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Review

Health risk assessment of heavy metals in water, air, soil and fish

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The study and application of health risk assessment techniques are crucial in order to understand the risk of exposure to heavy metals and other harmful pollutants. It entails evaluating the risks of exposure at various concentrations and with reference to certain standard values approved by World Health Organization (WHO) and United States Environmental Protection Agency (USEPA). Investigation of water contamination with heavy metals has become the prime focus of environmental scientists in recent years. Effluent discharges into aquatic system affect living organisms within the receiving environment. The concentrations of these metals were mostly assessed at 50th, 75th and 95th percentile and various exposure evaluated. This review covers studies in water, air, soil and fish samples. Air risks assessment was not given the needed attention and children were more susceptible to the hazard than adult, especially lead toxicity, resulting in health complications. Heavy metals bio accumulates over time, and lethal upon exposure at low concentrations. This review will assist risk managers to minimize the exposure at optimum level as well as for the government to formulate policies in safe guarding the health of population.

Key words: Health risk assessment, pollution, heavy metals, water, air, soil, fish.

INTRODUCTION

Water is an essential component of life, fresh water constitute about 3% of the total water on the earth surface, only 0.01% of this fresh water is available (Hinrichsen and Tacio, 2002), with two thirds of the earth's surface covered by water and the human body consisting of 75% of it, it is evidently clear that water is one of the prime elements responsible for life on earth.

Regrettably, even this small portion of fresh water is under pressure due to anthropogenic sources due to rapid growth in population and industrial activities (Li et al., 2009). Heavy metals are the main pollutants and elements of risk in drinking water (Enaam, 2013).

Investigation on water contamination by heavy metals has become the prime focus of environmental scientists

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in recent years (Fenglian and Qi, 2011). More attention should be given to toxic heavy elements because of bio accumulation and bio magnification potential, and their persistence in the environment. Some metals like copper (Cu), cobalt (Co) and zinc (Zn) are essential for normal body growth and functions of living organisms and are referred to as essential elements. Other elements are referred to as non-essential, high concentrations of these metals like cadmium (Cd), chromium (Cr), manganese (Mn), and lead (Pb) are considered highly toxic to human and aquatic life (Ouyang et al., 2002). A certain amount of Cr for instance is needed for normal body functions; but at the same time high concentrations may cause toxic effect such as liver, kidney problems and genotoxic carcinogen (Knight et al., 1997). Like Cr, Co is also one of the required metals needed for normal body functions as a metal component of vitamin B12 (Strachan, 2010). However, high intake of Co via consumption of contaminated food and water can cause abnormal thyroid artery, polycythemia, over-production of red blood cells (RBCs) and right coronary artery problems (Robert and Mari, 2003).

Generally, high concentrations of Mn and Cu in drinking water can cause mental diseases such as Alzheimer's and Manganism (Dieter et al., 2005). High Mn contamination in drinking water also affects the intellectual functions of 10-year-old children (Wasserman et al., 2006). Similarly, the Ni-sulfate and Ni-chloride ingestion can cause severe health problems, including fatal cardiac arrest (Knight et al., 1997). Pb is also a highly toxic and carcinogenic metal and may cause chronic health risks, including headache, irritability, abdominal pain, nerve damages, kidney damage, blood pressure, lung cancer, stomach cancer and gliomas. As the children are most susceptible to Pb toxicity, their exposure to high levels of Pb cause severe health complexities such as behavioral disturbances, memory deterioration and reduced ability to understand, while long-term Pb exposure may lead to anemia (Jarup, 2003).

Like other heavy metals, sufficient amount of Zn is also very significant for normal body functions. Its deficiency can lead to poor wound healing, reduced work capacity of respiratory muscles, immune dysfunction, anorexia, diarrhea, hair loss (Strachan, 2010). Cd exposure can cause both chronic and acute health effects in living organisms (Barbee and Prince, 1999). The chronic effects includes kidney damage, skeletal damage and itai-itai (ouch-ouch) diseases (Jarup et al., 2000). Experimental data in humans and animals showed that Cd may cause cancer in humans, diarrhea, hair loss, dermatitis (Acrodermatitis enteropathica) and depression. Cd exposure can cause both chronic and acute health effects in living organisms (Barbee and Prince, 1999). The chronic effects includes kidney damage, skeletal damage and itai-itai (ouch-ouch) diseases (Jarup et al.,

2000; Nordberg et al., 2002). Experimental data in humans and animals showed that Cd may cause cancer in humans (IARC, 1993). These heavy metals are not only found in water, but soil, food (eg fish) and air. The objectives of this review are therefore to study:

- (1) The levels of heavy metals in water, soil, fish and air.
- (2) To assess the health risks posed by the contaminated samples.
- (3) Propose general recommendations to the government and environmental management officials.

HUMAN HEALTH RISK ASSESSMENT

Human health risk assessment is considered as the characterization of the potential adverse health effects of humans as a result of exposures to environmental hazards (USEPA, 2012). This process employs the tools of science, engineering, and statistics to identify and measure a hazard, determine possible routes of exposure, and finally use that information to calculate a numerical value to represent the potential risk (Lushenko, 2010). A human health risk assessment involves four steps which are: hazard identification, dose-response assessment, exposure assessment, and risk characterization. Health risk assessment classifies elements as, carcinogenic or non-carcinogenic. The classification determines the procedure to be followed when potential risks are calculated. Non-carcinogenic chemicals are assumed to have a threshold; a dose below which no adverse health effects will be observed where an essential part of the dose-response portion of a risk assessment includes the use of a reference dose (RfD). Also, carcinogens are assumed to have no effective threshold. This assumption implies that there is a risk of cancer developing with exposures at low doses and, therefore, there is no safe threshold for exposure to carcinogenic chemicals. Carcinogens are expressed by their Cancer Potency Factor (Lushenko, 2010).

EXPOSURE ASSESSMENT

The daily environmental exposures to metals were assessed for carcinogenic and non-carcinogenic elements. There are two main exposure pathways: intake of the metals through water consumption, and by skin absorption through bathing. Calculations were done based on USEPA standards (The United States Environmental Protection Agency (USEPA), 1996). Assessment of non-carcinogenic risks can be achieved by estimating the hazard quotient (HQ). It is calculated as the quotient between the environmental exposure and the reference dose (RfD). HQ values were obtained for each element and exposure pathway. Subsequently, the

Table 1. Parameters used for estimating exposure assessment in Water (Liang et al., 2011; Wu et al., 2009; Liu et al., 2007).

Risk exposure factors	Values	Unit
Ingestion Rate (IR)	2.2	L/day
Exposure Frequency (EF)	360	Days/year
Exposure Duration (ED)	30	Years
Average Time (AT)	h/day	0.6
Average Body Weight (BW)	kg	70
Carcinogenic Potency Slope (CPS)	$\mu\text{g g}^{-1}\text{day}^{-1}$	Pb=0.009, Ni=1.7, Cd=0.6

hazard index (HI), which is defined as the total risk through health exposure pathway, was obtained by summing the HQ of each element. Finally, the total HI was calculated by summing the HI through oral and dermal routes (Hling and Hlderm, respectively) (USEPA, 1989). Values of HI under the unity are considered as safe (USEPA, 1989). The HQ is considered to be an estimate of the risk level (non-carcinogenic) due to pollutant exposure with respect to EDI (estimated daily intake) which is calculated from the following equation:

$$\text{HQ} = \text{EDI}/\text{RfD} \quad (1)$$

A summation of the hazard quotients for all chemicals to which an individual is exposed was used to calculate the hazard index (USEPA, 2011).

$$\text{HI} = \text{HQ}_A + \text{HQ}_B + \dots + \text{HQ}_n \quad (2)$$

Where HI is the hazard index; HQ_A is the target hazard quotient for A intake; HQ_B is the target hazard quotient for B intake, and HQ_n is the target hazard quotient for n intake.

Carcinogenic risk was evaluated by target cancer risk (TR). The method for estimating TR was provided in USEPA Region III Risk-Based Concentration (USEPA, 2011).

$$\text{TR} = (\text{MC} \times \text{IR} \times 10^{-3} \times \text{CPS} \times \text{EF} \times \text{ED})/(\text{BW} \times \text{AT}) \quad (3)$$

Where TR is the target cancer risk; MC is the metal concentration in the sample ($\mu\text{g g}^{-1}$); IR is the ingestion rate (g day^{-1}); CPS is the carcinogenic potency slope, (mg/kg bw day^{-1}); and ATc is the averaging time, carcinogens (days year^{-1}). The description and values of the parameters for exposure in water are shown in Table 1.

Another way to estimate Carcinogenic risks is by calculating the increase possibility of an individual to develop cancer as a result of exposure to the potential carcinogen over a lifetime. The estimated daily intake of toxin is converted by slope factor which is averaged by

direct exposure over a lifetime to the increased chances of an individual to develop cancer (USEPA, 1989).

$$\text{Risk} = \text{ADI} * \text{SF} \quad (4)$$

Risk is therefore a unit less of chances of an individual developing cancer when exposed over a lifetime and SF is the carcinogenicity slope factor (per mg/kg/day) and ADI is the acceptable daily intake. Risks values exceeding 1×10^{-4} are regarded as intolerable, risks less than 1×10^{-6} are not regarded to cause significant health effects, and risks lying between 1×10^{-4} and 1×10^{-6} are regarded generally as satisfactory range, but circumstances and condition of exposure determine the range of the value of the circumstance (Hu et al., 2012). Heavy metal evaluation Index (HEI) gives an overall quality of the water with respect to heavy metals (Edet and Offiong, 2002).

$$\text{HEI} = \sum \frac{\text{Mc}}{\text{MAC}} \quad (5)$$

Where Mc is the observed metal concentration and MAC is the maximum allowable concentration of the metal in the water guideline.

Analysis of exposures in samples

Several researches were carried out on human exposure to toxic metals and other pollutants through water, soil, fishes and other foods. Among the heavy metals analyzed in Langat River and Cempaka lake Malaysia, Cr had the HQ value greater than 1 for culture pond A and culture pond B, Langat River. While the HQ value for Pb ranged from 0.017 to 0.073 which were much lower than those measured in Tri states mining districts where the HQ ranged from 0.1 to 4.6 (Schmitt et al., 2006). In the study at Langat River and Cempaka Lake, Malaysia, although the observed values of HQ for Pb, Cd, Ni, Cu and Zn were lower than the safe standard of 1, but ΣHQ of these metals (HI) were higher than 1. The calculated

HI ranged from 0.24 to 1.88 which indicates that 71% of stations are in the risk level while Bandar and Jugra were the only stations analyzed with HI values of less than 1.

In Pakistan, the calculated chronic daily intake (CDI) values for consumption of drinking water suggest that in Jijal-Dubair area, people have consumed surface water contaminated with heavy metals, the maximum CDI values were 0.10, 0.02, 0.62, 3.76, 0.21, 0.23, 0.12, and 1.09 $\mu\text{g}/\text{kg}/\text{day}$ for Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn, respectively. But in Jijal-Dubair area, the consumed ground water had maximum CDI values of 0.03, 0.09, 0.98, 3.64, 0.25, 0.40, 0.15, and 78.93 $\mu\text{g}/\text{kg}/\text{day}$ for Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn, respectively. CDI indices for heavy metal in the study area were found in the order of $\text{Zn} > \text{Cu} > \text{Mn} > \text{Pb} > \text{Cr} > \text{Ni} > \text{Cd} > \text{Co}$ (Said et al., 2011). In drinking water, the high CDI values of Zn, Mn and Pb may be attributed to the Pb–Zn sulfide mineralization, while that of Cr and Ni may have resulted from the mafic and ultramafic bed rocks hosting chromite deposits (Miller et al., 1991; Ashraf and Hussian, 1982).

In a study carried out in northwestern Bangladesh, the proposed HEI criteria for the surface water samples were as follows; low ($\text{HEI} < 150$), medium ($\text{HEI} = 150$ to 300) and high ($\text{HEI} > 300$). For the groundwater samples, the criteria was: low ($\text{HEI} < 40$), medium ($\text{HEI} = 40$ to 80) and high ($\text{HEI} > 80$). Using this scheme, 55, 36 and 9% of surface water samples showed low, medium and high contamination, respectively with respect to heavy metals, whereas, 50, 40 and 10% of the groundwater samples showed as less, moderately and highly contaminated (Mohammad et al., 2010). MPI is computed to analyze the status of the heavy metal contamination in the environment. MPI is calculated according to Usero et al. (1997) using the given equation:

$$\text{MPI} = (\text{C}_1 \times \text{C}_2 \times \dots \times \text{C}_n)^{1/n} \quad (6)$$

Where C_n is the concentration of the metal n in the sample

Concentrations of metals in water were used to assess human exposure through oral intake and bath. The total HI resulting from exposure to metals through water ranged from 0.64 to 0.66 for adults. In children, HI levels increased between 1.87 and 1.85 up-and downstream river areas, respectively (Renato et al., 2014). The non-cancer risk associated with the single oral exposure to Ti already exceeded the safety level at the upstream area ($\text{HQ} = 1.38$). Arsenic (As) was another element of concern. Although Renato et al. (2014) revealed that As levels were below the safety level, relatively high values of HQ associated with As exposure were also observed. The presence of As, a natural occurring element, may also be attributed to anthropogenic activities, such as the use of herbicides (Christ et al., 2012). The health risk for heavy metals in seafood is usually quantified by the

target hazard quotient (THQ) (Storelli, 2008). The THQ as shown in equation 1 is defined as the concentration of heavy metals divided by a reference dose (RfD). If the THQ is less than 1, the seafood has no health risk. Conversely, the health risk should be considered. Based on the THQ equation and the RfDs of heavy metals published by the United States Environmental Protection Agency (USEPA), the safety limits of Cr, Cu, Zn, Cd, Hg, As and Ni in seafood were 2.9, 39, 292, 1.0, 0.1, 2.9 and 19 mg/kg ww, respectively (USEPA, 2012). Body weight (55.9 kg) and daily consumption amounts of seafood (57.5 g/day) were obtained from the survey conducted by Wang et al. (2005). However, the RfD of Pb is not considered by the USEPA. Thus the consumptive standard of Pb in aquatic organisms (0.5 mg/kg ww) published by the General Administration of Quality Supervision, Inspection and Quarantine of China (AQSIQ) is used in this review (AQSIQ, 2001). Based on the safety limits of heavy metals, consumption levels of Pb and As in most mollusks from Hong Kong exceeded the criteria (Fang et al., 2008). Concentrations of heavy metals in fish from northwestern and southern Hong Kong all met the consumption standards (Cornish et al., 2007). Heavy metal levels in most seafood from Lingdingyang were higher than the safety limit (Shuai-Long Wang, 2013).

A study done by Mishra et al. (2007) in the Trans-Thane Creek area of Mumbai, measured the trace element in different types of marine organisms and reported the HQ values of 0.01 (50th percentile) and 0.005 (95th percentile) in case of the ingestion of Cd. The same study also revealed lower HQ values for Cr, Ni, Zn and Cu and suggested that consumption of fish samples were within the safe limit (Mishra et al., 2007). Similar to the findings, Tu et al. (2008) worked on the concentration of Cr, Cu, Zn and Cd, and measured HQ values of less than 1 which indicated that the local residents were not exposed to potential risk via consumption of shrimp. On the other hand, Schmitt et al. (2006) reported a higher range of HQ values for Cd (0.1 to 0.5) and Zn (0.1 to 12.6) in carp fishes. Samples of black-chin Tilapia, collected from Sukumo lagoon of Ghana, were analyzed for the concentration of heavy metals and the calculated values of HI indicated that the Tilapia did not pose any health risk to humans (Laar et al., 2011).

Heavy metal pollution of soil is regarded as one of the severe environmental challenges in many countries of the world (Facchinelli et al., 2001). Concentrations of As, Cd, Ni and Pb in the soil are among the heavy metals investigated. People experienced higher exposure to As, Cd, Ni and Pb due to their high concentrations in the soil under investigation or low RfD values, whereas they had little exposure to other four heavy metals (Cr, Cu, Zn and Hg). For instance, in the surrounding area of the Chenzhou lead–zinc mine in China, the HQ value of As, Cd, Ni and Pb accounted for 25.8, 13.8, 3.5, and 54.0%

of the entire HI value, respectively. By contrast, the total percentage of the other four heavy metals for the entire HI value was 0.2%.

Generally, the total hazard quotients of Pb, Ni, Cd and As accounted for 98.6% of the full HI value in the surrounding area of the Dabaoshan multi-metal mine (Zhiyuan Li et al., 2014). An emphasis was particularly given to Arsenic due to its reported cases of poisoning and cancer related issues in the region. The carcinogenic risk values for As at some mining areas even exceed 1×10^{-4} . As a whole, these carcinogenic risk levels are unacceptable or close to unacceptable limit. For every mining area, the carcinogenic risks of As for different populations vary greatly, generally in the order of adult females > adult males > children. The reason that the carcinogenic risk for children is less than that for adults lies in the shorter duration of exposure for children. Average As carcinogenic risk values (standard deviation) for antimony, coal, copper, gold, and lead-zinc mining areas are; 5.8×10^{-4} (7.6×10^{-4}), 1.3×10^{-5} (6.9×10^{-6}), 4.7×10^{-5} (7.1×10^{-5}), 1.1×10^{-5} (7.7×10^{-6}), and 1.7×10^{-4} (2.0×10^{-4}), respectively (Zhiyuan Li et al., 2014).

Exposure assessment in air

Not much emphasis was given to assess the risk of exposure of humans to heavy metals by air, as much literature gave emphasis to water and soil by ingestion and dermal (skin). Humans can become exposed to heavy metals in dust through several routes which include ingestion, inhalation, and dermal absorption. In dusty environments, it has been estimated that adults could ingest up to 100 mg dust/day (Hawley, 1985). Children are usually exposed to greater amounts of dust than adults (Centers for Disease Control and Prevention (CDCP), 2005). Exposure to high levels of heavy metals can result in acute and chronic toxicity, such as damage to central and peripheral nervous systems, blood composition, lungs, kidneys, liver, and even death. Lead levels in dust have been significantly associated with Pb levels in children's blood (Lanphear and Roghmann, 1997), and a blood lead level (BLL) greater than an intervention level of 10 µg Pb/dl has been associated with a decrease in IQ (CDCP, 2005). According to Anna et al. (2008), the potential health risk to children at all locations was eight times greater. This was partly attributed to the higher ingestion rate used (200 mg/kg/day) in estimating the risk and the smaller body size. Overall, the accumulative risks due to the metals are a major concern (HI > 1) at all locations except for the locations studied.

GAP IN GENERAL HEALTH RISKS ASSESSMENT

In developed and developing countries of the world, air

pollution is increasing at alarming rate prompting the industrialized nations to impose a special tax on industries whose emission is above the approved values by the government. Significant amount of toxic heavy metals had been found in air and dust, especially in industrial areas, and incineration of electronic wastes containing chips, capacitors, and diodes. But much was only given to oral and dermal routes of exposure. Also, target hazard quotient (THQ) is widely employed to evaluate the health risk, it has several apparent weaknesses among which are: (1) Only suspected targets are considered and determined while the other potential hazardous pollutants are ignored and not analyzed; (2) The relationship or mutual effects of different pollutants are ignored by the THQ, such as, lower pH values which results in precipitation of toxic metals in water.

Also, Selenium (Se) can diminish or lessen the toxicity of As and Hg, but few investigations have taken Se into consideration when the health risk of heavy metals is evaluated (Peterson et al., 2009; Ouédraogo and Amyot, 2013), there is a significant correlation between As and Au in soil and water samples obtained from mining areas; (3) The toxicities of heavy metals mainly depend on their bioavailability. For example, As is believed to be one of the most hazardous substances. However, As in fish is mainly present as non-toxic arsenobetaine (Zhang et al., 2012; Zhang and Wang, 2012).

Levels of methylmercury (MeHg) in aquatic organisms were generally quite low, but it is highly toxic to wildlife (Liu et al., 2012; Sherman et al., 2013). Therefore further risk assessment techniques for heavy metals should be improved, and other physical parameters be incorporated in the analysis.

CONCLUSION

This review gives an overall view on pollution levels and health risks posed by heavy metals in water, soil and fish and provide reasonable evidence on the utmost need to fully assess the risks of heavy metals and other pollutants to safeguard the health of the community. The knowledge of risks assessment shall be a priority considering continuous increase in heavy metal and general environmental pollution globally in water, air and soil. The continuous and/or periodical acquisition of data on the quality of the water bodies is essential for stakeholders in various countries; also data from soil and food substances needs to be studied. Air risks assessment, especially in industrial and populated regions needs to be carried out periodically. This will enhance proper monitoring and ensure safety of the citizens especially children who are more vulnerable to toxicity of heavy metals.

Conflict of Interests

The authors have not declared any conflict of interests.

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