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MINI-REVIEW

A Review on the Status of Mercury Pollution in Pakistan: Sources and Impacts

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Abstract

Mercury (Hg) contamination in environmental matrices and associated human exposure has been recognized as a critical long-lasting issue worldwide. However, studies are still elusive that summarized the overall status of Hg pollution and its impacts on public health in Pakistan. Hence, this review encompasses the environmental prevalence, potential sources, and human exposure tendencies to Hg contamination in Pakistan. Reviewed literature revealed jolting levels of Hg in various environmental samples, such as dust, soil, water, and air collected from the residential and industrial areas. Inhalation of Hg via dust particle was identified as the primary pathway for human exposure, while atmospheric deposition and gold mining are identified as the two primary sources of Hg contamination in the environment. Considering human exposure, the highest bioaccumulation of Hg was ranged from 5885 to 8698 µg/kg in hair samples collected from the residents of the Kashmir Valley, Pakistan. However, in the lower Himalayan regions, including Islamabad and Swabi, the concentration of Hg in hair samples was reported at 1085 µg/kg, slightly beyond WHO devised reference dose (RfD) of Hg (1000 µg/kg). This review revealed the worst scenario of Hg contamination in human biomatrices and environmental compartments in Pakistan, which needed immediate rehabilitation measures.

Mercury (Hg) is a well-known toxic heavy metal and ubiquitous in different environmental compartments, such as biosphere, lithosphere, hydrosphere, and atmosphere (Basu et al. 2018). The complex series of chemical transformation and multiple oxidation states allow Hg to persist in the environment (Zhang and Wong 2007). The predominant inorganic forms of Hg in the environment include mercuric salts, such as $HgCl_2$, $Hg(OH)_2$, and HgS, while prominent organic forms encompass CH_3HgCl and CH_3HgOH, and other organic forms present in small but important fractions include C_2H_6Hg and C_6H_5Hg (Zhang and Wong 2007). Previous studies reported various sources of Hg emissions, such as natural, anthropogenic, and reemitted. Anthropogenic sources of Hg include mining activities and wastes from combustion, urban, and industrial discharges (Basu et al. 2018). Atmospheric deposition is the central pathway for contamination of Hg in the environment (Zhu et al. 2018). As shown in Fig. 1, humans are exposed to Hg via various pathways, including internal exposure pathways (blood, urine, and breast milk) (Basu et al. 2018) and external exposure pathway (ingestion through water, food, and dust; inhalation through air) (Sundseth et al. 2017). Hg contamination lead to various health abnormalities related to genetic, reproductive, renal, neurological, and cardiovascular systems (Andreoli and Sprovieri 2017; Xu et al. 2018). Recent atmospheric concentration of Hg is associated with the industrial revolution, which increased the environmental contamination of Hg by tenfold (Zhang and Wong 2007; Driscoll et al. 2013). Annually, the anthropogenic sources emit approximately 2000–2200t of Hg into the environment (Seigneur et al. 2004; Burmistrz et al. 2016). However, approximately 2000t of Hg also releases into the environment from the natural sources, such as soil, forests,

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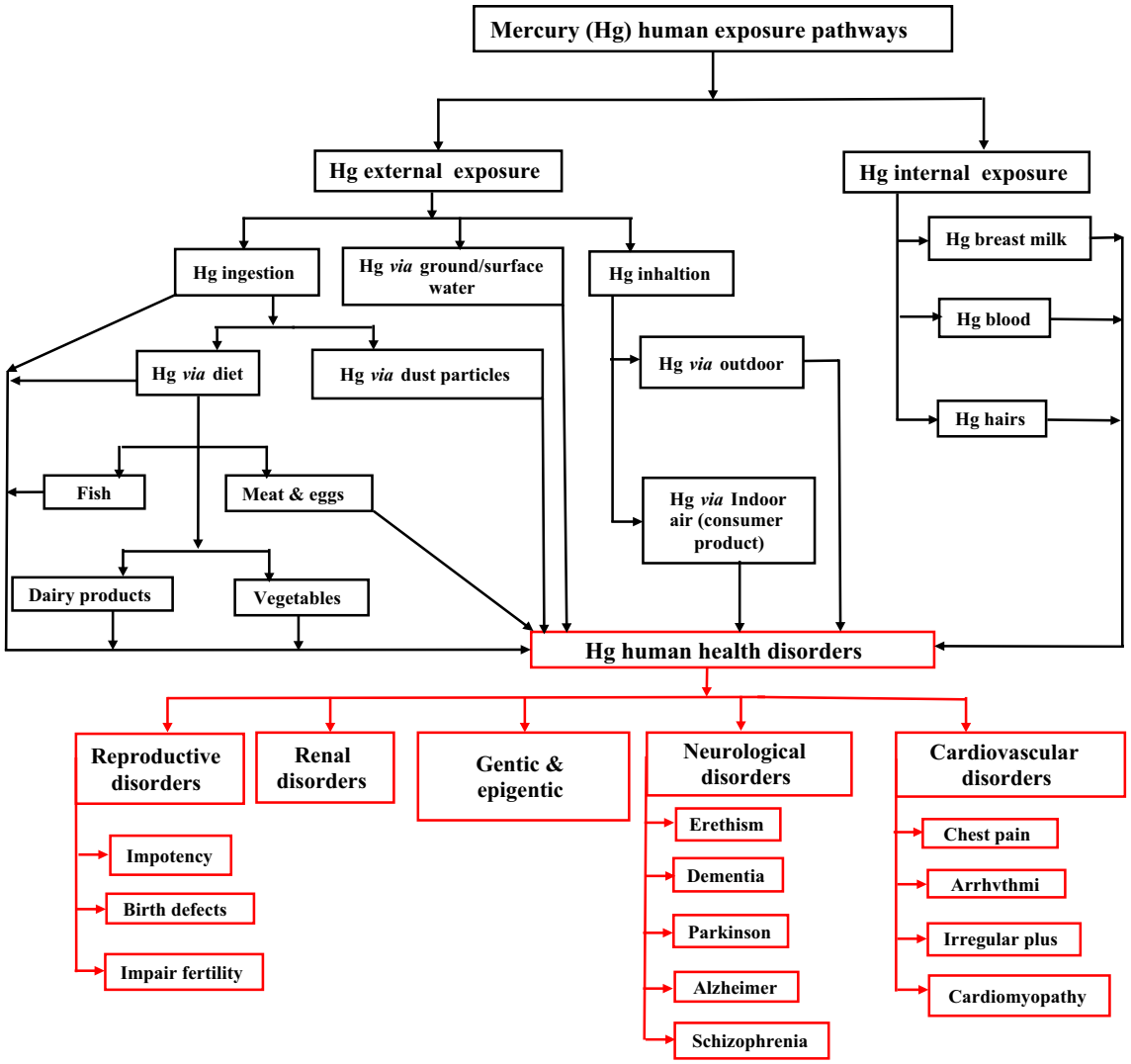

Fig. 1 General overview of mercury exposure pathways and associated human health abnormalities

lakes, open oceans, and volcanic emissions (Selin 2009) and approximately another 2000t of Hg reemits from the previous deposits (Zhang and Wong 2007). Globally, the total estimated Hg emission in 2009 ranged from 3290 to 4860 Mg compared with 2480 Mg in 2006, which implied an increase of up to 96%. The main driving forces enhance Hg emission may include expansion of coal-fired power plant for electricity generation in developing world, mainly in Asia (Giang et al. 2015). Therefore, this article is an attempt to summarise the contamination status and fate of Hg in various environmental matrices and human biomatrices in Pakistan. Moreover, the potential exposure pathways and health effects of Hg are also summarized in this study.

Methodology

A literature search was performed related to the main theme of this study: the contamination of Hg in various environmental compartments and human biomatrices in Pakistan, and associated potential exposure pathways and health effects, using databases of Science Direct, Web of Science, Google Scholar, and PubMed. Inclusion criteria were based on original research articles, peer reviews, empirical studies, short communications, and institutional reports for the years 1991–2018. The keywords used for search were: "Hg pollution, air, soil, dust, water,

sediments, soil along road side, quantification, Hg exposure pathways, gold miner's female/male, blood, hairs, urine and nails, Hg reference dose (RFD), total mercury (THg), organic mercury (OHg), inorganic mercury (IHg), distribution, status, analysis, acute/chronic health effects, disease incidences, Korea, Finland, Columbia, Japan, China, Malaysia, Hong Kong, Sweden, USA, Baltimore, San Francisco, Poland and Pakistan." These terms were used randomly with diverse combination and dissimilarities for literature search. The literature search was performed between August-December 2017 and December-January 2018. The data from research articles were extracted and analyzed using XLSTAT 2014 (Addinsoft, USA), Arc GIS 10.3 software (ESRI, USA), and Microsoft Excel 2016.

Mercury Levels in Different Environmental Compartments in Pakistan

Mercury Contamination in Dust

Pakistan is characterized by various land use and altitudinal settings that include Himalaya mountains, the Hindukush, and Karakoram long ranges in the north, the flat-lying Indus plain in the east, the upland Baluchistan plateau in the west, and the coastal Arabian Sea in the south (Fig. 2). On the basis of these characteristics, (Eqani et al. 2016a) divided Pakistan into five zones. Zone 1, the Swat Valley, is comprised of high mountain ranges of Skardu, Hunza, and Gilgit-Baltistan. Zone 2 is the Kashmir valley, which included mountain areas. Zone 3 is the lower Himalayan mountain ranges, which included Abbottabad, Nowshera, Swabi, Islamabad. Zone 4 is the Indus plain agriculture region, consisting of Bhakkr, Dera Ghazi Khan, Mianwali, Layha, Khairpur, and Sukkur along the Indus River. Finally,

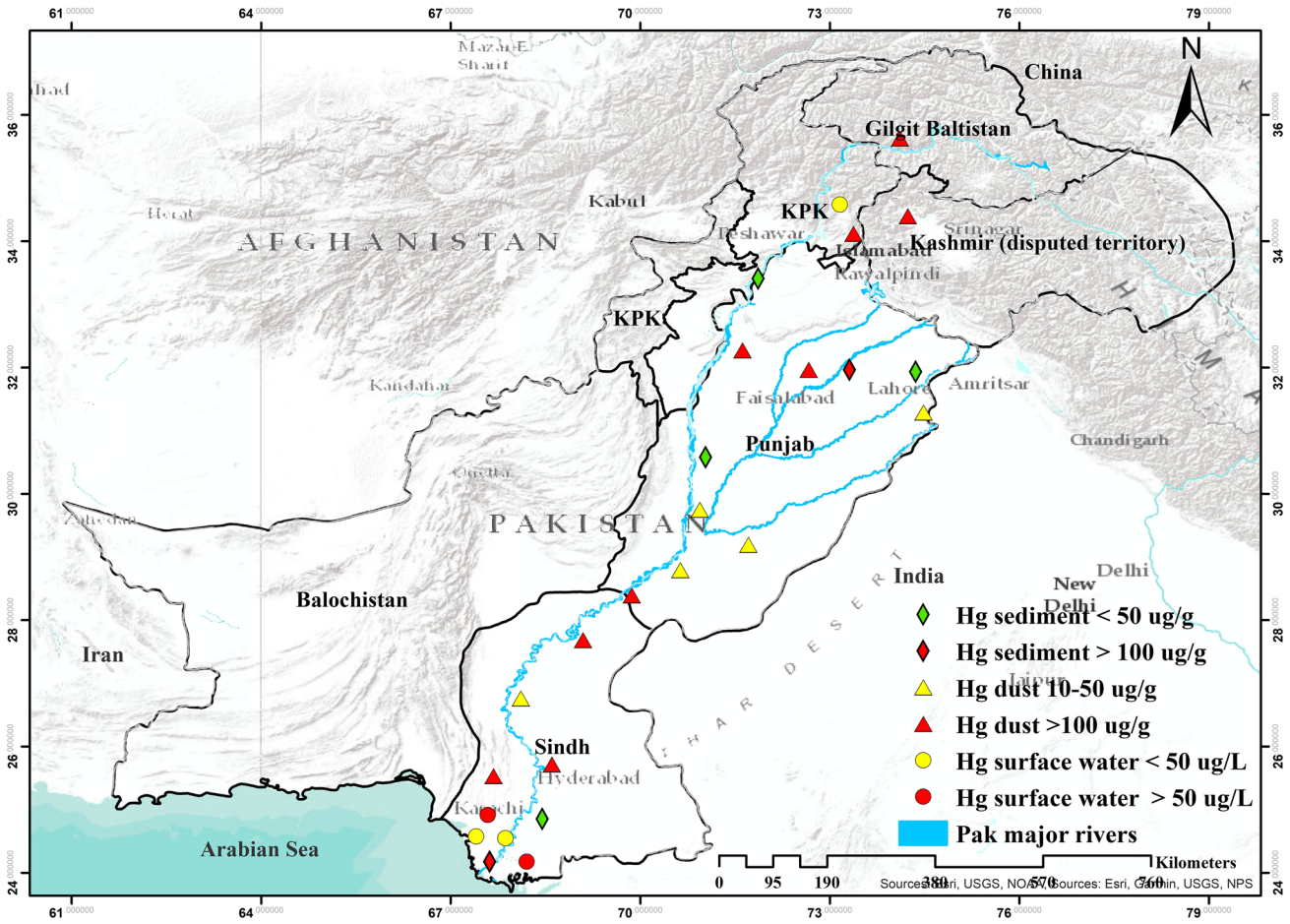

Fig. 2 Reported concentrations of mercury in different environmental matrices, such as sediments, dust, surface soil, surface water, and groundwater at various locations in Pakistan

zone 5 is the urbanized industrial areas of the Indus plain with high population density, including Sargodha, Lahore and Hyderabad. Eqani et al. (2016a) selected 22 sites, and the concentration pattern of THg in dust particles was varied according to different type of land uses in different zones. The highest concentration of THg ranged from 30 to 6120 µg/kg (median 3075 µg/kg) and was reported in major urban and agricultural land use regions. However, in agricultural areas around the Indus delta, the total mercury level ranged from 10 to 640 µg/kg (median 325 µg/kg). The highest level of THg 640 µg/kg was reported in Mianwali district due to the stone crushing and cement production units that may cause the release of Hg into the environment (Bhowmik et al. 2015; Eqani et al. 2016a). Regarding urban areas with significant industrial units, such as in Hyderabad, Lahore, and Sargodha, the maximum concentration of THg ranged from 120 to 6120 µg/kg (median 3120 µg/kg) in dust particles. In the northern mountainous regions, Gilgit-Baltistan and the lower Himalayan regions (Islamabad and Abbottabad), the concentration of THg was ranged from 90 to 450 µg/kg (median 270 µg/kg), and the maximum concentration was reported in Gilgit-Baltistan at 450 µg/kg (Table 1). The primary sources of Hg emission in these areas was mainly attributed to the agricultural and gold mining activities that may cause an increase in the levels of THg (Khan et al. 2012; Bhowmik et al. 2015; Biber et al. 2015; Eqani et al. 2016a). Due to the unique climatic conditions in this region, the geogenic process also contributes

Table 1 Hg contamination and associated various sources reported for groundwater surface water, sediments, surface soil, and dust samples collected from various locations in Pakistan

Location	Environmental matrices	Major Hg sources	Maximum Hg concentration	References
Karachi (Malir river basin)	Hg in groundwater (µg/L)	Sewage and industrial drain	56	Farooq et al. (2010)
Karachi		Sewerage and industrial effluent	10	Rahman (1996)
Karachi		Direct dispose of industrial effluent into recharge sources	10	Ashraf et al. (1991)
Kala Shah Kaku	Hg in surface water (µg/L)	Chlor-alkali plants and atmospheric deposition	6.7	Jamil et al. (2015)
Badin		Continuously carries industrial effluents and municipal wastes	55	Qureshi et al. (2015)
Tarbela	Hg in sediments (µg/g)	Continuously dispose of industrial and municipal wastes into river indus	14	Ashraf et al. (1991)
Chashma			17	
Lloyd			140	
Lahore			10	
Haleji Lake		Rivers and lakes continuously under pollution stress from urban and industrial sources	20	Tariq et al. (1994)
Karachi			2670	
Taunsa			30	
Lahore	Hg in surface soil (µg/g)	Vehicular traffic as the main nonpoint source and unregulated incineration and dumping sites of hazardous waste material along roadside are the key sources of Hg soil pollution	82	Khan et al. (2011)
Multan			64	
Bahawalpur			62	
Rahimyar Khan			75	
Ubaro			144	
Sukkur			89	
Moro			75	
Hyderabad			98	
Karachi			128	
Kala Shah Kaku		Chlor-alkali plants and atmospheric deposition	14.8	Jamil et al. (2015)
Kashmir valley	Hg in dust (µg/g)	Atmospheric deposition	210	Eqani et al. (2016a)
Gilgit-Baltistan and Swat valley		Gold refining activities	450	
Lower Himalayan mountains		Atmospheric deposition	310	
Indus Plain-Agriculture		Operational cement production units and huge scale stone crushing activities	640	
Indus Plain-Industrial		Dental clinic, chlor-alkali units industrial discharge and toxic agricultural runoff atmospheric deposition	6120	

toward the natural release of Hg (Azizullah et al. 2011). In the Kashmir valley, the concentration of THg in dust ranged from 10 to 210 µg/kg (median 110 µg/kg), and most of the contributing sources were of natural origin, including forest fires, geological processes, and climatic conditions. On the other hand, the long-range atmospheric transportation of Hg-contaminated dust from the neighboring countries, such as China and India. Within Pakistan, the atmospheric transportation of Hg from the southern area also contributed in Hg contamination toward the northern areas of Pakistan (Eqani et al. 2016a, b).

Mercury Contamination in Surface Soil

Mercury pollution in soil has attained significant attention from past couple of decades, because elevated Hg exposure via soil can cause serious health problems (Dong et al. 2010; Khan et al. 2011). Mercury can accumulate in human as well as in animal via direct ingestion of Hg-contaminated vegetables, food stuff, or inhalation of Hg-polluted soil particles and through dermal contact. Previous studies conducted on soil samples collected from roadside in Pakistan reported various possible sources of Hg contamination, including hospital waste, gastrointestinal tubes, batteries, fluorescent bulbs, switches, vehicle lights, and dumping of electronic waste. Khan et al. (2011) reported Hg contamination in surface soil along national highway-N5 from nine selected sites. The overall concentration of total Hg in surface soil ranged from 61.5 to 144 µg/kg (median 102.7 µg/kg). It was about 45 times higher than the acceptable concentration of Hg in soil (Khan et al. 2011). The highest concentration of Hg was reported at 144 µg/kg in surface soil of Ubaro city (Table 1), and this city is a junction point between two big provinces of Pakistan, Sindh, and Punjab. The main cities in Sindh province, such as Karachi (128 µg/kg), Hyderabad (98 µg/kg), Sukkur (89 µg/kg), and Moro (75 µg/kg), were reported with the high level of Hg contamination compared with the different cities in Punjab province, such as Lahore (82 µg/kg), Rahim Yar Khan (75 µg/kg), Multan (64 µg/kg), and Bahawalpur (62 µg/kg) (Khan et al. 2011). The total organic content in soil also increases the adsorption capacity of the soil for OHg (Ozaki et al. 2004; Zhang et al. 2013). According to the Khan et al. (2011), no correlation was found between total organic carbon (TOC) and Hg, indicating that the surface soil in Pakistan was contaminated with InHg instead of OHg.

Mercury Contamination in Sediments, Ground, and Surface Water

The Indus River is one of the most significant and essential rivers in Pakistan, originated from northern hilly regions, stretched from the northwest frontier and flanked through the agricultural and industrial regions of Punjab and Sindh, finally drained into the Arabian sea. The Indus River and its tributaries served as the major flux for heavy metals form industrial, municipal, as well as agriculture wastes that finally sank into the Arabian sea (Khan et al. 2011; Eqani et al. 2016a). The primary reservoirs of Pakistan included Tarbela, Lloyd, and Chashma, which were built along the downstream of the Indus River. The increasing population, industrialization, urbanisation, and waste dispersion were reported as the leading causes to increase the trace metal contents in these reservoirs (Azizullah et al. 2011). A study was conducted on sediments samples from different sites along the Ravi River by Tariq et al. (1994). The average Hg concentration was reported at 0.21 µg/g in Okara, 0.2 µg/g in Make, 0.13 µg/g in Lahore, 0.08 µg/g in Kamila, and 0.11 µg/g in Baloki. Another study was conducted by Ashraf et al. (1991) in the three significant reservoirs: Tarbela, Lloyd, and Chashma. The maximum concentration of Hg in sediments was 140 µg/g in Lloyd reservoir (Table 1), whereas that in Chashma was 17 µg/g and in Tarbela was 14 µg/g. However, the highest level of Hg (2670 µg/g) was reported in industrial wastewater from Karachi (Tariq et al. 1994; Boncompagni et al. 2003), and this wastewater was drained into the freshwater bodies directly without proper treatment. It also was identified as the primary source of Hg contamination in Karachi. The contamination of Hg in surface water from Taunsa Barrage on Indus River was observed as 20 µg/L and in sediments from Haleji Lake as 30 µg/g. The increasing level of Hg pollution in sediments may cause adverse effects on the environment. The translocation and bioaccumulation of Hg via food chain to human also were observed as one of the major issues. In Pakistan, the data about Hg contamination in water are very limited, and only few studies exist. According to Pakistan Council of Research in Water Resources (PCRWR), 5% of the surface water in the country is contaminated with Hg beyond permissible limits (Azizullah et al. 2011). According to Rahman (1996), the Hg concentration in groundwater samples from Karachi was reported at 10 µg/L, whereas the concentration were specifically high in Malir Basin, i.e., 56 µg/L (Ashraf et al. 1991; Farooq et al. 2010). Similarly, Qureshi et al. (2015) reported Hg concentration of 55 µg/L in surface water samples from Badin, Sindh. Generally, all previous studies showed Hg concentration in groundwater of Pakistan beyond the set standard of WHO, i.e., 1 µg/L (Table 1).

Mercury Levels in Human Blood

The total Hg levels in human blood samples collected from various countries, including Pakistan, are presented in Fig. 3a. The average concentration of THg in blood samples residents of San Francisco, Northern Quebec, and Pakistan were reported beyond the corresponding reference dose

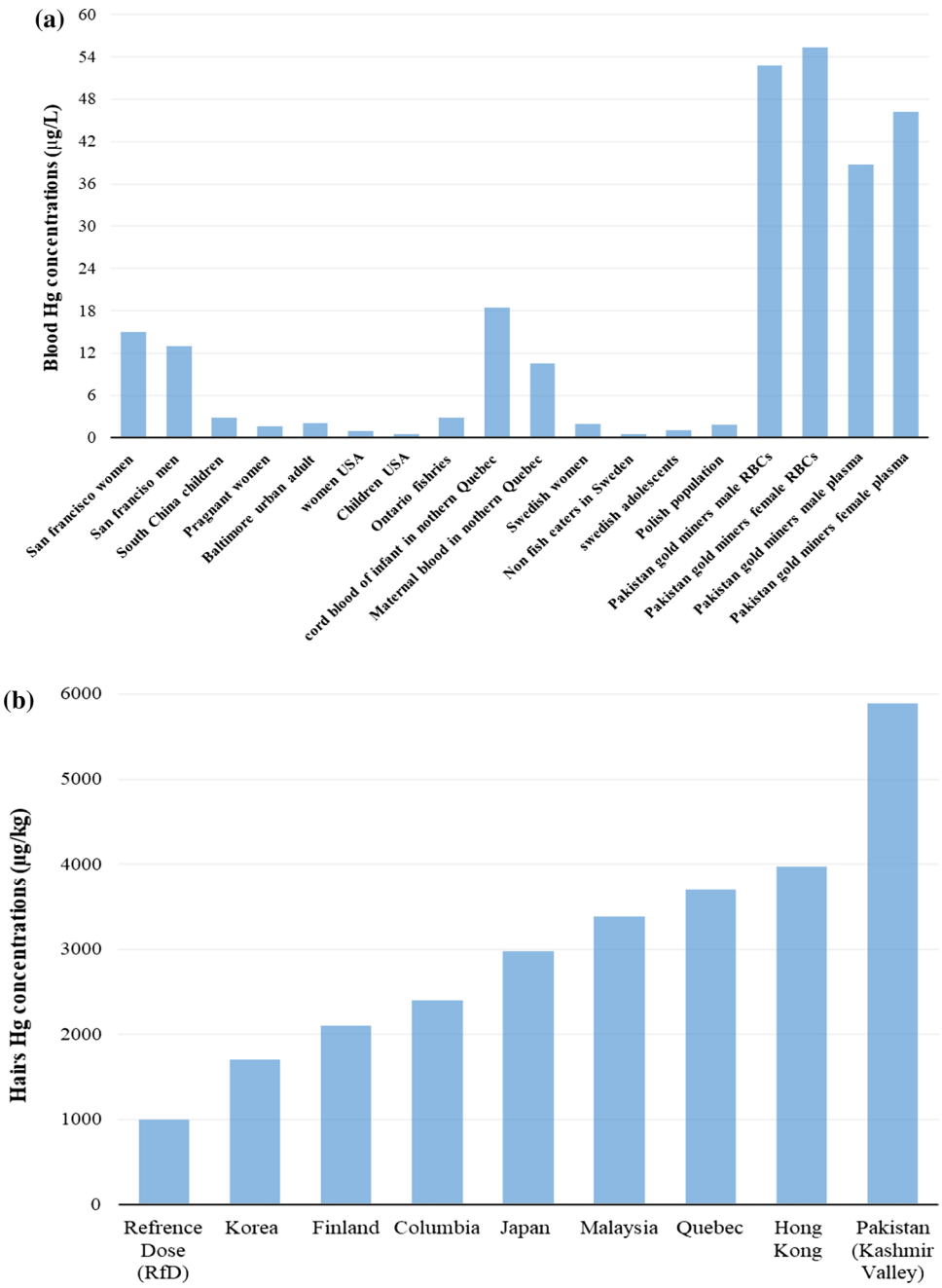

Fig. 3 The average mercury (Hg) concentration in human blood samples from different countries in comparisons with Pakistan (**a**), children from South China (Ip et al. 2004a, b), men and women from San Francisco (Hightower and Moore 2003), adult population from Baltimore (Weihe et al. 2005), women and children from USA (Schober et al. 2003), Ontario fishers (Cole et al. 2004), infant and mothers from Quebec (Cole et al. 2004), Swedish women (Björnberg et al. 2005), non-fish consumers from Sweden (Lindberg et al. 2004), Swedish adolescent (Bárány et al. 2002), Polish population (Lech and Sadlik 2004), pregnant women from Songyuan city, northeast China (Zhang and Wong 2007) in RBCs and plasma in gold miners males and females from Gilgit-Baltistan, Pakistan (Riaz et al. 2016). Comparison of mercury concentration in human hair samples from countries with those from Pakistan (**b**)

(RfD) of THg (5.8 µg/L) in blood, devised by USEPA (Riaz et al. 2016). The average concentration of THg in blood samples of gold miners in Pakistan was reported as male 52.79 µg/L in RBCs, females 55.32 µg/L in RBCs, male 38.7 µg/L in plasma, and female 46.2 µg/L in plasma. The comparison of THg concentration in blood samples between male and female showed higher levels in female compared with that of male (Table 2). The maximum concentration of THg in the blood was attributed to the direct inhalation of Hg fumes, and the female workers were at the high risk of Hg contamination due to their active participation in roasting of the amalgamated gold (Riaz et al. 2016). In blood samples, the lowest average concentration of Hg was reported at 0.19 µg/L in Sweden, whereas in other countries the concentration ranged from 1 to 2 µg/L, except in Hong Kong, China, where the concentration was as high as 3.53 µg/L (Ip et al. 2004a; Zhang and Wong 2007). The Hg level was almost three times higher in pregnant females from Songyuan, China, compared with nonpregnant females (Zhang and Wong 2007). It was reported that in children who consumed Hg-contaminated fish more than three times per week, the level of Hg contamination in their hair and blood samples was almost twice compared with children who consumed fish once per week (Ip et al. 2004a; Castaño

Table 2 Contamination status of organic (OHg), inorganic (InHg) and total mercury (THg) in human biometrics (blood, nails, hairs, and urine) collected from various locations in Pakistan

Location	Human biomarkers	Hg concentration Minimum–maximum and median	References
Hg concentration in human hairs (μg/kg)			
Kashmir valley	Hairs	116–8698 and 4407	Eqani et al. (2016a)
Gilgit-Baltistan and Swat valley		16–960 and 353	
Lower Himalayan mountains area		2.94–3255 and 1629	
Indus Plain-agriculture area		10–1129 and 569.5	
Indus Plain-Industrial area		188–5885 and 3036.5	
Gilgit Pakistan (THg male)		0.6–6.7 and 3.6	Riaz et al. (2016)
Gilgit Pakistan THg female)		1.1–4.03 and 2.6	
Hg concentration in human blood (μg/L)			
Gilgit Pakistan	InHg RBCs male	9.5–23.5 and 16.5	Riaz et al. (2016)
	InHg RBCs female	10.5–24 and 17.3	
	RBCs OHg male	25–50.5 and 35.2	
	RBCs OHg female	25.2–55.5 and 40.3	
	RBCs THg male	41–64.7 and 52.9	
	RBCs THg female	44.4–67 and 55.7	
Gilgit Pakistan	InHg plasma male	9.5–18.3 and 13.9	Riaz et al. (2016)
	InHg plasma female	10.3–19.8 and 15	
	OHg plasma male	18.5–34.4 and 26.5	
	OHg plasma female	24.7–39.2 and 31.9	
	THg plasma male	30.8–51.9 and 41.4	
	THg plasma female	36.8–52 and 44.4	
Hg concentration in human urine (μg/L)			
Gilgit Pakistan	InHg urine male	32.8–49.6 and 41.2	Riaz et al. (2016)
	InHg urine female	31.2–41.5 and 36.4	
	OHg in urine male	11.5–29 and 20.3	
	OHg in urine female	11.4–21.3 and 16.4	
	THg in urine male	49.5–76 and 62.8	
	THg in urine male	41.5–60. 4 and 51.2	
Hg levels human nails (μg/kg)			
Gilgit Pakistan	THg in nails male	0.5–6.3 and 3.4	Riaz et al. (2016)
	THg in nails female	0.97–7.9 and 4.4	

et al. 2015). The Hg-contaminated fish consumption population had a higher level of Hg concentration of ninefold in blood, sevenfold in hair, and 15-fold in urine as compared with the low Hg-contaminated fish consumption population (Johnsson et al. 2005; Zhang and Wong 2007). It also was reported in Swedish women that the Hg concatenation increase with increase in the consumption of Hg-contaminated fish (Björnberg et al. 2005).

Mercury Levels in Human Hairs, Urine, and Nails

The elevated concentration of Hg was reported in human hair samples collected from various areas of Pakistan, such as the Kashmir Valley, the Lower Himalayan region, the Indus Plain-Agriculture region, the Indus Plain-Industrial region, the Gilgit-Baltistan region, and the Swat Valley region. The concentration of Hg in the Gilgit-Baltistan and the Swat Valley were lower than the corresponding RfD of Hg in hair, i.e., 1000 μg/kg (Rice 2004; Eqani et al. 2016a). Due to the elevated consumption of Hg-contaminated fish, the high concentration of Hg was reported in hair samples from Munduruku, Brazil, and the State of Para. The Hg concentration in hairs of the children from gold-mining areas of Ecuador were reported six times higher than the RfD value, and it was predicted that the Hg emissions were mainly originated from the gold smelting activities. Similarly, Hg concentration in hair samples ranged from 1570 to 3970 μg/kg in Hong Kong, 2020 to 2980 μg/kg in Japan, 1100 to 1700 μg/kg in Korea, 3380 μg/kg in

Malaysia, 2400 to 3700 µg/kg in Quebec, 1330 to 2400 µg/kg in Columbia, and 2100 µg/kg in Finland (Fig. 3b). All of these reported concentrations were beyond the RfD values, indicating the population of these countries were at high risk due to the Hg exposure through frequent use of Hg contaminated fish (Zhang and Wong 2007). A study was conducted by Eqani et al. (2016a) on detection of Hg in hair samples collected from different land use areas in Pakistan. The average bioaccumulation of Hg in human hairs was reported as 188–5885 µg/kg (median 3036 µg/kg) in industrial areas, 116–8698 µg/kg (median 4407 µg/kg) in mountain areas of the Kashmir Valley and relatively lower concentration as 2.94–1085 µg/kg (median 544 µg/kg) in the Himalayan region, including Islamabad, Swabi, and Abbottabad. However, the relatively lower mean concentrations (300 µg/kg) were reported in hair samples collected from the Gilgit-Baltistan, the Swat valley, and agricultural areas of the Indus plain region and the low-lying southern region (Table 2). These values were much lower than that of RfD value of Hg for hair. The ingestion of dust particles was identified as one of the major exposure route for Hg contamination in occupational and general population of Pakistan. A positive correlation was reported between bioaccumulation of Hg in hair and its level in dust particles, $r = 0.5$ (Eqani et al. 2016a). Another study conducted by Riaz et al. (2016) reported the elevated Hg contamination in hairs of two different gold-miner groups of people, males and females. The Hg concentration in hairs of male gold miners ranged from 0.6 to 6.7 µg/kg with median value of 3.6 µg/kg, whereas in females ranged from 1.1 to 4.03 µg/kg (median 2.6 µg/kg). Albeit these reported levels of Hg in gold miner were under RfD value. The pattern of Hg contamination in Pakistan showed that the highly populated areas are potentially at a high risk of Hg contamination. Hg concentrations in term of InHg, OHg, and THg in urine and nails of gold miners of the Gilgit-Baltistan region in Pakistan were evaluated by Riaz et al. (2016). The concentration of different types of Hg were reported as follows: InHg in male 32.8–49.6 µg/L (median: 41.2 µg/L); InHg in female 31.2–41.5 µg/L (median: 36.4 µg/L); OHg in male 11.5–29 µg/L (median: 20.3 µg/L); OHg in female 11.4–21.3 µg/L (median: 16.4 µg/L), THg in male 49.5–76 µg/L, (median: 62.8 µg/L), and THg in female 41.5–60.4 µg/L (median: 51.2 µg/L). The concentration of THg mercury in urine of gold miner's male was higher compared with that in female, whereas overall the THg concentration in urine was much than RfD value of 17 µg/L set by USEPA and 50 ng/mL set by WHO (Riaz et al. 2016). THg concentration in nails was as follows: in male 0.5–6.3 µg/kg (median: 3.4 µg/kg) and in female 0.97–7.9 µg/kg (median: 4.4 µg/kg). The concentration of THg in the nails of female gold miners was higher than in male, indicating the potential risk to the future generation.

Conclusion and Recommendations

The atmospheric deposition and gold mining are the two primary sources of Hg contamination in Pakistan. The available data suggest that Hg contamination is ubiquitous in different environmental compartments in Pakistan. The atmospheric deposition was observed as the prior route of Hg contamination along the roadside, as well as in the industrial regions of Punjab province, Pakistan. Similarly, the residents around the industrial areas and along roadside exposed to the higher concentration of Hg via inhalation of dust particles. Therefore, it is necessary that the government of Pakistan and other environmental authorities must take part in elimination or reduction of Hg contamination from various environmental compartments. Furthermore, detailed studies are recommended for in-depth understanding the contamination of Hg in aquatic, terrestrial, and rice crop system in Pakistan. Studies also should be conducted on transportation, fate, environmental persistence, bioaccumulation, biomagnification, and persistence of Hg in different biological tissues and associated toxic effects. Further human biomonitoring studies are needed to investigate the harmful effects of Hg on infants via breast feedings if mothers contained high body burden of Hg via consumption of contaminated groundwater, fish, and other food stuff.

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Compliance with Ethical Standards

Conflict of interest Authors declared no conflict of interest.

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