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## PH DEPENDENCE ADSORPTION STUDIES OF MERCURY (II) ION ON BIO MASS – A REVIEW

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### ABSTRACT

Heavy metals adsorption on biomass being a physicochemical process involves three factors that play a vital role related to, the adsorption system, the type of metal and type of adsorbent. The mercury emitted from volcanoes, geothermal sources and topsoil enriched in mercury pertains to primary natural sources. Anthropological activities break the natural mercury cycle, and significantly increase the release of mercury into the environment thus enhancing elevated mercury concentrations in the water, the sediment, and fish. All forms of mercury play a key role in damaging/altering the tertiary and quaternary protein structure and alters the cellular function by attaching to the selenohydryl and sulfhydryl groups due to high affinity (-SH,-SeH) as known from their high stability constants. The amount of the Hg<sup>2+</sup> removed at a lower acidic pH is comparatively less than the amount of the metal ion removed at a higher acidic pH.

### KEYWORDS

Adsorption, Biosorption, Toxicity, Anthropogenic, pH and Adsorption isotherm.

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### INTRODUCTON

The expulsion of partly or un processed industrial water called as “waste water”, containing Metals and metalloids having atomic density greater than 5g/cm<sup>1,2</sup> along with other harmful substances into the hydrosphere is a major source of water pollution and a serious worldwide concern<sup>1,3,4,5,6</sup>. These toxics either get dissolved in water, lie suspended or get deposited on the bed disturbing both the aquatic and terrestrial ecosystems<sup>5,7</sup>.

Mercury and its compounds have received greater environmental concern due to their high toxicity, persistence in the environment, high bioaccumulation, and nonbiodegradability and can bind with organic and inorganic matter and form various composites which limit water use<sup>2,8,1,9,5,10</sup>.

The contamination of water is directly related to the degree of contamination of our environment<sup>11</sup> Toxicity causes either acute or chronic health effects.

It is therefore, essential to control and prevent the unsystematic discharge of these metals in the environment. For this reason, increased attention is being focused on the development of technical proficiency for the removal of metal bearing effluents before being discharged into water bodies and natural streams.

An MCL (metal contamination limit) is the legal threshold limit on the amount of a substance that is allowed in public water systems under the Safe Drinking Water Act.

In India the maximum concentrations allowed to discharge in inland surface water are 0.01 mg/L for Hg (II)<sup>12</sup> (WHO) recommends a maximum uptake of 0.3 mg/week and 1 µg/L or 0.001 mg/L as maximum acceptable concentration in drinking water<sup>13</sup>. Mercury acceptable concentration in drinking water according to the United States Environmental Protection Agency (USEPA) is 2 µg/L<sup>1</sup>. Fish containing more than 0.4 ppm Hg are unfit for human consumption.

Mercury a “d” block element is known to exist in nature in three forms (elemental, inorganic, and organic), with each having its own degree of toxicity causing gastrointestinal, neuro, and nephrotoxicity<sup>14</sup>. Methylation of inorganic (mercuric) forms of mercury to Methyl mercury by microorganisms of soil and water is the most frequently encountered compound of the organic form found in the environment, the two most highly absorbed species are elemental mercury (Hg<sup>0</sup>) and methyl mercury (MeHg).

### **Sources of mercury**

#### **Natural Sources of Mercury and its emission**

Concentrated mercury ores are frequently found where high-density rocks are forced to the surface in hot springs or volcanic regions. It naturally enters the environment through volcanic eruptions, forest fires, evaporation from oceans and topsoil, weathering of mercury-containing rocks, erosion, and, through tens of thousands of uncontrolled coal bed fire. Coal-bed fires have occurred since

prehistoric times and were initiated by natural causes including spontaneous combustion, lightning strikes and forest fires.

The mercury emitted from volcanoes, geothermal sources and topsoil enriched in mercury pertains to primary natural sources, whereas the re-emission of previously deposited mercury on vegetation, land or water surfaces is primarily related to land use changes, biomass burning, meteorological conditions and exchange mechanisms of gaseous mercury at air-water/top soil/snow-ice pack interfaces. The contribution of volcanoes varies over time depending whether they are in a degassing or eruption phase. Most of yearly natural source emissions, come from surface waters followed by terrestrial surfaces<sup>15,16</sup>.

#### **Anthropogenic sources of Hg**

Anthropological activities break the natural mercury cycle, and significantly increase the release of mercury into the environment thus enhancing elevated mercury concentrations in the water, the sediment, and fish<sup>17</sup>.

Persistent nature of Mercury in the environment is responsible for significantly elevated concentrations of mercury and it is submitted that past gold and silver mining events dating back to the 19th century have emitted historical amounts of mercury into the environment are responsible for the existing contamination<sup>15-17</sup>.

Anthropogenic sources of mercury are categorised into primary and secondary anthropogenic sources<sup>18</sup>.

The Primary anthropogenic sources involve the mobilization and release of geological origin of mercury to the environment, as an unintentional by product and the sources under this category are mining and extraction (both for mercury and for other minerals), The essential mining sources include gold, silver, mercury, and Lead mines. Stationary combustion of coal, burning of fossil fuels<sup>15,18-20</sup>.

Secondary anthropogenic sources are those sources where emissions occur by the purposeful use of mercury, including mercury use in industrial processes, in dental applications, or in artisanal and small-scale gold mining (ASGM) operations<sup>18</sup>,

ferrous and non-ferrous metals manufacturing Facilities, caustic soda production plants/ chlor-alkali plants, ore processing facilities, incinerators for Urban, medical and industrial wastes, cement plants, batteries, and pig-iron Production dental applications<sup>18</sup> and chemicals production<sup>15,16,18-20</sup>. Asia (especially China and India) alone accounts for about 50% of total anthropogenic mercury emissions<sup>15-18</sup>. Mercury appears as an impurity of copper, zinc, lead and nickel ores as well as in gold ores. Smelting processes to obtain these metals are known to be large sources of mercury released to the atmosphere, especially in developing countries<sup>15,20</sup>.

#### **Instruments/appliances**

Mercury-containing, measuring and control devices including batteries, paints, switches, electrical and electronic devices, thermometers, blood-pressure gauges, fluorescent and energy-saving lamps fluorescent tubes, compact fluorescent, high-intensity discharge lighting), sphygmomanometers, barometers and manometers are still manufactured in various parts of the world, although mercury-free substitutes are available for almost all such applications, with China responsible for world production of many of these products. Mercury-containing lamps remain the standard for energy-efficient lamps, and mounted around the world<sup>18</sup>.

#### **Mercury in industrial wastes**

The most important source that generates wastes containing mercury is the chlor-alkali industry ranking the third major mercury user worldwide. The chlor-alkali industry uses mercury to convert salt to chlorine gas and lye, which is used in soaps, detergents, plastics, and in the paper-making process<sup>15,18</sup>. Mercury, an additional source of waste during metal smelting process, and vinyl chloride monomer (VCM) an intermediate feed stock in the production of polyvinyl chloride (PVC)<sup>21</sup> the precursor of which again a compound of Mercury and the Hg/PVC ratio is in the range of 0.12–0.20 kg mercury per Mg of PVC produced<sup>15,18</sup>.

Petrol combustion contributed almost double of mercury emissions, as compared to diesel contribution<sup>15</sup>. Mercury is released into the

environment when Automobiles are scrapped and melted in mini-mills<sup>20</sup>.

#### **Toxicity of mercury**

Toxicity, a cluster of events, beginning with exposure, followed by distribution, metabolism, and ending with an interaction with cellular macromolecules (usually DNA or protein) and get expressed as a toxic end point, being a quantitative concept, almost any substance being harmful at some doses but, at the same time, causing no harmful effect at particular lower dose, between these two limits there is a range of possible effects, from subtle long-term chronic toxicity to immediate lethality<sup>14</sup>.

Exposure of humans and other organisms to toxicants would be in one of the ways: deliberate ingestion, job-related contact (industries), environmental exposure, as well as accidental and intentional (suicidal or homicidal) poisoning. The toxicity of a particular compound also depends and varies with the type of entry into the body, whether through the alimentary canal, the lungs, or the skin<sup>14</sup>. Elemental and inorganic mercury, are most efficiently absorbed by inhalation of mercury vapours, with about 80 % of the inhaled vapours directly entering the bloodstream and 90 % of ingested MeHg is absorbed through the intestine<sup>22,23</sup>.

Mercury is known to exist in three forms namely metallic mercury ( $\text{Hg}^0$ ), inorganic mercury salt ( $\text{Hg}^+$ ,  $\text{Hg}^{++}$ ), and organic mercury compounds (Methyl, ethyl, phenyl mercury)<sup>21</sup>. Different forms of mercury causes various degrees of toxicity, due to the variance in their absorption, transport, and decontamination in the body<sup>22,24</sup>.

All forms of mercury play a key role in damaging/altering the tertiary and quaternary protein structure and alters the cellular function by attaching to the selenohydryl and sulfhydryl groups due to high affinity (-SH,-SeH) as known from their high stability constants, which undergo reaction with methyl mercury and hinder the cellular, causing potentially impair function of any organ, or any sub cellular structure<sup>22,25,26</sup>. Inorganic and organic mercury compounds have higher affinity for selenol groups than for thiol groups.

Metallothionein and GSH are examples of thiol-rich intracellular proteins. Selenoproteins include GSH peroxidase, iodothyronine deiodinase, thioredoxin reductase, and selenoprotein. P.Cysteine residues on serum albumin and haemoglobin, transport ~90 % of  $Hg^{2+}$  and MeHg in plasma<sup>22</sup>. The basis for heavy metal chelation is that, though the mercury sulfhydryl bond is stable and divided to surrounding sulfhydryl consisting ligands, it also contributes free sulfhydryl groups to promote metal mobility within the ligands<sup>25</sup>. Due to the ionic nature of the mercury-- carbon bonds,  $Hg^{2+}$  is released in aqueous solution from the organic moieties, associated with the Hg atom<sup>23</sup>.

Metallic and inorganic mercury once absorbed, enter an oxidation-reduction cycle. Metallic mercury gets oxidized to the divalent cation in the red blood cells and lungs of humans and animals. Absorbed divalent cation from exposure to mercuric compounds can, in turn, be reduced to the metallic or monovalent form and released as exhaled metallic mercury vapour. Mercurous mercury ( $Hg^+$ ) disproportionate in the presence of protein sulfhydryl groups, to a divalent cation ( $Hg^{2+}$ ) and elemental form in the zero oxidation state ( $Hg^0$ )<sup>22,24</sup>. Most of the absorbed MeHg is incorporated into bile, secreted into the intestine, and reabsorbed through the enterohepatic circulation<sup>22</sup>, ethyl mercury, gets metabolized to  $Hg^{2+}$  more rapidly (the longer the alkyl chain of the organic mercury compound, the more it behaves as inorganic  $Hg^{2+}$ ), and therefore have different tissue accumulation, the highest levels of ethyl mercury are found in the kidney, with very little accumulating in neural tissues<sup>22</sup>.

Mercury is known to hamper the process of transcription and translation causing the disappearance of ribosomes, suppression of endoplasmic reticulum and the activity of natural killer cells<sup>25-27</sup>. The cellular integrity is also affected causing free radical formation. Studies show that, Detoxification of  $Hg^{2+}$  and MeHg species occur through complexation with GSH. GSH-mercury complexes facilitate systemic excretion through the liver (bile) and kidneys as well as cellular efflux of MeHg from astrocytes<sup>22</sup>. Excretion of alkyl mercury

occurs mostly in the form of faeces (90%), secondary to significant enterohepatic circulation. The biological half-life of methyl mercury is approximately 65 days.

#### **Metallic mercury**

Metallic mercury /elemental mercury ( $Hg^0$ ) is a liquid at room temperature with a vapour pressure of 0.00185 mm at 25°C showing its extreme volatility<sup>21,23</sup>, with no risk to life. However, in the vapour form, metallic mercury is freely absorbed through the Lungs causing damage<sup>22,24</sup>, because of its highly diffusible nature it passes through cell membranes as well as the blood-brain and placental barriers to reach target organs<sup>27</sup>. The chief target organ of mercury vapour being the brain, disrupting its function and affecting the nervous system<sup>24,26</sup> Enormous acute contact to mercury vapour, causes erosive bronchitis and bronchiolitis leading to respiratory failure accompanied by CNS symptoms such as tremor or erethism<sup>21,22,25</sup>. Chronic exposure to clinically significant doses of mercury vapour usually produces, Persistent neurological dysfunctions<sup>22</sup>. Once in the bloodstream, mercury undergoes catalase and peroxidase-mediated oxidation in red blood cells and tissues and is transformed into inorganic mercuric mercury ( $Hg^{++}$ ) and mercurous mercury ( $Hg^+$ ), a process that limits its absorption<sup>27</sup>.

#### **Inorganic mercury**

The mercurous mercury in the form of mercurous chloride ( $Hg_2Cl_2$ ) is little absorbed in the body<sup>13,14</sup> Salts of Mercuric mercury, are produced by the combination of mercury with divalent anions like chlorine, sulphur, or oxygen<sup>21,27</sup> Mercuric mercury in the form of mercuric chloride ( $HgCl_2$ ), is found to be highly poisonous and destructive, in the bloodstream it binds to sulfhydryl groups on erythrocytes, glutathione, or metallothionein or is transported suspended in plasma<sup>27</sup> Mercuric mercury accumulates in placenta, fetal tissues, and amniotic fluid. Numerous studies have also revealed that mercury generates oxygen free radicals mainly by activation of NAPH oxidase<sup>27</sup> Acute poisoning with mercuric salts (typically  $HgCl_2$ ) generally targets the gastrointestinal tract and the kidneys along with lung damage and Thyroid

dysfunction<sup>26,28</sup>. Extensive precipitation of enterocyte proteins occurs<sup>26</sup> Inorganic mercury does not readily cross cell membranes due to low lipid solubility or enter the brain because of the blood–brain barrier and hence central nervous involvement specifically brain dysfunction<sup>23,26,27</sup> related to inorganic mercury exposure is not known<sup>28</sup>. Once absorbed, the majority of  $\text{Hg}^{2+}$  accumulates in the kidney and liver<sup>22,24</sup>.

### Organic mercury

A covalent bond between mercury and the carbon atom of an organic functional group such as a methyl, ethyl, or phenyl group, results in the formation of Organic mercury compounds, also called organometallic. Methyl mercury ( $\text{CH}_3\text{Hg}^+$ ) is by far the most common form of organic Hg to which humans and animals are exposed and in the environment is predominantly formed by methylation of inorganic mercuric ions by microorganisms present in soil and water<sup>21,25,27</sup>. The brain, the target organ for methyl mercury, interrupts intracellular calcium homeostasis<sup>25</sup>. Several studies support that MeHg-L-cysteine is transported across the blood–brain barrier by the L-type amino acid transporters used to transport neutral amino acids, and was first observed by Aschner *et al.*, who observed competitive inhibition of MeHg uptake in astrocytes by large, neutral amino acids<sup>22</sup>. Methyl mercury is a neurotoxic compound which is responsible for microtubule destruction, mitochondrial damage, lipid peroxidation and accumulation of neurotoxic molecules such as serotonin, aspartate, and glutamate<sup>25</sup>. Methyl mercury forms thiol (-SH) complexes with proteins 164 and amino acids [such as glutathione (GSH) and cysteine] in the liver<sup>22</sup>.

Mercury accumulates in fish due to contamination of the marine environment and diet<sup>21</sup>. As such, fish living in contaminated waters, or predatory fish living a long time, are more likely to be contaminated with mercury<sup>21</sup>. The Minamata catastrophe in Japan in the 1950s was caused by methyl mercury poisoning from fish contaminated by mercury discharges to the surrounding sea<sup>28</sup>.

### The Effect of pH

pH plays a vital role in the removal efficiencies of mercury (II) ions. With increase in acidic pH to a certain maximum extent (<6) the adsorption also increases<sup>29</sup>.

The pH influences the equilibrium, by affecting the speciation of the adsorbates in solution as well as the surface charge of the adsorbents, functional groups on the adsorbent surface, such as carboxyl, phosphate, sulphur groups and amino groups, chemical characteristics of the metal ions (adsorbates), such as solubility the concentration of competing hydrogen ions, and the interaction of the active binding sites on the biosorbent<sup>30-34</sup>.

The amount of the  $\text{Hg}^{2+}$  removed at a lower acidic PH is comparatively less than the amount of the metal ion removed at a higher acidic pH<sup>6,31,41,34-37</sup>.

With the increase in acidic pH (i.e.) as, the adsorbates solution tends to become slightly acidic in nature (pH<6), the deprotonating the metal binding sites (increases, increasing biosorption<sup>29-32,34,38</sup>.

At lower acidic pH higher concentration of  $\text{H}^+$  ions can protonate the sorption sites of biosorbent or compete with  $\text{Hg}(\text{II})$  ions thus decreasing the removal of  $\text{Hg}^{2+}$ <sup>29-32,34,39,40</sup>.

At  $\text{pH} \geq 6$  (crucial zone of precipitation) the metal ions get out of the solution (mocking adsorption) due to the formation of metal hydroxide species such as soluble  $\text{Hg}(\text{OH})^+$  and/or insoluble colloidal precipitate of  $\text{Hg}(\text{OH})_2$ <sup>29,39,41</sup>.

The hydrolysis of mercury may be involved in the increase of the pH value<sup>38,41</sup>.

**Table No.1: Percentage of metal ion adsorbed by the various biosorbents at different pH**

S.No	Adsorbate	Adsorbent/biosorbent	Modifying agent/activant	Conditions	pH	% / quantity adsorbed	Model used to calculate adsorption capacity	Ref
1	Hg	S. glaucescens and G. corticata,	original	----	5 7	92.5% 97.3%,	Freundlich Langmuir	(35)
2	Hg	Dried <i>M. rouxii</i> IM-80 biomass (fungal biomass)	original	----	5.5	95.4%	----	(31)
3	Hg	Unmodified <i>Triplochyton scleroxylon</i> sawdust	original	----	2.57	59% 88% / 1.76mg g <sup>-1</sup>	Freundlich Langmuir	(29)
4	----	Modified (base treated) <i>Triplochyton scleroxylon</i> sawdust	NaOH	2M NaOH, air dried and oven dried	47	25% 94% /1.88 mg g <sup>-1</sup>	----	(29)
5	Hg	Un modified Peanut Hull Powder	original	----	4-6	Max. adsorption	Freundlich Langmuir Temkin	(32)
6	----	Chemical-Modified Peanut Hull Powder	Mercapto acetic acid, acetic anhydride, acetic acid, concentrated sulphuric acid and 17.5% NaOH	Oven dried 40oC for 2-4 days	4	Max. adsorption	----	(32)
7	Hg	Sheep bone charcoal	original	----	3	Max. adsorption	Freundlich Langmuir	(39)
8	Hg	Raw almond shell	original	65°C for 24 h.	5	Max. adsorption	Freundlich Langmuir Dubinin– Radushkevich	(42)
9	----	Chemically activated almond shell	ortho-phosphoric acid	nitrogen atmosphere at 500°C for 1 h	5	Max. adsorption	----	----
10	Hg	CS	glutaraldehyde	----	2,3,4, 5,6	25%,34%,45% %,56%,58%	Freundlich Langmuir	(29)
11	----	CSm	glutaraldehyde and functionalised with Magnetic	----	2,3,4, 5,6	29%,44%,54% %,57%,60%	----	----

12	Hg	<i>Macrocystis pyrifera</i>	0.2 mol/L CaCl <sub>2</sub> solution at PH 5.0.	Oven dried 50°C for 48 hr	6	Max. adsorption	Freundlich Langmuir	(38)
13	----	<i>Undaria pinnatifida</i>	0.2 mol/L CaCl <sub>2</sub> solution at pH 5.0.	Oven dried 50°C for 48 hr	6	Max. adsorption	----	(38)
14	Hg	Palm Shell Powder	original	----	7	Max. adsorption	Freundlich Langmuir	(43)
15	Hg	Novel Biocarbon Technology	Conc. sulphuric acid	160 ± 5 °C for 6 h	5.5.	96.5 %	Freundlich Langmuir	(37)
16	Hg	Protonated Pistachio Hull Wastes	HCL	24 hours dried at 35°C overnight	3.0 9.0.	28% 92%	----	(34)
17	Hg	Functionalized Activated Carbon Derived from Areca NutWaste	Succinic anhydride dissolved in acetone.	microwave irradiation at 60°C for 60 min	5	>70%	Freundlich Langmuir	(30)
18	Hg	ON peat	original	----	2 10	99 % 92 %	----	(6)
19	Hg	OT peat	original	----	2 10	99% 95%	----	(6)
20	Hg	LS peat	original	----	2 10	99% 96%	----	(6)
21	Hg	RH peat	original	----	2 10	96% 95%	----	(6)
22	Hg	LN peat	original	-----	2 10	97% 96%	-----	(6)
23	Hg	MS peat	original	-----	2 10	98% 98%	-----	(6)

## CONCLUSION

The present review is aimed to highlight the effectiveness of biosorption Process through the recently published studies regarding the use of improvised natural biomasses, which being cost effective and environment friendly are used for removal of metal ions like mercury from aqueous solutions. The studies proved that the functional groups present in the biomass material plays a key role in biosorption. The pH of the solution is one of the most important factors affecting metal ion biosorption, by affecting the metal ion solubility as well as biosorbent total charge. The removal of

metal ions is almost negligible at highly acidic pH values and increases by increasing the solution pH up to a certain limit. The isothermal models like Langmuir, Freundlich, and Temkin have been widely employed for modelling the biosorption process.

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## CONFLICT OF INTEREST

We declare that we have no conflict of interest.

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