Microbial and plant-assisted heavy metal remediation in aquatic ecosystems: a comprehensive review

Shyamalina Haldar[1] · Abhrajyoti Ghosh[2]

Abstract

Heavy metal (HM) pollution in aquatic ecosystems has an adverse effect on both aquatic life forms as well as terrestrial living beings, including humans. Since HMs are recalcitrant, they accumulate in the environment and are subsequently biomagnified through the food chain. Conventional physical and chemical methods used to remove the HMs from aquatic habitats are usually expensive, slow, non-environment friendly, and mostly inefficient. On the contrary, phytoremediation and microbe-assisted remediation technologies have attracted immense attention in recent years and offer a better solution to the problem. These newly emerged remediation technologies are eco-friendly, efficient and cost-effective. Both phytoremediation and microbe-assisted remediation technologies adopt different mechanisms for HM bioremediation in aquatic ecosystems. Recent advancement of molecular tools has contributed significantly to better understand the mechanisms of metal adsorption, translocation, sequestration, and tolerance in plants and microbes. Albeit immense possibilities to use such bioremediation as a successful environmental clean-up technology, it is yet to be successfully implemented in the field conditions. This review article comprehensively discusses HM accumulation in Indian aquatic environments. Furthermore, it describes the effect of HMs accumulation in the aquatic environment and the role of phytoremediation as well as microbe-assisted remediation in mitigation of the HM toxicity. Finally, the review concludes with a note on the challenges, opportunities and future directions for bioremediation in the aquatic ecosystems.

Keywords Aquatic ecosystem · Heavy metals · Human health · Phytoremediation · Water-treatment plants

Introduction

Metals form the natural constituents of the ecosystem. Twenty-three metals out of 35 naturally existing metals possess high specific density above 5 g/cm^3 with atomic weight greater than 40.04 and are generally considered as heavy metals (HM; Li et al. 2017a, b). In aquatic ecosystem, metals are introduced either by natural processes such as volcanic eruptions, weathering of soils and rocks and/or from various anthropogenic activities including mining, processing, industrial effluents containing the metals and/or substances that contain metal pollutants. Although the HM

are distributed in the ecosystem always, their exposure to the humans occurs particularly through numerous anthropogenic activities (Engwa et al. 2019). These non-biodegradable HM and metalloids adversely affect human health causing damage to multiple organs including liver, lungs, bones, kidney and even brain by forming nonspecific complexes at high concentrations (Islam et al. 2018).

The heavy metal pollution in aquatic ecosystem occurs either through (1) localized pollution occurring through point sources, where pollutants derive from single, recognizable sources or (2) nonpoint sources, where pollutants come from dispersed sources and are difficult to be identified (https://www.lenntech.com/aquatic/metals.htm; accessed on 18th October, 2019). The natural weathering of ore bodies and the little metal particles coming from coal-burning power plants via smokestacks in air, water and soils around the factory form the point-source.

Mining activity is the dominant source for metal pollution in freshwater due to the use of acid mine drainage (AMD) system to release HM from the ores due to the high solubility

✉ Abhrajyoti Ghosh
  abhrajyoti.ghosh@jcbose.ac.in

[1] Department of Biochemistry, Asutosh College, University of Calcutta, Kolkata 700026, India

[2] Department of Biochemistry, Centenary Campus, Bose Institute, P-1/12 C.I.T. Scheme VII-M, Kolkata 700054, India

of those metals in acid solutions. AMD is subsequently followed by the dispersal of acid solution in groundwater, containing high levels of metals (https://www.lenntech.com/aquatic/metals.htm; accessed on 18th October, 2019). Due to low pH, the solubility and the mobility of the metals increase and get transported easily or get locked up in the bottom sediments. The accumulated metals are being consumed and get stored in the tissues of the aquatic organisms disturbing their lives and thereafter also get transported through food chain into the other ecosystems. Therefore, decontamination of the metal-polluted environment is considered as a technical challenge to the ecologists. For this purpose, the development of effective methods for bioremediation of HM is one of the prerequisites (Mallick et al. 2019). Therefore, this review aims to provide an insight into the heavy metal pollution in the aquatic system in India; their effects on the aquatic organisms and the other ecosystems; followed by the established mechanisms and the future potentials of heavy metal bioremediation strategies.

HM accumulation in aquatic systems: present scenario in India

Biochemistry of heavy metal

To understand the toxicity of the HM, it is of immense importance to have robust knowledge on the concentrations and oxidation states of the HM at which stage they become harmful in the ecosystem. The toxic effects of HM are dictated by the chemical nature, concentration, oxidation states and bioavailability. Multiple external factors including the presence of other metals, pH of the medium, amount of organic matter present, presence of phosphates and sulphates influence the biological interactions of HM in a microhabitat (Diaz et al. 2020). This HM poisoning is caused by the interference of the HM with the normal metabolism of the living system. The ingested HM are converted to their most steady oxidation states (Silver: Ag^+, Arsenic: As^{3+}, Cadmium: Cd^{2+}, Chromium: Cr^{3+}, Mercury: Hg^{2+}, Lead: Pb^{2+}, Zinc: Zn^{2+}) at low pH of stomach which form stable in dissociable complexes with the proteins and the enzymes (Jaishankar et al. 2014).

Eg. As^{3+} can inactivate 200 enzymes such as dihydrolipoyl dehydrogenase and thiolase by binding to thiol/sulfhydryl groups of proteins while As^{5+} replaces phosphate group which forms an important source of energy in numerous biochemical pathways. Similarly, the toxicity level of Cr depends on its valence states in the range as $Cr^{3+} > Cr^{6+}$. Of these, Cr^{3+} is found ubiquitously in the environment while Cr^{6+} occurs in ground water and surface water In case of inorganic Hg, both mercurous (+1) and mercuric (+2) states exist in the environment that are

found to occur within living organisms as organic methylmercury (Tchounwou et al. 2012). Iron-mediated toxicity has been attributed to Iron [Fe(II)], that reacts with oxygen producing free radicals that damage macromolecules causing cell death (Chamnongpol et al. 2002). In addition, Nickel (Ni) and Cupper (Cu) are toxic in their + 2 valence states where they can bind and damage DNA (Guillamet et al. 2008).

Seas/Oceans

India is a peninsular land with Arabian Sea on the West, Bay of Bengal in the East and Indian Ocean in the South. The mainland of India is connected to these water bodies through thirteen ports along the nine coastal states and are under constant influence of anthropogenic activities. This results in accumulation of heavy metals along the estuaries, coastal sediments, and the water bodies. Studies from Arabian Sea coasts and harbours from Gujarat, India show the accumulation of Cd, Ni, Pb and Zn higher than the effect range median values, and quite similar to the urban sites around the world indicating their potential toxicity to the aquatic environment (Sundararajan et al. 2017). Anthropogenic activities and weathering of rocks were identified as the major causes for these HM accumulation (Upadhyaya et al. 2014). Another study also identified cobalt (Co), manganese (Mn), Fe and a decreasing gradient accumulation of these HM from nearshore to shelf (Laluraj and Nair 2006). Similar gradient concentrations for Hg in the sediments along the western continental marginal high of India was found to increase from the higher depth of the oceans towards the surface indicating increasing anthropogenic activities towards the shores (Chakraborty et al. 2016; Ram et al. 2003). Significant enriched levels of chromium (Cr) and Pb were identified from the sediments of nearshore area, off Calicut, southwest coast of India (Srinivas et al. 2017). Determination of geo-accumulation index for the sediments from the Ribandar solar saltern near the Mandovi estuary identified Fe, Mn, Ni, Co, Pb and Zn during the salt-making season. These observations were attributed to the barge transport of ferromanganese ore to the Mormugao harbour (Pereira et al. 2013). Besides these, several studies have reported the occurrence of HM among the different aquatic organisms including fishes, oysters, turtles, algae and bacteria from Arabian Sea and Indian Ocean calling for an urgent attention for the checking of HM contamination in the aquatic bodies through anthropogenic activities (Obaidat et al. 2015; Chen et al. 2018; du Preez et al. 2018). However, till date studies from the Indian coastal parts of the Indian Ocean are limiting as most of these studies concentrated on the other coastal sides of this ocean.

Rivers

The rivers of India form a vital component in the lives of the people by providing potable water, cheap transportation, electricity, and the livelihood for many people nationwide and hence all the major cities are located on the banks of the river. Seven major rivers along with their multiple tributaries make up the river system of India. However, most of the rivers are polluted due to industries, untreated sewage and solid wastes. A recent research in 2018 conducted by Central Water Commission revealed that 42 rivers in India have at least two toxic heavy metals beyond the permissible limit (https://cwc.gov.in/main/downloads/status_trace_toxic _materials_indian_rivers.pdf; accessed on 31st October, 2019). Of these, five HM including Cr, Cu, Fe, Pb and Ni are found to pollute the Ganga river while the other six rivers Arkavathi (Karnataka), Orsang (Gujarat), Rapti (Uttar Pradesh), Sabarmati (Rajasthan and Gujarat), Saryu (Uttarakhand and Uttar Pradesh) and Vaitarna (Maharashtra) possessed excessively high concentration of the four pollutants.

A study by Singh and Kumar in 2017 showed high concentrations of heavy metals including Fe, Mn, Ni, Co, Cd and Pb from numerous geogenic and anthropogenic sources in the Ajay river (Singh and Kumar 2017; Kumar and Singh 2018). High pollution load index and high concentrations of Cd, Cr, and Pb in the Sundarban rivers have been one of the principal reasons of mangrove deterioration in this heritage site (Roy et al. 2018). Even the presence of silver nanoparticles in the Hooghly river estuary due to boat activities have been found to negatively affect the aquatic life (Antizar-Ladislao et al. 2015; Kumar Sarkar et al. 2004). Similar anthropogenic activities with the inflow of urban and industrial effluents led to the accumulation of Cu, Cr, Fe, Mn, Ni, Pb, and Zn in the sediments of Kabini river, Karnataka (Hejabi and Basavarajappa 2013). Hazard index values of Cd, Co and Cr > 1 in addition to the presence of Fe, Co, Ni and Mn was reported from the river Damodar suggesting potential health risk for the children in the states of Jharkhand and West Bengal (Pal and Maiti 2018; Bhattacharjee et al. 2005; Banerjee and Gupta 2013).

In South India, similar toxic metals were found in the sediments of Coleroon River Estuary and Swarnamukhi River Basin in Tamil Nadu (Venkatramanan et al. 2015; Patel et al. 2018). India regarded as a source for major Hg pollution following China and the currently established Hg pollution sites in India are Kodai Lake, Kodaikanal, Tamil Nadu, and Thane Creek, Mumbai (Bhave and Shrestha 2018). Alarming level of heavy metals have been also reported from Gomti river, Uttar Pradesh (Gupta et al. 2014). Even the so-called pristine areas along the river Pachim (a tributary of the Brahmaputra river) was found to be influenced by anthropogenic activities and the recorded levels of Fe and Co were the highest as compared to other rivers of the country. In

addition, the values for Cr, Mn, Cu and Se each exceeded the world average value (Hussain et al. 2006). In addition, distribution of radioactive metals including 238U, 234U, 232Th and 230Th in the river basin around Kaiga and its estuarine region were found and were reported to have come from the adjacent Arabian Sea (Balakrishna et al. 2001; Karunakara et al. 2001).

Ground water

Groundwater, water found underground in cracks and spaces in soil, sand and rock, accounts for 98 percent of the Earth's fresh water, providing the main source of usable water for a huge mass of population all over the world (https://www. ngwa.org; accessed on 1st April, 2020). About 50% of the total population uses groundwater for drinking and 43% of agricultural water use depends on groundwater (https ://www.fao.org/nr/aquastat; accessed on 1st April, 2020). However, at present, concerns are raising for groundwater as this is being contaminated with pesticides, HM, residual waste products, pharmaceutics, cosmetics and other biological agents deriving from natural and anthropogenic sources (Kurwadkar 2017).

The HM in the soil percolate through the subsurface into the groundwater. These HM origin from atmospheric deposition of metal-containing particles, improper disposal of waste residues of thermal plants, mining sources, manufacturing units, industrial estates, run-off from the roads, metal-containing fertilizers and pesticides in commercial agriculture and other anthropological activities like smelting, transportation and military operations (Gall et al. 2015).

In North India, 17.07% of the water samples from forty-one locations in Udhampur district of Jammu & Kashmir, India showed high concentration of radon above the reference range suggested by United Nation Scientific Committee on the Effects of Atomic Radiations (UNSCEAR) (Kumar et al. 2018). However, this was attributed to the geology of the regions from where the water originated. Accumulation of Cr, Ni, Cd, Pb, Zn and As in groundwater from sub-urban areas of Kanpur, the leather-city of India with a huge number of tanneries have been reported. In a study by Dotaniya et al., the effluents from the tanneries were found to contain 1.53–57.3 ppm Cr, 0–0.12 ppm Ni, 0–0.02 ppm Cd, 0–0.07 ppm Pb, 0–0.48 ppm Zn and 0–0.03 ppm As. This was attributed to the use of high amount of salts of chromium sulphate for tanning processes and contamination of other industrial eluents; discharged into the same sewage channel (Dotaniya et al. 2017). Similarly, high amounts of Co, Cr, Fe, Ni, Pb and Zn have been reported from the groundwater of the industrial estates and thermal plants from different regions of northern India like Ghaziabad district, Uttar Pradesh, due to poor management of pond (mostly unlined ponds) and filtration of ash slurry through soil dissolving

and immobilizing the HM along with (Bhutiani et al. 2016; Verma et al. 2016; Chabukdhara et al. 2017). Not only the industrial estates, but groundwater from wetlands as from Ropar wetland of Punjab showed the presence of Cd and Cr higher than Bureau of Indian Standards (BIS) limits (0.003 and 0.05 mg/L, respectively) while mean non-cancer hazard quotient was > 1 mg/L for cobalt (5.09–7.63 mg/L). Similarly, mean hazard indices for cumulative risks for all the HM (6.00–10.11 mg/L) was also recorded to be > 1 mg/L. Taken together, these observations point out to the higher risk of non-cancerous health problems in residents of the study area (Sharma et al. 2019). Higher concentrations of Uranium (> 341 µg/L) from shallow ground water tables was reported in the central, southern, and southwestern parts of the Punjab and it was found to be the highest during post-monsoon periods due to leaching of bicarbonates from the plant-roots and increased oxic conditions in the groundwater (Sharma et al. 2018).

In South India, one such industrial area from Peenya in Bangalore has accounted for six eco-toxic metals in the order Cr > Fe > Pb > (Cu) > Ni > Cd from thirty groundwater sampling stations with metal index being 10.36 and heavy metal index being 146.32 (critical value = 6.0 and 100.0, respectively). Similarly, an increasing concentration of trace elements from west to north east along the Palar and Cheyyar river basins of South India was observed (Rajmohan and Elango 2005). These data highlighted the negative influence of urban activities on groundwater in these regions and made the groundwater unacceptance of for drinking. This scenario demands immediate enforcement of environmental laws and sustainable management of the pollution (Shankar 2019).

High amounts of Hg, As, Cd and Ni were reported from groundwater samples of highly industrialized areas of Thane, Maharashtra (Bhagure and Mirgane 2011). The rain water washing of the air borne and soil pollutants and contaminating the aquifers might be a probable cause for these contaminations. Interestingly, the residential areas around Kurichi Industrial Cluster of Coimbatore district from Tamil Nadu, declared as critically polluted region, showed non-detectable amounts of HM in groundwater. However, alarming presence of Arsenic in Singanallur, Gandhipuram and Selvapuram and Cadmium contamination in Singanallur and Kurichi Industrial cluster have been reported (Mohankumar et al. 2016).

In eastern India, from the state of West Bengal a large number of districts particularly Nadia, Murshidabad and Midnapur are highly exposed to heavy metals like As and Mn leading to numerous health problems for the population of eastern India (Bhowmik et al. 2015; Rahman et al. 2015; Sankar et al. 2014). An estimate of 150 million people are at high risk from As-contaminated groundwater in West Bengal and it's neighbourhood, Bangladesh with 26 million people from West Bengal itself (Bacquart et al. 2012; Chakraborti et al. 2009).

Hydrogeochemical studies from the Bhagirathi-Hooghly alluvial plains have revealed that the release of As from the soil into the groundwater is influenced by both the geomorphology and anthropogenic (i.e. unsewered sanitation) processes (Bhowmik et al. 2013). Oxidation of arsenic-bearing pyrite minerals, reductive dissolution of Fe(III)-oxyhydroxides and competitive exchange with phosphate ions are the three processes for releasing As from sediment to groundwater in West Bengal (Maity et al. 2011; Bose and Sharma 2002). The dissolved organic carbon (DOC) and Fe mineral phases influence the mobility of As in groundwater in West Bengal (Majumder et al. 2013). These observations led to the classification of the state into three zones as highly affected (9 districts in eastern side of Bhagirathi River), mildly affected (5 districts in northern part) and unaffected (5 districts in western part) As-areas and a total number of 107 blocks grouped as arsenic-affected (Roychowdhury 2010).

Besides West Bengal, in north-eastern regions like Assam, high amount of Al, As, Cu, Fe, Mn, Ni, Pb and Zn above the permissible limits have been reported in groundwater from various districts including Jorhat, Barpeta, Dhemaji, Kamrup and Tezpur (Mridul and Prasad 2013; Jain et al. 2018; Haloi et al. 2012; Buragohain et al. 2010; Patel et al. 2019). The observation of positive correlation between Fe and As in the Ganga–Brahmaputra alluvial plains of South Assam and Manipur provides a useful tool in groundwater planning and management in both the states (Gupta and Singh 2019).

In addition, the analyses of groundwater from different mining areas of India have identified HM (Al, As, Barium [Ba], Cr, Cu, Fe, Pb, Manganese [Mn], Ni, Selenium [Se] and Zn) with concentrations exceeding the risk threshold stipulated by World Health Organization (WHO) indicating carcinogenic risk (Tiwari et al. 2016; Tiwari and De Maio 2018; Singh et al. 2018; Giri et al. 2012). This is attributed to the discharge of wastewater from the coal mines along with the leaching of the HM from the fly ashes; and their admixture with the fresh water (Maiti et al. 2019; Prasad and Mondal 2009). For numerous groundwater samples in the capital Kolkata, the guideline values for Fe and Mn, exceeded as reported by McArthur et al. (2018). This was attributed to the Paris Green, an arsenical pesticide, manufactured between 1965 and 1985 and to the remnant of the pollution-plume stemming from a single factory site. In addition, Mn and Pb have been found in suspecting quality from the tube-wells of Kolkata.

Effects of heavy metal accumulation in aquatic systems

The bioaccumulation of non-biodegradable HM in the aquatic environment through food webs adversely affect the biota by getting transferred from soils to all the trophic

levels. In addition, the persistence of the HM in the aquatic environment for elongated period of time results in long-term risks to the living world even after the removal of the point sources; thereby forming a significant source for eco-toxicology (Babin-Fenske and Anand 2011). HM in aquatic environment forms particulates that sink from the surface to the bottom sediments; thereby affecting the aquatic organisms including microorganisms, the diatoms and the macro-phytes; all that reside in the lower sediments and alter their composition (Jaiswal et al. 2018). HM affect the microorganisms by reacting as redox catalysts in the production of reactive oxygen species (ROS) leading to malfunctioning of biomolecules; disrupting the enzyme activities and ion-regulation and by producing harmful and non-functional DNA and protein adducts (Liu et al. 2017).

The risk of metals entering a food chain is dependent on the mobilization of metals and therefore its availability in the soil where the metallic cations remain bound to negatively charged particles such as clay and organic matter (Rajakaruna and Boyd 2008). The metals enter the soil solution after detaching from these particles and accumulate in plants and other soil-organisms. The behaviour, physiology and diet of the animals determine the accumulation of the metals while the specificity of the metals define the effects of them on the particular species. However, the effects vary for the adapted organisms (metallophytes, hyperaccumulators and the metal-tolerant/resistant species) (Gall et al. 2015). The HM are categorized as essential (micronutrients) and non-essential classes on the basis of their nutritional requirement in the organisms. The former group of HM including Cu, Fe, Mn, Ni and Zn can be tolerated in little amounts but become toxic in excess while the nonessential metals such as Hg, Pb, Cd and As are not involved in normal biological functions and thus results in toxicity at a very fast rate. However, both the groups have a very high potency of being transferred from lower to higher trophic levels (Bourioug et al. 2015).

The consumption of drinking water contaminated with HM is the major source for human exposure to HM in addition to the consumption of crops grown in HM-contaminated environments. The major health concerns related to HM exposure includes cardiovascular disorders, respiratory problems, neuronal damage, hepatic and renal problems including proteinuria, skin corrosion and irritations, blood vessel damage, digestive problems, anaemia and risk of cancer and diabetes (Jaiswal et al. 2018). The basic mechanism involved for HM-toxicity is their accumulation in soft tissues if they remain in non-metabolized state and results in generation of reactive oxygen species (ROS) culminating into oxidative damage of the tissues (Rehman et al. 2018). In chronic incidents of toxicity, the symptoms appear after many years of exposure.

Of all HM, As is the most toxic due to it's high stability and solubility and thereby is easily absorbed by the

digestive tract, abdominal cavity and muscles in the human body. As drinking water is a major source of inorganic As; fish and meat are the chief sources for organic As and surprisingly sea foods contain several times more As than any other food-sources. Cd is a found as a complex in saewater; can rapidly cross biological membranes, bind to the ligands forming stable Cd-complexes. Due to this, Cd causes bioaccumulation in the fishes by either passive diffusion through the gills or through the marine food chain via the planktons and other marine microbes and thereby entering fish through the diet. Similarly, in a number of communities, the main exposure to Pb is through consumption of fishes. Besides Pb, Cd and Hg are transported to humans through fish-meat, processed fishes and other fish products (Bosch et al. 2016).

Organic Hg compounds are toxic as compared to inorganic due to their ready accumulation and high stability in the fish and human tissues. However, the latter can be converted to organic form through photomethylation or bacterial methylation. This methylmercury is the main toxic chemical form of Hg with a longer half-life; readily absorbed into the food chain; can pass through the cell membranes including blood–brain barrier; form a stable organometallic compound with a high affinity for the sulfhydryl groups of the amino acids and therefore is accumulated in the marine food chain. Thus, the fishes at higher trophic levels accumulate higher amount of Hg with a very high scale of biomagnification as opposed to As which is accumulated more in the lower trophic levels. HM stimulates the synthesis of specific enzymes in fish altering the metabolism resulting in cellular toxicity, necrosis and tissue death. These histopathological modifications produce irreparable changes in biota. The organs in the fish which are affected most include gills, muscles, digestive system and the brain. Bioavailable of Hg in the humans occurs through the muscular tissue of fish.

Besides fish, Cd has been shown to have negative impacts on the biochemical and physiological properties of several sea turtles. Dose-dependent exposure of Cd on the histopathology of liver tissues of Chinese three-keeled pond turtle, *Chinemys reevesii*, found in lakes, rivers and ponds in eastern Asia from Japan to southern China was found to induce ultrastructural changes in the mitochondria of liver (Huo et al. 2017). Studies have also shown the decreased success in the rate of fertilization and hatching of eggs in green sea turtle (*Chelonia mydas*) due to HM exposure in the aquatic systems (Sinaei and Bolouki 2017). Even low concentrations of HM have been detected in both blood and the eggs of the turtles that feed on algae and plants indicating the maternal transfer of HM. Therefore, the concentrations of HM are species, location and trophic level dependent which make the interpretation of their effects difficult; implicating the necessity of more research in the fields of trophic level disparities and spatial scale studies which will subsequently

establish links between aquatic pollution and it's effects on fish health and food safety for humans.

HM even adversely affect the macrophytes which aid to phytoremediation by inhibiting the synthesis of chlorophyll and reducing the production of sugar and proteins by increasing sugar consumption during metal stress and due to the formation of peptide-metal complexes (Sharma et al. 2015).

Phytoremediation to alleviate the heavy metal toxicity in aquatic systems

In the last two decades, exhaustive labours have been undertaken towards the (1) detection of heavy metal sources for water-pollution, (2) reduction of pollution sources and (3) alleviation of the polluted water resources. Alkaline precipitation, coagulation, electrochemical removal, evaporative recovery, filtration, ion exchange columns and membrane technologies, reverse osmosis as well as physicochemical methods including extraction, immobilization, stabilization and soil-washing are presently the most adopted technologies for the removal of HM (Ojuederie and Babalola 2017). However, these conventional techniques are non-economical and have negative effects on aquatic ecosystems. Therefore, the cost-effective, eco-friendly and environmentally sustainable strategy of phytoremediation has been a well-practised approach for the same (Praveen and Pandey 2019). This green remediation technology is one of the attractive research foci in modern times with respect to waste water treatment. Artificial wetlands (AW) is the most effective phytoremediation technique for treating wastewater. The removal of pollutants from aquatic systems by accumulating in plant biomass is called rhizofiltration (Rezania et al. 2016).

There are two mechanisms of phytoremediation: direct and explanta. In direct phytoremediation, the pollutants are absorbed through roots and are translocated to upper parts while in explanta phytoremediation (rhizoremediation), the pollutants are confined in the rhizosphere only where they are transformed/mobilized by the phytochemicals like carboxylates or microbial enzymes. HM phytoremediation occurs either through stabilization of the metals in the roots or by extraction of HM from the environment by roots and translocating to leaves and stems to be stored. The former plant species are called excluders while the latter are called hyperaccumulators. The hyperaccumulators have a very high tolerance for metals as greater than 10,000 mg/kg of Zn and Mn, 1000 mg/kg of Ni and Cu and 100 mg/kg of Cd in the leaves. A high density of plants with well-maintained strategies that would decrease the distance between the rhizosphere of neighbouring plants usually accelerate the phytoremediation. The plants themselves modulate the mobility of the pollutants by altering the pH, organic composition and redox potential of the environment. The plants usually follow any one of the following mechanisms of phytoremediation including phytostabilization/phytosequestration, phytoaccumulation/phytoextraction, phytovolatilization, phytofiltration and formation of hydraulic barriers (Sharma et al. 2015). (1) In phytostabilization, the pollutants are converted to a stable complex with the organic acids; precipitated in the root zone and sequestered through vacuoles; e.g., signal grass (*Brachiaria decumbens*) uses phytostabilization as a mechanism for phytoremediation. (2) Contrastingly, in phytoaccumulation the plant extracts the pollutants and translocates to aerial parts; and later on the pollutants are removed through harvesting the plants. Phytovolatilization is the process the pollutants are taken up from the soil and removed through evaporation; e.g., Chinese brake (*Pteris vittata*) absorbs As from the soil in the elemental form that gets converted into gaseous form by the metabolic processes within these plants and finally released into the atmosphere. (3) Phytofiltration is the process of absorption, concentration and precipitation of HM by the roots from the aqueous system; e.g., The macrophytes, floating plants and the ornamental plants such as pearl grass (*Micranthemum umbrosum*) remove HM by phytofiltration. During phytofiltration, the plants are grown in hydroponics and as they develop dense roots, they are being transferred to polluted areas. (4) Hydraulic barriers (e.g. vegetative caps and riparian buffer strips) control the transfer of the pollutants; leaching of the pollutants into the soil and check the soil erosion and usually the tall trees are involved in this process which can absorb huge volumes of water. The process of phytoremediation is also helped by the natural and synthetic chelating molecules which absorb, stabilize and degrade the HM. Among the natural chelating molecules which are synthesized by the plants in the stressed and toxic environment, siderophores which are produced in Fe-contaminated environments and chelate Fe, Zn and Cd atoms are the best-known HM natural chelating agents. Besides this, organic acid anions such as acetate, citrate, malate, oxalate, salicylate, succinate, tartrate and phthalate released as anions form chelates with HM and the membrane-bounded transport proteins transporting anions from the root surfaces helping in the HM detoxification. Peptide ligands and metallothioneins are also synthesized by the plants in response to HM toxicity.

At present, more than 400 plant species have been reported as metal hyperaccumulators, constituting less than 0.2% of all angiosperms (Dai et al. 2017). The role of *Brassica sp.* including *B. juncea* L., *B. juncea* L. Czern, *B. napus* L. *and B. rapa* L. species as moderate accumulators of Cd and Zn have been studied vividly in different systems including hydroponics (Mourato et al. 2015). *Thlaspi sp.*, *Arabidopsis sp.* and *Sedum alfredii sp.* are the most important plant species recognized as hyperaccumulators; of which

Thlaspi sp. can accumulate more than one HM (Bothe and Slomka 2017). Eg. *T. goesingense* and *T. ochroleucum* for Ni and Zn while *T. rotundifolium* for Pb in addition to Ni and Zn and *T. caerulescens* for all these three HM along with Cd. This significant tolerance and hyperaccumulation of HM by *Thlaspi* sp. are attributed to the root-architecture; presence of transport channels and carriers in the root membrane and organic ligands in the root exudates. In comparison to *Thlaspi* sp., where HM accumulation occurs in the vacuoles; for *Arabidopsis* HM accumulation is reported in the mesophyll tissues of the leaves. However, competition in the uptake of different HM and the concentration of accumulated HM in different parts of the plants very between species. *Alyssum serpyllifolium* sp. *lusitanicum* belonging to Brassicaceae has been also identified as an efficient species for phytoextraction of Cr and Cu in addition to Pb and Zn (Kidd and Monterroso 2005). As accumulation has been well reported from the various fern species including *Pteris* genus and Brake fern that can accumulate up to 7500 mg As/kg (Eze and Harvey 2018).

The seaweeds provide the most profitable and approachable material as biosorbent in the present day (Rezania et al. 2015). Among the various aquatic plant species, HM accumulation from the polluted water have been reported from calamus (*Lepironia articulata*), duck weed (*Lemna minor* L.), macrophyte (*Eiochhornia crassipes*), pennywort (*Hydrocotyle umbellate* L.), sharp dock (*Polygonum amphibium* L.), water dropwort [*Oenathe javanica* (BL) DC], hydrilla (*Hydrilla verticillata*), water hyacinth (*Eichhornia crassipes*) and water lettuce (*Pistia stratiotes*). The ability to tolerate HM without toxicity and accumulate them in the different parts for these plants have been well documented in hydroponics systems and pot-culture experiments (Sricoth et al. 2018). The duck weed was found to be a good accumulator of Cd, Selenium (Se) and Cu; a moderate accumulator of Cr; while water hyacinth removed Ag, Cd, Cr, Cu, Pb and Se from waste water and As, Cr and Hg in hydroponics. Hg and Pb were accumulated in high concentrations by water dropwort and calamus respectively. The duckweed (*Lemna gibba*) is regarded as a bio-indicator for HM as it transfers heavy metals from the polluted water to the plant and therefore can be used for monitoring the transfer of metal through different trophic levels (Sharma et al. 2015).

The hydrophytes including *Elsholtzia argyi*, *Elsholtzi splendens*, *Gladiolous* sp., *Isoetes taiwaneneses Dwvol* and *Echinodorus amazonicus* were found to be successful in remediation of Cu in different concentrations from hydroponics as well as waste water (Sun et al. 2016).

Due to uncontrollable growth, water hyacinth has been reported to be a challenging species globally but due to it's high nutrient absorbing capability it has been a model plant for phytoremediation (Rezania et al. 2015). In addition to it's use in waste water treatment plants, it has been used as a source for developing by products including animal and fish feeds, biogas, composting and power plant energy. A number of studies have shown the use of water hyacinth for removal of HM in the following rate as Cu > Zn > Ni > Pb > Cd from different water sources including wastewater from simulated wetland, synthetic waste water, composting wastewater, industrial wastewater and artificial waste water. The mechanism involves the uptake of HM by the roots of water hyacinth, translocation and accumulation of HM in the aerial parts. Therefore, harvesting the plants can permanently remove these HM from the water sources and the HM can be recovered from the ashes after burning the plants. Similarly, high phytoextraction capabilities for *Miscanthus* sp. which is a perennial bio-energy crop from tropical and sub-tropical countries showed effective absorption and translocation abilities of all HM (except As) at low concentrations indicating the potential of this plant to remove HM from marginally contaminated water-bodies (Bang et al. 2015). Submerged aquatic plants including *Potamogeton malaianus*, *Nymphoides peltata*, *Eichhornia crassipes* and *Hydrilla verticillate* were found to be potent accumulators for HM including Cu, Cr, Mn, Ni, Zn, Co, Pb, Titinium (Ti) and Vanadium (V) from moderately contaminated lakes (Bai et al. 2018). The seagrasses *Cymodocea nodosa* and *Posidonia oceanica* and the wetland macrophytes *Apium nodiflorum*, *Arundo donax*, *Nasturtium officinale*, *Phragmites australis* and *Typha domingensis* also showed the species-specific bioaccumulation and detoxification capacity. *Eichhornia crassipes* was found to be a dominating plant species from As-contaminated areas from West Bengal, India that could accumulate As (597 mg/kg) in the shoot along with *Lemna minor* and *Pistia stratiotes* which two accumulated As in the leaves and fronds (Singh et al. 2016). The *Lemna* sp., and *Myriophyllum aquaticum* exude oxalates and polyphenolic substances that form complexes with HM ions (Mkandawire 2013). In this respect, it is to be mentioned that the macrophytes can be used for HM removal in both living and dead states from the aquatic ecosytems; the processes being referred to as bioaccumulation and biosorption respectively for living and dead states (Sood et al. 2012).

Among the aquatic species, the algae including *Hydrodictyon reticulatum* (Microalgae), Diatoms (*Bacillariophyceae*), *Pithophora* sp. (*Cladophoraceae*), *Phormidium* sp. and *Oscillatoria* sp. (Cyanobacteria) were found to be dominant in As-contaminated area with *Hydrodictyon reticulatum* to be the most dominating plant species accumulating 430 mg/kg As. The maximum amount of As amounting to 760 mg/kg was accumulated by diatoms (Singh et al. 2016). This study indicated that as the macrophytes can be exploited for removing HM from aquatic systems while As-accumulating algae may provide a bioresource for elucidating algae-mediated As-detoxification and bioindication processes. Even the accumulation of algae in

the roots of paddy have reduced the exposure of the roots of the metal-contaminated sites; thereby decreased the As and HM uptake. Wetlands with grass (*Polygonum* sp.), reeds (*Phragmites australis*) and bulrush (*Typha latifolia*) have proved to be very efficient to improve the quality of the water of the streams receiving Uranium contamination from the mines (Sharma et al. 2018).

Microbial remediation to alleviate the heavy metal toxicity in aquatic systems

Microbial bioremediation is reliant on the metabolic capacities of the microorganisms to utilize environmental pollutants and convert them to non-hazardous forms by redox reactions. However, the microbial remediation is greatly influenced by the type species, screening conditions and the genetic constituents (Liu et al. 2017). On the other hand, the HM can also influence the microbial composition by nurturing the formation of new species; resulting in a competition with the indigenous flora. The bioremediation processes can be either in-situ or ex-situ. In-situ bioremediation process involves the facilitation of the growth of microbial population by providing nutrients in the contaminated sites or by introducing recombinant microbial strains in the polluted sites to utilize the toxic substances while in ex-situ bioremediation, the contaminated media is transferred to a site away from the original source for treatment (Rayu et al. 2012; Azubuike et al. 2016).

Due to the prevalence of multiple mechanisms of enduring HM in the microorganisms; they have been exploited widely to sequester and precipitate HM from the contaminated environments. There are four mechanisms utilized by microbes for HM remediation. (1) Enzymatic conversion of toxic metals to non-hazardous forms (bioreduction); (2) Modification of metabolic pathways to prevent uptake of metals; (3) Efflux of accumulated HM from intracellular components; (4) HM-quenching by anionic functional groups (COO^-, OH^-, PO_4^-, SO_4^-, $RCOO^-$) of the biomolecules on the cell wall surfaces; intracellular proteins (phytochelatins and metallothioneins) and siderophores. The presence of exopolysaccharides (EPS) in *Agrobacterium* sp., *Alcaligenes faecalis*, *Xanthomonas campestris*, *Bacillus* sp., *Zymomonas mobilis*, *Leuconostoc* sp., *Pseudomonas* sp. and *Acetobacter xylinum* aid to biosorption and biomineralization of HM. This is utilized for commercialization of EPS modified with anionicity to be used as biosorbent. Eg. alginate (*Pseudomonas aeruginosa*, *Azotobacter vinelandii*), galactopol (*Pseudomonas oleovorans*), fucopol (*Enterobacter sp. A47*), gellan (*Sphingomonas paucimobilis*), hyaluronan (*Pseudomonas aeruginosa*, *Pasteurella multocida*, *Streptococci* attenuated strains) and xanthan (*Xanthomonas campestris*). Due to this EPS which helps in the formation

of biofilms, the microbial consortia where the group of microbes function synergistically degrade HM more efficiently as compared to single strains. A mixture of *Viridibacillus arenosi* B-21, *Sporosarcina soli* B-22, *Enterobacter cloacae* KJ-46 and *E. cloacae* KJ-47 was found to be more efficient in bioremediation for Pb, Cd and Cu contaminated soil than individual bacterial culture grown for 48 h (Kang et al. 2016). Similar observations of higher Cu-absorption by a bacterial consortium was obtained from Cu-contaminated river in Brazil and was utilized to design fixed-bed bioreactor (Carpio et al. 2014).

The bacterial species that have been found to be efficient in absorbing HM including Cr, Cu, As, Zn and Cd belong to *Alpha-proteobacteria* (*Rhodobacter sphaeroides*), *Beta-proteobacteria* (*Achromobacter* sp., *Burkholderia* sp.), *Gamma-Proteobacteria* (*Pseudomonas veronii*, *Pseudomonas putida*, *Enterobacter alkaligenes*, *Enterococcus faecalis, Enterobacter cloacae, Stenotrophomonas maltophilia*), Actinobacteria (*Kocuria flava*; *Mycobacterium* sp.) and *Firmicutes* [*Bacillus cereus*, *Bacillus flexus*, *Bacillus licheniformis*, *Bacillus subtilis*, *Bacillus thuringiensis, Sporosarcina ginsengisoli*] (Mallick et al. 2018; Jebeli et al. 2017; Kamika and Momba 2013). Among these, *Rhodobacter sphaeroides* along with *Rhodospirillum rubrum* and *Rubrivivax gelatinosus* have revealed the existence of various mechanisms of resistance and/or tolerance to HM exposure (Kis et al. 2015).

Pseudomonas aeruginosa has been found to be efficient in the removal of cesium from waste water (Kang et al. 2017). Radioactivity resistant cyanobacteria, *Nostoc punctiforme* and *Chroococcidiopsis thermalis* associated with the removal of radionuclides (^{238}U and ^{226}Ra) and Cd were isolated from hot-springs (Heidari et al. 2018). Since, ^{238}U cannot be disintegrated into non-toxic small components, the bacterial bioremediation of this HM includes the transfer of ^{238}U from toxic to non-toxic forms and immobilizing the HM from the pathway. These are achieved by biosorption, biochelation, bioreduction, bioprecipitation and biomineralization by the production of extracellular polysaccharides (as *Pseudomonas* sp. and *Citrobacter* sp.) and low molecular weight organic acids including biogenic colloids. HM is internalized by metabolic pathways and is sequestered in the cell organelles like cell membrane, cell wall and vacuoles (Mkandawire 2013).

Actinobacteria, that includes phenotypically diverse microorganisms and exhibit varied metabolic and physiological properties including the production of secondary metabolites and therefore have been important candidates for bioremediation (Alvarez et al. 2017). About 35 genera and 10 orders from the class Actinobacteria have been found to be metal-tolerant. The species including *Propionibacterium freudenreichii* shermanii JS and *Bifidobacterium breve* Bbi99/E8, *Frankia* species, *Amycolatopsis* sp., *Lentzea* sp., *Saccharothrix* sp., *Corynebacterium* sp., *Mycobacterium*

sp., *Nocardia* sp., *Arthrobacter ramosus* and *Rhodococcus* sp. have shown high adaptation to HM indicating their role in HM remediation. Of these, *Streptomyces* sp. has been studied extensively for their use both in the form of metabolically active cells along with their products and dead cell forms for the HM remediation. Though the mechanisms have not been elucidated; biosorption, bioaccumulation, efflux of HM, reduction and biomineral production have been reported as plausible mechanisms for HM bioremediation in *Streptomyces* sp. It is noteworthy to mention that Actinobacteria are efficient to remove co-contamination of HM with other pollutants; which otherwise is difficult to remove as HM inhibit biodegradation of organic pollutants.

The fungi can tolerate and detoxify HM through active accumulation, adsorption into mycelia and spores, intracellular and extracellular precipitation and valence transformation, thereby acting as biocatalysts for the bioremediation of HM (Ojuederie and Babalola 2017). Eg. Yeast (*Saccharomyces cerevisiae*) is an efficient organism for biosorption of HM through the mechanism of ion exchange. Besides yeast, *Aspergillus versicolor*, *Aspergillus fumigatus*, *Acremonium* sp., *Fusarium flocciferum*, *Gloeophyllum sepiarium*, *Penicillium chrysogenum*, *Pleurotus ostreatus* sp., *Rhizopus oryzae*, *Trichoderma* sp. have been found to be efficient in absorbing Ni, Cu, Pb and Cr (Xu et al. 2015). Chitosan, a bioactive polymer, produced from the fungus, *Cunninghamella elegans* was found to be as an effective adsorbent of Cu^{2+}, Zn^{2+} and Pb^{2+} from the polluted water, with the adsorption ability being directly proportional to the concentration of the metals and thus could be recognized as a bioactive, eco-friendly and economically sustainable substance.

Algae produces the largest biomass with a high sorption capacity in comparison to other microbial biosorbents. Even the algae, biosorption efficiency through the same ion exchange mechanism of 15.3–84.6% was observed. Similarly, the brown marine algae can efficiently remediate Cd, Ni and Pb by bisorption through the chemical groups such as carboxyl, sulfonate, amino and sulfhydryl groups on their cell-surfaces. The different algae which have shown efficiency in degradation of Pb, Cu, Cr, Zn, Fe, Mn, As include *Spirogyra* sp., *Cladophora* sp., *Spirullina* sp., *Hydrodictylon* sp., *Oedogonium* sp. and *Rhizoclonium* sp. Marine and freshwater algae are capable to integrate U into aragonite ($CaCO_3$) mineral structures (Mkandawire 2013).

At present, sediment microbial fuel cells (SMFCs) are emerging as substitute remediation technique in HM-polluted aquatic systems by inserting anode electrode into the aquatic sediment allowing the indigenous microorganisms to generate electricity from sediment organic matter directly attaching to the anode electrode. SMFCs are utilized for remediation of HM in aquatic systems. Though, incorporation of SMFCs with the wetland plants to simultaneously remediate HM and generate electricity has not been studied exhaustively, addition of macrophytes to SMFC producing rhizodeposits and exudates producing sustainable bioelectricity as well as can accumulate HM; producing oxygen as a by-product for effective building of biocathode (Kabutey et al. 2019).

Conclusion and future challenges

HM are irretrievable, persistent and toxic pollutants of alarming environmental concern. Limitless use of HM to increase the productivity of the soil along with the disposal of industrial wastes into the aquatic systems consequently affecting the terrestrial and aquatic lives including humans is a major public health concern. The ill-effects on health are the consequences of dearth of knowledge, inadequate awareness of the problem and lack of proper management and regulation; thereby demanding for effective monitoring techniques to detect the areas and sources of HM, levels of HM-contamination and formulating methods to alleviate the contamination. To this end, phytoremediation and microbial bioremediation are advantageous over other conventional methods towards removal of HM from the environment due to cost-effectiveness and environmental sustainability. However, in spite of these methods being regarded as green approaches; few drawbacks have been incurred in regards to the disposal of the huge amount of wastes produced as an outcome of phytoremediation which is time-consuming and pain-staking. Therefore, immediate measures are to be taken to find out ways to remove the wastes. Secondly, in spite of significant progresses through experimental approaches, phytoremediation is regarded as an emerging green technology till date concerned to the metabolic functions and the growth patterns of the plants. Thirdly, there is a gap in efforts towards commercialization of the phytoremediation techniques due to the biomass disposal problem coupled with the seasonal growth of the aquatic plants (macrophytes) thereby calling for more experimental work to evaluate this process and beginning its application at commercial level. For example, an application of biomass disposal such as compost in metal-enriched soils could cause alterations in the soil microbial population by changing pH, decreasing the solubility of heavy metals, altering nutrient availability, and finally by increasing allochthonous microbial biomass. Furthermore, seasonal growth of aquatic macrophytes complicates the scenario as they contribute immensely in HM phytoremediation. Together, these limitations restrict transfer of phytoremediation technology from the lab to the field. To alleviate this shortcoming, eco-sustainable models may be developed along with the involvement of more regions for phytoremediation aiding to wetlands conservation. In this respect, genetic modification and bioprospecting

of threatened wetland plants may be considered as the vital targets in near future.

Author contributions AG conceived the idea. SH and AG wrote and revised the manuscript.

Compliance with ethical standards

Conflict of interest The authors declare that there is no conflict of interest.

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