Ecofriendly remediation technologies for wastewater contaminated with heavy metals with special focus on using water hyacinth and black tea wastes: a review

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Abstract Treatment of water contaminated with heavy metals is challenging. Heavy metals are non-degradable, persistent in the environment, have a high dispersion capacity by water, can bioaccumulate, and represent risks to human and environmental health. Conventional treatment methods have disadvantages; however, adsorption in biomass is a highly promising method with high efficiency and low cost that avoids many of the disadvantages of conventional methods. Black tea (BT) wastes and water hyacinth (WH) have attracted attention for their ability to remove heavy metals from wastewater. Utilizing these approaches can remove contaminants and effectively manage problematic invasive species and

wastes. The conventional uses of BT and WH were efficient for removing heavy metals from wastewater. Due to the unique and distinct properties and advantages of biochar and nano-forms of biosorbents, the use of BT and WH in these forms is promising to achieve sustainable heavy metals removal from wastewater. However, more study is needed to confirm preliminary results.

Keywords Polluted wastewater · Wastewater treatment · Wastes · Biochar

Introduction

One of the most concerning environmental issues today derives from the contamination of various environmental components (soil, water, and air) which triggers ecological and anthropological health hazards as a result of exposure to toxic levels of a variety of substances (Brevik et al., 2020; Gujre et al., 2021; Pujari & Kapoor, 2021). Industrial advances have been associated with an increase in the introduction of pollutants into the environment (Peng et al., 2020). Among these, water contamination by metals is noteworthy.

Heavy metals are non-degradable, persistent in the environment, show a high dispersal capacity by water, and a considerable bioaccumulation rate in plants (Geng et al., 2019; Ogunkunle et al., 2020), fish (Vieira et al., 2020), other animals, and humans (Shokr et al., 2016). Given the proper conditions, they can accumulate to

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levels that are toxic (Brevik et al., 2020). Their transfer via the food web to humans is a true risk; therefore, scientific and technological investigations targeted at finding solutions to this dilemma are very appropriate (da Silva Correia et al., 2018; Saroop & Tamchos, 2021). Heavy metals (HMs) usually reach the environment via several industrial activities, agricultural management practices, and inappropriate waste disposal (Brevik et al., 2020). Although humans, as well as all living organisms, need variable quantities of some HMs such as iron, zinc, copper, and chromium for suitable growth and development, these HMs can be toxic in high concentrations (Joseph et al., 2019). HMs can enter plant, animal, and human tissues through inhalation, consumption, and dermal contact, causing harmful effects (Zhou et al., 2018; Rehman et al., 2020). HMs are of rising interest due to their widespread contamination and toxicity to biota and particularly to the native microbial community, which plays a vital role in preserving ecosystems through nutrient cycling and contaminants removal (Grenni et al., 2019). Therefore, it is important and urgent to explore efficient and economical ways for HM remediation to protect the environment (Zhou et al., 2018; Kumar & Khan, 2021).

It is important for humanity to have a modern lifestyle, but people have also realized the importance of good health. If we want to live a healthy life, it is very important to have a balanced environment, because there is a very close relationship between the environment and human health (Brevik et al., 2019; Patil et al., 2019). Water is a natural resource that is essential for life on earth and upon which the life cycle and global biodiversity depend (Goher et al., 2019). The freshwater resource is required for almost all drinking and irrigation water needs; thus, the quality of water is a serious problem due to the expansion of industrial, agricultural, and leisure activities as well as poorly structured drainage and sewage systems (Abdel-Satar et al., 2017; Mokarram et al., 2020).

The world population is growing and has an impact on the environment. As the population grows, the demand for natural resources, including water, also grows. This means population growth has a direct effect on the quality of the water supply, and water problems have grown with the population (Patil et al., 2019). In addition, industrial and urban activities in developing countries have increased in recent years, which contributes to increased water pollution (Joseph et al., 2019). Global water scarcity is caused

by the physical scarcity of this resource and increasing water quality deterioration in many countries which reduces the amount of safe water available for use (Mateo-Sagasta et al., 2017; Pfister et al., 2020). For most people in developing countries, human health is often negatively influenced by the consumption of contaminated water (Adelodun et al., 2020). Also, in these countries, the effects of increased pollution are remarkably problematic as there is poor treatment of contaminated water (Joseph et al., 2019).

Unfortunately, this vital and important resource is subjected to many contaminants, including HMs, which are considered one of the riskiest contaminants resulting from population growth and urbanization. Their environmental release has been going on for a lengthy period and is a continuing problem that affects the health of billions (Mateo-Sagasta et al., 2017; Goher et al., 2019; Pandey & Tiwari, 2021). The separation of pollutants from waste streams has become a critical issue, and before releasing them into the environment, it is necessary to purify wastewater (Yap et al., 2017). Water pollution is mainly caused by the disposal of untreated wastewater by various industries (Nag et al., 2018). Therefore, water pollution is a worldwide challenge in both developed and developing countries (Mateo-Sagasta et al., 2017). As a result, interest has increased, and more comprehensive regulatory standards have been applied regarding the release of HMs and other pollutants into our natural environment (Yap et al., 2017; Cai et al., 2020).

Finally, according to World Health Organization (WHO), providing safe water, sanitation, hygiene (WASH), and waste management is essential to protect human health and prevent infectious and transmissible diseases outbreaks, such as the recent outbreak involving novel coronavirus disease (COVID-19) (Steffan et al., 2020; WHO, 2020). Thus, wastewater treatment is necessary, especially under COVID-19 conditions, where water shortage can present an obstacle in many countries for purification and cleansing purposes.

Although WH is not a new species used for HM removal, it exists in abundance in many countries and causes several ecological problems. In addition, new forms of WH and BT, such as biochar and nano-biochar, have unique and distinct properties (such as large surface area) which have proven efficient at removing HMs from aqueous solutions. However, these new forms are not well studied. Hence, because of this and the need to find more ecofriendly

materials to treat HM contaminated water, this review highlights the remediation of wastewater focusing on the use of black tea and water hyacinth in their different forms.

Wastewater pollution

Huge amounts of contaminated wastewater are released into the environment due to population growth, urbanization, and industrialization (Rezania et al., 2015), approximately $359.4 \times 10^9 \text{ m}^3 \text{ year}^{-1}$ (Jones et al., 2021). According to the United Nations, 80% of all industrial and urban wastewater in developing countries is released into the environment with no treatment (Joseph et al., 2019). In developing countries with water scarcity, wastewater is used for irrigation, which poses a health risk to farmers and consumers; at the same time, it provides nutrients for cultivated systems (Werner et al., 2018). Wastewater may include non-biodegradable HMs such as nickel (Ni), copper (Cu), cadmium (Cd), chromium (Cr), lead (Pb), arsenic (As), mercury (Hg), and zinc (Zn) and an appreciable amount of beneficial nutrients of agricultural importance (Turan et al., 2018). Therefore, environmentally friendly technologies to treat wastewater and allow use of the valuable nutrients are gaining attention among researchers around the world (Rezania et al., 2015).

During the past few decades, the term “heavy metals” has been extensively applied. It is often employed as a group label for metals and semi-metals (metalloids) that are related to contamination and potential toxicity or ecotoxicity. Legal regulations often establish a list of “HMs” to which they apply; these lists vary from country to country, and the term is sometimes used without even specifying which HMs are covered (Duffus, 2002). HMs are metallic elements with a high atomic weight and a density that is at a minimum five times higher than that of water. They are not biodegradable and cannot be broken down. They are therefore permanently hazardous materials (Paul, 2017; Goher et al., 2019). Increasing the hazard represented to and by water bodies is a major concern. HMs are considered one of the most critical contaminants in the environment because of their potential toxicity, persistence, and bioaccumulation. Joseph et al. (2019) added that HMs are of specific concern because of their toxic and carcinogenic

features, as well as their other harmful impacts on human health. HM pollution is furthermore a problem as many of the drinking water treatment practices in developing countries, involving chlorination, cooking, and solar sterilization, are ineffective at removing HMs (Joseph et al., 2019).

HMs come from a variety of sources; they can be natural due to weathering and erosion or the result of anthropogenic agents (Brevik & Burgess, 2015). Natural events such as atmospheric deposition and volcanic eruptions have contributed to HM pollution. Given the intense human activity that characterizes the modern world, the natural sources of HMs resulting from leaching and weathering of rocks in the environment are usually of little concern (Tchounwou et al., 2012; Paul, 2017; Elbehiry et al., 2019; Goher et al., 2019). HMs can additionally come into the environment by biogeo processes (Chowdhury et al., 2016). Despite approved regulations and updated legislation, metal ions are generally discharged as toxic contaminants in aqueous effluents from various metalworking, coal mining, chemical, electrolytic plating, and pharmaceutical operations (Humelnicu et al., 2020). Metallurgical industries, paints, pigments, coatings, fertilizers, and pesticides containing HMs, petroleum combustion, coal-fired power plants, electronics, microelectronics, galvanization, battery, pharmaceutical, chemical, plastics, fabric and electroplating industries, refineries, paper mills, nuclear power plants, and high voltage lines are also sources of HM pollution (Paul, 2017). In addition to industrial activities, the photo-engraving process, also known as metal engraving, photochemical processing, or chemical milling, can involve the risk of metal ions entering water sources in both different concentrations and combinations. Huge amounts of agricultural wastes are produced globally every year. A part of these agricultural wastes is directly cleared into water bodies, producing severe water contamination. Also, kitchen wastes introduced into untreated or under treated wastewaters may have several detrimental environmental effects, including the introduction of HMs. If the tolerance concentration of these metals is exceeded, they become dangerous to humans and other biological systems. Thus, water pollution by HMs is a serious problem in many developing countries as the qualities of drinking water and the aquatic environment are severely affected. It is, therefore, necessary to carefully monitor HMs and control

them at levels of concentration that comply with the environmental regulations established for each type of water source (Chowdhury et al., 2016; Paul, 2017; Goher et al., 2019; Elbasiouny et al., 2020; Humelnicu et al., 2020; Mostafa et al., 2020).

Based on European water legislation, the yearly mean environmental quality standards (AA-EQS) for inland surface water are < 0.25 and $1.2 \mu\text{g L}^{-1}$ for Cd and Pb, respectively. For human health protection, the highest allowable concentrations of Pb and Cd in natural water are 2.5–65 and $0.72\text{--}1.8 \mu\text{g L}^{-1}$, respectively. The World Health Organization guide values for Cd and Pb in drinking water are 0.003 and 0.01 mg L^{-1} , respectively (Yap et al., 2017; Elbehiry et al., 2018; Grenni et al., 2019).

Many government agencies have enforced strict rules and regulations as HMs are among the high priority pollutants and are becoming some of the most serious environmental problems (Karkra et al., 2017). Metals such as cobalt, copper, chromium, iron, magnesium, manganese, molybdenum, nickel, selenium, and zinc have been reported as essential nutrients needed for various biochemical and physiological functions. Other metals such as aluminum, antimony, barium, cadmium, gold, indium, lead, mercury, nickel, platinum, silver, strontium, tin, titanium, vanadium, and uranium have no recognized biological roles and are considered non-essential metals (Tchounwou et al., 2012). Many of these HMs are considered trace elements, which are present in small amounts (ppb to < 10 ppm) in different environmental matrices.

Heavy metal-induced toxicity and carcinogenicity include many physiological problems, some of which are not well understood. However, it is known that each metal has unique chemical and physical properties that give it ecological mechanisms of action (Tchounwou et al., 2012). Acute human exposure to high levels of HMs can cause dangerous and deadly diseases like cancer, Parkinson's, Alzheimer's, and anemia, as well as causing damage to the gastrointestinal, nervous, skeletal, and dermal systems (Brevik & Burgess, 2015; Goher et al., 2019). The disposals of municipal waste, untreated wastewater from various industries, and agrochemicals in open waters and rivers have reached an alarming level that continuously increases HM content and deteriorates water quality (Ali et al., 2016). Due to the abovementioned reasons, the growing demand for clean water in many parts of the world and the remediation of water pollutants has now become a critical issue (Yap et al., 2017).

Wastewater treatment

In contrast to organic pollutants, HMs are not biodegradable and can bioaccumulate through the food web causing various diseases. Therefore, HMs must be removed from water or soil to address pollution problems. One of the most important goals of remediation is to reduce the bioavailability of HMs and thus their accumulation and toxicity in plants and animals (Wang & Liu, 2018; Elbasiouny & Elbehiry, 2019). WHO has compiled a list of the top 10 chemicals of concern, which include many HMs: air pollution (not a chemical but on the WHO list), arsenic, asbestos, benzene, cadmium, dioxin, and dioxin-like substances, insufficient or excess fluoride, lead, mercury, and highly hazardous pesticides (WHO, 2011). Arsenic, cadmium, lead, and mercury are typically considered HMs. To recycle critical metals from industrially contaminated wastewater, they must be first removed from the aqueous solution (Vollprecht et al., 2019).

There is a need to develop methods that are inexpensive and result in less secondary waste generation. Microorganisms have been used as a biosorbent to remove HMs and organic compounds from wastewater. Both living and dead microbial cells are used to transform or adsorb HMs and their products and can be a highly efficient bioaccumulator (Gupta & Balomajumder, 2015). Many researchers have used inexpensive adsorbents from agricultural waste and obtained excellent adsorption capacity for removing HMs (Kumar & Chauhan, 2019). The development of environmentally friendly and efficient techniques for treating wastewater is one of the attractive modern research areas. Phytoremediation is a possible method of removing contaminants from wastewater and is known as a green remediation technology. Today, the goal is to seek a sustainable approach in the development of wastewater treatment capacity (Rezania et al., 2015).

Methods of heavy metals removal from wastewater

Traditional treatment methods vs ecofriendly methods

Emphasis has been placed on treating contaminated aqueous solutions before they reach natural water bodies (Ahmad et al., 2018). Treatment technologies to remove HMs from aqueous solutions include bioremediation, chemical precipitation, electrocoagulation, ion-exchange resins, membrane separation, adsorption,

reduction, evaporation, reverse osmosis, coagulation, flocculation, and extraction with solvents. Each of these methods has its advantages and disadvantages; however, most of them are expensive, inefficient when there is a need to remove HMs from large quantities of water, and have constraints such as high energy consumption, non-selectivity, and the use of chemical products with the corresponding need for appropriate disposal of the toxic wastes generated (Doula, 2006; Ahmad et al., 2018; Grenni et al., 2019). Among the various HM removal technologies, adsorption is considered the most promising given its high efficiency, ease of use, simple design, and low cost (da Silva Correia et al., 2018; Lin et al., 2019; Patil et al., 2019). Adsorption is referred to as a reliable modern technology and preferred by many industries. Materials including activated carbon, peat, fly ash, sewage sludge ash, zeolites, woody biomass, and other biomaterials have been reported to have a high ability to remove certain metals such as lead and cadmium from wastewater (Yap et al., 2017).

The HMs can be absorbed and immobilized by living organisms in aquatic systems and eventually accumulated in solid wastes, leaving purified aqueous solutions. Several submerged plants such as *Lemna gibba* (duckweed), *Veronica anagallis-aquatica* (blue water well), *Eichhornia crassipes* (water hyacinth), and *Ludwigia stolonifera* (water primrose) have been used to efficiently remove HMs from aqueous systems. The major advantages of this approach include the low cost and ease of processing; it simply requires that the plants grow naturally (Saleh et al., 2020). As a result, phytoremediation technology is increasingly being used to treat wastewater contaminated with HMs. The mechanisms of this process include removal of the dissolved element(s) from the aqueous medium through adsorption on the roots or translocation in the plant shoot; finally, the elements' stabilization occurs through physical and/or chemical interactions (Elbehiry et al., 2020; Saleh et al., 2020). Numerous terrestrial and aquatic plants have been investigated; in most instances, it is still necessary to assess their performance and efficacy under a variety of conditions. There is also a need to investigate unexamined plants for potential purification of aquatic environments (Elbehiry et al., 2020; Saleh et al., 2020).

Using low-cost adsorbents in wastewater treatment

Continued growth in this area requires improved performance with manageable costs to increase

industrial profit margins. Agricultural wastes have received special attention because of their wide and abundant availability around the world (Thines et al., 2017; El-Ramady et al., 2020). Low-cost adsorbents can be effective materials for treating contaminated wastewater. Although activated carbon, clays, alumina, zeolites, and solid wastes have been applied in adsorption for a long time because of their porous structures, the main problem limiting their application is the production of risky secondary solid wastes. On the other hand, many inexpensive adsorbents have a low adsorption capacity which requires research to modify their structure and improve their performance before they can be used in widespread applications (Pourrahim et al., 2020). The use of agricultural wastes as sorbents can also diminish environmental problems relevant to green wastes disposal and management. A huge variety of green wastes have been studied for HM sorption such as corn cobs, wheat bran, rice husks, tree leaves and barks, and aquatic weeds (Malakahmad et al., 2016).

Activated carbon is widely applied in wastewater treatment as an effective adsorbent (Grenni et al., 2019). It is amorphous with high porosity and a very large surface area which allows HMs or other molecules to be adsorbed (Tatarchuk et al., 2019). Nevertheless, there are some difficulties in its rejuvenation, and this makes it costly to treat contaminated water, which is a particular problem for developing countries. Therefore, low-cost materials with proficient adsorption capacities that are locally available are required. Natural resource conservation and the optimized use of non-renewable energies have motivated the recycling of organic wastes as an alternate to dumping and incineration. The discovery of new green remediation approaches utilizing plants or vegetable wastes is a growing issue. The development of new sorbents for HM removal from water, based on solid waste (e.g., plant biomass), may be a valuable alternative to reduce treatment expenses (Grenni et al., 2019).

Black tea and water hyacinth

Black tea wastes

Tea is one of the most common beverages in the world. Around 3.5 million tons of tea are consumed annually worldwide (Mohammed, 2012). Tea is an infusion

made from dehydrated green tea leaves or black tea powder. Black tea (BT) is mainly made from green tea leaves using the CTC (crush, tear, curl) method which creates a bulky, fibrous waste. Thousands of tons of BT waste are generated and disposed of in tea shops, restaurants, and houses every day. This issue does not stop with the waste generated after consumption and from the production process, and BT wastes are often disposed of in surrounding water bodies (Mohammed, 2012; Memić et al., 2014; Malakahmad et al., 2016; Indira et al., 2018; Hussain et al., 2018). In 2016, worldwide tea production was 5.73 million tons, with BT accounting for most of the overall tea production (65%) (Gao et al., 2021). In Turkey about 30,000 t of tea factory wastes are disposed of annually in the small bays near the Black Sea; in Malaysia, about 100,000 t are generated annually through the withering production process; and in India, 190,000 t of tea factory wastes are produced annually from the production of approximately 857,000 t of tea (Hussain et al., 2018). Thus, a large volume of spent tea leaves or tea leaf residue is discharged into the environment within daily tea consumption, including ready-to-drink fast teas (Mohammed, 2012; Memić et al., 2014; Malakahmad et al., 2016; Indira et al., 2018; Hussain et al., 2018). The two most widespread tea types (green and black) are differentiated based on the degree of fermentation. The green tea leaves are dried and heated after harvesting but not fermented, thus deactivating enzymes and impeding oxidative changes, whereas BT leaves are also fermented (Memic et al., 2014). Tea preparation waste should be distinguished from BT waste or spent tea waste that remains in tea houses and shops. Black tea is consumed by more people than green tea because of its low cost and plentiful availability (Indira et al., 2018).

Tea leaves have insoluble cell walls in addition to specific functional groups that are able to uptake pollutants; therefore, the tea leaves can potentially be used as pollutant scavengers in aqueous solutions (Weng et al., 2014). The main components of tea leaves are cellulose, hemicelluloses, tannins, lignin, and proteins, and the functional groups in them are essentially hydroxyl, aromatic carboxylate, amino, sulfonic, oxyl, and phenolic groups which enable the physicochemical interactions that sorb substances including HMs (Weng et al., 2014; Malakahmad et al., 2016). According to Hussain et al. (2018), BT has 37.2% cellulose, 14.7% lignin and structural

proteins, 12.1% hot water-insoluble proteins, and 9.6% hot water-soluble polysaccharides and proteins. The mean particle diameter of BT leaves is 0.38 mm, and mean surface area is $1.34 \text{ m}^2 \text{ g}^{-1}$ (Hossain et al., 2005). Weng et al. (2014) added that Fourier transform infrared spectrometer (FTIR) analysis showed that the chemical character of the cellulosic material in BT is an indicator of the presence of surface hydroxyl ($-\text{OH}$) and $\text{C}-\text{H}$ groups. The $-\text{C}=\text{C}$ groups carboxyl ($-\text{C}=\text{O}$) expand, which may be a result of the lignin aromatic $\text{C}-\text{C}$ bond.

Thus, the use of BT leaves as a low-cost sorbent is of interest. Some research has focused primarily on the conformational aspects of its removal or sorption capabilities. Others have been concerned about kinetics, an essential physicochemical factor in the evaluation of the fundamental sorbent quality, and the application of the adsorption process (Mohammed, 2012). There is some research on the use of factory tea wastes as adsorbents; however, there is limited work on the use of BT waste as a low-cost sorbent and its effectiveness at reducing the concentration of HMs (Malakahmad et al., 2016).

Hossain et al. (2005) reported that BT leaves are an attractive low-cost adsorbent for the removal of $\text{Cr}(\text{VI})$ as a result of their high adsorption capacity for this ion. Both adsorption and reduction processes are included in this removal, but the adsorption process is dominant at low pH levels (Hossain et al., 2005). Tea waste can bind considerable amounts of Cu and Pb from aqueous solutions, with the highest adsorption capacity at solution pH between 5 and 6 (Amarasinghe & Williams, 2007). In addition, the tea waste was a better adsorbent than coconut shell-based granular activated C. However, few studies are available about BT ecofriendly materials for HM adsorption from aqueous solutions; therefore, more studies are recommended in this area (Amarasinghe & Williams, 2007). Mozumder et al. (2008) investigated tea leaves waste as an adsorbent to remove $\text{Cr}(\text{VI})$ from aqueous solution. The adsorption of $\text{Cr}(\text{VI})$ on tea leaves was highly pH-dependent, where the removal percentage of $\text{Cr}(\text{VI})$ decreased from 97 to 10% with increasing pH from 1 to 5. An FTIR study suggested that amine functional groups were present on the tea leaves surfaces, which may participate in the metal binding (Mozumder et al., 2008). Weng et al. (2014) found that BT waste has a higher Cu adsorption capacity than activated C with

a maximum adsorption capacity of 43.18 mg l⁻¹ at pH 4.4. Tea waste has progressively gained interest as a cost-effective adsorbent for removal of several types of contaminants from aqueous solutions.

Kabir et al. (2021) used alkali-treated tea waste as a novel cost-effective natural adsorbent to remove chromium (Cr(VI)) from polluted water. They reported that, because of environmental complications caused by improper dumping of tea waste, recycling of these wastes can be a solution as a valuable alternative bio-material to scavenge HMs from aqueous solutions. The use of untreated tea wastes as adsorbents, on the other hand, might lead to a low adsorption capacity and increased effluent load. As a result, an appropriate modification procedure is necessary to enhance the chemical functions that make the BT waste an effective adsorbent. Alkaline treatment is one of the most widely used chemical modification procedures to increase the adsorption properties of organic wastes. The alkali modification treatment considerably alters the morphological, molecular, and supramolecular structure of cellulose, which can increase its physical stability, reactivity, and ion-exchange aptitude by adding functional groups like -OH, -COOH, and -C=O (Kabir et al., 2021).

Water hyacinth wastes

Water hyacinth (WH) (*Eichhornia crassipes*) belongs to the Pontederiaceae family and is amongst the most troubling aquatic plants globally (Rezania et al., 2015; Yu et al., 2018; da Silva Correia et al., 2018; Neris et al., 2019). Water hyacinth mainly originated in the American tropics and spread into all tropical regions (Priya & Selvan, 2017). It has been introduced in several countries as an ornamental plant because it has an appealing appearance (Kumar & Chauhan, 2019). The growth of WH on the water surface can decrease the amount of sunlight infiltrating the water which is critical to many photosynthetic organisms. As a result, WH slows the photosynthetic organisms' growth rate, disrupting the ecological equilibrium. Furthermore, its expanded coverage on the surface of water bodies limits oxygen transfer into the water. Water hyacinth has long roots suspended in the water; this root structure can provide proper media for the aerobic microorganisms that work in a sewage system. These microorganisms utilize organic materials and nutrients in the wastewater and transfer them into

inorganic substances that can be consumed by the plants. Usually, WH grows rapidly due to a lack of natural enemies or competitors in non-native countries. Up to two million WHs can found per hectare of water, corresponding to approximately 270–400 t of plant biomass. As an invasive species, it can become uncontrolled in places such as ponds or irrigation systems. In addition, it obstructs the navigation of rivers (Rezania et al., 2015; Yu et al., 2018; da Silva Correia et al., 2018; Carreño-Sayago, 2020). Thus, for these reasons and the eutrophication that results when the fungal and bacterial populations die off, many countries have focused on removal of WHs (Yu et al., 2018; da Silva Correia et al., 2018; Liang et al., 2018).

The removal of HM ions from aquatic ecosystems by particular aquatic plants is being investigated with increasing interest (Vlyssides et al., 2001). WH has the potential to remove aquatic pollutants (Rezania et al., 2015), which reduces its negative environmental impact (da Silva Correia et al., 2018). Du et al. (2020) reported that WH not only accumulates a wide range of HMs (i.e., non-selective uptake), but it also enhances their uptake and translocation in the more polluted areas, demonstrating that it has a strong ability for HM removal from water. Despite this, HM concentrations in the edible component of the WH (i.e., leaves) were well below the maximum tolerated values as an animal feed because of the low translocation factor into the leaves. The characteristics of WH (e.g., high growth rate, pollutant absorption effectiveness, low operating expenses, and renewability) make this plant a suitable material for phytoremediation in wastewater treatment, including industrial and domestic effluents (Rezania et al., 2015; Kumar & Chauhan, 2019). In addition, Priya and Selvan (2017) reported that WH is suitable for use in wastewater treatment due to its high biomass production rate, high pollution tolerance, and heavy metal and nutrient absorption abilities. The biomass of WH has an appreciable quantity of cellulose (35%) and hemicellulose (30%) which make it an alternative to treat water contaminated with HMs. Adsorbents are developed for HM removal based on the metals' interactions with the functional groups in the adsorbents. Thus, the presence of hydroxyl (OH), amino (NH₂), and carboxyl (C=O) groups in the cellulose enable the adsorption of different HMs by cation exchange. In laboratory experiments, dried and crushed WH biomass has

removed < 70% of the HMs in contaminated water (Carreño-Sayago, 2020). The oxygen of the OH group in the vegetable cellulose of WH has a pair of electrons that can be shared with the HM cations. One aspect of optimizing HM removal is through the chemical or physical transformation of WH biomass (Carreño-Sayago, 2020).

Zhu et al. (1999) demonstrated the potential of WH biomass for the phytoremediation of As(V), Cd(II), Cr(VI), Cu(II), Ni(II), and Se(VI) under controlled conditions. The highest accumulation was found for Cd and Cr, there were moderate levels of accumulation for Se and Cu, and the lowest level of accumulation was for As and Ni. They concluded that WH biomass had high bioconcentration factors for the studied elements when exposed to low concentrations of these elements. Lu et al. (2004) concluded that WH biomass was a moderate accumulator of Cd and Zn and can be beneficial to treat water contaminated with low levels of HMs. Kabata-Pendias and Mukherjee (2007) mentioned that WH has a strong ability to accumulate Pd. Mahamadi and Nharingo (2010) used WH biomass in a batch sorption experiment to study the biosorption of Cd^{2+} , Pb^{2+} , and Zn^{2+} ions in binary and ternary systems under acidic conditions ($\text{pH} = 4.84$) and a temperature of 30 °C. The mixed action of these metals was found to be incompatible following the order $\text{Pb}^{2+} > \text{Cd}^{2+} > \text{Zn}^{2+}$, while competitive biosorption behavior was recorded for the combinations Pb–Cd and Pb–Zn as described by the Langmuir competitive model. They concluded that Pb^{2+} ions could be efficiently eliminated from aqueous solution in the occurrence of Cd^{2+} and Zn^{2+} ions, but the elimination of the Cd^{2+} and Zn^{2+} ions would be repressed in the presence of Pb^{2+} . Neris et al. (2019) conducted kinetic and isotherm investigations of Pb^{2+} , Ni^{2+} , and Zn^{2+} ions adsorption in single and tri-element aqueous systems using modified alkali-treated WH biomass because of its higher ion adsorption capacity than acid-treated WH biomass and WH biomass washed with water. Analysis by FTIR confirmed that $-\text{OH}$ and $-\text{COO}^-$ groups were the key functional groups in Zn^{2+} adsorption, while $-\text{COO}^-$ groups were a key in Pb^{2+} and Ni^{2+} biosorption. The pseudo-second order was the best kinetic model that fit the single and tri-element experimental data for all investigated ions. The selectivity order of WH in single and tri-element systems was $\text{Pb}^{2+} > \text{Zn}^{2+} \geq \text{Ni}^{2+}$. Hemalatha et al. (2021) studied the

remediation Zn and Cr ions from electroplating industry wastewater by dried WH biomass (leaves, petiole, roots) through batch and column experiments. They considered influential parameters including biosorbent dosage, initial concentration of HMs, initial pH, and contact time. The WH root showed maximum Zn removal of 98.9%, and WH stem showed Cr removal of 96.4%. Thus, the results indicated that the dried WH biomass was a good and low-cost adsorbent.

Therefore, WH is a promising and potentially eco-friendly method of HM remediation from aqueous solutions. However, this needs more study because most of the previous research confirmed that surrounding conditions control the removal percentage, and this percentage differs among metals.

Black tea biochar

Biochar is a C-rich form of charcoal created by pyrolysis at high temperature (350 to 800 °C) under O-limited conditions (Liu et al., 2020). Biochar is composed of mineral ash and higher functional groups which makes it a potential environmental sorbent for ionic contaminants (Nyamunda et al., 2019). Gao et al. (2021) stated that, after pyrolysis treatment, BT is rich in C, O, N, and P. Pal and Maiti (2019) used biochar from BT waste (pyrolyzed at 300, 500, and 700 °C) as a potential amendment to immobilize Cd in a sediment-water meso-microcosm study. Ten percent BT biochar removed 67.7% of Cd, which increased to 87% when they added WH roots. Pal and Maiti (2019) used field emission scanning electron microscopy (FESEM) to study the morphology of BT biochar, showing a surface with a heterogeneous structure with the potential to adsorb Cd ions (Fig. 1). Khalil et al. (2020) used tea waste biochar to remove Cr (VI) from wastewater with 99.3% effectiveness (Table 1). They also used kinetic and isotherm modeling to show that the pseudo-second order and Langmuir (monolayer sorption) models offered the best sorption fit for Cr(VI) and the functional groups involved in this process were $-\text{OH}$, COO^- , and $-\text{NH}_2$, according to FTIR analysis.

Water hyacinth biochar

WH biochar has been used in many applications, such as adsorbing HMs, as a soil amendment, and for bio-energy production (Allam et al., 2020). Converting

Fig. 1 SEM micrographs of BT biochar **a** before and **b** after Cd adsorption (Pal & Maiti, 2019)

WH tissues into biochar to remediate wastewater could be a win-win strategy for both removing pollutants and the effective management of a highly problematical invasive species (Yu et al., 2018; da Silva Correia et al., 2018).

Based on SEM micrographs, WH biochar has a non-homogeneous, coarse, and porous surface that has many different (in size, shape, and depth) pores (Allam et al., 2020) (Fig. 2). The micropores in the surface of WH biochar are produced during the pyrolysis process when substances are volatilized; the volatiles are trapped in the biomass and expand its microstructure. WH biochar has displayed potential for HM adsorption from different types of wastewaters (Liu et al., 2020). The use of WH biochar for HM removal is the most recent and focused trend as a profitable pathway for the use of WH (Gaurav et al., 2020). The WH biochar generated at different pyrolysis temperatures had shown promise for high removal efficiency of HMs such as Cd with biochar created at 300, 500, and 700 °C (Li et al., 2016; Liu et al., 2020) and Cu^{+2} and Zn^{+2} with biochars created at 300 and 600 °C (Nyamunda et al., 2019). A significant challenge with the use of biochar in aqueous solutions is its low density in powdered form (Liu et al., 2020). Some researchers overcame this problem using modifications such as alginate capsules (Nyamunda et al., 2019) or magnetic modification (Liu et al., 2020). This is a promising way to sustainably utilize this invasive plant (Liu et al., 2020). Use with algae is another option to increase the efficiency of WH biochar (Shen et al., 2018). A complex of WH biochar-derived pellets immobilized

with *Chlorella* sp. was used in Cd bioremediation. The removal efficiency of Cd(II) by the complex was optimized by controlling numerous parameters, such as the pellet materials, algal culture age, and light intensity. The WH leaf biochar pellet was selected as the optimal carrier because the high surface hydrophilicity of biochar leads to high immobilization efficiency (Shen et al., 2018). In addition, Yu et al. (2018) used WH biochar supported by ZnO nanoparticles to enhance the removal of Cr(VI) from aqueous solution. They characterized the surface morphology of biochar with SEM, and the biochar surface obtained at 500 °C was relatively smooth; however, there were many heterogeneous pores and cavities that formed on the biochar surface at 700 °C. This resulted in the creation of more active adsorption sites for Cr(VI). Furthermore, the rough biochar surface increased the surface area, which is also useful for contaminant adsorption. They noticed that the specific surface areas and pore volumes of biochar obtained at 750 °C are larger than those created at 700 °C, and the Cr(VI) removal efficiency of biochar obtained at 750 °C is a little higher than that of 700 °C. They attributed this result to the narrow pore width (2.4 nm) of biochar obtained at 750 °C, which may reduce the probability of inner mass transfer.

Discussion and conclusions

The cost of remediation actions is a source of concern. Removing HMs using low-cost materials is

Table 1 Comparisons of heavy metals removal efficiency/adsorption capacity of water hyacinth and black tea waste

Metal(s)	Removal efficiency/sorption capacity	Material(s)	Reference
Cd and Cu	Removal efficiency was 15.9% for Cu and 91.4% for Cd	Water hyacinth roots	Zheng et al. (2016)
Cr	Removal efficiency ranged from 25–72% to 35–96%	Water hyacinth	Bandara et al. (2020)
Cr	Removed 80% from standard solution and 99% from water after 20 days at 0.5 ppm	Water hyacinth	Saha et al. (2017)
As	Water hyacinth roots removed up to 50 mg As g ⁻¹ after exposure of 1500 mg As l ⁻¹ to 30 g of dried roots over a 24-h time period.	Water hyacinth roots	Al Rmalli et al. (2005)
As	Maximum removal was 37% with 0.1 g of biomasses and an As(V) concentration of 0.1 mg/L at pH 6	Water hyacinth	Romero-Guzmán et al. (2013)
Cd, Cu, Pb, and Zn	Removed 61% of Cd, 59% of Cu, 49% of Pb, and 42% of Zn; removal efficiencies were 98% for Cd, 99% for Cu, 98% for Pb, and 84% for Zn	Water hyacinth	Sekomo et al. (2012)
Cr	Maximum removal was 13.05 mg/g, 26.74 mg/g, and 39.21 mg/g for stems, roots, and leaves, respectively, at pH 1; removal efficiencies of 31.5%, 40%, and 34%, respectively	Water hyacinth	Aggarwal and Arora (2020)
Cu	Removal efficiency from an aqueous solution containing various concentrations of Cu (1.5, 2.5, and 5.5 mg/L) was 97.3% over 21 days	Water hyacinth	Mokhtar et al. (2011)
Cd and Cu	Removal of Cd and Cu ranged from 87.69 to 95.59% and 6.67 to 35.48% in pharmaceutical wastewater. Average bioconcentration factors for Cd and Cu were 583.83 and 734.41, respectively.	Water hyacinth	Ajayi and Ogunbayo (2012)
Cr, Cu, and Zn	Maximum reduction of 94.78% for Cr, 96.88% for Zn, and 94.44% for Cu	Water hyacinth	Mahmood et al. (2005)
Cd	Maximum Cd(II) removal efficiency of 92.45%. Immobilization efficiency was 89.3%, and bioaccumulation capacity was 13.81 mg g ⁻¹	Water hyacinth with algal cells	Shen et al. (2018)
Cu	The sorption capacity of Cu was 22.7mg g ⁻¹ at an initial pH of 5.5.	Water hyacinth roots	Zheng et al. (2009)
Cr	The removal efficiency was 95% of Cr(VI) from a solution at neutral pH.	Water hyacinth biochar+30% Zno	Yu et al. (2018)
Cr	Maximum removal of 99.3% of Cr(VI); 197.5 mg g ⁻¹ Cr at 120 mg L ⁻¹ initial Cr concentration	Tea waste biochar	Khalil et al. (2020)
F	The fluoride removal efficiency was 91.79% and 98.29% for TWB and CMB, respectively	Tea waste biochar and chemically modified biochar	Roy et al. (2018)
Pb	Removal rate for alkali treated green tea (GT) was 98.54%, GT was 64.18%, and GT residue was 70.73% at 50 mg/L of Pb	Alkali treated GT residue, GT, and GT residue	Yang and Cui (2013)
Pb and Cd	Adsorption efficiency for Pb was 125 mg g ⁻¹ (72%) and Cd 142.9 mg g ⁻¹ (72.14%) at pH 8	Residual tea waste	Joshi et al. (2020)

Table 1 (continued)

Metal(s)	Removal efficiency/sorption capacity	Material(s)	Reference
Cd, Co, and Zn	Adsorption capacity for metals was 15.39 mg g ⁻¹ for Co, 13.77 mg g ⁻¹ for Cd, and 12.24 mg g ⁻¹ for Zn	Black tea waste	Mohammed (2012)
Ni and Zn	Maximum capacity of 90.91 mg Ni/g and 166.67 mg Zn/g at pH 5, contact time 250 min, and 20 g L ⁻¹ adsorbent concentration	Black tea waste	Malakahmad et al. (2016)
Cd, Pb, and Ni	94–100% removal for Pb, 86% for Ni, and 77% for Cd	Tea waste	Mahvi et al. (2005)
As	Optimum arsenic removal was 92.5% at pH 7	Tea waste	Shaikh et al. (2011)

promising in practice (Kabata-Pendias, 2011). Many studies have demonstrated the use of low-cost materials (such as BT wastes) and aquatic plants (such as WH) for HM removal from contaminated wastewater (Table 1).

The efficiency of using WH and BT for the removal of HMs from aqueous solutions varied widely among the studied metals, but overall, the removal efficiency tended to be high (Table 1). In the case of WH, Carvajal-Flórez and Cardona-Gallo (2019) attributed this variation to its rapid growth rate, high biomass content, and dense root system. Gaurav et al. (2020) reported that WH does not only contain cellulose, hemicellulose, and lignin in different proportions than biomasses such as rice straw, sugarcane bagasse, and wood but has other properties such as being C-rich and porous, with the ability to hold high levels of water and a high specific surface area. In addition, the abovementioned functional groups are also present in BT (see “Wastewater pollution”) and have a high potential to adsorb HMs from the environment (Pal & Maiti, 2019).

The use of biochars has received increasing interest as a low-cost and ecofriendly amendment and remediation tool (El-Bassi et al., 2020). The properties of WH and BT biochar and nano-biochar created by pyrolysis at 500 °C for 4 h (nano-biochar was ground in a ceramic mortar to reach to nano-size) are shown in transmission electron microscopy (TEM) images in Figs. 3, 4, and 5. TEM showed the connected pores on the surfaces of WH and BT biochar and nano-biochar. Particle size has a critical role in the treatment of BT raw material, and particle size distribution also affects the adsorbent surface area; the smaller the particle size, the larger the surface area for adsorption on a

per mass basis (Hussain et al., 2018). This structure is responsible for ion-exchange behavior which enhances the potential to use BT as a metal scavenger from wastewater and other solutions (Hussain et al., 2018).

Due to its unique structure and many excellent physicochemical properties, tea waste has been used extensively to remove a wide variety of environmental pollutants such as organic pollutants and HMs (Li et al., 2015). Pal and Maiti (2019) reported that physical changes occur in BT surface area and on surface pores when it is converted into biochar, which might cause dispersion of metal ions through these surface pores. The presence of abundant micro-meso, meso-, and macro-pores on BT surfaces indicated strong potential for metal adsorption. In addition, Pal and Maiti (2019) stated that the biomass type, pyrolytic temperature, furnace type, and residence time determined the final surface area and surface functional groups in BT biochar.

The studies included in this review and in Table 1 used WH or BT as raw or modified material or in biochar form. Studies on modification of these sorbents to the nano-form are very scarce. Among those that do exist, Maheswari et al. (2020) used surface-modified nano-WH and duck weed (DW) to remove Pb(II) from aqueous solution. Their results showed that WH had a higher removal efficiency (98%) than DW at pH 5.3 and an adsorbent dosage of 0.5 g L⁻¹ at 20 g L⁻¹ metal ion concentration. Investigation of modified nano-WH and DW by energy-dispersive X-ray spectroscopy (EDX), X-ray diffraction (XRD), Brunner-Emmett-Teller (BET) with mapping images, and thermogravimetric analysis (TGA) confirmed the high pore density of these nano-sorbents. The surface area of WH (261 m² g⁻¹) was higher than M-DW (192 m²

Fig. 2 SEM micrographs of WH biochar surfaces at several magnifications (Allam et al., 2020)

g^{-1}). It was also confirmed that the surface modification successfully increased the cellulose fiber crystallinity as a result of the elimination of hemicellulose

and lignin. This high crystallinity of the cellulose fiber might increase the domain size of these modified adsorbents. Thus, these changes are responsible

Fig. 3 TEM micrographs of biochar surfaces created with black tea wastes at 500 °C

for improving the WH efficiency in removing Pb(II) from aqueous solution.

Due to their physicochemical properties nano-materials, including nano-sorbents and nano-membranes, they are

attractive for a wide variety of uses in many fields including wastewater treatment (Samaddar et al., 2018). These promising bioremediation methods have many advantages such as low-cost, efficient sorption, and low

Fig. 4 TEM micrographs of biochar surfaces created with water hyacinth wastes at 500 °C

Fig. 5 TEM micrographs of **a** black tea nano-biochar and **b** water hyacinth nano-biochar surfaces. Both biochars were created at 500 °C

energy consumption that may allow sustainable wastewater treatment to be achieved. However, a better understanding of how to most effectively use such materials requires additional studies that investigate HM removal under different conditions and an expansion of these studies to include more metals. Thus, studies are recommended to evaluate the potential ability of WH or BT biochar and nano-biochar forms to eliminate HMs from wastewater. Finding ways to do this would be especially beneficial in developing countries, where the resources to adequately address water pollution issues do not currently exist.

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Declarations

Conflict of interest The authors declare no competing interests.

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