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Review

Deleterious effects of heavy metal pollution in the environment

Amartya Khan Chand¹ and Chandra Tilak Patel²*

¹Section of Chemistry, Women's College, A.M.U, Aligarh (U.P), India.

²Aquatic Toxicology Research Laboratory, Department of Zoology, A.M.U, Aligarh (U.P), India.

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Heavy metal pollution in environment is of great concern because of their accumulative and persistent nature. Among different heavy metals, Hg does not play any essential role in the body of living beings but after methylation, it acquires very dangerous form which is known as MeHg. Hg in air is within the limits and not harmful but in water and fishes the concentration of THg and MeHg content exceeds or is close to permissible limits. Water and food is the main source of Hg accumulation in human beings. Both Hg and MeHg have deleterious effects on all living beings. However, at some places around the world, efforts have been made to reduce the Hg concentrations which resulted in substantial improvements. Therefore the purpose of this review is to highlight different concentrations of Hg in the environment.

Key words: Mercury, MeHg, accumulation, fishes, human beings.

INTRODUCTION

A clean and pollution free environment is one of the prerequisite for a healthy life. However, our environment contains infinite number of pollutants in the form of heavy metals, smoke, dust particles, pesticides, etc. Once they are released into the environment, they circulate between the air, water and soil and change the physical, chemical and biological properties of the environment which endanger the life of masses. It is said that environmental deterioration begins with the modernization. These days the heavy metals are of particular concern because of their persistent, non-biodegradable and bioaccumulative nature. Heavy metals reach the environment by either natural or anthropogenic activities like volcanic eruptions,

*Corresponding author. E-mail: khan.chand@yahoo.com

ABBREVIATIONS: EFSA, European Food Safety Authority; JECFA, Joint Expert Committee on Food Additives; NRC, National Research Council; NTP, National Toxicology Program; SSSGMD, Social Scientific Study Group on Minamata Disease.

weathering of rocks, soil erosion, emissions from automobiles, burning of coal, mining, industrial activities, trash incineration, etc. Heavy metals are those elements which have their atomic weight between 63.546 and 200.590 (Kennish, 1992) and specific gravity greater than 4.0 (Connel et al., 1984). However, being a heavy metal has little to do with density but concerns mainly with the chemical properties (Javed, 2013). No doubt metals are required in trace amounts to the living beings which include Co, Ni, Fe, Cu, Mn, Mo, V, Sr and Zn, while Hg, Cd and Pb are of no significance to humans and their excess results in serious ecological problem. Numerous studies have already been conducted on harmful effects of essential metals such as: Cu, Ni, Fe, Co, Mn, Cr and Zn (Javed and Usmani, 2011, 2012a, b, 2013a, b, c; Taweel et al., 2012; Emere and Dibal, 2013).

It is only recently that man became aware of how heavy metals in the aquatic environment could create deleterious effects on the aquatic flora and fauna, and hence influence human health. The potential hazards of heavy metal pollution were triggered off by the Minamata disease caused by the consumption of Hg contaminated shell fish and fin fish from Minamata Bay (Nita, 1972). This suffering is a result of the wrongful and negligent acts of the Chisso Corporation who dumped mercury into the sea water and poisoned the people of Japan. The Chisso Corporation was once a fertilizer and carbicle company, and gradually advanced to a petrochemical and plastic-maker company. It dumped an estimated 27 tons of mercury compounds into Minamata Bay. Since then, there has been a rise in concern for the evaluation of heavy metal concentrations in fishery products. Therefore monitoring the concentration of Hg is of utmost importance in order to avoid the recurrence of such major disaster. Early warnings about MeHg toxicity given in Table 1.

Mercury (Hg) is the non-essential heavy metal which comes neither in the list of macroelements or microelements nor have beneficial properties that can support life. Hg is a naturally occurring element that exists in several forms. These different forms can be categorized as metallic Hg (elemental Hg), inorganic Hg and organic Hg. Metallic Hg appears as silver white metal and exists in liquid state at room temperature, but some amount evaporates at room temperature and changes into colorless and odorless vapors. Inorganic Hg compounds are formed by combining salts of the Hg with different elements such as chlorine, sulfur, oxygen. Most of the inorganic Hg compounds exist in the form of crystals or white powders except mercuric sulfide. Organic Hg is formed when Hg combines with carbon. One of the most important properties of Hg is biotransformation in which inorganic Hg transforms into organic Hg particularly methyl mercury (MeHg) by microorganisms or other naturally occurring processes. Over the past 100 years, there has been a 30-fold increase in Hg deposition due to industrialization, 70% of which is from anthropogenic sources.

Hg enters into the environment as a result of both naturally occurring as well as anthropogenic activities. Once minerals are exposed to the wind and water, they broke down into rocks and soil and release Hg into the environment. Volcanic activities also contribute Hg to the environment. Human activities include combustion of fossil fuels, mining, smelting and this comprise about 80% of released Hg. Metallic and inorganic Hg enters the air from the emission of coal fired power plants, burning municipal and medical waste. From air, both of these forms transported to water via rain or snow contaminating the fresh/marine water resources. MeHg is released into the environment by microorganisms. This form is highly dangerous because it gets accumulated into the living tissues of aquatic organisms and is not easily eliminated.

Once MeHg enters the aquatic ecosystem, it accumulates in tissues of the fish and when this fish is eaten by other predatory fishes, it accumulates and further magnifies into their body tissues. Consequently the highest amount of Hg will present in largest and oldest fish for instance, salt water fish (especially Sharks and Sword fish).

MERCURY IN AIR

As a natural element, Hg is ubiquitous in the environment and approximately 10,000 tons originates from degassing of earth's crust, which further added by approximately 20,000 tons/year due to anthropogenic activities (Hansen and Dasher, 1997). Hg emissions from the coal smoke are the main source of anthropogenic discharge and Hg pollution in atmosphere. It is estimated that the mercury emissions will increase at a rate of 5% a year (Zhang et al., 2002). Besides this municipal waste combustion (5.6%), mercury-cell chlor-alkali plants and hazardouswaste incinerators (4% each), stationary internal combustion engines (ICEs) (3.5%), industrial, commercial and institutional (ICI) boilers (3.3%) and lime manufacturing (3.0%) and medical waste incineration (1%) (Murray and Holmes, 2004) further add to the environmental Hg concentration. Gold mining activities also used Hg to remove it from ore. Emissions from all these sources go up in the air which finally comes to the water via rain or atmospheric deposition.

MERCURY IN WATER AND FISHES

Freshwater is limited on earth and is under constant threat of deterioration. Kannan et al. (1998) has reported that concentration of total mercury (THg) and MeHg in filtered water samples collected from the canals and creeks of the Florida Bay are 3-7.4 ng/L (mean 4.6 ng/L) and <0.002-2.3 ng/L (mean 0.474 ng/L), respectively. THg levels varied little in all these streams, MeHg levels varied considerably among locations. Greater than 1 ng/L, MeHg concentration was found in Shell Creek, Trout Creek, and water from the culvert that controlled the canal C111 flow. The concentration of both THg and MeHg tended to be higher in near-surface than in nearbottom waters of canal C111 (Kannan et al., 1998). THg concentrations in canals and creeks were found within the range of 2 to 15 ng/L for coastal estuaries waters (Schroeder, 1989; Stein et al., 1996). However, according to the report of Fitzgerald and Clarkson (1991) and Mason et al. (1995), concentrations of Hg were higher in Florida waters than in open ocean waters of the North Atlantic and Pacific Oceans but were comparable to those reported for the Baltic and North seas (Schmidt, 1992; Coquery and Cossa, 1995). The U.S. EPA (1985) Hg water quality criterion for protection of freshwater is 12 ng Hg/L, and for sea water it is 100 ng/L. The water quality criterion for Hg proposed for Minnesota's freshwater is 7 ng/L while a value of 2 ng/L has been established for Wisconsin waters (Glass et al., 1990). The Hg concentrations reported in Florida waters were below the U.S. EPA tolerance limit, but close to or higher than those established in Minnesota and Wisconsin. MeHg accounted for <0.03-52% (mean 10.4%) of THg in estuarine waters (Kannan et al., 1998). MeHg in coastal water of Qatar was 5% of THg (Al-Madfa et al., 1994).

The proportion of MeHg was higher, with an average of 25% and ranged up to 80% in freshwater areas (Gill and Bruland, 1990). MeHg accounted for 6-13% of the total dissolved Hg in inland surface waters from Sweden (Lee and Hultberg, 1990). In anoxic lake water, the percentage of MeHg was as high as 58% of the THg (Gilmour and Henry, 1991). The proportion of MeHg in water depends on many variables such as acidity, dissolved organic carbon, sulfate, and hydrological and geochemical factors (Gilmour and Henry, 1991).

Paul (1987) reported a Hg level of 0.135 to 0.200 µg/g in muscle of common edible fish species from the Stanley reservoir at Tamil Nadu, India. Avvadurai and Krishnasamy (1989) also examined the Hg level of 0.05 to 0.27 µg/g in fish from swamp contaminated with sewage and hospital waste water at Madras, India. Kannan et al. (1998) studied the THg and MeHg concentrations in muscle of fishes such as hardhead catfish, gafftopsail catfish, sand seatrout, sand seaperch, pin fish, white grunt, lane snapper, spot, pig fish, brown shrimp collected from coastal waters of Southern Florida. The range of accumulation of THg were between 0.11-10.1 µg/g d.w. (mean, 1.14 µg/g d.w.) and MeHg ranged from 0.06-4.5 µg/g d.w. (mean, 1.05 µg/g). It had also been observed that among various fish species, hardhead catfish, gafftopsail catfish and sand seatrout contained the highest concentrations of Hg. All these contain about 83% of MeHg of the THg. When cat fish was collected from Gordon River, it was found that 20% of its Hg was present in the form of MeHg. But THg concentration was highest in cat fish (10.1 μ g/g) from the same river followed by those from Hillsborough channels $(4.98 \mu g/g)$, Tampa Bay $(2.09 \mu g/g)$ and Florida Bay (2.64 $\mu g/g$). The accumulated levels of Hg in fishes of Florida were much beyond the permissible limits, therefore the state of Florida issued an advisory in (1989) that consumption of top - level predatory fish, for instance largemouth bass (Micropterus salmoides), Bowfin (Amia calva), and Gar (Lepisosteus lepisosteus) were prohibited throughout Florida (Cabbage, 1989; Hand and Friedemann, 1990; Royals and Lange, 1990).

Similarly, in other field studies, accumulation of large amount of different heavy metals in the tissues of fishes in different amount along with Hg were also reported which follows the order Fe > Zn > Pb > Cu > Cd > Hg in(Barbara and Malgorzata, 2006). They also reported that the accumulation of Hg in fish was very low, below 1 µg/g d.w., and its accumulation depends on pollution and may vary in different species of fishes inhabiting the same water body. Agarwal et al. (2007) examined the accumulation pattern of mercury in muscle of different species of fishes from the river Gomti, Lucknow, India in the order: Mastacembelus armatus > Clarias batrachus > Mystus cavasius > Notopterus notopterus > Rita rita ~ Heteropneustes fossilis > Channa punctatus > Labeo rohita. In M. armatus, Hg accumulated to 0.2774 µg/g. But the levels that were reported in all the fishes were

below the recommended guidelines set for Hg (0.30 μ g/g) by U.S Environmental Protection Agency (U.S EPA, 2001); 0.50 µg/g (wet wt. of fish) for human consumption as recommended by World Health Organization (WHO, 1990), United Kingdom and United States as well as the maximum residue limit as per the Prevention of Food Adulteration (PFA) Act 1954, India amended in 2002. The conclusion which was derived from these results was that the Gomti river water was not contaminated with Hg to cause any health risk. Similar Hg content (0.041-0.117 µg/g and 0.054-0.183 µg/g respectively) was also reported by Mirlean et al. (2005) and Viana et al. (2005). Burger et al. (2004) examined levels of Hg in muscle of Florida gar (Lepisosteus platyrhincus) from Lake Okeechobee in south-central Florida. The mercury levels were lowered than from many waters in the U.S. and also the standard guidelines. Has-Schon et al. (2008) tested the Hg concentration in fishes brown trout (Salmo trutta), souffie (Leuciscus turskyi), nase (Chondrostoma phoxinus), Dalmatian barbelgudgeon (Aulopyge hügeli) inhabiting Busko Blato reservoir and reported below recommended levels of Hg in all tissues (muscle, gills, liver, kidney and gonads) of fishes with the exception of muscle of brown trout. Jabeen and Chaudhry (2010) also reported high levels of Hg in muscle of Cyprinus carpio inhabiting waters of Indus River. Amundsen et al. (2011) reported high levels of Hg in the muscle $(0.77 \ \mu gg^{-1})$ of white fish (Coregonus lavaretus) inhabiting in waters of subarctic watercourse. Bhupander et al. (2011) examined the quality of fishes from East Kolkata Wetlands and reported Hg accumulation in different species such as Puntius ticto (0.7 µg/g d.w.), Oreochromis nilotica (0.5 µg/g d.w.), Labeo rohita (0.4 µg/g d.w.), Channa marulius (0.4 µg/g d.w.), Catla catla (0.3 µg/g d.w.), Hypophthalmichthys molitrix (0.3 µg/g d.w.) and Oreochromis mossambica (0.1 µg/g d.w.). Accumulation in Puntius ticto was beyond the permissible limits, however accumulation in O. nilotica, L. rohita and C. marulius was reported to contain Hg close to permissible limits. The concentration of Hg in fish tissues is directly proportional to age and size of fish, that is, it increases with increase in age and size which in turn is related to the affinity of this metal in muscle tissue (Green and Knutzen, 2003; Voigt, 2004). Shao et al. (2011) studied 18 freshwater fish ponds around the Pearl River Delta (PRD) and reported the concentrations of THg and MeHg in fish muscles which ranged from 7.43-76.7 to 5.93-76.1

In fish muscles which ranged from 7.43-76.7 to 5.93-76.1 ng/g wet wt, respectively, with significant linear relationships (r=0.97, p<0.01, n=122) observed between THg and MeHg levels in fish. Risk assessment indicated that the consumption of largemouth bass and mandarin fish would result in higher estimated daily intakes (EDIs) of MeHg than reference dose (RfD) for both adults and children. Recent studies conducted on Minamata Bay regarding the concentration of Hg in water and fish showed that Japanese stingfish (*Sebastiscus marmoratus*) and Bambooleaf wrasse (*Pseudolabrus* japonicas) are monitored annually for Hg pollution in Minamata Bay, Japan. The average THg concentration in the muscle of these two species was 0.36 and 0.20 mgkg⁻¹ wet wt, respectively, between 2008 and 2010. This was higher than levels elsewhere in Japan (0.125 and 0.038 mgkg⁻¹ wet wt, respectively). The FDA (2001) and EPA (2004) suggested that a proportion of Hg accumulated in fish was derived from seawater. Matsuyama et al. (2013) reared young red sea bream (Pagrus major) over a 2-year period in Minamata Bay and Nagashima (control) to evaluate the uptake of mercury from seawater. There was no difference in Ha accumulation in the muscle of red sea bream between Minamata Bay and Nagashima was reported. Thus, the results of his study suggested that the majority of Hg accumulated in fish muscle was not from seawater.

ADVERSE EFFECTS OF HG ON HUMAN BEINGS

For humans, the route of exposure of Hg can be air, water and food, out of which the oral exposure that is through water and food is the major source. According to Agency of Toxic Substances and Disease Registry (ATSDR, 1999), very low levels between 10 and 20 ng of Hg per cubic meter of air have been reported in urban outdoor air and it was considered safe to breathe while in non-urban areas it was much lower, that is, 6 ng/m³ or less. According to the report of Food and Drug Administration (FDA), most people are exposed, on an average to about 50 ng of Hg per kilogram of body wt. per day (50 ng/kg/day) in the food they eat. This is equivalent to 3.5 μ g of Hg per day for an adult of average wt. This level is not harmful to humans (ATSDR, 1999).

According to Green Peace researchers, what accumulates in fish can accumulate in human. From the above discussion, it has been noticed that Hg concentration in water and sea food particularly in fishes is the major source of Hg accumulations for humans. Fishes serve as a delicacy and are widely consumed for its polyunsaturated fatty acids (PUFA) content which is essential for good health. But as per the reports of various workers, consuming fishes from the contaminated water bodies becomes toxic because they are reported to contain heavy load of Hg and causes number of adverse effects to the living beings (human). Hg is considered by WHO as one of the top ten chemicals or groups of chemicals of major public health concern.

A recent study by the National Research Council (NRC) in its 2000 report on the toxicological effects of MeHg, concluded that the population of the offsprings of those women who consume large amount of fish and sea food during pregnancy are at highest risk. It can adversely affect a baby's growing brain and nervous system. The primary health effect of MeHg is that it impairs neurological development. Therefore, cognitive thinking, memory, attention, language, and fine motor and visual spatial skills may be affected in children who were exposed to MeHg as fetuses (WHO, 2012). More than 60,000 children born each year are at risk (USGS, 2000). This study is same as that of the National Academy of Sciences showed that people who are regularly exposed (chronic exposure) to high levels of mercury (such as populations that rely on subsistence fishing), that is, between 1.5/1000 and 17/1000 children, showed cognitive impairment (mild mental retardation) caused by the consumption of fish containing Hg. These included populations in Brazil, Canada, China, Columbia and Greenland (WHO, 2012).

Renal effects have been reported, ranging from increased protein in the urine to kidney failure (WHO, 2012). Li et al. (2013) investigated blood mercury (B-Hg) concentration of residents living in the vicinity of Chatian mercury mine (CMM) in southwestern China to assess the possible effects on renal function. It evaluates the effects of gender and age (children, <18 years; adults, 18-60 years; elderly, >60 years) on the B-Hg and serum creatinine (SCR) and serum urea nitrogen (SUN) levels. In the CMM, elevated levels were found for B-Hg, SCR, and SUN with mean values at 6.09 µg/L, 74.21 µmol/L, and 13.26 mmol/L, which were significantly higher than those in the control area, thus revealing that Hg exposure can cause human renal function impairment. Similar studies were conducted on rats and mouse by inducing Hg salts and it is concluded that it impairs renal functions due to biochemical alterations, since the increase in SCR and SUN are two of the most sensitive markers of renal disease (Augusti et al., 2008; Rumbeiha et al., 2000)

Neurological and behavioral disorders may also be observed after ingestion of different Hg compounds. Symptoms include tremors, insomnia, memory loss, neuromuscular effects, headaches and cognitive and motor dysfunction. Mild, subclinical signs of central nervous system toxicity can be seen in workers exposed to an elemental Hg level in the air of 20 μ g/m³ or more for several years. Besides this, elemental Hg and MeHg are toxic to the central and peripheral nervous systems. The inorganic salts of Hg are corrosive to the skin, eyes and gastrointestinal tract, and may induce kidney toxicity if ingested. Central Nervous System (CNS) toxicity includes erethism with symptoms of shyness, emotional lability, nervousness, insomnia, memory impairment, and inability to concentrate. Other CNS symptoms may include encephalopathy, peripheral neuropathy, Parkinsonian symptoms, tremor, ataxia, impaired hearing, tunnel vision, dysarthria, headache, fatigue, impaired sexual function, and depression. It also had its effects on gastrointestinal system which include nausea, vomiting, diarrhea, and colitis. Dermal toxicity includes allergic dermatitis, chelitis, gingivitis, stomatitis, and excessive salivation (Mark Hyman, 2004). But lately, keeping in mind its severe health concerns and rising concentrations, efforts are being made worldwide to reduce the emissions and releases to the environment. This includes reduction from point sources and overall reductions

 Table 1. Important early warnings about MeHg toxicity and recognition of MeHg toxicity.

Year(s)	Event	Reference(s)
1865	First published record of fatal occupational MeHg poisoning	Edwards (1865)
1887	First experimental studies on MeHg toxicity	Нерр (1887)
1940-1954	Poisoning cases in workers at MeHg fungicide production plants	Franke and Lundgren (1956); Hunter and Russell (1954)
1952	First report on developmental MeHg neurotoxicity in two infants	Engleson and Herner (1952)
1956	Discovery of a seafood-related disease of unknown origin in Minamata, Japan	SSSGMD (1999)
1968	Official acknowledgment of MeHg as cause of Minamata disease	SSSGMD (1999)
1986	First epidemiology report on adverse effects in children related to maternal fish intake during pregnancy in New Zealand	Kjellstrom et al. (1986)
1997	Confirmation from prospective study in the Faroe Islands on adverse effects in children from MeHg in maternal seafood intake during pregnancy	Grandjean et al. (1997)
2000	NRC supports exposure limit of 0.1 µg/kg per day	NRC (2000)
2003	Updated JECFA exposure limit of 1.6 µg/kg per week	JEFC (2003)
2004	European Union expert committee recommends that exposures be minimized	EFSA (2004) European
2005	European Union decides on a ban on mercury exports	Union (2007) UNEP
2009	International agreement on controlling mercury pollution	(2009)

Source: Taken from Philippe et al. (2010).

seen with the decreased use of mercury-containing products and processes utilizing mercury and proper waste management. These measures will ultimately reduce the Hg levels in fish as environmental levels go down. In some species of fish, this reduction may be seen quite quickly, while in other species, levels will decrease more slowly as a factor of their size, age and diet (UNEP, 2013).

Global emissions of mercury to the air in 2010 from human activities were estimated at 1,960 tonnes. Although it is difficult to compare emissions' estimates for individual years, total anthropogenic emissions of mercury to the atmosphere appear to have been relatively stable from 1990 to 2010 (UNEP, 2013). However, there has been a large shift in regional patterns. Economic growth has driven an increase in anthropogenic emissions in Southern and Eastern Asia, which now account for about half of global emissions. Emissions in Sub-Saharan Africa and in South America are slowly rising (together accounting for about 30% of global emissions), while emissions are declining in North America and Europe (about 8% of global emissions altogether) (UNEP, 2013). The largest anthropogenic sources are associated with artisanal and small-scale gold mining (ASGM) and coal burning.

In 2008, United States of America (USA) introduced its Hg Export Ban Act, which bans the export of Hg from the USA from 1 January 2013. It also includes provisions on long term Hg management and storage. Because the USA is one of the world's top Hg exporters, implementtation of the act will remove a significant amount from the global market (US EPA, 2012). The European Union (EU) banned mercury exports in 2011. The EU stopped all forms of Hg mining in 2001, as recently as 2008 it was the world's biggest exporter, responsible for up to a

quarter of the global supply. Only a few countries such as Canada and the USA have taken steps to set national standards specifically for Hg emissions from coal-fired plants. The USA has recently finalized the Hg and Air Toxics Standard which aims to reduce mercury emissions by 20 tonnes by 2016, a total of 70% reduction in emissions from the power sector (Sloss, 2012). In the EU, further Hg reduction will be achieved through the new Industrial Emissions Directive adopted in 2010; however, specific reduction or control requirements of mercury may still be required. In another study, it has been reported that efforts are being made to control the Hg pollution from the Great Lakes region, which have resulted in substantial and measurable improvements and that additional emissions controls will have multiple benefits for fish, wildlife, and people who consume fish from the Great Lakes region.

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