dynamics of Hg in the mining district of San Martin de Loba is that Hg not only affects people who are in direct contact with the metal in the mines. Humans living in urban areas are also affected when Hg is released into the atmosphere through burning processes of amalgamation, or directly above the ground where the miners carry contaminated mud on their shoes from the mines to the streets and houses of their municipalities.

Conclusions

Mercury pollution in the mining district of San Martin de Loba is extensive and covers environmental compartments and humans. Hg in fish from marshes located within the district possesses high risks for human health, as several species have Hg levels greater than those recommended by international agencies. This Hg is transported to marshes through effluent channels where Hg is directly accumulated in sediments; from here, Hg biomagnification starts and moves forward through the food chain until reaching humans, who are exposed to Hg by fish consumption or by breathing elemental Hg during amalgam burning, and therefore susceptible to high risks of Hg-related health problems. While steps toward new technologies for gold extraction are being made, it is urgent to implement programs to prevent and mitigate environmental pollution by Hg.

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Conflict of interest None.

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