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Mercury levels in human population from a mining district in Western Colombia

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ABSTRACT

A biomonitoring study was carried out to examine the adverse impacts of total mercury in the blood (HgB), urine (HgU) and human scalp hair (HgH) on the residents of a mining district in Colombia. Representative biological samples (scalp hair, urine and blood) were collected from volunteered participants ($n = 63$) to estimate the exposure levels of THg using a Direct mercury analyzer. The geometric mean of THg concentrations in the hair, urine and blood of males were 15.98 μg/g, 23.89 μg/L and 11.29 μg/L respectively, whereas the females presented values of 8.55 μg/g, 5.37 μg/L and 8.80 μg/L. Chronic urinary Hg (HgU) levels observed in male workers (32.53 μg/L) are attributed to their long termed exposures to inorganic and metallic mercury from gold panning activities. On an average, the levels of THg are increasing from blood (10.05 μg/L) to hair (12.27 μg/g) to urine (14.63 μg/L). Significant positive correlation was found between hair and blood urinary levels in both male and female individuals. Thus the present biomonitoring investigation to evaluate the Hg levels and associated health issues would positively form a framework for further developmental plans and policies in building an ecofriendly ecosystem.

Introduction

Mercury (Hg) is regarded as the sixth most toxic element on earth and it is a naturally occurring element with varied chemical forms (elemental, organic and inorganic) (Hui-Wen et al., 2011) and of significant ecological and public health concern. Natural processes like degassing of the earth's mantle/crust, evasion from soils, vegetation, wildfires, volcanic eruptions and geothermal activities are the sources of Hg (Riaz et al., 2016). On the other hand, Hg also occurs in occupational environments due to its extensive use in gold panning, pharmacology, industries and agriculture, enhancing its presence resulting in a lethal situation (WHO, 2003). Ultimately, Hg can enter human bodies through respiratory or digestive tracts and dermal absorption (Eqani et al., 2016). The transformation of inorganic to organic Hg (methylmercury MeHg) is regarded as the most toxic form and is more often intensified by bioaccumulation and biomagnification routes in the aquatic food webs

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(Ouédraogo et al., 2015). MeHg, with its lipophilic property, is highly neurotoxin and penetrates the blood–brain barrier to the central nervous system which can be fatal (WHO, 1990; Aschner et al., 1992; Chien et al., 2010). Serious occupational and complicated health issues in humans have elicited a global action called the Minamata convention (2013), which aims to protect human health and the environment by stringent plans to reduce mercury exposure and bring in harmonious living (Esteban et al., 2015).

Rising concerns of the threats to human health and environment were the motivating force behind conducting intensive and specific human monitoring programs to assess the exposure levels of Hg and the environmental risks for the populations that live in close proximity to highly polluted regions (Angerer et al., 2007). Over the past few decades, several studies have been conducted (Albert et al., 2010; Tian et al., 2011; Liu et al., 2014; Faial et al., 2015; Riaz et al., 2016; Ma et al., 2016) to evaluate and quantify the Hg levels in human matrices namely hair, blood and urine to estimate the magnitude of its contamination. Human biomonitoring (HBM) is regarded as the most recognizable method for measuring human exposure to toxic elements and hence serves to be a valuable tool for the protection of human health.

In this regard, the aim of the present study was to investigate the levels of mercury in different biological samples (scalp hair, blood and urine) of humans from the mining district of San Juan, Department of Choco, Colombia, an area impacted by gold mining.

1. Materials and methods

1.1. Study area

The mining district of San Juan is located in the south central part of the Department of Chocó (Fig. 1) in Colombia along the San Juan River basin (05°09′–05°21′N; 76°33′–76°41′W) approximately covering an area of 8619 km^2. Colombia offers immense mineral potential and the Department of Chocó is considered to be the country's most resource rich provinces. The country ranks fifth in gold production and produces about 54,000 kg of gold a year, mainly in the Department of Chocó (24,500 kg), Antioquia (19,000 kg) and Bolivar (5700 kg) representing 91% of the annual gold production (Güíza and Aristizábal, 2013). The Department of Chocó mainly encompasses of Afro-Colombian population and gold panning is their foremost economic activity. Major environmental and health hazards in the region are the use of mercury in the ore beneficiation process to extract the precious metal.

1.2. Questionnaire survey

During March and May 2011, a total of 87 residents living in the San Juan District from different population groups namely mine workers, gold and platinum dealers, agriculturalists, pensioners, students, pregnant women and children were involved for the present study. Participants were invited to provide data voluntarily related to an extensive questionnaire for a quick assessment of demographic information including age, sex, height,

Fig. 1 – Map showing the location of the study area in the mining district of San Juan, Department of Choco, Colombia.

weight, profession, alimentation, closeness to the mining activities, health status and their feedback was tabulated.

1.3. *Studied population*

All the samples were collected based on the recommendations put forth by the National Institute of Health of Colombia (INS, 2011). The voluntary individuals (n = 63) included miners (n = 16), gold smiths (n = 5), commercial dealers (n = 23) and general public n = 19). Human scalp hair (5 cm the scalp region) from individuals were collected and stored in plastic bags at room temperature until further analysis. It was also made sure that the participant's hair was clean, dry and free of dyes. In the case of blood samples, approximately 3–5 mL of venous blood per person were collected using 10 mL disposable syringes, transferred to EDTA plastic tubes as anticoagulant, and subsequently stored at 4°C immediately for further analyses within 96 hr. 24 hr urine samples were collected in 2.5 L polyethylene bottles that were prewashed with nitric acid and deionized water.

1.4. *Analytical procedure*

The collected samples were analyzed in the Pharmacology and Toxicology Laboratory (Laboratorio de Farmacologia y Toxicologia), Universidad de Antioquia, Colombia. The total mercury level was determined based on the US EPA method 7473 (SW-846) (USEPA, 2007) using a Direct mercury analyzer (DMA 80 Milestone Series) atomic absorption spectrophotometer. A standard reference material NIST-SRM 3133 Lot no: 061204 - Mercury (Hg) standard solution was analyzed every five samples to ensure quality assurance and control. The entire analysis produced a recovery percentage of (hair — 113.17%; urine — 108.97%; blood — 100.86%).

1.5. *Statistical analysis*

Pearson's correlation matrix was generated using varimax normalized values for the whole data set by means of STATISTICA (version 12.0) to understand the inter-relationship of Hg concentrations in human hair, blood, urine and its dissimilarity in male/ female populations.

2. Results and discussion

Hg exposure levels in different population groups of males and females are illustrated in Figs. 2a–c and 3a–b respectively, based on their occupational activities like miners, gold smiths, dealers and others. Males presented higher exposure to Hg (17.05 μg/g) than the females (7.57 μg/g) probably due to the fact that males consume more fish per Kg body weight at each meal compared to females (Diez et al., 2008). Moreover, gender differences in the metabolization of MeHg and frequent hair treatment (*e.g.*, artificial hairweaving and hair coloring/dyeing) by females might result in the elimination of MeHg (Knobeloch et al., 2007).

2.1. *Hg in hair*

Even though scalp hair Hg (HgH) analysis involves a number of factors like hair growth, color and use of synthetic dyes, external contamination and pretreatments, they are considered to be ideal biomarkers as they provide a well-defined time-integrated data because the average human hair growth is approximately 1 cm/month (WHO, 1990; Sakamoto et al., 2016). Moreover, scalp hair has frequently been recognized as a biomarker for evaluating methyl mercury (MeHg) exposure (McDowell et al., 2004), since this toxic organic form is incorporated into hair follicle in proportion to its content in blood.

In the present study, hair Hg concentrations (μg/g) were observed comparatively higher in the male populations (15.98) than the females (8.55). Variabilities among individuals are based on their susceptibilities in mercury accumulation and genetic differences (Castaño et al., 2015). However, previous studies of similar kind also suggest that Hg concentrations in hair were highly associated with the sea food consumption (Chien et al., 2007; Liu et al., 2014) and various occupational exposures. The hair Hg concentrations were also observed to be varying in accordance with age due to the change in dietary and nutrition patterns with increasing age (Nakagawa, 1995). Infants and children recorded a value of 1.11 μg/g (boys) 4.45 μg/g (girls), where it is primarily sourced from breast or bottle feeding, rapid development, prenatal exposure, shared dietary patterns from mothers and their exposure to mercury via contaminated soils like hand to mouth activities and playing time (Health Canada, 2004; Tian et al., 2011; Safruk et al., 2015). In males, Hg levels increased in the lower age group of (20–40 yr) to 22.86 μg/g and was found to be gradually decreasing in the higher age group from 13.76 μg/g (40 to 60 yr) to 0.08 μg/g (60 to 80 yr) due to the slower rate of mercury metabolism and less dense hair (Janicka et al., 2015). In case of females, individuals from 40 to 60 years presented higher levels of Hg (21.88 μg/g) in hair due to high consumption of fish. Based on the profession, miners (males: 13.59 μg/g) and dealers (males: 26.79 μg/g; females: 21.87 μg/g) presented chronic levels of hair Hg due to their occupational exposure in gold panning activities where the mercury vapor is irreversibly adsorbed on human hair (Queipo Abad et al., 2016). Moreover, specific diets could also result in definite effects in terms of limiting Hg absorption and accumulation (Passos et al., 2003; Golding et al., 2013). In addition, the interaction between different factors plays an important role in determining the Hg concentrations in the studied matrices.

2.2. *Hg in blood*

Male and female participants presented an average (μg/L) blood Hg value of 11.29 and 8.80 respectively, which is an indicator of internal Hg burden (Halbach and Welzl, 2010). Less blood mercury levels than in the hair and urine are ascribed to the fact that the biological half-life of Hg absorbed into blood is 2–4 days when 90% is excreted through urine and feces (Ha et al., 2017). Blood Hg levels (μg/L) were higher in the male miners (11.86) and dealers (11.70) as the concentrations increase rapidly even after a brief exposure (Ye et al., 2016). In accordance with the age groups, the older males (90.14 μg/L) presented chronic levels of blood Hg suggesting that older people have been consuming more fish during their lives (Liu et al., 2014), which is considered to be a major source for MeHg. Moreover,

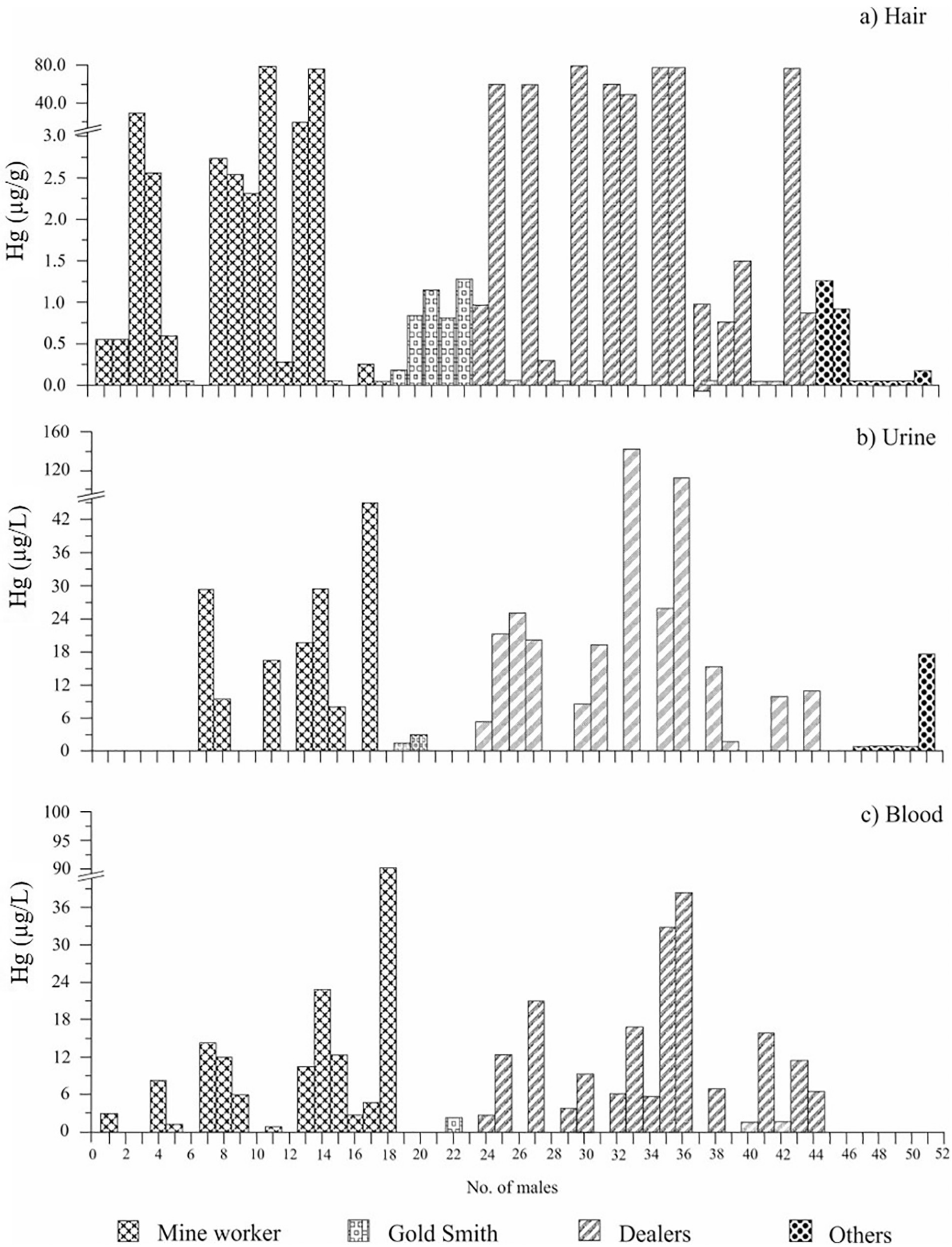

Fig. 2 – a–c. Mercury concentrations in hair (a), urine (b) and blood (c) of the male populations based on their occupation.

long exposures to inorganic Hg due to the local occupational activities also prove to be important criteria for high levels observed in individuals as in females of age 40–60 yrs. (13.81 μg/L).

2.3. *Hg in urine*

Urine samples from the mine workers were easily available hence this matrix was measured to assess the level of

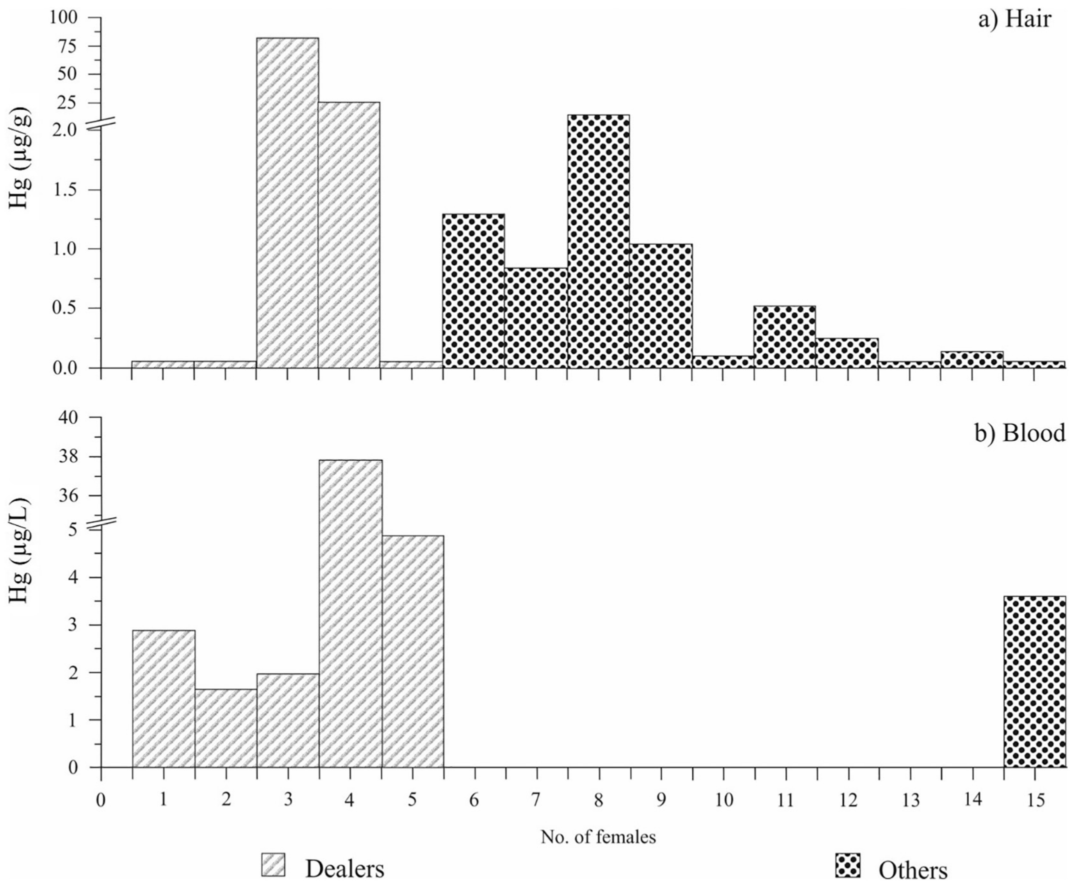

Fig. 3 – Mercury concentrations in hair (a) and blood (b) of the female populations based on their occupation.

exposure to Hg° vapor. Chronic concentrations of Hg (μg/L) observed in the urine samples ((avg.) male: 23.89; female: 5.37) exhibit nearly 90% of inorganic and metallic mercury reflecting the present or recent exposure level (Ye et al., 2016). High HgU in the male miners (22.47 μg/L) and dealers (32.53 μg/L) affirms that they were severely exposed to mercury vapor from the smelting process of gold and mercury. Extremely high HgU concentration (3662 μg/L) was recorded from Chinese mercury mine workers in Guizhou Province by Sakamoto et al. (2007), ~90 fold higher than the health-based occupational exposure limit level (50 μg/g creatinine) set by WHO (1991). Extremely high levels of Hg observed in working men indicate long term exposure causing adverse effects to the body, which is also well evidenced in the study area where the smelters and local populations are exposed to toxic Hg° vapor due to gold panning activities (Klassen, 2007).

2.4. Statistical analysis

Correlation studies for the biomarkers (hair, urine, blood) in males and females were carried out ($p < 0.05$). Significant positive correlation was observed between hair and blood in both males ($r^2 = 0.59$) and females ($r^2 = 0.87$), representing the fact that mercury in hair is directly proportional to the amount of mercury in blood reflecting the body load of the metal (Cernichiari et al., 1995). Moreover, hair which is made up of a protein called keratin includes a large amount of sulfur containing amino acid cysteine. Therefore, Hg with great affinity for sulfur binds rapidly and as hair grows actively, Hg in the blood into the tissues and to the hair follicles (Faial et al., 2015). Relationship between urine and blood ($r^2 = 0.59$) the chronic exposures of Hg for long term and high consumption of seafood which is an important pathway of MeHg to humans.

2.5. Local health issues

The local residents of San Juan, Department of Choco in Colombia also presented various health issues due to the local artisanal gold panning activities (Table 1). Chronic and long term exposures may result in neurotoxicity affecting the central nervous system. The most vulnerable and targeted organs include the brain and kidneys (UNIDO, 2008). Moreover, prenatal exposure of Hg is another well pronounced health issue that could result in fetal-type MeHg intoxication during gestation (Chen and Hu, 2010; Grandjean et al., 2005). Physical disturbances like headache, body pain, memory loss, muscular cramps were common symptoms among the miners who are directly exposed to Hg° vapor.

3. Comparative studies

Comparing the Hg concentrations (Table 2) in human matrices (scalp hair, urine and blood) of the present study with that

Table 1 – Health related symptoms witnessed by the local residents due to Hg exposure.

Symptoms	*N* (%)	Mine workers	Dealers	Housewives	Jewelers	Others
Headaches	44.3	6	11	4	1	9
Memory loss	15.7	3	7	1	0	0
Cramps	27.1	6	7	1	1	4
Tremor	8.6	2	3	0	1	0
Hardening of hands	1.4	0	1	0	0	0
Less strength	1.4	0	0	0	0	1
Mood swings	2.9	1	0	1	0	0
Hemoptysis	1.4	0	0	0	1	0
Joints pain	1.4	0	0	0	1	0
Insomnia	1.4	1	0	0	0	0

N = Number of individuals.

of the other studies were conducted worldwide and it was observed that the inhabitants of Gilgit, Pakistan presented chronic levels of Hg (in μg/L) in urine (men 40.9; women 34.9) and blood (men 38.7; women 30.77). The factors responsible for the extreme levels of Hg are the century old artisanal mining practices of placer gold by the gold washers in the locality. Similar values of Hg exposures by the populations of Colombia and Pakistan suggest the fact that small scale mining in these countries largely to their economies (Riaz et al., 2016) and in turn the human health. Other studies in women of child-bearing age from various parts of the world represent Hg levels as a consequence of high seafood consumption which is matter of concern as it could affect the fetal development. The average Hg concentrations in the present study were found to be eight to fifteen folds higher than the reference limits set forth by USEPA (2001) and WHO (1990) respectively. Likewise, blood mercury levels were also two fold higher than the tolerable limit of 5.8 μg/L representing high burdens of Hg in humans.

Table 2 – Comparison of Hg concentrations in different populations from various parts of the world.

	Hair (μg/g)	Urine (μg/L)	Blood (μg/L)	References
Pakistan				
Men	2.7	40.9	38.7	Riaz et al. (2016)
Women	1.8	34.9	30.77	
Augusta Bay, Southern Italy	2.61	1.67	10.1	Bonsignore et al. (2016)
Korea				
Men	–	–	4.04	Park et al. (2014)
Women	–	–	2.99	
France, Amazonia				
Women	10.3	–	–	Chevrier et al. (2009)
French Guiana				
Women (Children < 12 years)	7.4	–	–	Cordier et al. (2002)
Seychelles Islands				
Women	6.8	–	–	Myers et al. (2000)
Germany				
Women	6.15	–	–	Bose-O'Reilly et al. (2008)
Japan				
Women	1.51	–	–	Ohno et al. (2007)
Italy				
Women	0.56	–	–	Diez et al. (2008)
Kazakhstan				
Men	0.823	–	–	Hui-Wen et al. (2011)
Women	0.416	–	–	
South China Sea				
Men	1.123	–	–	Jin-Ling et al. (2014)
Women	0.916	–	–	
Colombia, Bolivar				
Men	1.94 ± 0.9	–	–	Olivero-Verbel et al. (2011)
Women	1.40 ± 0.06	–	–	
Reference limits				
USEPA	1[a]	–	5.8[b]	USEPA (1997)
WHO	1.9	–	–	WHO (1990)
Present study (Colombia)				
Men	15.98	23.89	11.29	
Women	8.55	5.37	8.80	

4. Conclusion

In regard to the increasing global Hg levels due to anthropogenic emissions, this baseline study report provides a baseline report on the Hg exposure levels in human matrices (hair, urine and blood) of local residents in a mining district in Colombia. High HgU in male workers (32.53 μg/L) indicated their long term exposure due to occupational exposures and differences in metabolization of MeHg. The high mercury content in females is a matter of concern as there is direct negative consequence on fetus brain affecting its development during pregnancy. However, individual susceptibility also plays a vital role in mercury accumulation based on their genetic variations, metabolism, diet and nutrition patterns. The degree of Hg exposure from lower to chronic levels is due to the result of global pollution and occupational activities. Hence, this study from a mining district in Colombia proves to fill the gap in environmental health research by providing guidelines to reinforce stringent policies for the betterment of human societies and environment. Assessment of methylmercury (MeHg) from the exposure group, the relationship between consumption of local fish and Hg levels and further comparative investigation with a control group could help to improve the evaluation of the human exposure to Hg in this contaminated region. Health surveys and provisions are recommended for adequate protection of mine workers from the present exposure level.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at https://doi.org/10.1016/j.jes.2017.12.007. These data include the Google map of the most important areas described in this article.

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