

## Review

# Environmental impacts of mercury and its detoxification from aqueous solutions

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Accepted 29 November, 2006

There has been an increased concern over the level of heavy metals in the environment because of the serious environmental hazards these metals pose. Mercury is one of these heavy metals present in the environment. Mercury undergoes complex chemical and physical transformation once released to the air, land or rivers. The main sources of mercury are the natural and anthropogenic sources. Mercury may be emitted to the atmosphere as a gas or particulate matter, which may return to the earth's surface either dry by gravitational settling or wet by precipitation. While it circulates and changes its form, it becomes persistent. It tends to accumulate in sediments of water bodies. Mercury constitutes a considerable hazard to vertebrates including man and it is bioaccumulating in biota. The detoxification of mercury from aqueous solutions has been achieved by conventional methods such as precipitation, coagulation, reverse osmosis, ion exchange and adsorption using activated carbon. Recent advances in mercury remediation include the use of agricultural by-products and microorganisms as adsorbent. The application of biosorbents has proved to be a very good process for mercury remediation from aqueous solutions. This paper reviews the toxicity of mercury and its remediation processes.

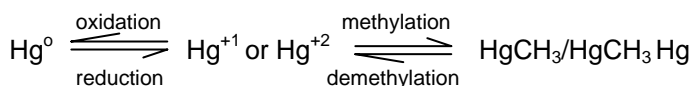
**Key words:** Mercury, detoxification, pollution, environment, heavy metals.

## INTRODUCTION

Mercury, amongst other heavy metals has attracted global concern due to its extensive use, toxicity, widespread distribution and the biomagnifications. A chemical whose concentration increases along a food chain is said to be biomagnified. The bioconcentrate of mercury in aquatic organisms such as oysters and muscles has been reported to be much greater than those contained in the environment in which they live (Baird, 1995). Mercury intoxication originated from the ingestion of fish and shell fish resulted in the death of several people (Kurland et al., 1960). The occurrence of mercury has been studied in a number of aquatic species in various parts of the world (Hattula et al., 1978; Phillips et al., 1982, Kehring and Malm, 1999).

Mercury circulates through the environment in different chemical forms and different physical states. In inorganic form, it exists in three oxidation states as elemental mercury ( $\text{Hg}^0$ ), monovalent mercury ( $\text{Hg}^{+1}$ ) or divalent mercury ( $\text{Hg}^{+2}$ ). Elemental Hg in liquid form is the type of mercury found in many consumers products e.g. household fever thermometer with the silver bulb. When open to the atmosphere elemental Hg vaporizes from its liquid state into the atmosphere. Mercury may also exist in organic forms (ie in combination with carbon containing compounds such as methyl mercury ( $\text{CH}_3 \text{Hg}^+$ ). Methyl mercury is the chemical formed when bacteria in soil or water convert deposited mercury through ingestion and absorption (Nescaum et al., 1998; Okoronkwo et al., 2006). The common mercury transformation is:

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The atmosphere is the environmental compartment with the largest influence on mercury transportation and fluxes (Peterson et al., 1995; Iverfeldt et al., 1996). The atmospheric mercury is primarily elemental mercury (between 90 and 95%), divalent mercury (3 - 4%) and methylated mercury (2 - 3%). The residence time of elemental mercury in the atmosphere is estimated to be between 1.1 and 1.4 years (Slemr, 1996) which allows not only long-range transportation. Most of the mercury deposited into the ocean from the atmosphere is in the form of  $\text{Hg}^{2+}$ . This reactive form is reduced mainly to elemental mercury by biological processes. The elemental mercury is then re-emitted back to the atmosphere to be ultimately deposited on the surface of soils. The recent studies on mercury contamination have shown that the anthropogenic sources of release have increase relative to the natural sources since the onset of industrial period in most region of the world. Mercury is injected into the environment through mercury compounds which are component of fungicides, wastes from dental clinics and hospitals and chemical industries (Ackefors et al., 1970; Holden, 1973).

Mercury contamination of aquatic foods has been observed to constitute a considerable hazard to many fish eating vertebrates including man (Ackefors et al., 1970). Mercury occurs in the biota as biaccumulating methyl mercury which has neuropathological and physiological properties (Peakall and Loveth, 1972; Holden, 1973). This paper therefore reviews the various sources of mercury, its environmental impacts and removal from the environment. Also, its detoxification from aqueous solution using agricultural wastes and other biosorbents are reviewed.

## SOURCES OF MERCURY TO THE ENVIRONMENT

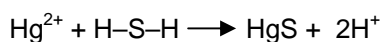
Mercury enters the environment as a result of natural and human activity (anthropogenic sources). Natural release of mercury into the environment refers to the mobilization and release of geologically bound mercury by natural biotic and abiotic processes with mass transfer of mercury to the atmosphere. This could be from erosion and degassing from mineralized soils (Lindarvist et al., 1991); volcanic eruption and geothermal activities contribute significantly to the natural emissions (Varekamp and Buseek, 1986). This release by volcanoes, evasion of mercury from the earth's subsurface crust which occurs through faults and fractures in bedrocks (Rasmussen, 1991). Anthropogenic mercury release into the environment refers to the mobilization and release of geologically bound mercury by man's activity with mass-transfer of mercury to the atmosphere. This includes large amount of mercury vapour released into the environment as a result of burning coal and fuel oil, both of which contains trace amounts of the element, as well as incinerating

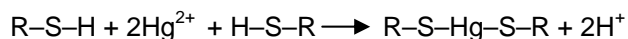
solid waste that contain mercury in products such as batteries (Bairds, 1995). Indeed, the incineration of municipal garbage has become a major source of environmental mercury pollution. When spent batteries are incinerated with other garbage, volatile mercury is released into the air. Furthermore, mercury may be released from municipal solid waste landfills as a trace component of landfills gas which is generated during decomposition of waste under anaerobic condition or in the liquid leachate flowing from site. This source of atmospheric mercury has increased substantially in recent times and now rivals the input from volcanoes, formerly the predominant source of air borne mercury. In the air, the vast majority of mercury is in the vapour state which can travel long distances before being deposited on land or in water ways.

The use of mercury amalgam in some industrial chloro-alkal plants in the process that converts aqueous sodium chloride into the commercial products, chlorine and sodium hydroxides (and hydrogen) by electrolysis. Here the recycling of mercury is not complete, and some find ways into air and into the rivers from which the plants cooling water is obtained or effluent is discharged. Although liquids mercury is neither soluble in water nor in dilute acid, apparently, it can be oxidized to soluble form by the intervention of bacteria that are present in natural water. Until recently, alkyl Hg was used as an agricultural seed dressing to prevent fungal disease in germinating seeds. This resulted in significant amounts of the metal being added to highly productive, intensively farmed agricultural soils. Steignes (1995) has previously reviewed the behaviors of mercury in soils. Other modern sources of mercury are small batteries for use in cameras and hearing aids.

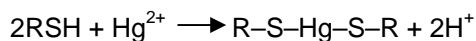
## ENVIRONMENTAL IMPACTS AND TOXICITY OF MERCURY

Although mercury vapour is highly toxic and not particularly toxic as the condensed free element. However, it is dangerous in the form of its cation and when bonded to short chains of carbon atom. Biochemically the mechanism of its toxic action arises from the strong affinity of the metal cation for sulphur. Thus, the sulphur hydryl groups SH which occurs commonly in enzyme that controls the speed of critical metabolic reactions in the human body readily attach themselves to ingests Hg metal cations or molecules that contains it. This generally affects the enzymes and makes it not to act normally and human health is affected adversely. The reaction of  $\text{Hg}^{2+}$  with sulphur hydryl units of the enzymes  $\text{R-S-H}$  to produce stable systems such as  $\text{R-S-M-S-R}$  is analogous to its reaction with the simple inorganic chemical  $\text{H}_2\text{S}$  (Bairds, 1995).





OR



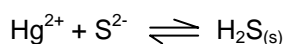
Mercury vapour can enter the body through inhalation and be carried by the blood stream to the brain where it penetrates the blood brain barriers. It disrupts metabolic processes in the brain causing tremor and psychopathologic symptoms such as shyness, depression and irritability. Divalent ionic mercury  $Hg^{2+}$  damages the kidney; organometallic mercury compounds such as dimethyl mercury  $Hg(CH_3)_2$  are also very toxic. The oxidation state favours its toxicity (Clak and Mura, 1977). Mercury intake into the human system through any means is harmful. The organic or inorganic mercury can both precipitate protein in a local reaction. In the gastrointestinal track, acute poisoning produces a sloughing away of the mucosa to an extent where piece of the intestinal mucosa can be found in the stools. This produces a large loss of fluids and electrolytes.

During recent years the importance of mercury in the food chain has become better understood. Inorganic mercury and aromatic mercury derivatives, arising from effluents from industrial processes are converted in the mud of lakes/sediments and rivers into soluble methyl mercury by the action of bacteria (Clak and Mura, 1977; Alloway and Ayres, 1997). The methylated form is very volatile and lipophilic which accumulate in the food chain of fish having high concentration of mercury that is harmful to human. Other heavy metals can be methylated in the environment but mercury appears to cause the most dangerous problems for human health (Fergusson, 1990).

### Detoxification of mercury from aqueous solution

A broad spectrum of mercury treatment technologies has been described in the technical literature, ranging from established full-scale applications to innovative approaches investigated to date only at bench or pilot scale. The literature however provides only limited information on actual full-scale treatment technology performance and almost no full-scale economic data or information on mercury recovery. Well established and widely reported full-scale technologies are precipitation, coagulation/co-precipitation, ion exchange and activated carbon adsorption (U.S E.A, 1997).

One of the more commonly reported precipitation methods for removal of inorganic mercury from wastewater is sulphide precipitation. In this process, sulphide (e.g., as sodium sulphide or another sulphide salt) is added to the waste stream to convert the soluble mercury to the relatively insoluble mercury sulphide form:



As with other precipitation treatment, this process is usually combined with pH adjustment and flocculation, followed by solids separation (e.g., gravity settling, filtration). The sulphide precipitant is added to the wastewater in a stirred reaction vessel, where the soluble mercury is precipitated as mercury sulphide. The precipitated solids can then be removed by gravity settling in a clarifier. Flocculation, with or without a chemical coagulant or settling aid, can be used to enhance the removal of precipitated solids.

Both inorganic and organic mercury can be removed by coagulation/co-precipitation for a variety of mercury containing waste waters (Patterson, 1985). Coagulants employed include, aluminum sulphate (alum), iron salts and lime. For alum and iron, the dominant mercury removal mechanism is most likely by adsorptive co-precipitation (Patterson, 1992). Here, one iron is adsorbed into another bulk solid formed, for example, by addition of alum and precipitation of aluminum hydroxide or by addition of an iron (ferrous or ferric) salt and precipitation of iron hydroxide. The adsorption process is isothermal and treatment performance can be enhanced by optimal bulk solids formation and by pH manipulation to optimize bulk solid surface change and soluble mercury speciation.

Mercury in the form of anionic complexes such as  $HgCl_3^-$ , can be treated by anion exchange resins. Resins containing the iminodiacetic acid group will exchange for cationic mercury selectively over calcium and magnesium, but copper and cobalt are also readily exchanged. The thiol resin, Duolite GT-73, is reported to be selective for mercury in any of its three oxidation states (Ritter and Bisler, 1992), ion exchange technology for mercury removal has historically been limited to the use of anion resins to treat industrial waste water that contains inorganic mercury in the complex mercuric chloride form, for the process to be effective, the chloride content of the waste water must be high, such as that generated by a chloro-alkali plant. This will yield negatively charged mercury chloride complexes. If the chloride content of the waste water is low, either chlorine or chloride salt could be added to improve removal process efficiency (Sorg, 1979).

Cation exchange of anion content of the wastewater is low (Sorg, 1979). Certain cation exchange resins (Amberlite IR-120 and Dowex -50W-X8) are reported to be effective for ion exchange treatment of mercury present in industrial wastewater (Patterson, 1985). Also, Duolite GT-73, a cationic resin, contains the thiol (-SH) group and reacts with ionic mercury, the thiol functional group has a high selectivity for mercury as well as bind certain other metal ions such as copper, silver, cadmium and lead.

Adsorption processes have the potential to achieve high efficiencies of mercury removal and/or low effluent mercury levels. The predominant adsorption process

**Table 1.** Freundlich isotherm parameters for mercury adsorption.

Adsorbent	K <sub>f</sub>	1/n	Reference
GAC	4.68	3.16	Namasivayam and Periasamy, 1993
BPHC	42.17	3.50	Namasivayam and Periasamy, 1993
Coal fly ash (pH 2.2)	1.014	0.053	Sen and De, 1997
Coal fly ash (pH 3.1)	1.094	0.333	Sen and De, 1997
Coal fly ash (pH 4.2)	1.230	0.361	Sen and De, 1997
MHBB	1.07	0.324	Deshkar et al, 1990
Sawdust	14.91	0.0757	Igwe et al., 2006.

GAC = Granular activated carbon, BPHC = Bicarbonate-treated peanut hull carbon, and MHGB = Modified Hardmickia binata bark.

utilized activated carbon. Metal hydroxides are also used as adsorbents. When metal hydroxides are employed for adsorptive treatment, the process is commonly termed coagulation or co-precipitation as earlier discussed. An inherent advantage of adsorptive treatment, particularly when the adsorbent displays isothermal or quasi-isothermal behavior, is that increased treatment efficiency results from incremental adsorbent dosage. Isothermal behaviour is observed when, for a fixed initial pollutant concentration, decreasing residual soluble concentrations are observed as the dosage of adsorbing treatment material is added. Unless adsorbent recovery is feasible, these incremental dosages also result in production of increased waste water treatment residuals, requiring ultimate disposal. Variables other than adsorbent type and dosage can also affect adsorption efficiency. Common variables include waste water pH and pollutant speciation.

Granular activated carbon (GAC) is the most commonly used adsorbent system for treating industrial waste (U.S. DOE, 1994; Igwe et al., 2005), pretreatment or modification of activated carbon with carbon disulphide to enhance mercury removal. Humenick et al. (1974) utilized an activated carbon that was presoaked in carbon disulphide and dried before being used as packed activated carbon (PAC). The enhanced mercury removal was attributed to chemisorptions reactions. Sulphur atoms have a high affinity for mercury, as evidenced by the K<sub>sp</sub> of HgS. The mercury removal mechanism proposed by Humenick et al. (1974) involves transport and diffusion to the carbon disulphide sites and subsequent formation of a chemical bond between a carbon disulphide molecule and the mercury ion.

An alternative adsorption material to activated carbon is starch xanthate, yielding mercury-starch xanthate. One modification is termed the metals extraction by xanthate Insolubilization and chemical oxidation (MEXICO) process, also known as the advanced MEXICO precipitation process (Macchi, 1985; Tiravanti, 1987).

Recently, research has focused on the use of other adsorbents. These adsorbents have been reported to per-

form in comparable fashion to activated carbon for mercury treatment. These adsorbents include bicarbonate-treated peanut hull carbon (Namasivayam and Periasamy, 1993), modified Hardmickia binata bark (Deshkar et al., 1990), coal fly ash (Sen and De, 1987), sawdust (Igwe et al., 2006), boiler fly ash (Okoronkwo et al., 2006b), coconut fiber (Igwe et al., 2005a) and iron felt (Grau and Bisana, 1995).

Table I presents mercury adsorption Freundlich parameter values for these adsorbents. The Freundlich adsorption equation is (U.S. EPA 1997):

$$\text{Log } x/m = \text{Log } K_F + 1/n \text{ log } C_e$$

Where  $x$  = the amount of solute (mercury) adsorbed,  $m$  = the amount of adsorbent required to adsorb  $x$ ,  $K_f$  and  $1/n$  = empirical constants (Freundlich parameters), and  $C_e$  = equilibrium concentration (mercury).

The Freundlich parameters  $K_f$  and  $1/n$  are equal to the intercept and slope of the line obtained by plotting  $\log x/m$  vs  $\log C_e$ . The Value of  $K_f$  is roughly an indication of sorption capacity and  $1/n$  is an indicator of sorption intensity. The favourable comparison between the values of  $K_f$  and  $1/n$  shows that these adsorbents are potential sorbents for mercury removal and detoxification.

Also, the use of micro-organism such as bacteria and fungi have been employed (Brunke et al, 1993; Chang and Hong, 1995; Von Canstein et al, 1999, 2001; Wagner Dobler et al., 2000, 2000a; Whiteley et al., 2001). Von Canstein et al. (2002), found out that the growth of micro-organisms in wastewaters containing ionic mercury depends on the Hg (II) bioavailability and toxicity. Nutrients containing sulphhydryl groups (e.g. yeast extract), as well as negatively charged ions (e.g. chloride), bind to ionic mercury and thereby alter its bioavailability and toxicity (Farell et al., 1990, 1993). The ability to grow in the presence of mercury (a process requiring several hours) has been shown to be a more important feature of mercury in resistant cells than short-term volatilization rates (a reaction of several minutes) for the application in a long-term bioremediation process.

## CONCLUSION

The re-emission of mercury is the mass transfer of mercury to the atmosphere by biotic and abiotic processes from a pool of mercury that was deposited to earth's surface after initial mobilization by either anthropogenic or natural sources. Also, the environmental mercury derived from natural sources – weathering processes, terrestrial and submarine volcanic activity makes mercury pollution a fact of life. However, man-made sources could be restricted to reduce mercury pollution in the environment. Mercury is highly toxic, persistent and bio-accumulates in food chain. Once deposited the chemical form of mercury can change through a methylation process into methyl mercury which is a highly toxic, more bioavailable forms that biomagnifies in the aquatic food chain.

Recent researches indicates that mercury removal may be enhanced through the use of oxidizing agents that convert elemental mercury to the ionized form or through the use of sorbents that adsorb the mercury onto solid particles. Microorganisms are also being employed.

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