REVIEW ARTICLE

Current Status and Development of Remediation for Heavy Metals in China

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Abstract: At present, the problem of heavy metal pollution is a hot topic in the world. There are significant differences in the types and concentrations of metal ions distributed in each contaminated sites. In China, due to the vast territory and diverse ecoenvironments, the pollution situation is complex and variable, and the composite pollution is particularly obvious. Overall, pollution in the southern provinces is relatively higher than in other provinces, and Cd, Hg, Pb, Cr, As and Ni are listed as the priority pollutants for control. The metals have different physical or chemical specificity that allows them to be treated differently. Toxic Cr(VI) needs to be reduced to non-toxic Cr(III) before removal, whereas Cd(II) can form an insoluble Cd compound precipitate under alkaline conditions. Nevertheless, the characteristics of the soil itself such as pH, humidity, mineral composition, *etc*., are the hurdles in the process of remediation. Therefore, this review systematically summarizes the characteristics of heavy metal contaminated soil in major areas of China. It also proposes appropriate restoration methods and schemes such as phytoremediation and microbial remediation, which provides a theoretical basis for the elimination of heavy metals from a polluted land.
Keywords: Heavy metal, soil pollution, remediation techniques

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1 Introduction

The problem of heavy metal pollution has attracted comprehensive attention since past several years worldwide (Hu et al., 2014). Due to the differences of geographical location, living and production modes, the levels and types of heavy metal contamination in different regions show significant differences (Chen et al., 2015). In China (Fig. 1), the areas affected by long-term mining activities result in the accumulation of various metal ions near the mining areas, considering Cd as the severe contaminant (Khan et al., 2008; Pan and Li, 2016). In farmland soil, wastewater elution, irrigation, improper and excessive employment of pesticides and fertilizers are the main factors causing the accumulation of heavy metals in the fields (Sungur et al., 2014). Cd, Hg, and As occurring in soil can easily accumulate in vegetables (Li et al., 2014; Zhang et al., 2018). Researchers assessed the levels of heavy metals in the surface lands and street dust of major roads in large cities and found that Hg, Zn and Cu existed in high concentration, followed by Cd and Pb (Wei et al., 2015; Bi et al., 2018). The analysis of heavy metals in the soils of the Pearl River estuary and the Yellow River estuary showed that the average content of Cu, Zn, Cr and Pb in the sediment samples exceeded the standard value (Yao et al., 2015; Liang et al., 2016). In addition, heavy metal contamination in northwestern Aibi Lake showed exceeded levels of Pb and Ni (Zhao et al., 2018).

Different heavy metals exhibit different physical or chemical properties. It has been reported in the literature that for Cu(II) in water, chemical reduction may be the most important mechanism leading to the immobilization of Cu(II), rather than by precipitation of hydroxide [$Ca(OH)_2$] method to remove Cu(II) (Li et al., 2017). Since Pb(II) is a positively charged metal cation, it is possible to electrostatically adsorb lead ions using a negatively charged material on the surface (Ren et al., 2016). The main principle for the removal of Cr(VI) is reduction and adsorption. The reductant reduces Cr(VI) to Cr(III) that can be removed both

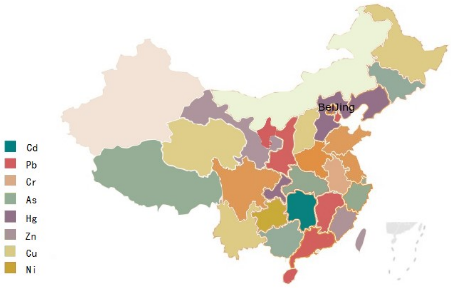

Figure 1. Distribution of Cd, Pb, Cr, As, Hg, Zn, Cu and Ni in various provinces of China.

by adsorption (Zhang et al., 2015; Qian et al., 2016). Cd can form an insoluble Cd compound precipitate under alkaline conditions or could be exchanged with an exchangeable alkaline earth element such as Ca to separate Cd (Chen et al., 2015). Hg(II) can be bound by chelating and then electrostatically attracted to surface-rich functional groups (Cui et al., 2015). The volatile metals such as Zn can be effectively removed by calcining at high temperature (Li et al., 2015). Ag^{+} can be removed and recovered by porous and efficient adsorbent (Wang and Wang, 2016).

So far, numerous methods have been developed to remediate heavy metals. Compost can serve as a soil amendment to reduce heavy metal pollution. However, it can only immobilize the heavy metals in soils, reducing the absorption and utilization by plants, while heavy metals remain in the soil. With the passage of time, serious secondary pollution may occur under environmental changes and the decomposition of organic matter (Huang et al., 2016). The electrokinetic method extracts metal ions from the soil matrix by applying an electric field of appropriate strength on the land. Nevertheless, the complicated applications, energy-extensive consumption, the need to consume a large amount of chemical reagents and the leaching solution is quite expensive, and thus is rarely applied in practice (Rosestolato et al., 2015). There are experimental biochars to fix Cd, Cu, Pb and Zn in polluted region that have shown encouraging results. The effectiveness of biochar on the other hand, is affected by the pyrolysis temperature, and the impact of current application rates and particle size pairs is still unclear and needs to be validated by long-term field trials (Lu et al., 2017). In last several years, the concept of phytoremediation has also been proposed. For example, *Psyllium* can be effectively used as a biological indicator for Cd and Sr in soil (Galal and Shehata, 2015). Additionally, *Spartina maritima* has been proven to be useful in phytoremediation, and it has great capacity to enrich heavy metals in its roots and rhizosphere. However, at present its accumulation mechanism for heavy metals is still unclear. The presence of excessive metals in the soil strongly inhibits seed germination, retarding plant growth and biomass, thus making plant restoration inefficient (Mesa et al., 2015; Sobariu et al., 2017). Many investigators have also begun to research the use of bacteria as a more effective aid to phytoremediation in order to reduce plant stress in the presence of contaminants. It has recently been discovered that selected bacterial consortia separated from the rhizosphere of *Spartina maritima* can improve plant growth responses and tolerance to metals. However, the response of bacterial-plant interactions is still poorly understood. The metal ions could disturb the natural population of microorganisms, which interrupts the bacterial species in charge of nutrient cycling with a succeeding opposite force on ecosystem functioning (Paredes-Paliz et al., 2016).

The 9th Conference on Metal Toxicity and Carcinogenesis held in 2016, discussed current and future studies on the effects of heavy metal exposure on human disease and health complications, showing that metals are an important public health issue leading to diseases (Wise et al., 2017). Nonetheless, as can be seen from the above content, different heavy metals have different physical or chemical properties. Also, the distribution of heavy metals in polluted areas is complex, so it is necessary to consider the peculiarities of metal ions and soils to carry out remediation. Many existing studies have focused on the exposure of single heavy metal pollution. The existing technologies are not suitable for repairing multiple heavy metal pollution points at the same time, which has the potential risk of secondary pollution, and large-scale use is also limited.

2 Characteristics of Soil Heavy Metal Contamination

2.1 Yangtze River Basin, Pearl River Delta and Coastal Cities

Soil heavy metal contamination varies from region to region in China (Table1). In general, the degree of soil heavy metal pollution in southern regions is higher than in other districts of China. The Pearl River Delta (PRD) has become a vital region for farming, commerce and manufacture progress over the past thirty years. Except for rocky decay, atmospheric sedimentation and ore resource are the crude imports of heavy metals in aquatic ecosystem. Electroplating factory, a textile plant, marine antifouling paint, wastewater irrigation, and exhaust emission are the dominant reasons of heavy metal contamination in sediment (Leung et al., 2014). The anthropogenic perturbations release waste into the aqueous ecosystem causing heavy metal pollution of water as a result of socio-economic development and urbanization (Xiao et al., 2013). The PRD has become a large-scale heavy metal reservoir. On account of topographic cause, the contamination condition in coastland is more serious than high seas because of intensive anthropogenic activity. Poisonous metal ultimately amasses in fish, that can bioaccumulate in humans causing health hazards if they are long-term eaten by consumers. Levels of Cu, Cr, Pb and Zn were 46.8, 87.6, 47.9 and 140 mg kg^{-1} that exceeded international

Table 1. A summary of mean concentrations of heavy metals in soils at different regions of China (mg/kg)

Region		Cr	Cd	Pb	Ni	Zn	Cu	Hg	As
Yangtze River Basin, Pearl River Delta and Coastal Cities	(Cai et al. 2015)	78.87	0.6	-	33.45	-	-	0.38	16.8
	(Bi et al. 2018)	-	0.17	27	-	131.9	27.8	0.5	7.8
	(Zhang et al. 2018)	-	-	31.2	14.7	-	16.6	0.13	12
	(Huang et al. 2018)	55.1	0.25	22.56	48.47	57.02	25.41	-	-
Mining areas	(Lü et al. 2018)	80.4	12.8	368.5	-	369.6	34.4	0.12	7.9
	(Li et al. 2015)	104	12.8	712	75	1688	239	0.6	71.7
Old industrial areas and developed metropolises	(Han et al. 2018)	112	0.66	50	52	210	160	0.38	6.5
	(Chai et al. 2015)	35	0.072	18.3	15.2	35	16.7	0.014	7.2
	(Lu et al. 2012)	-	0.136	20.4	-	69.8	22.4	0.073	7.85
	(Guan et al. 2018)	97.51	-	5.54	47.42	75.34	35.2	-	-
Plateau and Mountain areas	(Guan et al. 2018)	94.8	-	-	39.9	57.3	37.2	-	-
	(Guo et al. 2015)	31.37	0.08	16.71	15.61	24.53	14.7	-	-

standards greatly in the samples from embouchure and ambient littoral district settlings. Investigations revealed that a large proportion of heavy metals come from the human interruptions in the PRD region (Jin et al., 2014). In the middle reaches of Yangtze River, where harbors the technical and economic core of Hubei, has suffered serious heavy metal pollution over the past two decades. The content of As, Cd, Cr, Cu, Ni, Pb and Zn exceeding safety limits in Hubei; among them, carbonate-bound Cd and exchangeable Cd held 40.2% and 30.5% of total Cd, respectively, indicating that Cd has a high eco-risk in compounds (Wang et al., 2011).

Most areas of China such as Tianjin, Shandong, Shanghai and Guangdong are coastal cities that are dominated by human activities and man-made pollutants. Over the past decades, with the emission of industrial and domestic sewage and the abandonment of aquaculture and e-waste, nocuous pollutants including organics and heavy metals have augmented in environment. This has put a pressure on coastal and estuarial environments that has caused harmful effects on human health, wildlife habitats and ecosystem. Qingdao is an imperative port and auld industrial city where the industrial area is principally seated in the east coast of Jiaozhou Bay. Jiaozhou Bay discharges about 23.0 tons of wastewater annually, of which 40% comes from industry. In the eastern part of Jiaozhou Bay, the level of Cu, Zn, and Pb pollution was found to be moderate, but the contamination of Zn, Ni and Cd were serious in the rivers nearby (Deng et al., 2010). The increased level of metal contamination in the coastal environment may increase the risk of water and seafood, and the latter cause heavy metal poisoning if being eaten by humans. Domestic demand for seafood averages about 25 kilograms per person. Meanwhile, China is the world's fundamental exporter of marine products, and its export value increased by 47% between 2002 and 2004. Shellfish exported to other countries in 2002 was calculated at 9.6 million tons. Since the tragedies of Hg and Cd poisoning in Japan in the 1950s and 1960s, people have been concerned about metal pollution in the aquatic environment. Although fresh water system pollution incidents caused by industrial emissions occur frequently in China, it is generally believed that the marine environment seems as fewer influenced by contaminant on account of stronger attenuation ability of littoral sites. Nevertheless, with the decline of river drainage in current years, self-purification capacity of rivers may be reduced and heavy metals deposited in riverbed sediments may seep into groundwater. In addition, with the discharge of domestic sewage and industrial waste, metals may be transported into the sea through rivers and eventually amass in littoral districts. China's coastal and estuarial environments are confronting with rising pressure of heavy metal contaminants (Pan and Wang, 2012).

2.2 Mining Areas

The mining area in China is the most ample in Hunan Province where abundant mineral resources including Pb, Zn, As, Sb, Hg, coal and gold ores are found. Furthermore, there are more than 140 mineral deposits and exceeds 2700 years of mining history (Ma et al., 2015); it also has a reputation as the hometown of non-ferrous metals. In Hunan, Chenzhou is one of China's major Pb-Zn mine foundations and has a history of more than 1,000 years of metal mining. Also, Lengshuijiang possesses the world's greatest Sb ore containing the vast mining companies, smelters, refineries, and processing plants. Mining is considered the major source of heavy metals in the surrounding circumstance (Chen et al., 2017). Inadequate management, mineral extraction, slag treatment, heavy rainfall, and severe soil erosion may exacerbate shallow water and agricultural soil pollution in neighboring areas. This may cause heavy metals to migrate into drains, then posing potential health risks to local residents. Consumption of heavy metal-polluted crops will severely deplete iron, ascorbic acid and else essential nutrition in an organism. This leads to immunologic defense, intrauterine growth restriction and psychosocial function, damage and other disabilities related to malnutrition. Rice is a primary staple-diet for about 60% of the population and the main crop in China; the yield of rice is also relatively high in Hunan Province. At the same time, China is the topmost producer and consumer of rice and its output account for 30% of total rice production overall, which supports most of the world's food needs, especially in Asia (Wei et al., 2015). However, 20 million hectares occupy about 20% of the total area in farming that is heavily polluted by heavy metals and the rate is growing at 46700 hectares per year. Cd was detected in autochthonic rice. Natives who obtained Cd poisoning by intake of toxic rice had spine and legs hurt accompanied by a cough, anemia, renal insufficiency and result in death. "Cd rice" incident occurred in 2014 at Youxian that triggered significant social panics, and food security problems caused by heavy metal pollution that have aroused wide focus since the accident. According to a research, the mean daily intake of Cd through rice is 179.9 mg/day/person, which outdistance the permissible limit stated by the World Health Organization (Fan et al. 2017). Besides, vast Hg appended during processing of gold extraction leads to soil contamination around mines (Jiang et al., 2014). It has been reported that the average concentration of Hg in upper Miyun Reservoir reaches 30 fold preceding the background concentration. Miniature gold mining actions are chiefly handmade, low-tech and the rejects of ores are randomly emitted into water and soil, followed by air-slake and release of poisonous metals into the environment that are ultimately absorbed by animals and plants. Meanwhile, Pb may serve as an abandoned by-product of mining, along with Cu and As, which are released from ores combined with gold deposits. Cd exhibits a symbiotic connection with Pb and Cu as well. Elements of waste minerals can be released into the ground when they undergo weathering or leaching. The amount of heavy metals in the adjacent of abandoned oil and gas reservoirs is high, because the distance decreases when diggings increase. Soil contamination is on the rise due to retention, imperceptibility, and long-half-life of heavy metals. Their contaminations are centralized in south central, southwest regions and east coast developed areas due to Pb/Zn ore digging and processing, while less amount of fouling lies in northwest districts and Inner Mongolia (Zhang et al., 2013). Therefore, management should be paid attention after the production is stopped (Liang et al., 2014).

2.3 Old Industrial Areas and Developed Metropolises

The Songhua River is the third greatest river in China. It flows through heavy industrial districts of Heilongjiang and Jilin Province. There are two major sources of pollution about Harbin section in Songhua River; 1] the plenty of effluent from upper reaches: the investigation indicates that the daily average discharge capacity of the upper reaches of Zhu Shutun in the Harbin area is about 450 million tons; 2] secondly, the pollution about human and industrial activities in Harbin. The influents of Songhua Stalemate are scattered along the river and vast machinery and chemical industries produce a bulk of sewages containing numerous heavy metals. For example, compared to other crucial rivers and lakes in China, the density of Cr, Cd, Pb, Hg and As in Songhua section exceed the background value, the pollution level of surface sediments overtop the average, and the concentration of metals in surface sediments is higher than deep deposits. Furthermore, the combination of heavy metals and organics may increase their toxicity (Li et al., 2017). Most old manufacturing districts were constructed or converted to uptown or leisure area in Shenyang, and many heavy metals are prevalent in surface soil and street dust in these areas. Therefore, it is necessary to consider whether these areas are cleaned up so as not to cause adverse health effects on local residents. As and Pb come from manufacturing and transportation districts and may cause serious health issues to indigenes, particularly in children. The hotspots for children's health risks are chiefly around the Tiexi region, which is a representative technical area for Shenyang heavy industry. After 10 years of reconstruction, soil restoration is still inadequate; some heavy industrial areas are even prohibited and became minefields for use (Ren et al., 2015).

With the developing industrialization and fast urbanization processes since the reform and opening up in the 1970s, the soil was polluted by different levels of heavy metals that harshly and diffusely existed in the metropolis (Khan et al., 2008). Among them, transportation and industrial discharges are considered as the primary sources for amass of heavy metal in urban land, and the urbanization process could significantly enhance the content of Zn in the soil (Wang et al., 2018). Since the beginning of the 21st century, Xi'an has increased its urban footprint from 197.28 km^2 in 2000 to 531.31 km^2 in 2016 due to the implementation of the western development strategy and one belt and one road policy (Chen and Lu, 2018). Many cultivated land and gardens have become building sites; technical and commercial activities promote the accumulation of heavy metals in the soil. Automotive exhaust is an important origin of Pb in urban soils, which is attributed to the addition of anti-knock agents to petrol to cut bursting trends in vehicle motors. A large amount of Cu and Zn in lands are related to transportation discharge. Zn is a crucial agent in vulcanization course, accounting for 0.4-4.3% of tire portion. Besides, Zn is employed as an anti-oxygen and detergent additive in lubricants (Erturk et al., 2015). Cu is commonly utilized in vehicle brake systems and chalcopyrite car radiators, as the mechanical parts of the vehicle wear out, and Cu and Zn motes are expelled. The survey revealed that the contribution rates of natural, traffic, compounded and technical sources to pollution are 25.04%, 24.71%, 24.99%, and 25.26%, respectively. In the city park, about 80% of Cd and Zn existed in soil that is related to park's age or years since the construction, and high-level of Cd, Pb, Zn, and Cu are present in the land of neighborhood unit and entertainment venues (Wang et al., 2018). Among them, Cd and Hg are considered as the preferred metals for control because of their higher content at ground or higher threat for public health. The high concentrations latter has been designated as a priority control category in Beijing, which is a rapidly developing and historic city with over thousand years of geschichte. The reason is that in addition to coal combustion, industrial effluents, and wastes, HgS was used in red dyes in historic architectures and decorations that tardily migrated into ambient land via corrosiveness courses of wind and rain. It is estimated that over the past 500 years, more than 100 tons of HgS have been used to adorn ancient buildings in Beijing (Chen et al., 2015).

The use of the number of electronic devices such as computers, mobile phones, recreational machines, and domestic appliances is continuously increasing worldwide. However, the majority of e-waste is not properly assembled, classified and managed, except for recovering valuable materials, the residual parts are simply mixed with garbage for landfill or incineration (Zhang et al., 2012). Inappropriate and informal reclaim of e-waste, discharge poisonous metals into circumstance, most of which are Pb, Cd, Cr, As, Ni, Hg. GuiYu and TaiZhou cities have become the crucial e-waste machining areas, the formers are one of the world's largest e-recycling regions and approximately 100 thousand people are employed as e-waste recyclers. The content of Pb and Cd, along with PM2.5 quality are far exceed than other Asian areas (Xu et al., 2017), and the latter has circa 30 years history and 40000 laborers refer to e-waste processing and import (2.2 million ton/year). Soils in e-waste regions are usually polluted by compounds of heavy metals and organics. Therefore, the contamination conditions are intricate and multiple from farm land to e-waste locations.

2.4 Plateau and Mountain Areas

Yungui Plateau is one of the five great lake areas and a significant nonferrous metal production base in China. Economic growth and waste emission have generated heavy metal pollution in environment and waters on Yungui Plateau over the years (Lin et al., 2016). Guizhou has a topmost background level of Cr, Cd, As, Zn and Ni, which are 133.3, 1.244, 28.5, 135.9 and 66.7 mg/kg, respectively. Yunnan has the uppermost background value of Cu (48.2 mg/kg). The quantity of Cr, Ni, Cu, Zn, and Pb in dry-hot valley (DHV) region exceed the most farm land background values around the world and the DHV may provide a latent origin of heavy metals in Red River. The concentration of heavy metal in topsoil was controlled by soil and topographic elements, whereas in subsoil it was chiefly controlled by soil correlation factors. Therefore, high setting contents, irrational use of chemical fertilizers and pesticides as well as mining are causes of heavy metal pollution in the DHV region that has great geologic background values of heavy metal. According to research on DHV, the amount of Pb is 1.58-times and Cu is 1.44-times above the domestic level. Serious soil denudation generated by the vast sand content in grounds and ramps in the DHV may contribute to heavy metal contamination in the water system because of increased sediment loads. Moreover, hydroelectric development through stepped reservoir system in upstream of Red River may overwhelm section of the DHV, making it a possible origin of heavy metal pollution for downstream farming activity, fishing, and riverine ecosystem (Duan et al., 2015). Cheng River is the main source of heavy metals transported to lake entrance sediments. The higher density of Ni, Cr, and Zn in settlings accounts from host rocks; As, Cd, and Pb are derived from human activities, whereas Cu is eluted from both sources. According to a study, 65% of samples pollution are prevailingly caused by Pb, Cr, and As (Bai et al., 2011). Erhai Lake may also be contaminated by heavy metals that will further reduce the eco-environment of the lake and abate the ecological action. Rivers and streams in the Tianshan Mountains with a little effect by human activities, the contents of Ni, Co, As, Cr, and Cu are relatively higher than those from wells and lakes. This is because of the desert grasslands, dry riverbeds, exposed rocks, and forests with the human activities. Heavy metals accumulate in soils over long periods, and are easily delivered into water environment by the effects of weathering and rain erosion. So the amount of these elements in soil is lower than that in the water environment of the Tianshan Mountains. In contrast, quantities of Zn, Mn, Pb, Cd, and Hg in farmland soil samples are higher than those in wells. It might have come from agricultural production (use of herbicides, pesticides and chemical fertilizers), industrial emissions and sewage irrigation. In addition, the ability of self-purification in a water body is better than soil environment, which results in the higher quantity of heavy metals in soil environment than water at the same site (Zhang et al., 2013).

3 Remediation Methods of Heavy Metal Pollution

3.1 Chemical Remediation

Chemical immobilization is increasingly valued because of its ambient sustainability and lessening effect of the heavy metals by completely removing them from the surroundings. It can get rid of Cd in contaminated soil and ensure the safe production of crops; can widely be used in industrial sites, agricultural lands and heavily polluted soils (Sun et al., 2016). Selection of suitable fixative such as sepiolite and palygorskite during immobilization process can lower the activity of Cd in rice soils. Sepiolite cuts down the bioavailability of Cd in soils and inhibits the assimilation of microelements in the plant. Palygorskite can stabilize the soil polluted by Cd and Pb. Both the fixatives could be precipitated by carbonate or hydroxide after chelating Cd from the surface. The concentration of Cd in rice can be reduced from 0.72 to 0.18 mg/kg after this treatment meeting the national standard of China (Liang et al., 2014). Sorbents used for heavy metal exclusion have a strong ability to combine metals in deposits that is price moderate and slight influence method to restore soils contaminated by heavy metals. Therefore, the pollutants adsorptive power of the deposit can be significantly improved by their accretion. Sorbent combinative capabilities restrict the transferability of heavy metals in settlings, which in turn leads to a reduction in assimilating of benthic microorganisms. Alternatively, adsorbents can be physically blended or immediately used to surface deposits as sealing material. On the whole, settlings disposed with 10% natural calcium-rich attapulgite and sepiolite give rise to decreases of Cd about 56.2% and 34.2%, Pb about 81.5% and 77.4% in an acid-soluble fraction, respectively. In the same way, assimilation of Pb and Cd by benthic microbiota went down separately 33.5–53.3%

and 20.9–37.1%, correspondingly which emphasizes the possibility for employment in the naturally existent and calcium-rich clay mediums as an *in-situ* adsorbent to repair contaminants (Yin and Zhu, 2016).

Soil washing process is considered to be a powerful heavy metal repair technique. The dominating dislodge mechanism is desorption of contaminants in soils that are dredged in polluted-sites, by vigorously stirring and washing soil particles with scrubbing solution in slurry phase. Using low frequency ultrasound is very valid to improve the elimination efficiency of contaminants in adverse situations of rinsing process. Majority of evidence as for ultrasonic soil washing courses have been received via horn-type sonicators and bath-type sonoreactors, but the appliance of large-range sonoreactors for soil washing is very limited in quite small-scale (Park and Son, 2017). The development of rice (*Oryza sativa*) was enacted on acid-treated soils in comparison to untreated soils from the heavy metal polluted regions. Soils from the polluted area were amended by acid washing or blended with soils acquired from the undefiled district. Grain yield was not affected by the soil restoration technologies (Kim et al., 2017).

The commonly used technology for removing Cr(VI) from waste water is hydroxide precipitation on account of correspondingly simple, low-expense and liable to pH regulation. Metal hydrides can be eliminated by adding coagulants of alum, iron salt, organic polymer and lime, which notably enhanced the effectiveness of heavy metal obliteration (Fu and Wang, 2011). Cr(VI) was transformed into Cr(III) by $FeSO_4$; largest sedimentation of Cr(III) emerged at pH 8.7 and chromate concentration decreased from 30 to 0.01 mg/L after adding $Ca(OH)_2$.

3.2 Physical Restoration

Electrokinetic remediation (EKR) is one of the most effective approaches to eliminating heavy metals for low permeability clay and silt soil, and it could control the flow direction of pollutants. Through the action of the current, metal ions in soils are transported to the electrode under the action of the electric field, latter by centralized collecting and disposing off (Kopp et al., 2018). *In-situ* treatment with the electrokinetic technique by hexagonal electrode configuration is for the sake of repair of As, Cu and Pb in polluted rice soils. The iron electrode was applied to avert the grievous acidulation of soils near the positive pole. Ethylenediamine Tetraacetic Acid (EDTA) was selected as a purified electrolyte to improve the leaching of Cu and Pb. Experimental results showed that this system removed 44.4% As, 40.3% Cu, and 46.6% Pb after 24 weeks of running. The removal rate of Pb(II) and Cr(III) in the soil can eventually reach more than 90% (Jeon et al., 2015). Therefore, EKR is economical and feasible *in-situ* restoration technology, which does not interfere with the soil layer and shortens the repair time. However, the repair effect is highly dependent on the metal type, voltage gradient, soil pH, buffer properties, electrode material and spacing. Electric heating reclamation can also effectively repair the heavy metal in soils. It uses the electromagnetic wave generated by high-frequency voltage to heat the soil, remove the pollutants in soil particles, and accelerate the separation of some volatile heavy metals from land to achieve the purpose of repair.

Vitrification can be used to remove heavy metal pollutants by high-temperature treatment of contaminated areas to form oxide solids. In this process, it could not only volatilize or destruct heavy metals, but also organic pollutants (El-Sonbaty and El-Hadedy, 2015). Most soils can be vitrified, with low energy requirements and low cost, which can be carried out on site or *in situ*. The *in situ* process uses electrodes inserted vertically into the contaminated area to allow the current to pass through the soil. It could not provide enough electrical conductivity if the soil is too dry, while flake graphite and glass frit must be put between electrodes. Grooves supply premier fluxion route of current. Individual melt can handle up to 1000 tons of polluted soil at a depth of 20 feet, typically handling 3 to 6 tons per hour. The larger area is formed by merging multiple separate vitrified areas (Radic et al., 2010). The *ex-situ* treatment includes excavation, pretreatment, mixing, feeding and melting, exhaust gases gather and management, molding or casting of molten products. Glassy materials with certain properties can be acquired by using agents together with sand, clay or natural soil. Glass crap can be cyclically utilized as cleaning filler, aggregate or other reusable resources.

3.3 Bioremediation

Bioremediation is an environmentally-friendly heavy metal repairmen technology that does not produce secondary pollution. Its cost is generally lower than the expense of traditional physical and chemical restoring methods (Fig. 2). Microorganisms can stabilize toxic hexavalent Cr through enzymatic reaction taking place in cytoplasm of the cells (Cheung et al., 2006; Cheung and Gu, 2007). Rhizosphere *Arthrobacter* and non-rhizosphere *Bacillus* were screened out from contaminated cropland soil in the catchment of mining region. Both can reduce the utilization of metals and increase the pH of low-pollution soils when inoculated into the polluted site. The former increases phytomass and valid iron in rice and red ramie, and the latter reduces the metal content of rice shoots, but increases the metal content of red ramie. Microorganisms can influence the bioavailability of heavy metals via metabolism, which play a critical role in keeping soil's function (Wen, 2016). Biological adsorption by means of biomaterials (sea-weed, green algae, and alginate derivatives) to remove or recover metal ions from aqueous solutions, is cost-efficient and minimizes the chemical or biological sludge. These biological adsorbents diffusely exist and are easily biodegradable. Purslane is reported to be a latent low-expense absorbent for removing Pb ions from water and lichens, which removes Pb(II) from aqueous solutions (Gong et al., 2005). Biomineralization is a natural way to produce complex structure of inorganic materials including mesoporous silica nanoparticles (MSPS). It is formed by metal-mediated minerals and has characteristics of low toxicity, large surface area and pore volume (Ge et al., 2012). Recently, researchers investigated the mechanism of urea producing bacteria involved in mineral precipitation that can be applied to bioremediate the heavy metals pollution. As of the urease enzyme catalysis, the pH of soil increases and produces carbonate. The mineralization of metal ions surrounding cell membranes will be induced when pH is 8-9. X-ray diffraction study verified the function of bacterial aroused calcite sedimentation in bioremediation of Pb in tailings, and phosphorus amendments were used to lessen the relative bioavailability of Pb in polluted soils (Govarthanan et al., 2013).

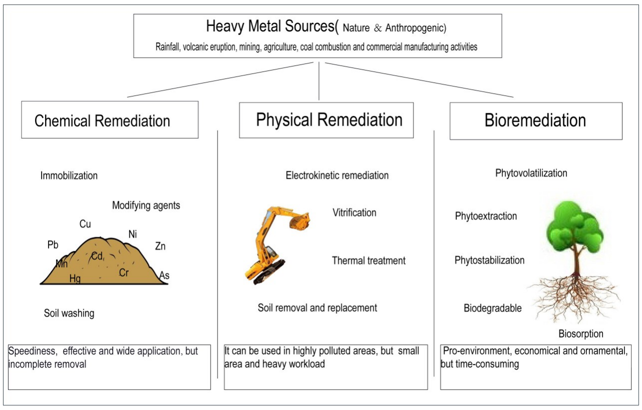

Figure 2. Comparison of different heavy metal contaminated soil remediation methods: physical, chemical and biological.

Phytoremediation uses the inherent feature of plants to regulate, degrade or dislodge heavy metals from soils. Hyper-tolerance is the basis of hyper-accumulation. Therefore, the selected plants should be metal-tolerant and must have the capability to grow dense root systems, could prevent corrosion and reduce permeation to groundwater. The repair process depends on the depth and level of contamination, soil factors, plant species and the length of the root system (Shao et al., 2010). Many plants can be used for cleaning up contaminated sites with promising results obtained under laboratory conditions (Yu and Feng, 2016; Yu and Zhang, 2017; Yu et al., 2018). *Miscanthus x giganteus* was substantiated by its strong ability to grow well in heavy metal polluted districts, which can weaken transferability and enhance accumulation of metals in roots and restrict them to pass on aerial tissues. It has been confirmed that *M. x giganteus* is an excellent species for phytostabilization because of its ability to decrease transferability and availability of Cd and Zn, as well as its dedication to aggrandize organic carbon amounts of soils (Al Souki et al., 2017). Studies have shown that mangroves growing in coastal rivers, estuaries and gulf intertidal have a strong ability to retain Cr, Pb, Zn, Cu and Ni from tidal waters, freshwater rivers and rainwater runoff while maintaining the stability of the adjacent coastal terrain (Zhou et al., 2010). Therefore, forestation and restoration of mangrove forests are conducive to accumulation of heavy metals in surface settlings, while reducing mobility and bioavailability of them.

3.4 Combined Remediation

Phytoremediation unites with advisable microbial and biodegradable chelating agent, which can restore heavy metal polluted soils up to a point. Treatment of soils with Pb- and Cd- resistant bacteria (JB12) and ethylenediamine-N,N'-disuccinic acid (EDDS), promoted a significant increase in biomass yield and an accumulation amount of Pb and Cd in tall fescue and red clover growing in treated lands. The Plant-associated microbial remediation is on account of the intrinsic relationship between soils, microorganisms and plants, which can make full use of the advantages of both to enhance the remediation effect of contaminated-soils. In addition, it is expected to achieve wide-range repair combined with the apple of chelating agents in polluted areas (Jin et al., 2013).

Chemo-phytostabilisation is the course of using soil conditioners and plants to immobilize heavy metals. A study found that the bioavailability of Pb, Cd, and Zn could be significantly reduced by using sludge and inorganic modifiers and sowing tall fescue to correct contaminated soil (Grobelak and Napora, 2015). Phosphorous compounds are commonly used to accelerate Pb sedimentation to reduce its transferability and motivate ion exchange processes. Cd can be fixed by using CaO, $Ca(H_2PO_4)_2$ or K_3PO_4 and sewage sludge bond inorganic improver. Reducing the bioavailability of metals is a vital factor and soil additives are used to facilitate the growth of plants and stabilization of metals. The application of sewage sludge has significantly increased soil adsorption capacity of metal ions. Tall fescue is a suitable choice for phytostabilisation because Pb, Cd, and Zn are chiefly aggregated in root systems.

The applications of electric restoration united with phytoremediation can purify heavy metal contaminated soils. In operation, potato tubers are cultivated in plastic containers loaded up with Zn, Pb, Cu and Cd-polluted soil and pull-in current. The biomass of plants treated by alternating current reached 72%, and the cumulative amounts of Zn, Pb, Cu, and Cd in potato roots were found to be 822.5 ± 102.3, 113.7 ± 5.0, 58.9 ± 7.2 and 15.0 ± 3.8 mg/kg, respectively (Aboughalma et al., 2008). Metal ions dissolve under acidic conditions near the positive electrode and migrate toward the cathode. Nonetheless, soil pH change is the prime element to normalize metal distribution between the solid and liquid phase. The dissolvability of metals augmenting with soil pH descending, but acid levels and vast dissolved metals may limit plant growth. Biomass yield of plants is one of the significant aspects in plant restoration, as the application of electric field increases the production of biomass. This technology is a new technique to optimize the phytoremediation of heavy metal in soils.

The restoration of heavy metal polluted soils by screening bond with soil washing has confirmed to be valid. Washing of Pb, Cd, Zn and As in soil particles $>$ 2 mm in size, the removal rates of Pb and Cd were 75%-87%, Zn and Cu were 61%-77%, As and Cr reduction was $<$ 45%. There is a significant difference in eliminating efficiency heavy metal at lands resting with soil particle size. The combination of soil washing and particle separation transfer metal into a small amount of soil, significantly reducing the volume of pollutants. Optimal soil particles and washing time enables high efficiency, low energy consumption and cost in removing heavy metals (Liao et al., 2016).

Microorganisms – plant system enhances the adsorption of heavy metals by plants and significantly improves the remediation effect. Plants play an important role in cleaning up pollutants, especially metals and metalloids in sediment (Yu and Feng, 2016; Yu and Zhang, 2017; Yu et al., 2018). The triumphant application of this technology depends not only on the choice of plants, but also the link between rhizosphere bacteria and plant roots. It would contribute to increasing the metal-resistance and absorption of metals in seedlings by applying resistant microbes to soils contaminated by heavy metals. Soil polluted by Zn and Cd was managed with rhizosphere bacteria and then planted with *Ligustrum quihoui Carr.* The content of Zn in the aboveground and underground part was divided into 1308.7 and 2,524.3 mg/kg, and there was no symptom of injury for seedlings. This indicates that the microorganisms can assist plants to absorb heavy metals under the case of unitary metal contamination, and transfer them in underground part to aerial part, which is beneficial to subsequent disposing of plants (Ying et al., 2009).

3.5 Other Repair Technologies

Nano-remediation has employed novel nanomaterial (NM) as an adsorbent or catalyst to remove heavy metals from soils and received extensive attention in recent years (Gong et al., 2018). Nanometer materials have a smaller particle size (1-100 nm) than the typical bacterial cell size (1000 nm), which possess great ability to fix or eliminate harmful contaminants and increase the absorption and accumulation of pollutants in plants. Furthermore, they also improve the microbial degradation rate of pollutants and sequentially enhance the bioremediation effect. The application of nanometer particle size of Zero-Valent Iron (nHAP) obviously reduced the level of metal ions in the shoots and roots of pakchoi grown in contaminated soils. Applying 30 g/kg nHAP, the concentration of Pb, Zn, Cu, and Cr in a bud was 53.4%, 32.1%, 32.2%, and 30.9%, respectively (Li et al., 2014). Infected water and soil treated with nZVI separately (100 mg/ 10 mL water or soil), the content of Ni were reduced by 97% and 85% (Rathor et al., 2017).

Permeable reactive barriers combine modified montmorillonite are deemed to one of the most prospective *in situ* restoration techniques on account of high-efficiency, low operation and warranty costs and energy demands. The results showed that for reactive zone filled with quartz sand, total As wipe off 98.57% was obtained within the first 60 mm and therewith declined laxly to 88.84% at exit. For reactive zone filled with quartz sand and iron, analogous space variations tendency was discovered in quartz sand layer, and total As eliminating subsequently rose to $\geqslant$ 99.80% in iron layer (Luo et al., 2016). Furthermore, ferrous sulfide can external adsorb and co-precipitate with As on account of doughty appetency of Fe (hydroxide) to As, the oxidizing agent (usually oxygen or aerated water) is supplied to subterranean water to cause sedimentation and adsorption of Fe (III). The concentration of As can be significantly reduced to less than 10 mg/L by preprocessing of sorbents with acid, alkali, hydroxide (H_2SO_4, HCl, NaOH, and KOH) or wiping with iron oxide minerals (goethite, akaganeite) (Shakoor et al., 2015). In most high aquifers, sulfide can be stabilized in a strong reducing environment, and its removal capacity is not affected by pH change. In addition, microbial activity in aquifers can immensely irritate sulfate reduction and increase generation of ferrous sulfide. Through field practice in the middle of Datong Basin in China, the concentration of As in groundwater decreased from 593 to 136 mg/L and removal rate was up to 77% after 30 days (Pi et al., 2017).

Biochar comes from plant residues, animal wastes or sludge, and reserves the ability to adsorb heavy metals. Charcoal was placed in the solution including low consistency of Cd, Pb, Cu, and Zn for 0.25 h or 48 h, the removal efficiency of four type metals were 80% or over 90%. It should be considered that metal ions eluted from biochar are affected by the pH level when charcoal is put into use, because the state of the metal affects its availability in the soil (Fig. 3). Washing rate is restrained to less than 10% if the pH of the solution immersed with metal-adsorbed charcoal exceeded 3.5, while adsorbed heavy metals are lixiviated from charcoal after-touching with a solution having a pH $<$ 1.5. In consequence, although the heavy metals adsorbed on charcoal are left in the ecological environment for ages, it rarely causes the metal ions in the charcoal to be eluted to the environment and give rise to re-contamination (Miura and Shiratani, 2018).

Portable X-ray fluorescence (PXRF) spectroscopy provides the potency for fleet *in-situ* repair and non-destructive analysis of entire metal detection. It has been extensively applied in evaluating the pollution level of heavy metals in soil of late years. PXRF has also been used to measure a series of edaphic factors (soil acidity and alkalinity, texture and salinity) by using blanket elemental concentrations and

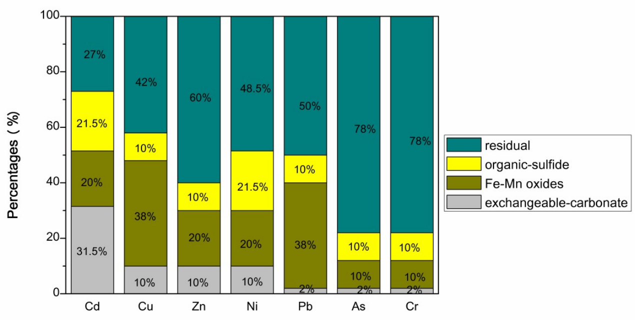

Figure 3. Percentages of four fractions for Cd, Cu, Zn, Ni, Pb, As and Cr in soils.

multiple linear regression models (Tian et al., 2018).

The use of both adsorption and magnetic properties about zero-valent iron (ZVI) could help to eliminate adverse aspects allied with root position fixation of contaminants. A precondition for using ZVI to remove heavy metals is superficial corrosion because metal adsorption dependes on the reciprocity between the dissolved portion and Fe^{2+} or Fe^{3+} products of ZVI oxidative dissolution. After disposed of with the ZVI in polluted sediments, the toxicity unit (TU) removal efficiencies achieved to 97.3%, 97.2% and 97% for Hg, Cu, and Zn, respectively. In conclusion, the toxicity of heavy metals in contaminated sediments was significantly reduced after the ZVI was applied and magnetic separation was performed (Feng et al., 2018).

4 Summary and Future Perspectives

The characteristics and specificity of heavy metal contamination in China can be summarized as the following five points:
1). Heavy metal contaminated soils are widespread, but existing repair techniques are limited and generally suitable for small-scale heavy metal removal.
2). The situation of heavy metal pollution is complicated, but usually, a single repair technique is adopted.
3). Soil heavy metal pollution varies across China, but there is no targeted approach to repair it.
4). Presently, for heavy metal remediation, the main application is still chemical or physical repair based methods.
5). Some emerging repair technologies are still lab-scale or in a small range, without actual or large-scale applications.

5 Conclusions

The heavy metal pollution of soil in China is specially caused by Cd, Pb, Hg, As, Ni, and Cr that are most frequent hazardous materials which have serious impacts on mankind and the ecosystem. Pollution type, concentration and physical or chemical states of heavy metals found in polluted soils depend on the activities and operation in contaminated sites. Other influencing factors may include soil or groundwater chemistry and local transport mechanisms. Therefore, we need to find an effective correction method to mitigate or eliminate the toxic substance from a contaminated area. In recent years, phytoremediation has irreplaceable advantageous in control of heavy metal pollution. It has increasingly attracted attention because of low cost, simple management, and operation, small damage to soils, eco-friendly and beautify environment in process of field treatment. Meanwhile, microbes have wide variety and distribution, rapid propagation, small volume and various types of metabolism, which can degrade or convert heavy metals and take a significant role in the treatment of heavy metal pollution. Some emerging technologies have also been proved to have obvious effects on the repair of heavy metals today, but most of them are still limited to laboratory conditions and need to be verified by practice. In view of the current pollution situation in various regions of China, it is a good choice to combine phytoremediation technology with microbial remediation methods or other effective restoration measures. This proposal can be extended to other areas contaminated by heavy metals. In practice, mixtures using two or more heavy metal repair techniques will achieve relatively low cost and better repair effect.

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