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Mercury pollution by gold mining in a global biodiversity hotspot, the Choco biogeographic region, Colombia

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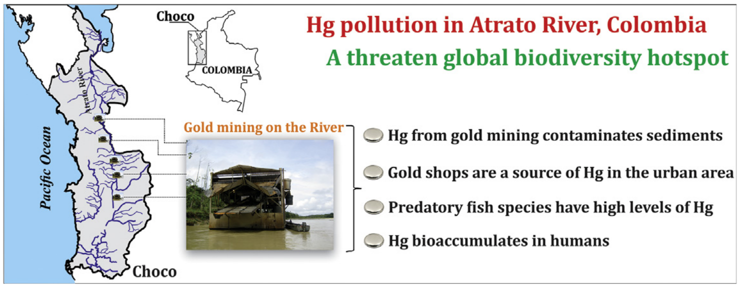
HIGHLIGHTS

- Hg content in Atrato River was evaluated in human hair, fish, sediments and air.
- Median Hg values in hair of people from Quibdo is almost twice that from Paimado.
- Carnivorous fish from Atrato River constitutes a health risk to people.
- Forty four percent of sediment samples showed moderate Hg pollution.
- Gold shops increase Hg in air compared to background levels.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) is a harmful pollutant released into the environment from gold mining activities, representing a risk to human health and the ecosystems. The aim of this study was to assess the levels of total Hg (T-Hg) in human hair, fish, sediments and air; and to determine fish consumption-based risks for T-Hg ingestion in the Choco biogeographic region, a global biodiversity hotspot located at the Colombian Pacific. Mercury concentrations in hair were measured in two locations, Quibdo, the state capital, and Paimado, a riverine community. The median T-Hg value in human hair in Quibdo was 1.26 μg/g (range: 0.02–116.40 μg/g), whereas in Paimado it was 0.67 μg/g (range: 0.07–6.47 μg/g). Mercury levels in examined locations were weakly associated with height ($\rho = 0.145$, $P = 0.024$). Air T-Hg levels in Quibdo were high inside gold shops being up to 200.9-fold greater than the background. Mercury concentrations in fish from Atrato River were above WHO limit (0.5 μg/g), with highest levels in *Pseudopimelodus schultzi*, *Ageneiosus pardalis*, *Sternopygus aequilabiatus*, *Rhamdia quelen* and *Hoplias malabaricus*, whereas the lowest appeared in *Cyphocharax magdalenae* and *Hemiancistrus wilsoni*. Based on fish consumption, these last two species offer low risk to human health. Sediment samples from fifty different sites of Atrato River showed low T-Hg concentrations, with little variability between stations. However, contamination factors revealed a moderate pollution in 44% of sampling sites along the river. In conclusion, Hg pollution is widespread in the Biogeographic Choco and governmental actions must be taken to protect the population and preserve its biodiversity.

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1. Introduction

Mercury (Hg) continues to be the primary tool for gold extraction in the developing world. Colombia is not the exception and its use and presence in several environmental matrices in this country has been documented in humans (Olivero-Verbel et al., 2011, 2016), air (Olivero Verbel et al., 2014), macrophytes (Olivero-Verbel et al., 2015), water (Olivero-Verbel et al., 2015) fish (Olivero-Verbel and Caballero-Gallardo, 2013; Olivero-Verbel et al., 2009; Olivero-Verbel et al., 2015), and birds (Olivero-Verbel et al., 2013), among other matrices.

During gold recovery Hg directly contaminates soil, water and air, finally reaching sediments where it is converted to methylmercury (MeHg), absorbed by plankton, whereby entering the food chain (Selin, 2009). Once there, MeHg bioaccumulates in predator species, including humans, who resulted exposed to high levels through their diet (Mahaffey, 1999). Humans also get directly exposed from Hg vapor inhalation, in any case, both Hg species are highly to the nervous system (Aschner and Aschner, 1990), causing sensory and mental disturbances, motor and cognitive dysfunction, ataxia, constriction of the visual field, audition problems (Harada, 1995), as well as deleterious effects on the renal, pulmonary, cardiovascular, digestive and immune system (Bernhoft, 2012; Frodello et al., 2000; WHO, 2016), among others.

The release of Hg into the environment from gold mining represents a public health problem. However, in countries like Colombia, without doubt, extracting the precious metal is a huge factor of destruction of the environment. A large percentage of gold mining is informal and the territory where mining is developed corresponds to areas of high importance for biodiversity, leading to a threat to forests, water bodies and living organisms. Colombia hosts several megadiverse ecosystems, hotspots areas of high species richness (Reid, 1998), being the Choco region one of the most complex ecosystems of the planet, registering 4584 species of spermatophytes, 793 birds (Rangel-Ch and Rivera-Díaz, 2004), 188 reptiles (Castaño et al., 2004), 139 amphibians, 196 freshwater fish (Mojica et al., 2004), 206 mammals (Muñoz-Saba and Alberico, 2004), and 176 beetles (Amat-García et al., 2004). This high variety of environments is attributed to its strategic location between two seas (Forero and Gentry, 1989), conditions that allow the formation of eight life zones (Forero, 1982), making it especially valuable for research.

Despite the immense potential of Choco, massive and uncontrolled gold mining activities are causing dramatic loss of biodiversity, a problem proceeding at a striking speed, becoming a major concern worldwide, as forest destruction is not only eliminating one of the world genome banks, but also leaving Hg pollution as a legacy, with little future for coming generations. Moreover, it is well known that these mining operations usually bring diverse social and public health disturbances that make the environmental problem a lot more complex (Castellanos et al., 2016; Tubb, 2015), and difficult to approach and solve. Accordingly, the main goal of this study was to determine the extent of T-Hg pollution in humans, fish, sediments and air in the region, aiming to establish environmental and population risks derived from Hg exposure to initiate counter measurements to approach several edges and establish research and intervention priorities.

2. Materials and methods

2.1. Study area

This study was conducted in the Choco region, at the Pacific coast of Colombia. Hair samples were collected in Quibdo and Paimado (Fig. 1). Quibdo (5°4132″N and 76°39′29″W) is the capital of the Department of Choco, located on the banks of Atrato River, one of the main rivers of Colombia, with high influence on the Embera National Natural Park, and near to a large number of indigenous reserves. Paimado (5°28′58″N and 76°44′23″W), on the other hand, is located on the Quito River, one of main Atrato's effluents, severely destroyed by illegal gold mining on its watershed (Fig. 2).

2.2. Hair collection

A total number of 360 human hair samples were collected from Choco: 248 from Quibdo and 112 from Paimado inhabitants. Samples were collected during November 2015 to July 2016. A sample of approximately 100–200 mg of hair from the occipital scalp of each voluntary was removed using ethanol-cleaned scissors and processed according to the methodology reported elsewhere (Olivero-Verbel et al., 2011, 2015, 2016). The participants were interviewed by trained health professionals who carefully explained the objectives of the study and gathered sociodemographic information, as well as possible factors linked to Hg exposure. A written informed consent was signed by each voluntary after receiving detailed explanation of the study, and its potential consequences prior to enrollment. In the case of children, these were signed by their parents (Olivero-Verbel et al., 2015, 2016). The study was approved by the Ethical Committee of the University of Cartagena, as part of the Colombian Observatory for Mercury.

2.3. Fish and sediment collection

A total of 258 fish, belonging to 16 species and different trophic levels were caught in February 2016 with the help of local fishermen at 11 different locations in Atrato River (Fig. SM1) (5°49′4.6" N and 76°41′18.4" W - 8°6′37.8 "N and 76°57′16.9" W), covering traditional fishing spots, swamps and mouths of several affluents. After collection fish were stored in ice, and the length and weight of each specimen were recorded, a dorsal muscle tissue sample removed using plastic knives, transported to the laboratory on ice, lyophilized (Labconco FreeZone 2.5) and kept at −20 °C until analysis.

Sediment samples were collected in fifty different sites along of Atrato River (Fig. SM2) by lowering an Eckman grab from a boat. At each station, at least three to four subsamples were pooled to make a composite sample of approximately 500 g. Each sample was placed in plastic bags, labeled and packed in ice, transported to the lab, stored at −20 °C, freeze-dried (Labconco Freezone 2.5) at −50 °C for 20 h (Olivero-Verbel et al., 2015), homogenized and them kept at −20 °C until analysis.

2.4. Mercury analysis in hair, fish, and sediments

Total Hg (T-Hg) in hair, fish, and sediment was determined using a RA-915^{+} Zeeman mercury analyzer with RP-915P software (Lumex, St. Peterburgo, Russia) with a pyrolysis unit (RP-91c). Total Hg quantification was carried out employing calibration curves constructed by measuring the absorbance given by different weights of certified materials, such as IAEA-085 (human hair) and IAEA-086 (human hair) from International Atomic Energy Agency, Analytical Quality Control Services, Wagramer Strasse 5, P:O. Box 100, A-1400 Viena Austria; DORM-2 (dogfish muscle) from National Research Council of Canada, Institute for National Measurement Standards M12, Montreal Road, Ottawa, Ontario Canada K1A 0R6; and PACS-2 (marine sediment) from National Research Council of Canada, Measurement Science and Standards, 1200 Montreal Road, Building M – 12, Ottawa, Ontario K1A 0R6, for hair, fish and sediments, respectively. Curves were considered optimal if the R^2 was

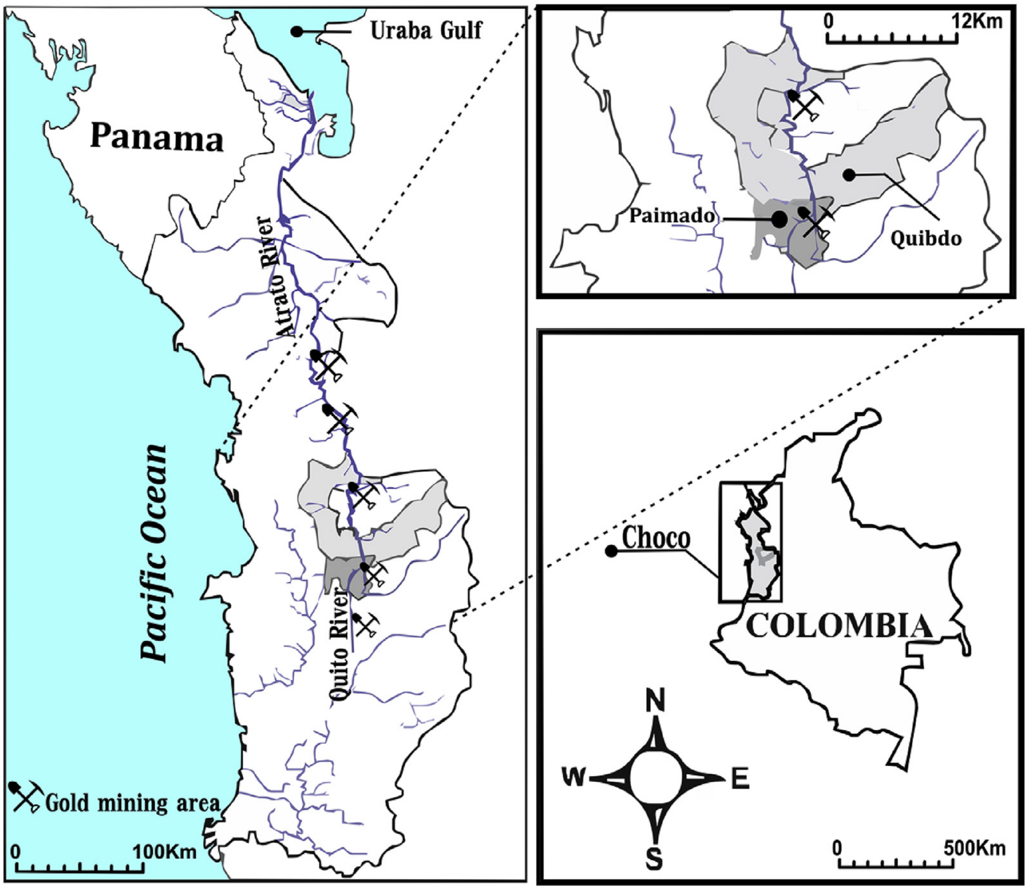

Fig. 1. Map of the study site.

$\geq$0.99. All samples were measured at least by duplicates (Olivero-Verbel et al., 2015). The minimum level of quantification for hair, fish and sediments was 0.005 μg/g. The percentage recovery for IAEA-085 was 99.9%, IAEA-086 was 98.4%, DORM-2 was 97.5% and for PACS-2 was 99.0%.

2.4.1. Risk-based consumption limits

Risk factors were calculated according to the guidelines of the United States Environmental Protection Agency (USEPA, 1989, 2000), previously reported by different authors (Copat et al., 2013a, 2013b). 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 (RfD) set by the US-EPA for MeHg. The risk was calculated using the estimated daily intake of Hg per meal (E) with this formula $E=(MS^*C)/W$; where MS is the standard portion size of 230 g for adults; C is the MeHg mean concentration in fish ($0.90 \times$ T-Hg); and W is the body weight of 70 kg for adults. The Hazard Quotient (HQ), the ratio between exposure (E) and the reference dose (RfD, 0.1 μg/kg/day for MeHg) (USEPA, 1989), indicates that systemic effects may occur when $HQ > 1$.

The allowable number of fish meals of a specific meal size that may be consumed over a given period of time (CRmw) was also evaluated. For non-carcinogenic effects that would not be expected to cause any chronic systemic effects, the CRmw in meals/week (USEPA, 2000) was calculated as $CRmw = 49/(C^* MS)$. Based on an average adult body weight of 70 kg (USEPA, 1994) the MeHg USEPA Acceptable Daily Intake (ADI) can be approximated 7 μg/day (Hosseini et al., 2013).

2.4.2. Risk for metal levels in sediments

The degree of pollution associated with the concentration of a metal present in sediments was determined by the contamination factor (CF), the ratio between the metal content in the sediments and its background level. In this study the reference value used for Hg in the earth crust was 0.085 μg/g (Lide, 2008). According to (Hakanson, 1980), $CF < 1$ refers to low contamination; $1 \geq CF \geq 3$ means moderate contamination; $3 \geq CF \geq 6$ indicates considerable contamination and $CF > 6$ suggests very high contamination. In addition, T-Hg values were compared to two numerical sediment quality guidelines (SQG), the probable effect concentration (PEC, level above which adverse effects are expected to frequently occur) and the threshold effect concentration (TEC, level below which adverse effects are not expected to occur (Mac Donald et al., 2000).

2.5. Mercury determination in air

Measurements of Hg in air were performed using a spectrometer with background correction—Zeeman atomic absorption mercury spectrometer RA-915+ (Lumex Ltd, Russia) as described elsewhere (Olivero-Verbel et al., 2006; Olivero Verbel et al., 2014). The equipment uses a built-in pump to automatically pass airflow through the spectrophotometer, which has a detection limit of 2 ng Hg/m^3. Mercury monitoring took place during morning hours with at least five readings on each place. Measurements were performed at twenty different sites in Quibdo, and the background readings were carried out in a rural area near the City. In addition, different internal sections of some gold shops located in downtown Quibdo were monitored in the absence of amalgam burning.

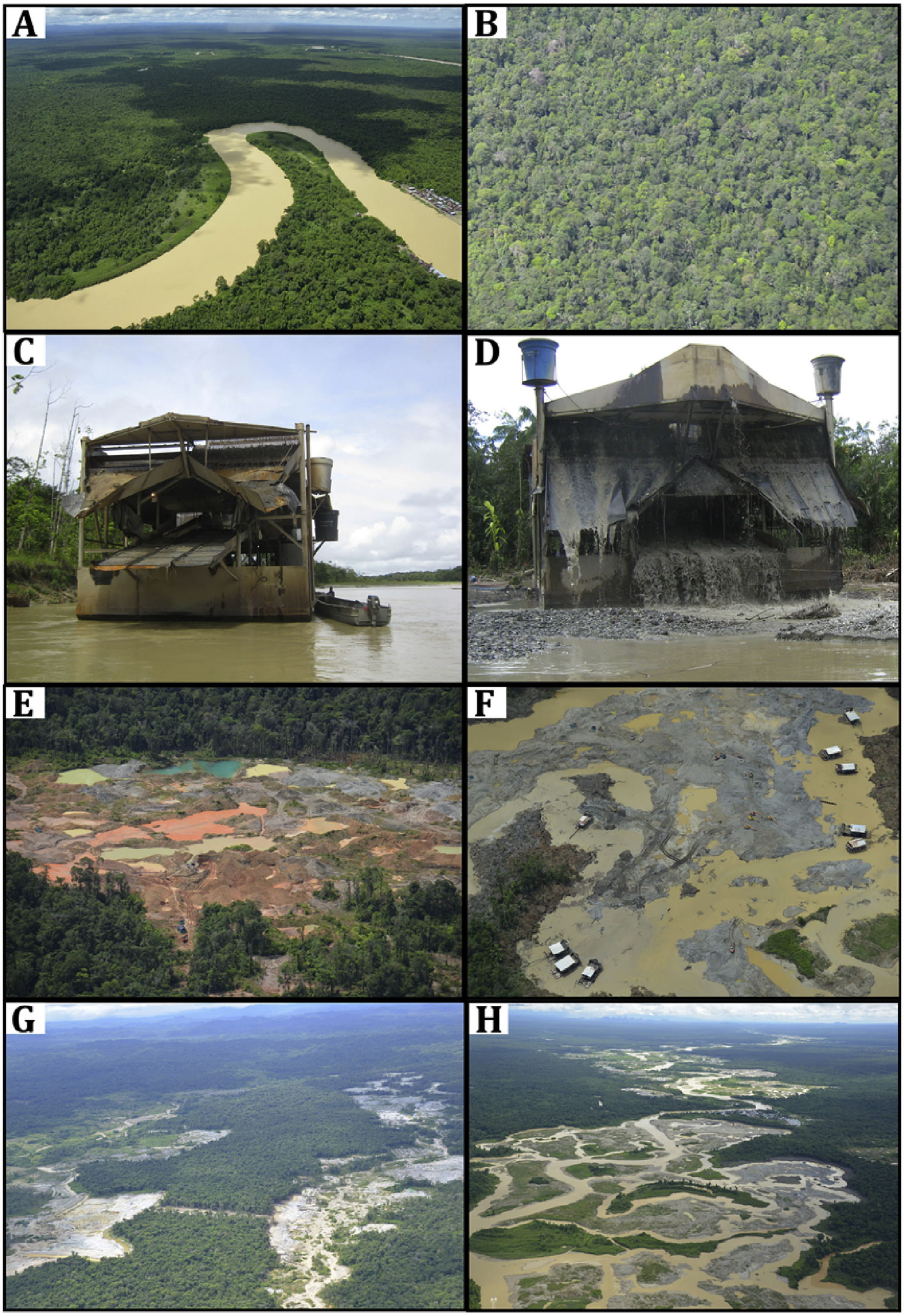

Fig. 2. Watershed destruction on Quito River. A, B. Virgin forest; C, D. Heavy machinery used for gold mining on riverbed; E-H. Forest and river destruction by gold mining.

2.6. Statistics analysis

Data are presented as the median or mean ± standard error. Changes in outcome mean measurements between two groups were tested by Student's t-test. The Mann-Whitney was employed when the data did not follow normal distribution. Chi-Square test was utilized for testing relationships on frequency distributions. In the present study, a level of 1 μg/g was used to distinguish hair Hg concentrations below or greater this reference value. Spearman correlation analysis was conducted to determine associations between T-Hg and other variables. The significance threshold was set to $P < 0.05$.

3. Results

3.1. Sociodemographic data

The socio-demographic characteristics of the volunteers in sampling sites are summarized in Table 1. The average age for the participants from the two locations were significantly different ($P < 0.005$), although frequency distributions were similar in terms of gender ($P = 0.215$) and fish consumption ($P = 0.136$), but different regarding education status ($P < 0.001$), and dental amalgam filling ($P = 0.010$).

3.2. Human hair mercury concentrations

Central tendency statistics for volunteers in Quibdo and Paimado are presented in Table 2. There were significant differences of hair Hg concentrations between Quibdo and Paimado (median value, 1.26 vs. 0.67 μg/g, $P < 0.001$). In Quibdo the average concentration of T-Hg in hair was 5.3-fold (6.72 μg/g) greater than the median, indicating some individuals have high T-Hg levels, reaching up to 116.4 μg/g.

In this study, 1.0 μg/g in hair was used as limit criteria for T-Hg in hair to protect human health. Data showed that 52.8% of volunteers from Quibdo and 33.9% from Paimado had greater hair mercury concentrations than the threshold level according to USEPA (USEPA, 2005) ($\chi^2 = 11.06$, $P < 0.001$, Table 3).

According to Spearman correlation data (Table 4), T-Hg levels correlated with the height of volunteers from Quibdo ($\rho = 0.145$, $P = 0.024$), but the relationship was weaker for those from Paimado ($\rho = 0.182$, $P = 0.057$).

3.3. Fish mercury concentrations and risk-based consumption limits

General morphometric variables and average muscle T-Hg concentrations in fish from Atrato River are depicted in Table 5. Total Hg values varied between 0.01 μg/g and 3.88 μg/g. The mean T-Hg concentration in all studied samples (16 species, 258 specimens) was 0.40 ± 0.03 μg/g. The highest average value for T-Hg (2.01 ± 0.51 μg/g) was detected on *Pseudopimelodus schultzi*, whereas the lowest (both with 0.06 ± 0.01 μg/g) was recorded in *Hemiancistrus wilsoni* and *Cyphocharax magdalenae*. Significant statistical differences for mean T-Hg concentrations were found between species (Kruskal–Wallis test, $P < 0.001$). In average, T-Hg concentrations decreased in the order (Fig. SM3 and Table 5) *Pseudopimelodus schultzi* (Bagre sapo) > *Ageneiosus pardalis* (Doncella) > *Sternopygus aequilabiatus* (Beringo) > *Rhamdia quelen* (Barbudo) > *Hoplias malabaricus* (Quicharo) > *Cathorops melanopus* (Bagre blanco) > *Centropomus undecimalis* (Róbalo) > *Caquetaia umbrifera* (Mojarra negra) > *Caquetaia kraussii* (Mojarra amarilla) > *Pimelodus* sp. (Gunguma) > *Pimelodus punctatus* (Charre) > *Prochilodus magdalenae* (Bocachico) > *Spatuloricaria atratoensis* (Guacuco palo) > *Leporinus muyscorum* (Denton) > *Hemiancistrus wilsoni* (Guacuco corroma) ≈ Cyphocharax *magdalenae* (Boquipompo).

Table 2
Total mercury concentrations in hair (μg/g) of inhabitants of Atrato watershed.

Sampling Site	n	Median (range)	Mean ± SEM	U [a]	p-value
Quibdo	248	1.26 (0.02–116.40)	6.72 ± 0.89	10735	<0.001
Paimado	112	0.67 (0.07–6.47)	0.87 ± 0.08		
Atrato River Basin	360	0.92 (0.02–116.40)	4.90 ± 0.63		

[a] Mann–Whitney *U* test.

Table 3
Frequency distribution of total mercury concentration in hair of people from Quibdo and Paimado.

Parameter	Quibdo	Paimado	Statistic	p-value
<1 μg/g, n (%)	117 (47.2)	74 (66.1)	$\chi^2 = 11.06$	<0.001
>1 μg/g, n (%)	131 (52.8)	38 (33.9)		

χ^2, Chi-square.

Table 4
Spearman correlations between total mercury concentration in hair and some general characteristics of studied sample (significance level in parentheses).

Characteristic	Quibdo	Paimado
Age (years)	−0.035 (p = 0.585)	0.078 (p = 0.421)
Height (cm)	0.145 (p = 0.024)	0.182 (p = 0.057)
Fish intake[a]	0.021 (p = 0.745)	0.109 (p = 0.252)

[a] Number of times (meals) a week that fish is part of the diet.

Table 1
Demographic characteristics of participants in the study.

Characteristics	Quibdo (n = 248)	Paimado (n = 112)	Statistic	p-value
Age, n[a] (yr, mean ± SEM)	240 (35.9 ± 1.1)	109 (41.81 ± 2.0)	t = 2.810	<0.005
Gender, n (%)				
Male	70 (28.6)	25 (22.3)	$\chi^2 = 1.537$	0.215
Female	175 (71.4)	87 (77.7)		
Fish intake (meals/week), n (%)[a]				
0	3 (0.8)	4 (3.6)		
1–4	208 (88.9)	89 (79.4)	$\chi^2 = 5.541$	0.136
5–10	21 (9.0)	17 (15.2)		
>10	3 (1.3)	2 (1.8)		
Education, n (%)				
None	14 (6.6)	27 (24.1)		
Primary school	27 (12.6)	54 (48.2)		
High school degree	58 (27.1)	24 (21.4)	$\chi^2 = 101.9$	<0.001
Technical school	15 (7.0)	3 (2.7)		
College degree	100 (46.7)	4 (3.6)		
Dental amalgam filling, n (%)				
No	119 (54.1)	77 (68.8)		
Yes	101 (45.9)	35 (31.2)	$\chi^2 = 6.595$	0.010

[a] Number of data; t, t-value; χ^2, Chi-square.

Table 5
Species, trophic level, morphometric parameters and total mercury concentrations in muscle of fish collected from Atrato River.

Common name	Scientific name	Trophic level	n	Weight (g)	Length (cm)	T-Hg (µg/g, fw)[b]
Quicharo	*Hoplias malabaricus*	4.5	46	496.9 ± 19.2	34.6 ± 0.4	0.62 ± 0.08 (0.09–1.96)
Bagre blanco	*Cathorops melanopus*	4.3	6	1103.3 ± 425.0	46.3 ± 5.7	0.47 ± 0.12 (0.24–0.96)
Robalo	*Centropomus undecimalis*	4.2	3	892.0 ± 387.7	45.3 ± 7.3	0.40 ± 0.03 (0.36–0.46)
Barbudo	*Rhamdia quelen*	3.9	21	221.2 ± 29.8	28.3 ± 1.0	0.68 ± 0.10 (0.10–1.75)
Doncella	*Ageneiosus pardalis*	3.8	23	221.9 ± 34.3	29.0 ± 1.4	0.95 ± 0.16 (0.17–2.50)
Mojarra negra	*Caquetaia umbrifera*	3.8	4	343.5 ± 66.1	26.2 ± 2.0	0.29 ± 0.10 (0.06–0.45)
Bagre sapo	*Pseudopimelodus schultzi*	3.7	3	1246.0 ± 156.0	45.4 ± 1.8	2.01 ± 0.51 (1.19–2.94)
Mojarra amarilla	*Caquetaia kraussii*	3.4	44	187.9 ± 9.4	21.3 ± 0.5	0.24 ± 0.04 (0.03–1.14)
Charre	*Pimelodus punctatus*	3.3	14	153.5 ± 10.4	27.7 ± 0.7	0.20 ± 0.05 (0.04–0.60)
Gunguma	*Pimelodus* sp.	3.3[a]	3	364.3 ± 18.8	36.7 ± 0.3	0.21 ± 0.03 (0.16–0.25)
Beringo	*Sternopygus aequilabiatus*	3.3	6	551.8 ± 117.9	87.1 ± 16.2	0.87 ± 0.60 (0.12–3.88)
Denton	*Leporinus muyscorum*	2.2	6	364.0 ± 25.2	32.8 ± 0.8	0.08 ± 0.01 (0.04–0.11)
Bocachico	*Prochilodus magdalenae*	2.2	26	522.2 ± 73.9	32.7 ± 1.2	0.14 ± 0.03 (0.02–0.75)
Guacuco palo	*Spatuloricaria atratoensis*	2.2	16	188.1 ± 30.8	45.8 ± 3.2	0.12 ± 0.02 (0.02–0.30)
Guacuco corroma	*Hemiancistrus wilsoni*	2.2	14	269.2 ± 28.5	30.4 ± 1.2	0.06 ± 0.01 (0.01–0.15)
Boquipompo	*Cyphocharax magdalenae*	2.0	23	181.3 ± 29.5	20.0 ± 0.5	0.06 ± 0.01 (0.02–0.19)
Total			258			

FishBase (http://www.fishbase.org).

[a] This species does not report trophic level in FishBase; therefore, the assigned trophic level was reported for species of the same genus.

[b] Mean ± standard error of the mean (range), fw: fresh weight.

Hazard quotients and CRmw values in fish from Atrato River are presented in Fig. 3. Highest HQ (>10) in fish were reported in species such as *Hoplias malabaricus*, *C. melanopus*, *C. undecimalis*, *A. pardalis, R. quelen, P. schultzi* and *S. aequilabiatus;* and lowest HQ values were observed in *H. wilsoni* and *C. magdalenae*. However, all monitored species displayed HQ values above 1, which indicates that systemic effects may occur if populations are exposed through the diet to these species. In addition, maximum allowable fish consumption rate in meals/week (CRmw) is shown in Fig. 3. The results indicate that the species that can be eaten more than three times a week with low risk for human health are *L. muyscorum, H. wilsoni* and *C. magdalenae* being the first one of the most commercially available in the region.

3.4. Sediment mercury concentrations and SQGs

Concentrations of T-Hg in sediment samples from Atrato River were relatively low, with little variability (Table 6). Sediment sampling sites with T-Hg levels greater than 0.10 µg/g were registered near Quito River, at Quito River mouth (Site 2) (0.12 µg/g) and Quibdo (Site 3) (0.13 µg/g) and in locations receiving mining effluents, Pune River (Site 8) (0.12 µg/g), Buchado (Site 17) (0.11 µg/g) and San Jose (Site 18) (0.14 µg/g). Mercury levels in sampled sediments were greater than the background values (0.085 µg/g) in twenty-two out of the fifty sampling points.

In this study SQGs included CF, TEC and PEC. The sites with greater CF values were San Jose (1.65), Quibdo (1.53), Pune River (1.41) and Quito River (1.41), all located either upstream or midstream the river where most gold mining places are located. However, Hg concentrations measured in sediments from all sampling sites were always lower than TEC (0.18 µg/g) and PEC (1.06 µg/g) values for Hg (Table 6).

3.5. Air mercury levels in Quibdo

The results of air T-Hg monitoring in Quibdo are given in Fig. 4. In general, the concentrations of Hg in air were above the background level (up to 122.5 ng/m^3). These increased from the south to

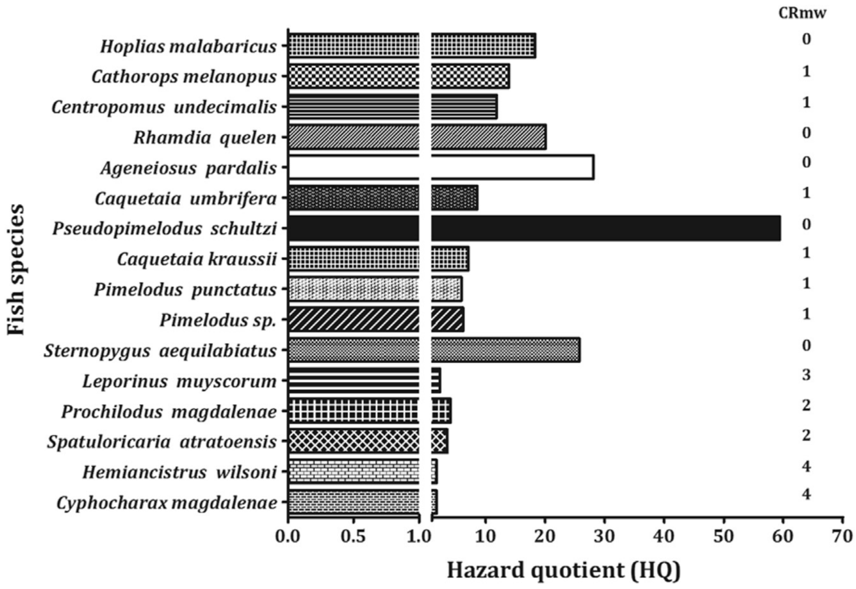

Fig. 3. Hazard quotient (HQ) and maximum allowable fish consumption rate in meals/week (CRmw) for fish from Atrato River, Choco (Colombia).

Table 6
Total mercury concentrations and pollution index in sediment samples from Atrato River.

Station	Sampling site[a]	GPS Location		T-Hg	Contamination factor (CF)	Pollution Level
		N	W	(μg/g, dw)		
1	Cabi	5° 40′ 30,1″	76° 39′ 49,7″	0.05	0.59	No pollution
2	Quito	5° 40′ 31,4″	76° 39′ 51,0″	0.12	**1.41**	Moderate
3	Quibdo	5° 41′ 44,4″	76° 39′ 42,1″	0.13	**1.53**	Moderate
4	Sanceno	5° 44′ 7,4″	76° 41′ 18,5″	0.08	0.94	No pollution
5	Guayabal	5° 45′ 15,5″	76° 40′ 23″	0.09	1.06	Moderate
6	Negua	5° 49′ 4,6″	76° 41′ 18,4″	0.03	0.35	No pollution
7	Tangui	5° 53′ 55,1″	76° 42′ 53″	0.07	0.82	No pollution
8	Pune	5° 53′ 55,1″	76° 43′ 57″	0.12	**1.41**	Moderate
9	Bete	5° 59′ 26,5″	76° 46′ 47,7″	0.08	0.94	No pollution
10	Buey	6° 4′ 34,8″	76° 45′ 25,5″	0.09	1.06	Moderate
11	Bebarama	6° 3′ 18,3″	76° 41′ 30,5″	0.07	0.82	No pollution
12	Bebara	6° 7′ 33,7″	77° 43′ 57″	0.06	0.71	No pollution
13	Tagachi	6°13′4,8″	76° 43′ 9,6″	0.05	0.59	No pollution
14	Padua	6°17′10,7″	76° 45′ 23,5″	0.06	0.71	No pollution
15	Tigre	6°21′ 9,9″	76° 47′ 19,9″	0.10	1.18	Moderate
16	La Boba	6° 23′53,9″	76° 46′ 8,5″	0.09	1.06	Moderate
17	Buchado	6° 25′ 22,7″	76° 46′ 41,3″	0.11	1.29	Moderate
18	San Jose	6° 27,5′1,7″	76° 46′ 13,6″	0.14	**1.65**	Moderate
19	Veracruz	6° 29′ 9,8″	76° 49′ 10,9″	0.09	1.06	Moderate
20	San Miguel	6° 31′ 36,5″	76° 49′ 20,8″	0.09	1.06	Moderate
21	Murri River	6°33′16,9″	76° 50′ 47,8″	0.09	1.06	Moderate
22	Bellavista	6°33′39″	76° 52′ 50,1″	0.09	1.06	Moderate
23	Napipi	6°39′37,8″	76° 56′ 20,5″	0.08	0.94	No pollution
24	Opogado	6°48′26″	76° 58′ 24″	0.08	0.94	No pollution
25	Palaces	6°49′46,9″	76° 55′ 37,7″	0.07	0.82	No pollution
26	Montaño	6°56′41,3″	76° 56′ 22″	0.05	0.59	No pollution
27	Vigia de Curbarado	7°4′37,1″	76° 55′ 33,3″	0.05	0.59	No pollution
28	Curbarado	7° 9′ 34″	76° 57′ 26,8″	0.09	1.06	Moderate
29	Domingodo	7°10′42,2″	77° 1′ 44,2″	0.07	0.82	No pollution
30	Pedeguita	7° 15′ 32,3″	77° 3′ 8,3″	0.07	0.82	No pollution
31	Riosucio	7° 25′13,9″	76° 6′ 27,5″	0.06	0.71	No pollution
32	Truando	7° 25′ 34,8″	77° 6′ 45,3″	0.04	0.47	No pollution
33	La Larga	7° 29′ 37,6″	77° 3′ 54,7″	0.10	1.18	Moderate
34	La Honda	7° 34′ 22,4″	77° 7′ 30,3″	0.07	0.82	No pollution
35	Cacarica	7° 40′ 50,2″	77° 0,8′ 3,7″	0.06	0.71	No pollution
36	Tumaradó	7° 52′ 20″	77° 2′ 9,5″	0.08	0.94	No pollution
37	Leoncito	7° 55′ 26″	77° 00′ 34,2″	0.10	1.18	Moderate
38	Palo Blanco	8° 00′ 52,8″	76° 59′ 59″	0.08	0.94	No pollution
39	Marriaga	8° 6′ 37,8″	76° 57′ 16,9″	0.07	0.82	No pollution
40	Tarena	8° 10′ 46,3″	76° 57′ 14,9	0.09	1.06	Moderate
41	La Orqueta	8° 10′ 52,7″	76° 56′ 34″	0.08	0.94	No pollution
42	Candelaria	8° 10′ 43,5″	76° 54′ 38,1″	0.09	1.06	Moderate
43	El Roto	8° 11′ 27,9″	76° 55′ 55,6″	0.10	1.18	Moderate
44	Yerba Sal	8° 12′ 23,2″	76° 55′ 30,1″	0.09	1.06	Moderate
45	El Cocó	8° 3′ 45,1″	76° 56′ 9,1″	0.06	0.71	No pollution
46	Between Marriaga and La Montañita	8° 8′ 2,9″	76° 57′ 20,7″	0.10	1.18	Moderate
47	La Montañita	8° 9′ 3,2″	76° 57′ 18,1″	0.06	0.71	No pollution
48	Matuntugo	8° 12′ 5,3″	76° 54′ 59,4″	0.09	1.06	Moderate
49	El Calvo	8° 12′ 5,4″	76° 55′ 13,2″	0.07	0.82	No pollution
50	El Rotico	8°12′38,6	76°55′34,4″	0.08	0.94	No pollution

[a] Town, marsh or river-mouth effluent.

the center of the capital of Choco. In the suburban area T-Hg concentrations were 1.1–1.9-fold greater than the background level. However, in the downtown area, T-Hg average concentrations increased significantly with respect to the suburban readings ($P < 0.001$), reaching up to 200.9-fold inside gold shops in the absence of amalgam burning.

4. Discussion

Mercury is a highly toxic element. However, it is used in artisanal small-scale gold mining to extract gold and this is currently the main global source of anthropogenic mercury emissions. This activity generates effects on the environment and human health. Due to the informal character of artisanal and small-scale gold mining, the magnitude of its impact on human health and the extent of the problem are frequently studied in countries like Colombia. However, few studies have dealt with Hg pollution in areas considered global hotspots for biodiversity that must be protected for current and future generations.

4.1. Hair mercury

Mercury amalgamation as a process to extract gold from ore materials is widely utilized in many tropical countries, such as Colombia, with impacts reflected in the populations living near the mining areas (Cordy et al., 2011; Marrugo-Negrete et al., 2008; Olivero and Solano, 1998). This study demonstrated that 52.8% of the volunteers from Quibdo had hair mercury concentrations that exceeded the USEPA threshold dose of 1 μg/g (USEPA, 2005), whereas for people from Paimado, it was 33.9%, suggesting that Hg is being bioaccumulated by local residents. It should be emphasized that 13% of hair T-Hg values in the study area was greater than

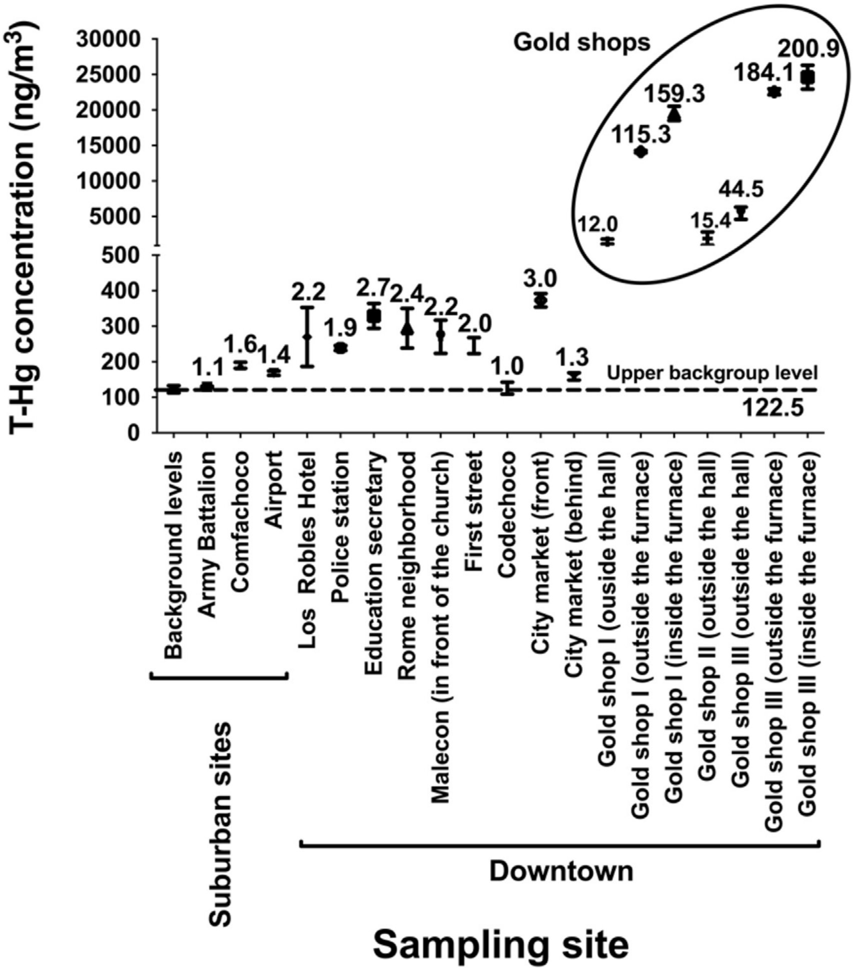

Fig. 4. Distribution of air T-Hg concentrations in Quibdo, Choco. Dotted lines correspond to the background level. The value on top of each point depicts the number of times the average T-Hg reading for the location exceeds the upper background limit.

10 μg/g, denoting that for some individuals Hg exposure may be a health concern. The average hair T-Hg concentration of volunteers in Quibdo (6.72 μg/g) was considerably greater than those reported for inhabitants from South of Bolivar, specifically in La Raya, 5.3 μg/g (Marrugo-Negrete et al., 2008); Achi, 2.4 ± 0.2 μg/g (Olivero-Verbel et al., 2011); and Montecristo, 2.2 ± 0.2 μg/g (Olivero-Verbel et al., 2011), among other places. However, it was lower than that observed in Caqueta River, at the Colombian Amazon with 17.3 μg/g (Olivero-Verbel et al., 2016).

A recent paper on Hg levels in another site of Choco, the San Juan region (Salazar-Camacho et al., 2017), showed a similar median concentration (1.16 μg/g, n = 81) to that reported here (1.26 μg/g, n = 360). This may indicate Hg pollution is homogeneously distributed along this territory. Those authors also measured methylmercury in hair, and highlighted that the portion of T-Hg present as methylmercury ranged from 62 to 92%, typical for methylmercury in hair in riverine populations (78%, Du et al., 2016; 67%, Hong et al., 2016; 80%, Malm et al., 2010).

The relationship of hair T-Hg concentration and age was significant ($P < 0.005$), suggesting an accumulation process is taking place within the population. These results are similar to those reported by Olivero-Verbel et al. (2011), in the southern Bolivar, Colombia. However, the same group (Olivero-Verbel et al., 2016) examined hair Hg levels in indigenous people from Caqueta River, at the Colombian Amazon, also impacted by gold mining, and found no effect of age on hair mercury concentration.

4.2. Fish mercury concentrations and risk-based consumption limits

In terms of Hg exposure routes, it is widely accepted that fish consumption is the main exposure pathway that contributes to overall body burden of Hg, in particular for residents living near rivers contaminated by artisanal mining activities. As carnivorous species are always at a higher trophic level than non-carnivorous species in a food chain. Several studies have reported that Hg concentrations in carnivorous fish are higher than in non-carnivorous species (Castilhos et al., 1998; Reuther, 1994). This was observed in fish collected from Atrato River.

On the other hand, *Hoplias malabaricus, Ageneiosus pardalis, Rhamdia quelen, Pseudopimelodus schultzi* and *Sternopygus aequilabiatus* depicted a T-Hg concentration higher than the maximum recommended by the WHO for human consumption (0.5 μg/g). Moreover, the results reported for T-Hg in *H. malabaricus* from area polluted by gold mining activities in Colombia such as Mina Santa Cruz marsh (0.322 μg/g) (Olivero and Solano, 1998), Ayapel marsh (0.277 μg/g) (Marrugo et al., 2007), Grande marsh (0.58 μg/g) (Marrugo-Negrete et al., 2008) were lower than those observed in this study (0.62 μg/g), while in the Amazon were higher (0.72 μg/g) (Olivero-Verbel et al., 2016) compared to this study.

4.3. Sediment mercury concentrations and pollution index

Elevated concentrations of Hg in sediments are usually found near immediate surroundings of gold processing sites, as a result of Hg physically lost by the miners during the amalgamation process. The sediments collected along Atrato River showed lower T-Hg concentrations, as those sites were located several kilometers downstream mining activities. This result is coincident with that reported for Malinowski-Tambopata River, Southeastern Peruvian Amazon (0.02–0.053 μg/g) (Moreno-Brush et al., 2016), also a rainforest with large precipitations and fast-flowing currents that allows a rapid Hg dilution and fugacity from contaminated sites (Navarro et al., 2009), resulting in low Hg concentrations in sediments downstream along the basin. It is clear that Hg pollution is usually very high within mining operation sites (Olivero-Verbel et al., 2015), but it usually decreases with the distance (Niane et al., 2014), generating low concentrations far from point sources (Chen et al., 2016; Haris et al., 2017). Other possible factors determining Hg concentrations in sediments, in addition to distance and dispersion due to heavy rainy events, include the type of organic matter (Maia et al., 2009) and the river depth (Van Straaten, 2000), among others.

4.4. Air mercury concentrations

In general, the presented results demonstrate that in Quibdo, T-Hg air levels in suburban areas are considered low. However, in places near downtown where amalgam burning is taking place, the concentrations reach maximum values inside gold shops where amalgam burning takes place. Inside gold shops, high T-Hg levels in air were recorded in the absence of amalgam burning (up to 24610 ± 614 ng Hg/m^3), suggesting exposure during active burning may be very high. However, the concentrations described in this study are lower than those reported in a study carried out in five municipalities of Antioquia (Segovia, Remedios, Zaragoza, El Catre, and Nechi), where urban air mercury levels were 300 ng/m^3 (background level) and in gold shops it reached 1 million ng/m^3, with values of 10.000 ng/m^3 in residential areas (Cordy et al., 2011). The health problem is not only for gold shop workers, exposure also occurs outdoors when air extractors are turned on or when entrance doors are open, representing a potential risk for Hg exposure in the surrounding communities (Steckling et al., 2011).

Data presented here detailed Hg pollution in several environmental compartments of one of the global biodiversity hotspots in the biogeographic Choco. Immediate actions should be carried out by the government, local authorities and the community, not only to avoid further Hg accumulation in humans through the diet, but also preventing Hg effects in the fragile biota, as well as other harmful consequences of gold mining, such as forest destruction and contamination.

5. Conclusions

This is the first report regarding Hg concentrations in Atrato River. In summary, hair mercury concentrations in people from this biodiversity hotspot were similar to those found other gold mining areas in Colombia. Fish have biomagnified Hg, especially those from high trophic levels, although for all species average HQ was greater than one, suggesting diet is a potential source of Hg contamination for the population. Mean Hg levels in sediments was 0.081 μg/g, with concentrations below TEC (0.18 μg/g) and PEC (1.06 μg/g) values, although according to CF, 44% of the samples had moderate pollution. Dilution due to heavy raining may be a key factor to the relatively low observed Hg concentrations. In gold mining areas such as Choco, Hg exposure also occurs by inhalation of Hg vapors from burning amalgams at gold shops, and that possesses a risk for urban residents, but also for the nearby forest. Data presented here showed gold mining releases Hg into the environment, reaching sediments, air, fish and humans, making Hg pollution a widespread problem in Choco, threatening its biodiversity and human health.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.chemosphere.2017.10.160.

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