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A human health risk assessment of methylmercury, arsenic and metals in a tropical river basin impacted by gold mining in the Colombian Pacific region

Carlos Salazar-Camacho [a], Manuel Salas-Moreno [a], Siday Marrugo-Madrid [c], Roberth Paternina-Uribe [b], José Marrugo-Negrete [b,**], Sergi Díez [c,*]

[a] Biosistematic Research Group, Biology Department, Faculty of Naturals Sciences, Universidad Tecnológica Del Chocó, Quibdó, Colombia
[b] Faculty of Basic Sciences, Universidad de Córdoba, Carrera 6 No. 76-103, Montería, Córdoba, Colombia
[c] Department of Environmental Chemistry, Institute of Environmental Assessment and Water Research, IDAEA-CSIC, E-08034, Barcelona, Spain

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ABSTRACT

The Atrato River basin is one of the most biodiverse areas worldwide, and paradoxically, it is one of the sites in Colombia with the highest environmental impact from gold mining. This study assessed the distribution of Hg, As, Pb, and Cd in 47 fish species (n = 1372) and the accumulative human health risk in inhabitants (n = 2325) from 13 municipalities located along the Atrato River basin. The results revealed that Hg and As in fish present a high potential human health risk based on their mean concentrations. Estimated daily intake (EDI) calculations showed that humans could present detrimental health effects, while that target hazard quotient (THQ) above 1 showed that the exposed population might experience noncarcinogenic health risks, mainly from the accumulative effects of Hg (80.4%) and As (18.2%). The species that would most affect the health of the inhabitants are carnivorous *H. malabaricus*, *A. pardalis, P. schultzi, R. quelen,* and *C. kraussii,* which are among the fourteen most consumed in the region. These species had values of estimated weekly intake (EWI) above the provisional tolerable weekly intake thresholds for MeHg (PTWI of 1.6 and 3.2 μg/kg bw/week for adults and children, respectively) in 7 of the 13 localities evaluated. According to the surveys, the calculated weekly allowable fish amount (MFW) showed that carnivorous fish may generate adverse effects on the consumers because the allowed MeHg is about 2 times higher than the upper reference limit. Other results indicate a significant carcinogenic health risk, mainly from As, in 8 of the 13 localities evaluated. Due to the high rates of unsatisfied basic needs and the monetary poverty in the region, the possibility that inhabitants can replace fish as the principal source of protein is low. Therefore, a food guidance is required to avoid risks, obtain nutritional benefits, and sustain fish populations.

1. Introduction

Metal contamination in the environment is of great concern due to its bioaccumulation, biomagnification, and eventual toxic effects on biological systems. The growing impact of metal toxicity is an unavoidable reality from an environmental, ecological, and nutritional point of view (Nagajyoti et al., 2010; Rahman et al., 2019). Metal contamination in aquatic ecosystems is a problem of great importance, because some elements such as mercury (Hg), lead (Pb), arsenic (As), and cadmium (Cd), are toxic even at low concentrations and have great bioaccumulation capacity. The U.S. Agency for Toxic Substances and Disease Registry (ATSDR) ranks those as four first, second, third, and seventh respectively in its 2017 Priority List of Hazardous Substances (ATSDR, 2017). Mercury is one of the most critical metals since it is a global pollutant that affects the health of humans and ecosystems due to its high toxicity, persistence, high bioaccumulation, and great atmospheric distribution capacity (Cheng and Hu, 2012; Cao, 2019). Gold mining, specifically its extraction by amalgamation with Hg, is the anthropogenic activity that produces the highest levels of Hg contamination in soil and water, this activity is estimated to generate the emission of about 880 tons of Hg

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* Corresponding author.
** Corresponding author.
E-mail addresses: jmarrugo@correo.unicordoba.edu.co (J. Marrugo-Negrete), sergi.diez@idaea.csic.es (S. Díez).

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each year (UNEP, 2013; Kocman et al., 2017; Obrist et al., 2018). Other elements associated with gold minerals such as Cd, Pb and As, can be dispersed due to erosion and chemical weathering of tailings (Da Silva et al., 2004).

Gold mining in Colombia uses around 200 tons of Hg, which generates emissions between 30- and 70-tons of Hg (Telmer and Veiga, 2009; UPME, 2014). In the department of Chocó, gold exploitation has been for many years a significant socioeconomic activity in many communities; there are places where this activity is the basis of the economy, at the same time, these activities have generated environmental impacts on water sources such as the Atrato River. It is estimated that 150 tons of Hg are used in the exploitation of gold, where a large amount of mining waste goes to the Atrato River or its tributaries, contaminating the sediments (UPME, 2014; Palacios et al., 2018). In the river sediments, an important part of this Hg has been transformed to methylmercury (MeHg) (Gutiérrez et al., 2020), that due to its high affinity for S-containing molecules and its ability to cross membranes, it can bioaccumulate in freshwater fish species at toxic concentrations (Zillioux, 2014; Cariccio et al., 2019; Buck et al., 2019). MeHg is the most toxic form of Hg, which can have harmful effects on the human body if fish is consumed in high quantities (Fuentes-Gandara et al, 2018), causing damage to the nervous system in the early stages of development, leading to alterations in structure and function (Clarkson et al., 2003; Cao, 2019).

Despite the large number of years of gold mining activity in Chocó, little research shows the impacts that this practice has had on the human health of the communities that inhabit the Atrato River basin (Salazar et al., 2017, 2021; Gutierrez et al., 2020). Therefore, the objectives of this study were: (1) to determine the concentrations of MeHg and toxic metals (i.e. Hg, As, Cd, Pb) in muscle tissue in fish species of high consumption and commercial interest from the Atrato River basin, (2) determine the target hazard quotient (THQ) and total THQ to assess the non-carcinogenic and carcinogenic risk of metals from the consumption of fish; and (3) to evaluate the potential health risks associated with the consumption of contaminated fish with MeHg, either establishing the estimated weekly intake (EWI) and comparing it with the PTWI or by estimating the pollution index (Pi) in the fish, to provide information on the safety of fish consumption in the inhabitants along the municipalities of this basin. The results of this research provide important data regarding the degree of risk to human health in the inhabitants living along the banks of the Atrato River basin, this information could be the starting point to establish public strategies that control the intake of fish and can reduce the health risk in these communities based on the T-622 judgment of 2016.

2. Materials and methods

2.1. Study area

The Atrato River basin is in the west of Colombia, in the Chocó and Antioquia departments (Fig. 1). It is born in the Cerro del Plateado in the municipality of "El Carmen de Atrato", western Andes mountain range, and ends in the Gulf of Urabá, in the Caribbean Sea; has an area of 35,700–36,400 km^2, a length of 750 km, a variable width between 150 and 500 m and a depth of 31–38 m. The basin is constituted by great water bodies, forests, wetland marshes, lands used for agriculture, pastures, and many rural communities (Palomino et al., 2019). It has one of the most abundant average flows worldwide, approximately 4137 m^3/s; with an average annual precipitation of 5000 mm/year reaching up to 12,000 mm/year and an annual average temperature of 26 °C (Restrepo, 2006; Palomino et al., 2019; Velásquez and Poveda, 2019). It receives more than a hundred rivers along its course, among the most importants are Truandó, Quito, Beté, Bojayá, Bebaramá, Tagachí, Buchadó, Bebará, Neguá, and Cabí; of which Quito and Neguá are the largest contributors of Hg-contaminated mining waste to the Atrato River basin (Codechocó, 2012; UNODOC, 2016).

The inhabitants of the Atrato River basin base their economy on gold mining, agriculture, livestock raising, and fishing. Our study was conducted in 13 areas of the tropical geographic basin of the Atrato River. The areas were: Quibdó(QD), Carmen Atrato (CA), Bagadó (BO), Rio Quito (RQ), Medio Atrato (MA), Riosucio (RS), Bojaya (BY), Murindó (MO), Cañasgordas (CG), Dabeiba (DB), Vigía del Fuerte (VF), Turbo (TB), and Unguía (UG), selected because they are important places where gold mining is practiced (Codechocó, 2012; Sentencia T-622, 2016). The fish samples were captured in the Atrato River, as well as in marshes and rivers, and the tributaries of these, within the mentioned areas. Fish are from representative and consumed species, and are present in the fishery throughout the year. Most of the municipalities studied along the Atrato River basin, are located in areas of difficult access, with a warm humid climate, where it rains all year, very jungle areas. Likewise, the presence of illegal armed groups, hindered the sampling work and population surveys, and required months of previous work before studying these areas.

2.2. Sampling

All fish samples were collected by fishermen employed by the project using trammel-net, cast nets, fishing pens, and fishing rods between June and December of 2019. In total 1372 samples of 47 different species were collected, the carnivorous species (N = 1014) were *Oligoplites saliens, Geophagus crassilabris, Caranx hippos, Micropogonias furnieri, Trichiurus lepturus, Chloroscombrus chrysurus, Eugerres plumieri, Caranx crysos, Trachelyopterus insignis, Haemulon boschmae, Umbrina broussonnetii, Centropomus undecimalis, Gerres cinereus, Caranx hippos, Scomberomorus sierra, Trachelyopterus fisheri, Pimelodella chagresi, Sternopygus macrurus, Brycon moorei, Andinoacara pulcher, Brycon amazonicus, Oreochromis mossambicus, Pimelodus* sp, *Cynopotamus atratoensis, Pimelodus maculatus*, *Hoplias malabaricus, Ageneiosus pardalis, Trachinotus falcatus, Pseudopimelodus schultzi, Lesporinum muyscorum, Trachelyopterus fisheri, Gymnotus henni, Andinoacara biseriatus, Ctenolucius beani, Caquetaia umbrifera, Caquetaia kraussi, Astyanax fasciatus, Pimelodus punctatus,* and *Rhamdia quelen*; and the non-carnivorous (N = 358) were *Colossoma macropomum, Oreochromis niloticus, Symphysanodon berryi, Mugil incilis*, *Cyphocharax magdalenae, Hemiancistrus wilsoni, Hypostomus hondae,* and *Prochilodus magdalenae*. The fish samples were packaged in polyethylene bags, were placed in ice-cold styrofoam coolers, and transported to the Toxicology and Environmental Management laboratory of the University of Córdoba (Colombia). Subsequently, the fish samples total length and weight were measured, and then a portion of the dorsal muscle, of approximately 10 g, was cut out with a ceramic knife of ceramics and immediately stored and frozen until Pb, Cd, As, Hg, and MeHg were determined (UNEP, 1990). Fish species were identified using specialized classification keys (Fishbase, 2020), and thanks to the expertise of fishermen and ichthyologist involved in the sampling campaign.

2.3. Analysis of THg, MeHg, As, Pb, and Cd

The analytical method used to determine Hg concentration was based on thermal decomposition with detection by atomic absorption spectrometry using a Direct Mercury Analyzer (DMA-80 TRICELL, Milestone Inc, Italy) as stated by the EPA method 7473 (EPA, 1998). For Cd and Pb analysis, samples were digested with HNO_3/HCl (1:3 v/v) using Method 3051 A (EPA, 2007) and according to the procedure described by Karadede and ÜnlÜ (2007), respectively. Cd and Pb analyses were performed using a Thermo Elemental Solaar S4 - graphite furnace method. The analysis of As was carried out by calcining a mixture of 1 g of fish sample with $Mg(NO_3)_2$ at 550 °C in a muffle furnace, then 1 mL concentrated HNO_3 was added and heated to dryness, later it was dissolved with HCl 4.5 N, it was filtered with a 0.45 μm filter, then it was refilled to 25 mL with distilled water (Szkoda et al., 2006). A Thermo Scientific iCE™ 3500 AAS Atomic Absorption

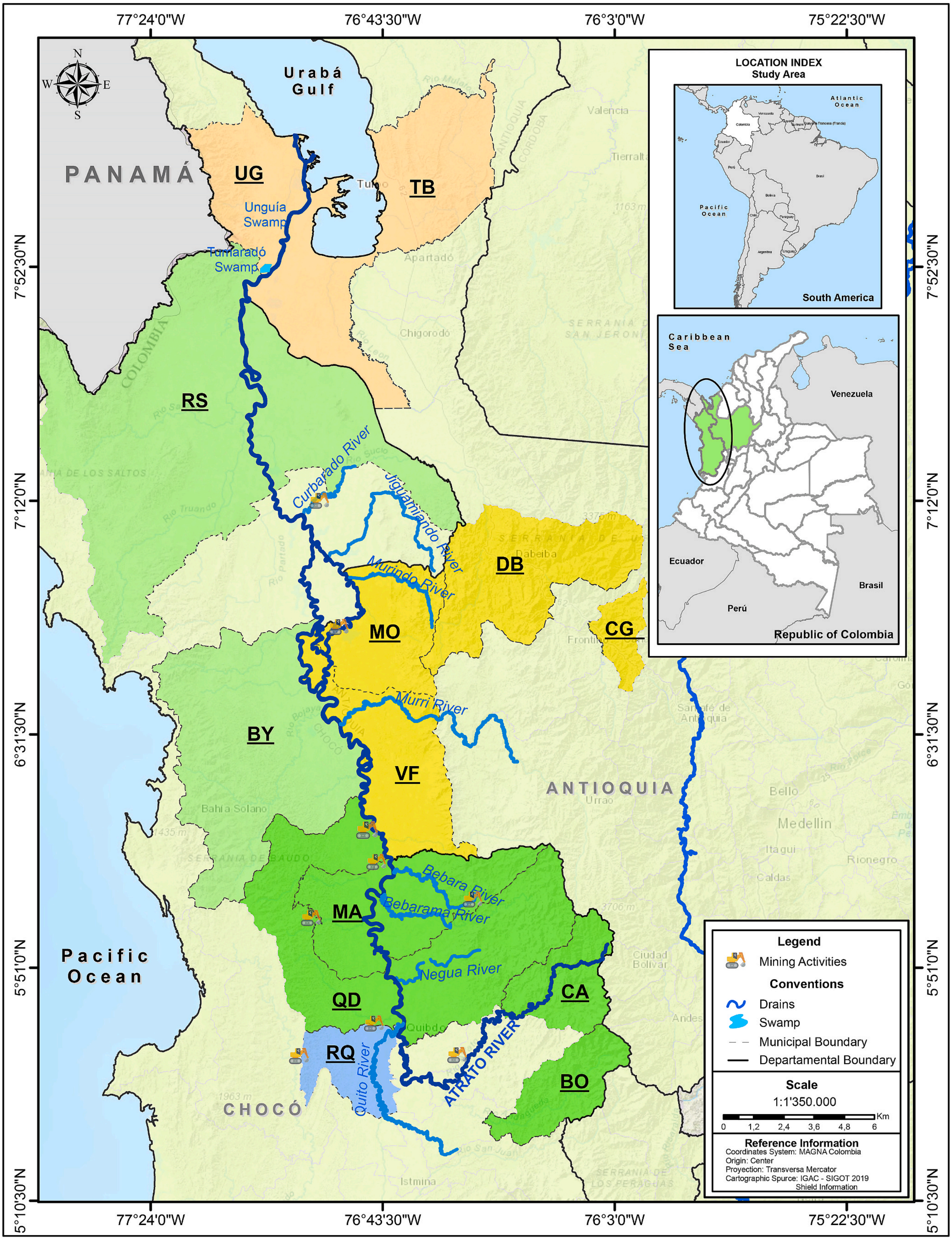

Fig. 1. Map of the 13 municipalities studied influenced by the Atrato River basin (Chocó-Colombia): Carmen Atrato (CA), Bagadó (BO), Rio Quito (RQ), Quibdó (QD), Medio Atrato (MA), Bojayá (BY), Vigía del Fuerte (VF), Murindó (MO), Dabeiba (DB), Cañasgordas (CG), Riosucio (RS), Unguía (UG), and Turbo (TB).

Spectrometer coupled to a VP100 Continuous Flow Vapor Generator (Waltham, MA, USA) (HGAAS; Standard Methods SM 3114, 2017) was used for the analysis of As. The methods were validated with IAEA-405 and DORM-2, and the recovery average percentages ($n = 3$) was 96.2%. MeHg was extracted from fish samples with hydrobromic acid and toluene. The extract was then mixed with a solution of L-cysteine. An aliquot of 100 μL was taken from the aqueous phase for analysis in DMA 80 TriCell Milestone (Cordeiro et al., 2013). The method was validated with DORM-2 and the recovery percentage was 99% ($n = 3$). The detection limits for the different metals were 0.014 μg/g for Hg, 0.006 μg/g for Cd, 0.010 μg/g for Pb, 0.016 μg/g for As and 0.007 μg/g for MeHg.

2.3.1. Estimated daily intake (EDI)

To evaluate the risk by daily intake of metal(loid)s from fish consumption, it was assumed that the ingested dose was equal to the absorbed pollutant dose (USEPA, 1989); cooking had no effect on the pollutants (Chien et al., 2002) and people who lived in Atrato River basin eat 256 g fish per day. Therefore, the EDI of above-mentioned elements for adults was calculated as follows:

$$EDI = \frac{C\ x\ C_{cons}}{Bw} \tag{1}$$

where C is the concentration of the elements in fish (μg/g ww), C_{cons} is the average daily consumption of fish in the local area (256 g/day), and Bw represents the body weight according to surveys in each municipality.

2.3.2. Determination of target hazard quotient (THQ)

The THQ (HQ/RfDo) represents the risk of noncarcinogenic effects. If THQ is less than 1, it indicates that the hazard quotient (HQ) is below the reference dose (RfDo), and therefore daily exposure at this level is unlikely to cause adverse effects during a person's lifetime. THQ calculation was performed using the assumptions from the integrated US EPA risk analysis (USEPA, 2000). THQ was determined using the following equation (Chien et al., 2002):

$$THQ = \frac{EFr\ x\ ED_{tot}\ x\ FIR\ x\ C}{RfDo\ x\ B_w\ x\ ATn}\ x\ 10^{-3} \tag{2}$$

where EFr is the exposure frequency (350 days/year); $EDtot$ is the exposure duration (30 years); FIR is the food ingestion rate (g/day), and 10^{-3} is the unit conversion factor (kg/g); C is the element concentration in fish (μg/g ww); $RfDo$ is the oral reference dose (mg/kg-day); Bw is the average adult body weight according to surveys in each municipality; and ATn is the average exposure time for noncarcinogens (365 days/year × number of exposure years, assuming 30 years). The total THQ (TTHQ) was expressed as the sum of the THQ values for each element (Chien et al., 2002):

$$Total\ THQ\ (TTHQ) = THQ_{Hg} + THQ_{As} + THQ_{Pb} + THQ_{Cd} \tag{3}$$

2.3.3. Carcinogenic risk assessment (CR)

CR is the possibility of an individual developing any type of cancer in its lifetime due to exposure to carcinogenic hazards (Adimalla, 2020; Zhaoyong et al., 2019). Carcinogenic health risks for an individual element over a lifetime was calculated according to the following equation (USEPA, 1989, 2002):

$$CR = HQ\ x\ SF = \left[\frac{EFr\ x\ ED_{tot}\ x\ FIR\ x\ C}{B_w\ x\ ATn}\ x\ 10^{-3}\right]\ x\ SF \tag{4}$$

$$TCR = \sum CR \tag{5}$$

Where HQ (mg/kg/day) is the chronic daily dose of toxic elements received through the fish intake, CR is the carcinogenic risk, TCR is the total carcinogenic risk, and SF is the slope factor (mg/kg/day), 1.5 for As and 0.0085 for Pb. According to USEPA (1989), when CR and TCR values are less than 1×10^{-6}, the risk is regarded negligible, and if CR and TCR exceed the 1×10^{-4}, there is likely to be a risk to human health.

2.3.4. Assessment of human health risk related to methylmercury

In this assessment, 2325 volunteers replied to a questionnaire in which they indicated their gender, educational status, average body weight, and dietary habits. In particular, the questionnaire included questions about the habitual intake of fish, focusing on the frequency they consumed fish, including the number of fish meals consumed per week and the type of fish. The general characteristics of the population surveyed (i.e. general population (GP) and women of childbearing age (WCHA)) are presented in Table 1.

The potential risk of human exposure to MeHg was assessed with the estimated weekly intake (EWI - μg/bw/week) using the equation described by UNEP (2010):

$$EWI = \frac{IR\ x\ C}{Bw} \tag{6}$$

Where C is the median concentration of MeHg (μg/kg) in fish, IR is the weekly intake (g/week) of fish, and Bw is the bodyweight of the person (kg). The IRs were calculated taking into account the consumed portion of fish (g/day) and the frequency of consumption (days/week) in the thirteen municipalities of the Atrato region.

The concentration of MeHg that the consumed fish species should contain to avoid exceeding the provisional tolerable weekly intake (PTWI) (FAO/WHO, 2017) was calculated by the following equation:

$$[MeHg]_{permissible} = \frac{C\ x\ PTWI}{EWI} \tag{7}$$

Where C is the median concentration of MeHg (μg/kg) in fish and PTWI is a reference value of 1.6 μg/kg bw/week for women of childbearing age and children, and 3.2 μg/kg bw/week for the adult population. Considering the MeHg concentrations of the fish consumed by the inhabitants of the Atrato region, we estimated the maximum amount of fish that a person can consume weekly (MFW) without producing harmful health effects, according the following equation:

$$MFW = \frac{PTWI\ x\ IR}{EWI} \tag{8}$$

Finally, to calculate the degree of Hg contamination in the most consumed fish species, we used the formula proposed by Zhang et al. (2019):

$$P_i = \frac{C_i}{S_i} \tag{9}$$

Where Pi is the pollution index, C_i and S_i are the median concentration of the metal in the fish muscle and the value of the evaluation criteria, respectively. Two reference limits were used: 200 μg/kg ww (WHO, 2008) for vulnerable populations such as children under 15 years of age and women of childbearing, and a threshold of 500 μg/kg ww (WHO, 1990) for the adult population.

2.3.5. Data analysis

Kolmogorov-Smirnov ($n \geq 50$) and Shapiro-Wilk ($n < 50$) tests were used to assess whether data followed or not a normal distribution. The Kruskal-Wallis test was employed to evaluate the differences among Hg, As, Pb, and Cd concentrations between fish species. Spearman's test was performed to evaluate the correlation between the concentration of the elements and the trophic level of the fish. A p-value of 0.05 was chosen to indicate statistical significance. THg, MeHg, As, Pb, and Cd concentrations were expressed as μg/kg ww of fish. The statistical analyzes were carried out using the R Project statistical program version 3.6.1.

Table 1
General characteristics of the general population (GP) and women of childbearing age (WCHA) groups in the Atrato River basin, Colombia (n = 2325).

		GP						WCHA			
Municipality	**weekly fish intake (days)**	**n (male)**	**n (female)**	**Age (years)**	**Range**	**Weigth (kg)**	**Range**	**n**	**Age (years)**	**Weigth (kg)**	**range**
Carmen de Atrato	1.9 ± 0.0	40	91	40.8 ± 18.9	15–84	61.8 ± 11.2	42.1–105.9	59	30.5 ± 10.2	60.6 ± 13.0	42.1–105.9
Bagadó	3.0 ± 0.1	76	93	48.1 ± 18.7	15–90	69.9 ± 15.2	40.2–170.5	48	32.2 ± 9.2	68.3 ± 12.6	42.6–98.9
Río Quito	2.6 ± 0.4	54	108	44.2 ± 17.6	15–92	71.4 ± 15.9	41.0–161.9	66	33.3 ± 9.5	72.0 ± 12.0	48.1–108.5
Quibdó	2.6 ± 0.5	59	197	43.0 ± 17.5	15–89	73.9 ± 17.1	40.6–168.4	125	31.1 ± 9.2	73.5 ± 17.5	43.8–136.6
Medio Atrato	2.7 ± 1.1	87	119	42.2 ± 17.8	15–88	69.8 ± 16.0	41.1–175.6	74	32.0 ± 10.5	67.5 ± 15.4	43.3–120.1
Bojayá	3.4 ± 0.6	70	86	40.9 ± 16.3	15–80	72.6 ± 15.5	43.5–144.4	60	30.1 ± 9.6	69.5 ± 18.4	43.5–144.4
Vigía del Fuerte	2.7 ± 2.4	95	134	42.9 ± 17.9	15–82	71.3 ± 13.2	41.8–115.7	85	32.5 ± 9.9	69.5 ± 13.2	44.6–101.6
Murindó	2.8 ± 2.1	69	105	42.9 ± 18.0	15–89	68.1 ± 14.4	40.0–113.5	76	30.3 ± 10.7	66.8 ± 16.3	41.3–109.7
Dabeiba	1.9 ± 0.0	65	113	42.4 ± 19.8	15–93	65.0 ± 13.7	38.9–113.3	75	28.5 ± 9.9	64.8 ± 14.6	39.1–113.3
Cañasgordas	2.0 ± 0.4	43	125	50.7 ± 20.7	16–99	66.5 ± 12.5	38.8–98.5	59	30.7 ± 11.0	64.4 ± 12.4	40.1–91.2
Riosucio	2.8 ± 0.4	54	106	38.8 ± 17.2	16–83	68.2 ± 14.8	40.2–106.9	84	28.6 ± 9.8	66.7 ± 14.7	43.4–106.3
Unguía	3.0 ± 0.8	54	92	41.2 ± 18.4	15–90	71.8 ± 16.2	30.4–124.9	64	30.8 ± 8.5	70.3 ± 13.0	43.8–109.2
Turbo	2.5 ± 0.2	78	112	40.4 ± 17.1	15–88	69.5 ± 15.4	42.9–113.9	81	30.6 ± 9.4	67.7 ± 15.2	43.5–112.8

The order of the municipalities is from upstream to downstream in the Atrato River.

3. Results

3.1. Metals and As in fish muscle

The Hg, As, Pb, and Cd concentrations, feeding habits, and trophic level of species collected in the Atrato River Basin are shown in Table S1 and Fig. 2. According to the classification made by Fishbase (2021), 47 fish species (n = 1372) were collected. Of these, 8 were piscivorous (238 individuals or 17.3% of the sample), 14 carnivorous (207 individuals or 15.1% of the sample), 8 omnivores with a tendency to carnivore (403 individuals or 29.4% of the sample), 2 omnivores with a preference for fish and plant material (44 individuals or 3.2% of the sample), 7 omnivores (122 individuals or 8.9% of the sample), 7 detritivorous (356 individuals or 25.9% of the sample), and 1 planktivore (2 individual or 0.15% of the sample). The most frequently collected species were *Prochilodusmagdalenae* (11.7%), *Astyanax fasciatus* (10.5%), *Hoplias malabaricus* (9.0%), *Hipostomus hondae* (8.9%), and *Rhamdia quelen* (8.2%). The concentrations of Hg were highest, followed by As, Pb, and Cd. Among the analyzed fish samples, the lowest concentrations of Hg were found in *Brycon amazonicus* (9.31 ± 2.35 μg kg^{-1}), *Oreochromis niloticus* (6.45 ± 7.41 μg kg^{-1}), and *Oreochromis mossambicus* (4.51 ± 5.75 μg kg^{-1}), and the highest concentration was registered in the piscivorous species *Ageneiosus pardalis* (688.85 ± 332.24 μg kg^{-1}). Of the total number of fish samples analyzed for Hg, 36.9% (506 individuals, 473 with carnivorous and 33 with non-carnivorous habits) exceeded the limit for populations at risk (set in 200 μg kg^{-1}) (WHO, 2008). Of these, 14.1% (201 individuals, 194 with carnivorous and 7 with non-carnivorous habits) surpassed the maximum recommended limit for human consumption (established in 500 μg kg^{-1}) (WHO, 1990). Carnivorous fish species with the higher number of samples exceeding these thresholds were *Hoplias malabaricus* and *Ageneiosus pardalis* with 102 and 61 samples respectively. The municipalities with the higher proportion of samples of *Hoplias malabaricus* exceeding both limits were MO, where all samples (n = 10) exceeded the threshold of 500 μg kg^{-1}, QD (n = 15 > 200 μg kg^{-1}, of these, n = 11 > 500 μg kg^{-1}), MA (n = 16 > 200 μg kg^{-1}, of these, n = 8 > 500 μg kg^{-1}). For *Ageneiosus pardalis* were MO (n = 21 > 200 μg kg^{-1}, of these, n = 19 > 500 μg kg^{-1}), VF (n = 6 > 200 μg kg^{-1}, of these, n = 5 > 500 μg kg^{-1}), QD (n = 12 > 200 μg kg^{-1}, of these, n = 10 > 500 μg kg^{-1}) and RS (n = 18 > 200 μg kg^{-1}, of these, n = 11 > 500 μg kg^{-1}). For non-carnivorous, the species with the higher number of samples exceeding these thresholds were *Prochilodus magdalenae* and *Leporinus muyscorum* with 11 and 7, respectively; and the municipalities with the higher proportion of samples exceeding the Hg WHO's limits were RQ (n = 6 > 200 μg kg^{-1}, of these, n = 4 > 500 μg kg^{-1}) to *Prochilodus magdalenae* and RS (n = 4 > 200 μg kg^{-1}) to *Leporinus muyscorum*. The levels of As, Pb, and Cd for all samples, ranged between < LD - 1,617.37 μg kg^{-1}, <LD - 1,111.27 μg kg^{-1}, and <LD – 59.09, respectively. None of the mean concentrations of the 47 fish species reported in this study were higher than the permissible maximal levels for As, Pb, and Cd in fish muscle of 1000, 300, and 100 μg kg^{-1}, respectively (FAO/WHO, 2002; FAO/WHO, 2009). However, some samples had mean concentrations close to half the permissible value for arsenic as *Caranx crysos* (514.58 ± 98.35), *Gerres cinereus* (503.19 ± 386.82), *Sphyraena guachancho* (485.63 ± 144.39), *Symphysanodon berryi* (484.06 ± 18.31), and *Scomberomorus sierra* (461.76 ± 78.13). Also, some specimens surpassed these limits. One sample of *Prochilodus magdalenae* (1,617.37 μg kg^{-1}), in the UG municipality, exceeded the arsenic threshold; two in UG (*Ctenolucius beani* –1,617.37 μg kg^{-1} and *Cyphocharax magdalenae* – 1,111.27 μg kg^{-1}), one in QD (*Astyanax fasciatus* - 407.69 μg kg^{-1}) and another in MO (*Astyanax fasciatus* - 427.85 μg kg^{-1}) exceeded the Pb threshold. The varying distribution pattern, especially for Hg and As, shows that the feeding habit of a particular fish species exerts an influence on the type of exposure to contaminants (Fig. 2a and b). The sample proportion with concentrations lower than detection limits (2.12 μg kg^{-1} for Cd, 8.68 μg kg^{-1} for Pb, and 5.42 μg kg^{-1} for As) was 83.5% for Cd, (56.2%) for Pb and 12.4% for As. Significant differences in Hg, As, Pb, and Cd concentrations (KW = $p < 0.05$) were found between fish species.

3.2. Most consumed fish species in the Atrato River basin

A survey applied to the inhabitants of the Atrato River basin showed

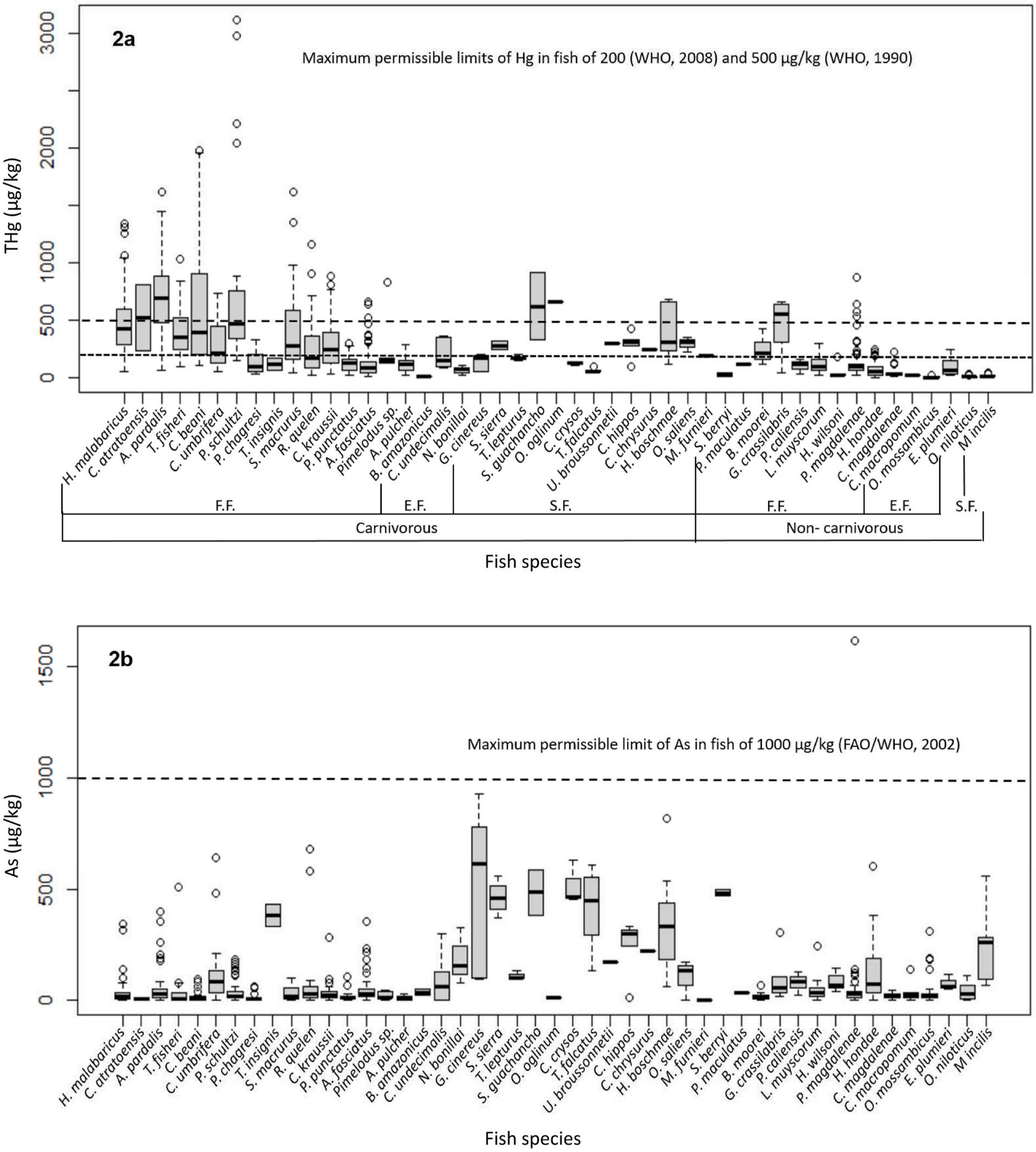

Fig. 2. Concentrations of Hg (2a), As (2b), Pb (2c) and Cd (2d) (μg kg^{-1} ww) in fish of the Atrato region. Boxes depict median values (P50), whiskers (P75) and circles are outliers. Dashed line refers to the FAO/WHO recommended thresholds. Freshwater fish (F.F), estuarine fish (E.F), sea fish (S.F.).

that, of the 47 fish species collected, 14 were the most commonly eaten by them (Fig. S1). Among these, six species of non-carnivorous habits stand out: *P. magdalenae*, *L. muyscorum*, *O. niloticus*, *O. mossambicus*, *Hypostomus hondae*, and *Colossoma macropomum*. It should be highlighted that the four species most consumed by people in the Atrato River basin are *P. magdalenae*, *H. malabaricus*, *Caquetaia kraussii,* and *A. pardalis* with 70, 38, 23, and 21% of the surveyed inhabitants (n = 2325).

3.3. Determination of human health risk by fish consumption

The general characteristics of the studied population in the Atrato River basin are shown in Table 1. In it can be seen that 63.7% (n = 1481) of the respondents were women and 36.3% (n = 844) men. In addition, it is shown that the average age is between 39 and 51 years (range: 15.0–99.3 years), and the average weight is between 62 and 74 kg (range: 30–176 kg). Based on the survey information of this study, the mean amount of fish consumed in the Atrato River basin was 256 g/day; and considering the mean frequency with which the inhabitants eat the most consumed fish (2.7 day/week), the total weight of fish consumed per year was more than 36 kg yr^{-1}. The most consumed items were *P magdalenae*, *H. hondae*, *H. malabaricus*, *R. quelen*, *C kraussii,* and *A. fasciatus* (average consumption of 2.8–3.4 day/week). The risk by fish consumption was calculated using the metal pollution indexes EDI,

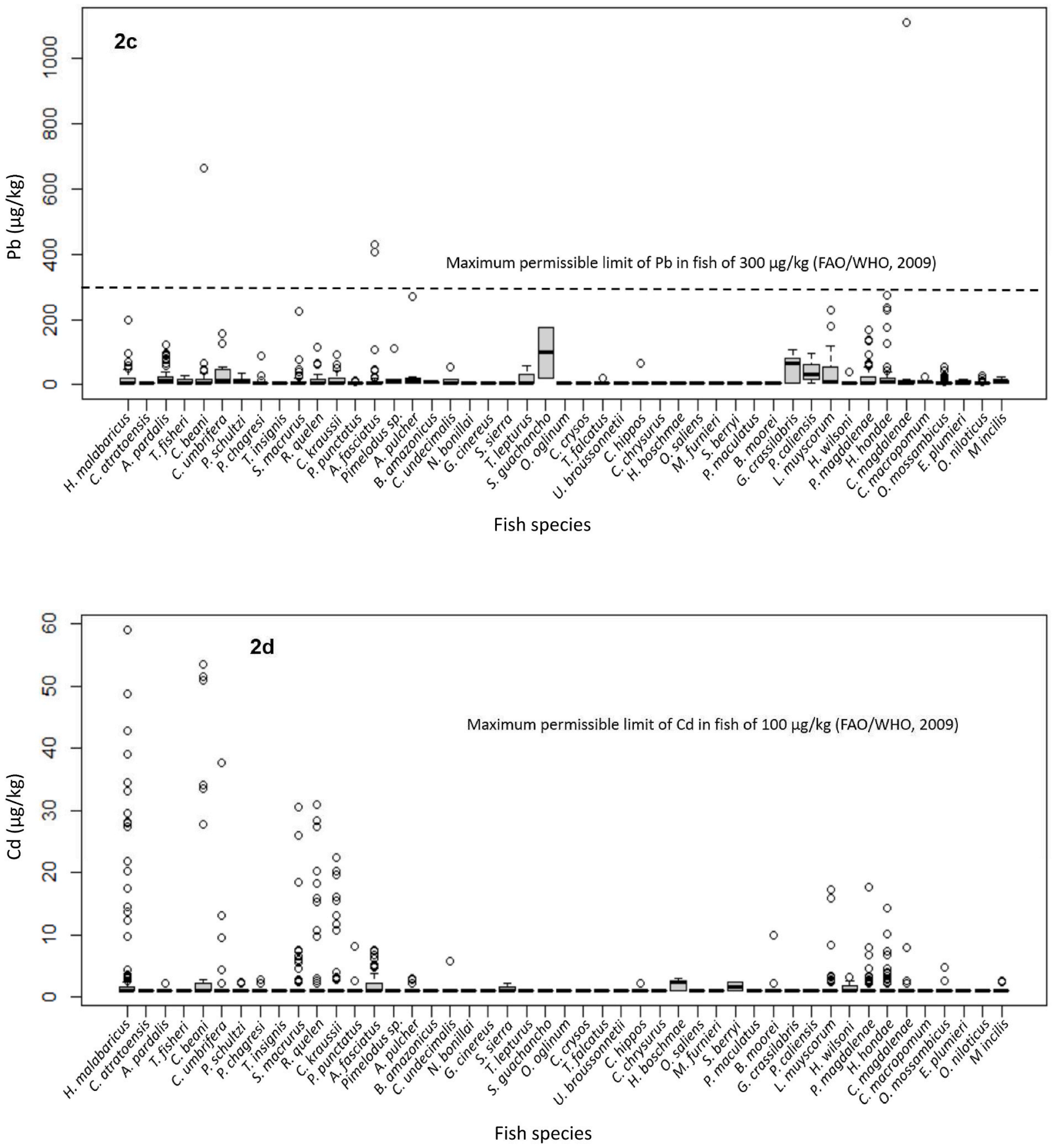

Fig. 2. (*continued*).

THQ, TTHQ, CR, and TCR to estimate the accumulation and risk levels of metals in the most consumed species (Table 2, Figure S1).

3.4. *Human exposure and health risk assessment through fish consumption*

The estimated daily intake (EDI) was calculated for each metal to assess the health risks based on the frequency of consumption of multiple species of fish with different levels of contamination. In this study, EDI average values were 0.893 µg/kg/day (range: 0.016–3.620) for Hg, 0.138 µg/kg/day (range: 0.010–1.038) for As, 0.048 µg/kg/day (range: 0.015–0.390) for Pb, and 0.006 µg/kg/day (range: 0.003–0.079) for Cd. *A. pardalis* showed the highest Hg EDI values (RQ 3.051, QD 2.634, VF 3.620, MO 2.633, RS 2.237 µg/kg/day) followed by *H. malabaricus* (MO 2.590, QD 2.056) and *P. schultzi* (QD 2.291, VF 2.110). The species with the lowest EDI values were *O. mossambicus* (DB 0.044, CG 0.030, CA 0.016 µg/kg/day)] and *O. niloticus* (DB (0.024, CA 0.022 µg/kg/day)]. The results also showed that *O. mossambicus*, *O. niloticus* in CA, DB, and CG, *H. hondae* in BO, VF, and RS, and *Colossoma macropomum* in CG were below the RfDo value for Hg (0.16 µg/kg/day). The other EDI values were above this limit. However, EDI values for *O. mossambicus*, *O. niloticus* in DB, *H. hondae, P. schultzi* in RS, and *Centropomus Undecimalis* in TB were above the limit for As (0.30 µg/kg/day). The other EDI values were below this limit. In the Cd and Pb case, all EDI values were lower than the RfDo.

3.5. *Assessment of noncarcinogenic health risk*

The target hazard quotients (THQ and TTHQ) were calculated to show the risk of noncarcinogenic effects (Table 2). If it is less than 1, the

Table 2
Estimated daily intake (EDI), estimated target hazard quotients (THQ), Total estimated hazard quotient (TTHQ), and carcinogenic risk of the most consumed fish for individual metals from fish consumption. The order of the municipalities is from upstream to downstream on the Atrato River. * Carcinogenic risk. EDI values in (µg/kg/day). Oral reference dose (RfD) values used were: Hg = 0.16 µg/kg/day. Cd = 1.00 µg/kg/day. Pb = 3.50 µg/kg/day and As = 0.30 µg/kg/day.

Fish species	Hg		Cd		As		Pb			As*	Pb*	
	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	TTHQ	CR	CR	TRC
Carmen de Atrato												
Oreochromis mossambicus	0.016	0.097	0.004	0.004	0.063	0.201	0.018	0.005	0.307	9.032×10^{-5}	1.465×10^{-7}	9.047×10^{-5}
Oreochromis niloticus	0.037	0.224	0.004	0.004	0.037	0.119	0.018	0.005	0.352	5.366×10^{-5}	1.465×10^{-7}	5.381×10^{-5}
Bagadó												
Astyanax fasciatus	0.122	0.130	0.008	0.049	0.100	0.320	0.160	0.004	0.503	1.439×10^{-3}	1.270×10^{-7}	1.439×10^{-3}
Hypostomus hondae	0.098	0.580	0.013	0.076	0.109	0.347	0.060	0.016	1.019	1.573×10^{-3}	4.866×10^{-7}	1.573×10^{-3}
Río Quito												
Holpias malabaricus	1.088	6.521	0.079	0.075	0.075	0.239	0.016	0.004	6.839	1.075×10^{-4}	1.268×10^{-7}	1.076×10^{-4}
Ageneiosus pardalis	3.051	18.287	0.004	0.004	0.173	0.553	0.035	0.010	18.854	2.487×10^{-4}	2.885×10^{-7}	2.490×10^{-4}
Rhamdia quelen	0.881	5.282	0.010	0.010	0.234	0.747	0.016	0.004	6.043	3.359×10^{-4}	1.268×10^{-7}	3.360×10^{-4}
Caquetaia kraussii	0.839	5.028	0.051	0.049	0.063	0.202	0.016	0.004	5.283	9.074×10^{-5}	1.268×10^{-7}	9.087×10^{-5}
Pimelodus punctatus	0.495	2.967	0.004	0.004	0.046	0.146	0.016	0.004	3.121	6.558×10^{-5}	1.268×10^{-7}	6.571×10^{-5}
Astyanax fasciatus	0.420	2.518	0.004	0.004	0.113	0.363	0.016	0.004	2.889	1.632×10^{-4}	1.268×10^{-7}	1.633×10^{-4}
Leporinus muyscorum	0.371	2.222	0.004	0.004	0.095	0.303	0.016	0.004	2.533	1.363×10^{-4}	1.268×10^{-7}	1.364×10^{-4}
Prochilodus magdalenae	0.403	2.415	0.004	0.004	0.052	0.168	0.016	0.004	2.591	7.551×10^{-5}	1.268×10^{-7}	7.564×10^{-5}
Quibdó												
Hoplias malabaricus	2.056	12.325	0.004	0.004	0.075	0.239	0.015	0.004	12.572	1.074×10^{-4}	1.225×10^{-7}	1.075×10^{-4}
Ageneiosus pardalis	2.634	15.784	0.004	0.004	0.084	1.269	0.015	0.004	17.061	1.211×10^{-4}	1.225×10^{-7}	1.212×10^{-4}
Pseudopimelodus schultzi	2.291	13.732	0.004	0.004	0.058	0.185	0.015	0.004	13.925	8.316×10^{-5}	1.225×10^{-7}	8.328×10^{-5}
Rhamdia quelen	0.938	5.623	0.004	0.004	0.091	0.290	0.015	0.004	5.921	1.307×10^{-4}	1.225×10^{-7}	1.308×10^{-4}
Caquetaia kraussii	1.396	8.634	0.004	0.004	0.096	0.308	0.015	0.004	8.950	1.386×10^{-4}	1.225×10^{-7}	1.387×10^{-4}
Pimelodus punctatus	0.411	2.460	0.004	0.004	0.076	0.243	0.015	0.004	2.711	1.092×10^{-4}	1.225×10^{-7}	1.093×10^{-4}
Leporinus muyscorum	0.262	1.573	0.004	0.004	0.147	0.470	0.015	0.004	2.051	2.117×10^{-4}	1.225×10^{-7}	2.118×10^{-4}
Prochilodus magdalenae	0.364	2.184	0.004	0.004	0.118	0.377	0.015	0.004	2.569	1.698×10^{-4}	1.225×10^{-7}	1.699×10^{-4}
Medio Atrato												
Holpias malabaricus	1.720	10.306	0.006	0.035	0.120	0.382	0.085	0.023	10.746	1.721×10^{-3}	6.957×10^{-7}	1.722×10^{-3}
Pseudopimelodus schultzi	1.622	9.723	0.005	0.029	0.063	0.202	0.050	0.014	9.968	9.082×10^{-5}	4.060×10^{-7}	9.123×10^{-5}
Rhamdia quelen	0.608	3.646	0.004	0.023	0.067	0.215	0.061	0.017	3.901	9.675×10^{-3}	4.959×10^{-7}	9.675×10^{-3}
Caquetaia kraussii	0.402	10.306	0.007	0.041	0.170	0.541	0.059	0.016	10.904	2.436×10^{-3}	4.777×10^{-7}	2.436×10^{-3}
Leporinus muyscorum	0.401	2.586	0.006	0.039	0.152	0.485	0.052	0.014	3.124	2.180×10^{-3}	4.209×10^{-7}	2.180×10^{-3}
Prochilodus magdalenae	0.292	1.752	0.006	0.037	0.144	0.462	0.034	0.008	2.259	2.078×10^{-3}	2.797×10^{-7}	2.078×10^{-3}
Bojayá												
Hoplias malabaricus	1.714	10.270	0.004	0.004	0.066	0.210	0.090	0.025	10.509	9.461×10^{-5}	7.370×10^{-7}	9.535×10^{-5}
Prochilodus magdalenae	0.481	2.880	0.004	0.004	0.056	0.180	0.063	0.017	3.081	8.086×10^{-5}	5.129×10^{-7}	8.137×10^{-5}
Vigía del Fuerte												
Holpias malabaricus	1.700	10.200	0.004	0.023	0.042	0.136	0.016	0.004	10.363	6.130×10^{-5}	1.270×10^{-7}	6.143×10^{-5}
Ageneiosus pardalis	3.620	21.670	0.003	0.023	0.120	0.384	0.054	0.015	22.092	1.728×10^{-3}	4.477×10^{-7}	1.728×10^{-3}
Pseudopimelodus schultzi	2.110	12.620	0.004	0.023	0.043	0.019	0.059	0.016	12.678	6.272×10^{-5}	4.817×10^{-7}	6.320×10^{-5}
Rhamdia quelen	1.640	9.820	0.004	0.023	0.180	0.574	0.016	0.004	10.421	2.586×10^{-3}	1.270×10^{-7}	2.586×10^{-3}
Caquetaia kraussii	1.450	8.680	0.004	0.023	0.072	0.230	0.016	0.004	8.937	1.036×10^{-3}	1.270×10^{-7}	1.036×10^{-3}
Leporinus muyscorum	0.350	2.080	0.004	0.023	0.209	0.668	0.145	0.040	2.811	3.005×10^{-3}	1.179×10^{-6}	3.006×10^{-3}
Prochilodus magdalenae	0.350	2.080	0.004	0.023	0.042	0.136	0.015	0.004	2.243	6.130×10^{-5}	1.270×10^{-7}	6.143×10^{-5}
Hypostomus hondae	0.081	0.490	0.004	0.023	0.130	0.424	0.016	0.004	0.941	1.909×10^{-3}	1.270×10^{-7}	1.909×10^{-3}
Murindó												
Holpias malabaricus	2.590	15.521	0.004	0.024	0.082	0.263	0.047	0.013	15.821	1.183×10^{-3}	3.833×10^{-7}	1.183×10^{-3}
Ageneiosus pardalis	2.633	15.781	0.004	0.024	0.105	0.336	0.065	0.018	16.159	1.511×10^{-3}	5.293×10^{-7}	1.512×10^{-3}
Caquetaia kraussii	1.904	11.412	0.004	0.024	0.037	0.120	0.084	0.023	11.579	5.383×10^{-5}	6.846×10^{-7}	5.451×10^{-5}
Prochilodus magdalenae	0.468	2.807	0.004	0.024	0.092	0.294	0.095	0.026	3.151	1.322×10^{-3}	7.763×10^{-7}	1.323×10^{-3}
Leporinus muyscorum	0.113	0.679	0.004	0.024	0.108	0.346	0.390	0.107	1.156	1.557×10^{-3}	3.181×10^{-6}	1.560×10^{-3}
Dabeiba												
Oreochromis mossambicus	0.044	0.262	0.004	0.004	0.399	1.274	0.017	0.005	1.545	5.734×10^{-4}	1.393×10^{-7}	5.735×10^{-4}
Oreochromis niloticus	0.024	0.146	0.004	0.004	0.327	1.046	0.017	0.005	1.201	4.706×10^{-4}	1.393×10^{-7}	4.707×10^{-4}
Cañas Gordas												
Colossoma macropomum	0.082	0.494	0.004	0.024	0.187	0.600	0.045	0.012	1.130	2.696×10^{-3}	3.711×10^{-7}	2.696×10^{-3}
Oreochromis mossambicus	0.030	0.180	0.004	0.024	0.134	0.427	0.072	0.020	0.651	1.920×10^{-3}	5.875×10^{-7}	1.921×10^{-3}
Oreochromis niloticus	0.085	0.507	0.004	0.024	0.230	0.738	0.071	0.019	1.288	3.302×10^{-3}	5.762×10^{-7}	3.303×10^{-3}
Riosucio												
Hoplias malabaricus	1.188	7.122	0.004	0.004	0.040	0.127	0.060	0.016	7.269	5.708×10^{-5}	8.884×10^{-7}	5.797×10^{-5}
Ageneiosus pardalis	2.237	13.409	0.004	0.004	0.126	0.404	0.016	0.004	13.821	1.819×10^{-4}	1.328×10^{-7}	1.820×10^{-4}
Pseudopimelodus schultzi	1.575	9.439	0.004	0.004	0.415	1.327	0.056	0.015	10.785	5.973×10^{-4}	4.598×10^{-7}	5.978×10^{-4}
Rhamdia quelen	0.416	2.496	0.004	0.004	0.148	0.473	0.051	0.014	2.987	2.130×10^{-4}	4.128×10^{-7}	2.134×10^{-4}
Caquetaia kraussii	0.928	5.120	0.004	0.004	0.043	0.137	0.047	0.009	5.270	6.159×10^{-5}	3.855×10^{-7}	6.198×10^{-5}
Prochilodus magdalenae	0.283	1.697	0.004	0.004	0.100	0.320	0.066	0.018	2.039	1.442×10^{-4}	5.389×10^{-7}	1.447×10^{-4}
Hypostomus hondae	0.101	0.607	0.004	0.004	1.038	3.319	0.016	0.004	3.934	1.493×10^{-3}	1.328×10^{-7}	1.493×10^{-3}
Unguía												
Holpias malabaricus	0.743	4.450	0.004	0.023	0.010	0.031	0.015	0.004	4.508	1.389×10^{-3}	1.261×10^{-7}	1.389×10^{-3}
Ageneiosus pardalis	0.314	1.882	0.004	0.023	0.010	0.031	0.188	0.052	1.988	1.389×10^{-3}	1.503×10^{-6}	1.391×10^{-3}
Centropomus undecimalis	0.601	3.602	0.004	0.023	0.010	0.031	0.082	0.022	3.678	1.389×10^{-3}	6.642×10^{-7}	1.390×10^{-3}
Caquetaia kraussii	0.241	1.442	0.004	0.023	0.091	0.290	0.015	0.004	1.759	1.303×10^{-3}	1.261×10^{-7}	1.303×10^{-3}
Prochilodus magdalenae	0.272	1.631	0.008	0.051	0.126	0.403	0.015	0.004	2.089	1.814×10^{-3}	1.261×10^{-7}	1.814×10^{-3}
Turbo												

(*continued on next page*)

Table 2 (*continued*)

Fish species	Hg		Cd		As		Pb			As*	Pb*	
	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	TTHQ	CR	CR	TRC
Holpias malabaricus	0.491	2.946	0.004	0.023	0.223	0.713	0.016	0.004	3.686	3.208×10^{-3}	1.305×10^{-7}	3.208×10^{-3}
Centropomus undecimalis	0.513	3.077	0.004	0.023	0.570	1.815	0.016	0.004	4.919	8.168×10^{-3}	1.305×10^{-7}	8.168×10^{-3}
Caquetaia kraussii	0.565	3.388	0.004	0.023	0.278	0.888	0.016	0.004	4.303	3.996×10^{-3}	1.305×10^{-7}	3.996×10^{-3}
Prochilodus magdalenae	0.211	1.267	0.004	0.023	0.074	0.350	0.016	0.004	1.644	1.064×10^{-3}	1.305×10^{-7}	1.064×10^{-3}

exposure level is less than the RfDo; indicating that daily exposure at this level is unlikely to cause adverse effects during a person's lifetime (USEPA, 2000; Chien et al., 2002). In this study, average THQ values were 5.469 (range: 0.097–21.670) for Hg, 0.459 (range: 0.019–3.319) for As, 0.012 (range: 0.004–0.107) for Pb, and 0.019 (range: 0.004–0.076) for Cd. The THQ from fish consumption based on the mean concentration of Hg, was less than 1 for all fish species in CA, BO, DB, and CG, thus as for *H. hondae* in VF, RS, and *L. muyscorum* in MO. For As, THQ index was higher than 1 for *A. pardalis* in QD, *O. mossambicus*, *O. niloticus* in DB, *P. schultzi*, *H. hondae* in RS and *C. undecimalis* in TB. For Cd and Pb all THQ values were less than 1. The relative contributions to the total THQ showed that Hg and As were the major risk contributors and accounted for 80.4% (Range: 12.2–99.5%) and 18.2% (Range: 0.1–87.1%) of the total THQ, respectively. The risk contribution of Cd and Pb was less than 2.0% (Range: 0.02–9.7%). With exception of the municipality of CA, *A. fasciatus* in BO, *H. hondae* in VF, and *O. mossambicus* in CG, all the total THQ (TTHQ) values exceeded 1. Also, although average THQ values for As, Cd, and Pb were less than 1, the average THQ for Hg far exceeded 1 (5.469). In general terms, the species with the highest values of EDI, THQ, and TTHQ, for Hg, were those with a carnivorous habit (e.g., *H. malabaricus, A. pardalis,* and *P. schultzi*); whereas for As both, carnivorous (*C. undecimalis and P. schiltzi*) and noncarnivorous (*O. mossambicus, O. niloticus, and H. hondae*) species showed EDI, THQ and TTHQ values above of their limits (Table 2).

3.6. *Assessment of carcinogenic health risk*

As and Pb elements present in the fish species of the Atrato River basin could induce carcinogen effects on the population due to fish consumption. Therefore, the carcinogenic risk for these elements must be determined. For this, the USEPA recommended using the carcinogenic (CR) and the total carcinogenic (TCR) indexes to assess this risk. Thus, if CR and TCR values are lower than 1.0E-06, they are regarded as negligible, whereas a CR and TCR above 1.0E-04 are likely to have adverse effects in humans (USEPA, 1989, 2002). In this study, average CR values were 1.215E-03 (range: 5.366E-5 – 9.675E-3) for As and 3.745E-07 (range: 1.255E-7 – 3.181E-6) for Pb. Average TCR value was 1.215E-03 (range: 5.381E-5 – 9.675E-3). Results of the assessment of the carcinogenic health risk for As through of fish consumption showed that CR values for the species *O. mossambicus*, *O. niloticus* in CA, *C. kaussii*, *P magdalenae*, *P. punctatus* in RQ, *P. schultzi* in QD and MA, *H. malabaricus*, *P. magdalenae* in BY, *H. malabaricus, P. schultzi, P. magdalenae* in VF, *C. kraussii* in MO, and *H. malabaricus*, *C. kraussii* in RS, are above 1.0E-06 limit but below the 1.0E-04 limit. The other species in the other municipalities CR are above both limits. In the Pb case, all CR values except *L. muyscorum* in VF, MO, and *A. pardalis* in UG, were low these limits (Table 2). The TCR values showed the same behavior that CR values for As. These results indicate a significant carcinogenic health risk, principally by As, through fish consumption. In general terms, EDI, THQ, TTHQ, CR, and TCR results show that the inhabitants of the Atrato River basin could present health problems due to fish consumption during their lifetime, depending mainly on the type of fish they consume.

3.7. *Risk assessment by MeHg in most consumed fish*

Median concentrations of THg, MeHg (μg kg^{-1}), and the MeHg percentages (%MeHg) of the fish species most consumed among the inhabitants of the Atrato River basin are described in Table 3. Results showed that three carnivorous species, *A. pardalis* in RQ, QD, VF, MO, RS; *H. malabaricus* in QD, VF, MO; and *P. schultzi* in QD, VF; and *C. kraussii* in MO have THg and MeHg values that exceeded the permissible Hg values established by the WHO of 500 μg kg^{-1} (WHO, 1990). *H. malabaricus* in MA and *R. quelen* en VF were close to this threshold (468.54 and 456.27 μg kg^{-1}, respectively). THg (and MeHg) concentrations in fish were higher in carnivorous species than in non-carnivorous ones, indicating the occurrence of biomagnification processes. The most abundant form of Hg accumulated in fish is MeHg (Marrugo et al., 2020; Salazar et al., 2021). In this study, the average MeHg percentage was 86.5% (range: 57.9–99.5%). The species *P. magdalenae*, which is the most frequently consumed fish in the Atrato River basin showed MeHg percentages between 84.4 and 95.1%. Additionally, the average concentration of THg and MeHg did not exceed the WHO (1990) threshold of 500 μg kg^{-1}, but it exceeded the WHO (2008) of 200 μg kg^{-1} (250.3 and 219.9 μg kg^{-1}, respectively).

On the other hand, the intake rate (IR) is the amount of fish consumed for an individual per unit of time and serves to consider the fish consumption preference of each person surveyed. If the IR value is over 100 g of fish per day, it must be considered as high consumption, especially when assessing a vulnerable population [e.g., children and women of childbearing age (WCHA)] (WHO, 2008). Hence, in this study, IR values above 700 g/week of fish were considered as high. As can be seen in Table 3, except in TB, *P. magdalenae* in all localities exceeded this value (819.2–1075.2 g/week), followed by *C. kraussii* in VF, MO, RS, UG, and TB (742.4–768.0 g/week), *H. malabaricus* in MA, BY, VF, MO, and RS (742.4–793.6 g/week), *R. quelen* in MA and RS (742.4–793.6 g/week), *A. fasciatus* in BO and RQ (716.8–768.0 g/week), *L. muyscorum* in MA and MO (768.0–793.6 g/week), *H. hondae* in BO and RS (716.8–742.4 g/week), *A. pardalis* in UG (742.4 g/week), and *P. punctatus* in QD (742.4 g/week) (Table 3). It is also important to mention that *C kraussii* in RQ, MA, *R. quelen* in QD, *H. malabaricus,* in UG and *H. hondae* in VF were close to this value (691.2 g/week). In addition, it has been stated that individuals have a high frequency of fish consumption (FIR) when it is equal to or greater than three times per week (Health Canada, 2019). In this study, the frequency of fish consumption in the population of the Atrato River basin was equal to or greater than 3 days/week for the species *P. magdalenae* (3.2–4.2 days/week) in all localities where this species was consumed (except in TB), *R. quelen* in MA and RS (3.1 days/week), *H. malabaricus* in MA, RS, and UG (3.0–3.1 days/week), *C. kraussii* in UG and VF (3.0 days/week), *L. muyscorum* in MA and MO (3.0–3.1 days/week) and *A. fasciatus* in BO (3.0 days/week). Values close to this limit (2.8–2.9 days/week) were observed for *H. malabaricus* in BY, VF and MO, *C. kraussii* in MO, RS and TB, *H. hondae* in BO and RS, *A. fasciatus* in RQ, *P. punctatus* in QD and *A. pardalis* in UG (Table 3). The estimated weekly intake (EWI) obtained in this study for the GP and the WCHA group (Table 3) showed that fish with carnivorous habits have higher EWI values, many of them surpassing the potential weekly intake (PTWI) thresholds for adult (3.2 μg/kg bw/week) and vulnerable populations (1.6 μg/kg bw/week). Fish species with the highest EWI levels were *A. pardalis, H. malabaricus*, *P. schultzi, R. quelen, and C. kraussii* (RQ, QD, MA, BY, VF, MO, and RS), which have values between 1.2 and 5.3 times the PTWI for WCHA group and between 1.0 and 2.6 times the PTWI for GP. Values for non-carnivorous fish ranged from 0.04 to 1.61 μg/kg bw/week for the GP and between 0.04 and 1.68 for the WCHA group in all municipalities,

Table 3

Estimate of the potential risk in the population by consumption of fish in the Atrato River basin. THg a MeHg (μg/kg) are mercury and methylmercury concentrations, FIR (g/day) is the food ingestion rate, IR (g/week) is the weekly intake of fish, EWI (μg/kg bw/week) is the Estimated Weekly Intake of MeHg, MeHg permissible is the permissible safety level (μg/kg), and MFW (g/week) is the estimated maximum amount of fish that can be weekly consumed per person. Pi: pollution index. The order of the municipalities is from upstream to downstream on the Atrato River. ^{a} For calculation the PTWI 3.2 μg/kg bw/week for the adult population and 1.6 μg/kg bw/week for vulnerable population were used. ^{b} For Pi calculation the threshold value of 500 μg/kg ww for adult populations was used. C For Pi calculation the threshold value of 200 μg/kg ww for vulnerable populations was used. The highlighted bold values are above those established in the case of MeHg concentrations, the weekly IR values that are above the MFW, and the Pi values that are in Pollution Degree 2, 3 and 4.

							GP			WCHA			
Fish species	THg	MeHg	%MeHg	FIR	IR	EWI	Permissible MeHg ^{a}	MFW ^{a}	EWI	Permissible MeHg	MFW ^{a}	Pi^{b}	Pi^{c}
Carmen de Atrato													
Oreochromis mossambicus	5.13	5.01	97.7	1.9	486.4	0.04	406.58	39,473.1	0.04	199.34	19353.3	0.01	0.03
Oreochromis niloticus	9.01	5.22	57.9	1.9	486.4	0.04	406.58	37,885.1	0.04	199.34	18574.7	0.01	0.03
Bagadó													
Astyanax fasciatus	33.26	28.71	86.3	3.0	768.0	0.31	296.88	7,941.5	0.32	142.29	3806.3	0.06	0.14
Hypostomus hondae	26.41	25.14	95.2	2.9	742.4	0.27	301.12	8,892.3	0.27	147.20	4346.9	0.05	0.13
Río Quito													
Holpias malabaricus	**303.46**	**264.94**	87.3	2.3	**588.8**	2.18	388.04	862.4	2.17	195.65	**434.8**	0.53	1.32
Ageneiosus pardalis	**851.05**	**801.42**	94.2	1.9	**486.4**	5.46	469.74	**285.1**	5.41	236.84	**143.7**	**1.60**	**4.01**
Rhamdia quelen	**245.82**	**205.54**	83.6	2.6	**665.6**	1.91	343.27	1,112.7	1.90	173.08	**561.0**	0.41	1.03
Caquetaia kraussii	**234.00**	**226.10**	96.6	2.7	**691.2**	2.19	350.56	1,010.5	2.17	166.67	**509.5**	0.45	1.13
Pimelodus punctatus	138.00	119.07	86.3	2.4	614.4	1.02	371.88	1,918.9	1.02	187.50	967.5	0.24	0.60
Astyanax fasciatus	117.19	99.40	84.8	2.8	716.8	1.00	318.75	2,298.7	0.99	160.71	1159.0	0.20	0.50
Leporinus muyscorum	103.42	99.02	95.7	2.6	665.6	0.92	343.27	2,307.5	0.92	173.08	1163.4	0.20	0.50
Prochilodus magdalenae	112.38	102.33	91.1	3.2	819.2	1.17	278.91	2,232.7	1.16	140.63	1125.7	0.20	0.51
Quibdó													
Hoplias malabaricus	**590.54**	**514.54**	87.1	2.6	**665.6**	4.63	355.29	**459.6**	4.66	176.88	**228.6**	**1.03**	**2.57**
Ageneiosus pardalis	**807.40**	**722.75**	89.5	2.4	**614.4**	6.01	384.90	**327.2**	6.04	191.41	**162.7**	**1.45**	**3.61**
Pseudopimelodus schultzi	**725.98**	**624.05**	86.0	2.2	**563.2**	4.76	419.89	**378.9**	4.78	208.81	**188.4**	**1.25**	**3.12**
Rhamdia quelen	**270.85**	**221.94**	81.9	2.7	**691.2**	2.08	342.13	1,065.5	2.09	170.14	**529.9**	0.44	1.11
Caquetaia kraussii	**409.37**	**376.08**	91.9	2.4	**614.4**	3.13	384.90	628.8	3.14	191.41	**312.7**	0.75	1.88
Pimelodus punctatus	118.50	110.17	93.0	2.9	742.4	1.11	318.53	2,146.4	1.11	158.41	1067.4	0.22	0.55
Leporinus muyscorum	75.76	65.48	86.4	2.6	655.6	0.59	355.29	3,611.7	0.59	176.68	1796.1	0.13	0.33
Prochilodus magdalenae	110.78	97.84	88.3	3.3	844.8	1.12	279.92	2,416.9	1.12	139.20	1201.9	0.20	0.49
Medio Atrato													
Holpias malabaricus	**468.54**	**432.99**	92.4	3.1	**793.6**	5.04	274.76	**503.6**	5.09	136.09	**249.4**	0.87	**2.16**
Pseudopimelodus schultzi	**442.02**	**423.65**	95.8	2.6	**665.6**	4.14	327.60	**514.7**	4.07	162.26	**261.6**	0.85	**2.12**
Rhamdia quelen	165.76	143.46	86.5	3.1	793.6	1.24	274.76	2,046.8	1.20	136.09	1059.9	0.29	0.72
Caquetaia kraussii	109.44	99.46	90.9	2.7	691.2	1.01	315.46	2,192.3	1.02	156.25	1085.9	0.20	0.50
Leporinus muyscorum	117.56	106.53	90.6	3.1	793.6	1.24	274.76	2,046.8	1.25	136.09	1013.8	0.21	0.53
Prochilodus magdalenae	79.08	72.86	92.1	3.9	998.4	1.07	218.40	2,992.7	1.08	108.17	1482.3	0.15	0.36
Bojayá													
Hoplias malabaricus	**447.63**	**400.17**	89.4	2.9	**742.4**	4.09	312.93	**580.6**	4.27	149.78	**277.9**	0.80	**2.00**
Prochilodus magdalenae	142.04	119.91	84.4	3.8	**972.8**	1.61	238.82	1,937.5	1.68	114.31	**927.4**	0.24	0.60
Vigía del Fuerte													
Holpias malabaricus	**602.51**	**474.48**	78.8	2.9	**742.4**	3.90	307.11	**609.3**	5.07	149.78	**234,4**	0.95	**2.37**
Ageneiosus pardalis	**1,006.26**	**956.37**	95.0	2.4	**614.4**	8.25	371.09	**238.3**	8,45	180.99	**116.3**	**1.91**	**4.78**
Pseudopimelodus schultzi	**586.04**	**459.35**	78.4	2.0	**512.0**	3.30	445.31	**496.4**	3.38	217.19	**242.2**	0.92	**2.30**
Rhamdia quelen	**456.27**	**446.45**	97.8	2.6	**665.6**	4.17	342.55	**510.7**	4.28	167.07	**249.1**	0.89	**2.23**
Caquetaia kraussii	**402.53**	**335.63**	83.4	3.0	**768.0**	3.62	296.88	**679.3**	3.71	144.79	**331.3**	0.67	**1,68**
Leporinus muyscorum	96.64	84.40	87.3	2.3	588.8	0.70	387.23	2,701.4	0.72	188.86	1317.5	0.17	0.42
Prochilodus magdalenae	96.78	86.87	89.8	3.4	870.4	1.06	261.95	2,624.6	1.09	127.76	1280.1	0.17	0.43
Hypostomus hondae	22.53	19.73	87.6	2.7	691.2	0.19	329.86	11,556.0	0.20	160.88	5636.1	0.04	0.10
Murindó													
Holpias malabaricus	**689.33**	**485.71**	70.5	2.9	**742.4**	5.29	293.71	**448.9**	5.40	143.97	**220.0**	0.97	**2.43**
Ageneiosus pardalis	**700.89**	**589.08**	84.0	2.4	**614.4**	5.31	354.90	**370.2**	3.80	173.96	**258.9**	**1.18**	**2.95**
Caquetaia kraussii	**506.85**	**471.98**	93.1	2.9	**742.4**	5.14	293.71	**462.00**	5.25	143.97	**226.5**	0.94	**2.36**
Leporinus muyscorum	30.17	25.10	83.2	3.0	768.0	0.28	283.92	8,687.2	0.29	139.17	4258.2	0.05	0.13
Prochilodus magdalenae	124.69	118.59	95.1	3.2	819.2	1.43	266.17	1,838.7	1.45	130.47	901.3	0.24	0.59
Dabeiba													
Oreochromis mossambicus	11.08	11.03	99.5	1.9	486.4	0.08	427.63	18,851.5	0.08	213.16	9396.7	0.02	0.06
Oreochromis niloticus	6.18	5.01	81.1	1.9	486.4	0.04	427.63	41,517.0	0.04	213.16	20694.6	0.01	0.03
Cañasgordas													
Colossoma macropomum	21.41	14.09	65.8	2.1	537.6	0.11	395.95	15,107.5	0.12	191.67	7313.0	0.03	0.07
Oreochromis mossambicus	7.76	6.85	88.3	2.4	614.4	0.06	346.46	31,075.0	0.07	167.71	15042.3	0.01	0.03
Oreochromis niloticus	21.97	14.82	67.5	1.6	409.6	0.09	519.69	14,363.3	0.09	251.56	6952.8	0.03	0.07
Riosucio													
Hoplias malabaricus	**361.95**	**344.08**	95.1	3.0	**768.0**	3.87	284.17	**634.3**	3.96	138.96	**310.2**	0.69	**1.72**
Ageneiosus pardalis	**538.18**	**467.70**	86.9	2.6	**665.6**	4.56	327.88	**466.6**	4.67	160.34	**228.2**	0.94	**2.34**
Pseudopimelodus schultzi	**419.56**	**412.90**	98.4	2.4	**614.4**	3.72	355.21	**528.6**	3.80	173.70	**258.5**	0.83	**2.06**
Rhamdia quelen	110.93	101.90	91.9	3.1	793.6	1.19	275.00	2,141.7	1.21	134.48	1047.3	0.20	0.51
Caquetaia kraussii	**227.58**	**218.72**	96.1	2.9	**742.4**	2.38	293.97	997.8	2.43	143.75	**487.9**	0.44	1.09
Prochilodus magdalenae	83.18	76.42	91.1	3.5	896.0	1.00	243.57	2,855.6	1.03	119.11	1396.4	0.15	0.38
Hypostomus hondae	37.80	27.40	72.5	2.8	716.8	0.29	304.46	7,963.6	0.29	148.88	3894.2	0.05	0.14

(*continued on next page*)

Table 3 (*continued*)

							GP			WCHA			
Fish species	THg	MeHg	%MeHg	FIR	IR	EWI	Permissible MeHg ^{a}	MFW ^{a}	EWI	Permissible MeHg	MFW ^{a}	Pi^b	Pi^c
Unguía													
Holpias malabaricus	**208.34**	**173.90**	83.5	3.0	**691.2**	1.67	332.55	1,321.8	1.90	146.46	**646.8**	0.35	0.87
Ageneiosus pardalis	88.13	70.52	80.0	2.9	**742.4**	0.73	309.61	3,259.4	0.74	151.51	**272.4**	0.14	0.35
Centropomus undecimalis	168.64	136.16	80.7	2.1	537.6	1.02	427.56	1,688.1	1.04	209.23	826.1	0.27	0.68
Caquetaia kraussii	67.53	55.79	82.6	3.0	768.0	0.60	299.29	4,120.0	0.61	146.46	2016.1	0.11	0.28
Prochilodus magdalenae	76.34	65.39	85.7	4.2	1.075.2	0.98	213.78	3,515.2	1.00	104.61	1720.1	0.13	0.33
Turbo													
Hoplias malabaricus	133.29	110.23	82.7	2.3	588.8	0.94	377.23	2,015.0	0.96	183.97	928.7	0.22	0.55
Centropomus undecimalis	139.20	93.28	67.0	2.6	665.6	0.89	333.70	2,381.1	0.92	162.74	1161.2	0.19	0.47
Caquetaia kraussii	153.30	112.41	73.3	2.8	716.8	1.16	309.87	1,975.9	1.19	151.12	963.6	0.22	0.56
Prochilodus magdalenae	57.22	50.42	88.1	2.3	588.8	0.43	377.23	4,405.2	0.44	183.97	2148.4	0.10	0.25

being the lowest value recorded in *O. niloticus*, about 80 and 40-fold lower than PTWI for GP and WCHA, respectively. Although most of the EWI values calculated for non-carnivorous were below the PTWI's, *P. magdalenae* in BY with a value close to PTWI for GP and WCHA group (1.61 vs 1.68 μg/kg bw/week), could pose a threat especially to the vulnerable population of this municipality. According to the surveys, the calculated weekly allowable fish amount (MFW) for *A. pardalis, H. malabaricus*, *P. schultzi*, *R. quelen,* and *C. kraussii* in RQ, QD, MA, BY, VF, MO and RS for GP, and additionally UG for WCHA group, was much lower than that consumed by the population (Table 3). These results suggest that in these localities, the amount consumed of this species is 1.0–2.6 times higher for GP and 1.1 to 5.3 times for WCHA group.

3.8. *Diagnosis of the population*

The pollution index (Pi) (Zhang et al., 2019) was used to show the contamination degree with Hg of each fish species in each municipality, taking as reference the permissible limits established by WHO (1990) (500 μg kg^{-1}) and WHO (2008) (200 μg kg^{-1}). Table 3 shows that when Pi values were calculated according to WHO (1990) limit, fish species *A. pardalis* in RQ, QD, VF, and MO, and *H. malabaricus* and *P. schultzi* in QD presented a slight pollution degree ($1 < \text{Pi} \leq 2$) (Material supplementary table S2). However, when the pollution index was calculated based on the WHO (2008) threshold, *H. malabaricus* in RQ, BY, and RS, *C. kraussii* in RQ, QD, VF, and RS and *R. quelen* in RQ and QD showed a slight pollution degree; *H. malabaricus* in QD, MA, VF, and MO, *A. pardalis* in MO and RS, *P. schultzi* in MA, VF, and RS, *R. quelen* in VF and *C. kraussii* in MO had moderate pollution degree ($2 < \text{Pi} < 3$); and *A. pardalis* in RQ, QD, and VF, and *P. schultzi* in QD had heavy contamination (Table S2).

4. Discussion

The Atrato River basin receives a large amount of domestic waste, sewage, agro-industrial and wood residues. This is due to the fact that 50% of the municipalities near the basin lack disposal services for wastewater and solid waste collection (DANE, 2018). Further, high amounts of sediments that may contain toxic elements reach the river due to gold mining operations that are practiced either on the banks of the river or along its tributaries and deforestation that devastate forested areas. Thus, this could explain the presence of metal(loid) such as Hg, As, Pb, and Cd in the sediments and fish species of the Atrato River. In this study, the concentrations of these elements in fish followed the order Hg > As > Pb > Cd. The highest mean concentrations of Hg appeared in fish with carnivorous habits, especially in freshwater fish (Fig. 2a) while, the highest for As were found in carnivorous species living in brackish waters (Fig. 2b), which is consistent with previous reports (Pei et al., 2019; Marrugo et al., 2020; Salazar et al., 2021). The mean Pb and Cd concentrations in fish muscle always were lower than the other metals studied. According to previous studies in the Atrato River (Palacios et al. 2018, 2020), the sediments were between non-polluted and moderately polluted for Hg (0.03–0.14 μg/g), moderately polluted for Cd (0.22 ± 0.06 μg/g), strongly polluted for As (3.53 ± 0.96 μg/g) and Pb (5.62 ± 1.00 μg/g). Nevertheless, concentrations de Hg, As, Pb, and Cd in sediments follow a tendency different to this described by fish (Pb > As > Cd Hg). To investigate this fact, a Spearman correlation test was performed between the concentration of the elements and the trophic level of the fish. The results show that the relationships were negative and non-significant for Pb ($rho = -0.0001$, $p = 0.9711$), negative and significant for As ($rho = -0.1468$, $p < 0.001$), positive with low value and non-significant for Cd ($rho = 0.0173$, $p = 0.5261$), and positive with high value and significant for Hg ($rho = 0.6162$, $p < 0.001$). In addition to trophic ecology, there are other factors such as exposure times (Kojadinovic et al., 2007), the content of intramuscular fat (Farkas et al. al, 2003), and physiological processes such as respiration or/and environmental conditions of metals in the fish habitat, that could determine the bioaccumulation of As, Pb and Cd in fish (Li and Gao, 2014; Yi et al., 2017). Similar tendencies of elements accumulation like those reported in this study were shown for fish species as *M. incilis* for Hg, As, Pb, Cd (Pinzón et al., 2020), *C. Undecimalis* for Hg, Pb, Cd (Fuentes et al., 2018; Burgos et al., 2017), *T. lepturus* and *E. plumieri* for Hg, Pb, Cd (Burgos et al., 2017), and *S. sierra* for Hg, As (Salgado et al., 2017). It is important to note that concentrations of Hg, Pb, and Cd reported by Burgos et al. (2017) for *C. undecimalis* and Pinzón et al. (2020) for *M. incilis* were greater than those found in our study.

A comparison of the results of this study with those of other studies in the world can be found in Table S3. Data showed a wide variation in metal(loid)s concentrations in fish muscles of different species and different concentrations in specimens of the same species in different localities. This phenomenon might be attributed to differences in sampling sites, regional sorption of metals by sediment, seasonal variations (Rahman et al., 2019; Skoric et al., 2012; Fu et al., 2013) and types of fish species caught. It should be noted that the Atrato River basin is the main freshwater source of fish as food in the pacific area of Colombia. Hence, concentrations of these four elements in muscles of all fish species should not exceed the maximum levels according to the standard values of reference (WHO, 1990; FAO/WHO, 2002; WHO, 2008; FAO/WHO, 2009). However, results indicate that although average concentrations of As and Pb in fish are lower than the maximum allowable levels, there were some specimens as such as *A. fasciatus* in QD and MO, *C. beani*, *C. magdalenae in UG* and *Haemulon boschmae, G. cinereus* in TB that surpassed or were close these values, so that inhabitants of those localities could be at risk through fish consumption.

The results of the EDI calculation of the four elements studied for the most consumed fish species in the Atrato River basin were compared with their respective RfDo values. It was shown that the EDI for Pb and Cd were all lower than their RfDo values (Table 2). These results indicated that the exposure to Pb and Cd from fish consumption might not have an adverse effect on human health. However, in the municipalities of RQ, QD, MA, Bojayá, UG, and TB all the calculated EDI were above RfDo value for Hg. In VF, MO, and RS only one species of those evaluated did not exceed the RfDo. In DB and RS two species exceeded the RfDo limit for As and one in TB (Table 2). Consequently, the daily intake of fish from these locations, especially *H. malabaricus, A. pardalis, C. undecimalis, P. schultzi, O. mossambicus, O. niloticus,* and *H. hondae* might cause detrimental health hazards to the population.

The THQ for Hg from fish consumption based on the average concentration for each species was less than 1 in CA, BO, DB, CG, and one species in VF, MO, and RS respectively. For As, the THQ were greater than 1 in DB, one species in QD and TB, respectively, and two species in RS (Table 2). For Pb and Cd, all THQ values were less than 1. These results suggest that the population of the Atrato River would not experience significant non-carcinogenic health risks from the intake of Pb and Cd through fish consumption. Nevertheless, they might experience a certain degree of adverse non-carcinogenic health effects by Hg from the intake of most of the commonly consumed species, and for As in QD, DB, RS, and TB from the intake of some species as *A. pardalis, O. mossambicus, O. niloticus, P. schultzi, H. hondae, and C. undecimalis.* Although most of the values of THQ for As were less than 1, it is possible to conclude that As and Hg had higher potential non-carcinogenic health risks compared to Pb and Cd in the study area. Concerning the relative contributions of Hg, As, Pb, and Cd to the TTHQ value (Table 2), Hg and As were the two major risk contributors accounted on average for 98.6% of the TTHQ. This suggests that although the inhabitants of the Atrato River are exposed to the cumulative effect of these four elements by fish consumption, the potential non-carcinogenic risk by fish intake is due to Hg and As. A similar trend of results, where As and Hg were among the two major risk contributors in the assessment of non-carcinogenic risk by fish consumption have been reported (Yi et al., 2017; Qian et al., 2020).

On the other hand, As and Pb were assessed for carcinogenic risk because these elements could induce cancer risk in the exposed populations (USEPA, 1989, 2002). Results of the carcinogenic health risks assessment of the of As and Pb through fish consumption were listed in Table 2. As it is shown, for each fish species, almost all the carcinogenic values of Pb are lower than the recommended limit of 1.0E−06. However, carcinogenic values for As were between 5.366E-05 and 9.675E-03. This indicates that unliked Pb, As might present a carcinogenic health risk through the fish intake in the study area (USEPA, 2002).

Globally, fish is a very important source of protein, as about 160 million tons are consumed per year. According to FAO (2020), per capita fish consumption (for a population of 7.6 billion people) is estimated at 20.5 kg/year. In Colombia, per capita fish consumption is 8.80 kg/year (MADR, 2021), which compared to the average consumption in the world is low. According to the National Survey of Nutritional Status (ENSIN, 2015), Colombians consume 5 times less fish than eggs, beef, or chicken. Fish consumption in the country is regionalized, presenting high consumption in areas where access to other protein products is low, as is the case for most of the inhabitants of the Atrato River basin. It is estimated that in riparian areas 90% of the consumption of protein of animal origin comes from fish, implying that the risk to health by consumption of fish should be high. As shown in Table 3, the highest proportion of Hg present in fish is MeHg. Many of these species that have high concentrations of MeHg, have a high intake (>700 g/week) and are highly consumed (>3 times/week) and others have ingestion rates higher than MFW. In addition, the food meal size in the Atrato region has a value of 256 g, a value higher than 2 times the portion size of 114 g agreed by the USEPA (2002). For the above reasons and because of the high neurotoxicity that MeHg presents for health, the frequency and the quantity consumed of species such as *H. malabaricus, A. pardalis, P. schultzi, C. kraussii,* and *R. quelen* in the Atrato River basin must be reduced. Instead, and taking care of the quantities, other species such as *P. magdalenae, O. niloticus, O. mossambicus,* and *A. fasciatus* could be consumed since their IR is lower than MFW.

Methylmercury was the major chemical form of mercury found in fish species of the Atrato River basin. It has been showed that after ingestion of the fish, MeHg is absorbed in the gastrointestinal tract and it is stored in the hair (Díez et al., 2011)(;), that can be easily analysed (Díez et al., 2007; Montuori et al., 2004) . After 3 or 4 days, MeHg is spread throughout the body, with the brain as the prime target organ. In addition, it is known that Hg passes through of the placenta to the fetus and it is due to its high affinity of the Hg for fetal hemoglobin (Kim et al., 2014). Also, it has been stated that fish consumption correlated significantly with maternal blood and cord blood Hg concentration in pregnant women, while that prior to pregnancy, this parameter correlated significantly with placental tissue Hg concentrations (Hsu et al., 2007). Also, it has been showed that Hg concentrations increased specially in placenta tissue with increasing frequency of fish consumption (Papadopoulou et al., 2021). This fact has been attributed to the placenta's function of retain toxins to protect the fetus from infections and harmful substances. Morrissette et al. (2004) found a strong dose relation between the frequency of fish consumption before and during pregnancy and Hg exposure in mothers and newborns. Moreover, Nyanza et al. (2020) studied the association between maternal exposure to As and Hg and adverse birth outcomes in gold mining areas (ASGM, n = 788) and non-ASGM communities (n = 173) in Northern Tanzania, and they found that a significant proportion of women in ASGM areas had adverse birth outcomes; and suggested that exposure to high levels of As [9.6 (5.1–15.9 μg/L)] and Hg [1.2 (0.8–1.8 μg/L)] contribute significantly to increase risk.

In our study, the WCHA group consumed fish with more frequency (1.3–2.1 times) that this described by Schulz et al. (2009) and in greater quantities that the recommended MFW values (Table 3). According to our results, the WCHA group in the Atrato River basin (especially in RQ, QD, MA, BY, VF, MO, RS and UG) are at risk to have high concentrations of Hg (and MeHg) in blood, cord blood and placental tissue affecting maternal, fetus and newborn's health. The results in this study provide essential information regarding the impact of fish consumption in the WCHA of the Atrato river basin. The local and national authorities must advise to WCHA and children to avoid eating fish that contain high levels of Hg such as *H. malabaricus, A. pardalis, P schultzi, C. kraussi and R. quelen*. Instead, they should recommend that these groups eat fish low in Hg such as *O. mossambicus and O. niloticus*. It also is important to emphasize that the authorities must conduct a large-scale investigation to evaluate Hg and MeHg concentrations at least in blood of vulnerable groups. Such information may help to: a) reduce the consumption of fish species containing high mercury concentrations, and b) diminish possible adverse health effects to vulnerable population.

Finally, the pollution index (Pi) was used to show the degree of Hg contamination of each of the fish species, using the WHO permissible limits of 200 and 500 μg/g ww. Similar as for EWI and MFW data from the WCHA group (Table 3), the Pi data showed that the vulnerable populations have a higher probability of being affected by fish consumption than the adult population. All these results suggest that inhabitants of the Atrato River basin must avoid the consumption of certain species of fish, many of which are preferred by the riverside residents and widely consumed in the local market; however, they can become a problem for human health due to the harmful effects of MeHg. However, in Colombia, there are no public policies that warn vulnerable groups of this risk and the Chocó department has a monetary poverty level of 68.4% (DANE, 2020); therefore, advising the population on what type and quantity of fish to consume will be a difficult task. However, according to these and the others results (EDI, THQ, TTHQ, IR, MFW), it is a task that must be done. Therefore, it is necessary to disseminate and discuss with the communities the risk of certain fish species, as a strategy to reduce their consumption, but preserving their benefits at the nutritional, social, cultural and environmental levels. Also, the inhabitants of this region should be advised to consume other sources of protein such as eggs and bushmeat (a very popular practice before the arrival of mining in the region) and limit consumption of some fish species such as *A. pardalis, H. malabaricus, P. schultzi, R. quelen,* or *C. kraussii*. Traditional fishing practices and a diet based on wild fish are cultural values that communities in this region still preserve; therefore, it is important to provide a fish consumption guide that highlights clear messages such as: "eat more fish that do not eat other fish", to avoid health risks, to obtain nutritional benefits, and to make a fish population more sustainable. In Europe and USA, the European Food Safety Authority (EFSA), the Environmental Protection Agency (EPA) and the Food and Drug Administration (FDA) have issued advice on the consumption of fish, with the aim of protecting the population from the adverse effects of MeHg (Evans et al., 2002; EFSA, 2012, 2015; EPA-FDA, 2017). In Colombia, there are no public policies that clearly warn the residents about the weekly consumption quantities of fish and the species with higher levels of MeHg, something that is even more difficult considering that these policies must be in accordance with the social, cultural, and environmental context of the populations in the study area. This river has been seriously impacted by illegal gold mining and the environmental, ecological, and social damage has been of great proportions, and its effects on the health and food security of the population are already evident. In summary, according to results from EDI, THQ, EWI, MFW and Pi indexes, the Atrato River basin population (particularly children and WCHA groups), must be protected from carcinogenic, neural damages, aborts and birth outcomes that could be caused by fish consumption with high levels of As and Hg species. Therefore, educational actions should be taken to establish environmental and health guidelines that allow riverside populations to consume fish safely, avoiding adverse health effects. The results of this work are the basis for initiating a first stage necessary to shape plans and public policies, according to Minamata Convention on mercury, to mitigate the impact of gold mining in the Atrato River basin.

5. Conclusions

This study analyzed the risk to human health from Hg, As, Pb, and Cd to which the inhabitants of the Atrato River basin are exposed through fish consumption. Health risk assessment showed that the EDI and individual THQ were sequenced: $Hg > As > Pb \approx Cd$. The total THQ exceeded 1 in the most consumed species from 12 of the 13 municipalities evaluated, suggesting that the population could experience adverse non-carcinogenic health effects through fish consumption, especially from Hg and As which were the major contributors to this parameter. As in fish showed a higher carcinogenic risk potential for human health than Pb, since in 11 of the 13 municipalities evaluated, a high proportion of the most consumed fish species showed CR values between $1.074\ 10^{-4}$ and $9.675\ 10^{-3}$. The highest MeHg concentrations were measured in carnivorous fish species, particularly in *H. malabaricus*, *A. pardalis, P. schultzi, R. quelen,* and *C. kraussii*. Significant risk for the population with the highest values for the mentioned fish species are suggested by EWI, MFW, and MeHg permissible values. Results suggest that the consumption of these species must be reduced in a large part of the basin, and could be replaced by other such as *O. niloticus, H. hondae*, and *O. mossambicus*. In summary, fish is a valuable source of protein that will not be quickly replaced in the diet of the inhabitants of the Atrato River basin. Therefore, guidance on the cost/benefit of continuing to consume fish must be established, so that the health risk analysis of this study provides a favorable complement to make recommendations.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2022.113120.

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