Remediation technologies and risk assessment of soil contaminated with heavy metals

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Abstract. Soil contaminated with heavy metals is an urgent issue worldwide because of its great influence on human health and the ecological environment. Therefore, the remediation of heavy metal contaminated soil is more important, as well as the corresponding assessment methods. This review introduces the main sources of soil contamination with heavy metals, and the soil-remediation methods, and focuses on the risk assessment methods. Study shows that the major soil-remediation methods are phytoremediation and biological techniques. The risk assessment methods include ecological and human health risks, the aim of which is to understand the soil pollution degree and evaluate the availability of the soil.

1. Introduction

Heavy metals are the most noticeable inorganic contaminants in soil, mainly because it is easy to accumulate and hard to be degraded and even harm human health through the food chain. Naturally, the content of heavy metals is low in soils, its high concentration is increased gradually with social development, leading to soil pollution. The increasing concentration of heavy metals such as Cr, Hg, Pb, and Cd in the soil would influence plant growth. Besides, these exposed heavy metals have impacts on abundant human diseases. Therefore, soil heavy metal pollution has caught public attention globally. As for China, it has undergone huge and rapid urbanization and industrial expansion, and releases abundant heavy metals into the environment, especially soils. It has been estimated that almost 20 million hectares of arable land have been polluted by heavy metals, among which Cd, Pb, As, Cr, and Hg are the most important heavy metal pollutants in soils in China [1] [2].

Soil heavy metal pollution threatens human health and the ecological environment. Therefore, it is of great significance to focus on the remediation of heavy metal contaminated land. Decrease and eliminate the heavy metal that accumulated in the soil is an effective measure to enhance soil safety. There are many remediation methods to reduce heavy metals from soils. Economic viability and effectiveness are key elements in identifying the applicability of remediation methods. Traditional physical-chemical methods have been widely studied and applied, moreover, remediation of polluted soil through microbial measure and phytoremediation has become a hot topic recently.

After applying these remediation technologies, it is essential to know whether the treated soil is safe for reuse. Various assessment methods are commonly applied to evaluate contamination and ecological risk in the areas, focusing on heavy metals pollution. This review introduces the main sources of heavy metals in the soil as well as the remediation technology, emphasis the measures for risk and danger assessment of heavy metal contamination in soil, providing an important reference for policymakers,
environmental engineers, and agrotechnicians regarding remediation measures and risk assessment methods.

2. Sources and remediation of soil contamination with heavy metals

2.1. Sources of heavy metals in the contaminated soil

The heavy metals in soil are commonly from anthropogenic sources and atmospheric deposition. The anthropogenic sources are the main reasons for heavy metal emission and include mining activities, application of fertilizer and pesticides, and sewage irrigation. The atmospheric deposition from industrial activities such as the burning of coal and petroleum has an intensive mark in soils concerning heavy metals.

Mining operations are the major source of heavy metals. For example, the mining of Zn-Pb ores leads to the comprehensive contamination of Zn, Cd, As, Cu, and the toxicity of Pb [3]. Taking the Diaojiang river basin in Guangxi as an example, the discharge of Pb-Zn mine wastewater from Nandan upstream caused the concentration of heavy metal in farmland to exceed the standard [4]. Moreover, the expansion of the nonferrous ore development scale releases heavy metal pollutants into the soil environment through slag, tailings, and sewage. Coal mining contributes to the pollution of Cu, Cr, Pb, and Zn. The abandoned mine is another important source of heavy metal pollution. For instance, the unused Gesenhad uranium mine in the east of Thuringia, Germany, yet could still be examined heavy metal pollution [5].

Farming practices could increase Zn, Cu, and Cd in the soil through sewage irrigation, livestock manure, and chemical fertilizers. Pesticides are widely used for controlling harmful weeds (herbicides), fungi (fungicides), bacteria (bactericides), and insect infestations (insecticides) in the agricultural field [6]. The long using time of Cu-containing reagents could increase the concentration of heavy metal Cu in the soils, resulting in the deterioration of plant growth and chlorosis. Meanwhile, fertilizers that contain phosphorus could lead to the concentration of F, Hg, and Pb in the soil [7]. Animal manure is a great fertilizer, but Cu and Zn as growth promoters are added to the diets and discharged into the environment, causing heavy metal pollution. Based on the records from the internet site of the National Bureau of Statistics of China, there was an important correlation between the Cd concentrations in agricultural soils and the amount of fertilizer application [8]. Most studies show that soil has buffer capacity, so the effects of fertilizers and pesticides on the soil would increase significantly over time, and heavy metal pollution could lead to land degradation.

Sewage irrigation makes a large number of harmful matters, particularly heavy metals, into the soil, leading to soil pollution of different degrees [9]. The heavy metal elements in sewage are difficult to degrade, when they accumulate in large quantities and exceed the environmental limit of the soil, they can damage human health and cause severe ecological problems. Wang et al. (2012) studied the heavy metals contamination in Beijing and Tianjin city cluster, China, which have a long history of sewage irrigation, evaluated the health risk, and indicated that Cd exceeded the environmental limit [10]. Kanwal et al. (2020) checked the impact of sewage irrigation on soil, and their study showed that heavy metals such as Pb, Cr, and Ni in wastewater could reduce the function of soil macrofauna populations, decline the soil fauna density and abundance, and cause harm to resident soil fauna and flora [11].

The atmospheric particulate deposition is another source of heavy metal pollutants in the environment, threatening human health. Schröder et al. (2010) founded that atmospheric deposition was an important way to lead to Pb accumulation in soils [12]. Peng et al. (2020) researched the sources of heavy metal in soil and maize in Northeast China, and the results indicated that Pb in atmospheric dust came from vehicle emissions and coal combustion [13]. Moreover, studies confirmed that atmospheric deposition used to be accountable for 43–85% of the complete As, Cr, Hg, Ni, and Pb inputs [14].

2.2. Remediation of soil contamination with heavy metals

A nationwide soil survey shows that concentrations of some heavy metals increasing at much great rates, in some parts of China, especially in the south, food crops generally exceed pollutant limits [15]. Faced
with the severe situation of soil heavy metal pollution, it is urgent to develop economical, efficient, and green remediation technology. Currently, there are a variety of remediation methods for contaminated soil, including physical, chemical, and biological processes. These measures reduce the risk by minimizing heavy metal concentrations in soil. Besides, the choice of the most effective soil remediation measure rests with the site characteristics, pollutant type and concentration, and the type of land use [16]. The advantages and disadvantages of these remediation methods are given in Table 1.

Physical remediation is the treatment of reversing or terminating damages to the soil applying physical methods including soil replacement, encapsulation, vitrification, and electrokinetic remediation. Substitution of soil refers to the replacement or partial replacement of polluted soil with clean soil, which dilutes the concentration of heavy metals in soil to restore soil functionality. However, this method is time-consuming, expensive, and appropriate for micro-regions with severe pollution. Encapsulation is to comprise polluted soil in a physical barrier system consisting of low permeability caps, enclosing underground barriers, and barrier floors [17]. This method is not a direct remediation treatment, but a measure to reduce the migration of heavy metals [18]. Also, encapsulation is limited to micro-region, shallow but heavily contaminated sites. Vitrification is a thermal remediation measure that reduces the mobility of heavy metals inside soil and converts contaminated soil into glass-like solids at high temperatures. This method does not apply to soils with high organic matter content and high moisture content. Electrokinetic remediation is to establish a suitable electric field gradient on both sides of the electrolytic cell containing saturated contaminated soil, and to separate heavy metals in the soil by electrophoresis, electroosmosis, or electromigration, to reduce pollution. This process may take a few days to several years, and it depends on the overall speed at which metal ions in soil are moving.

Chemical remediation is the process to remove contaminants using chemical reagents, reactions, and principles including immobilization techniques and soil flushing. Immobilization technology termed solidification/stabilization (S/S) can be used to fix heavy metals in the soil by adsorption, rather than removing or extracting contaminants from soil. Therefore, it is not a permanent solution as heavy metals are released into the soil under weathering conditions. The operation of solidification needs a large amount of binder, so its applicability is mainly affected by the applicability of the binding agent and transportation cost. Furthermore, the future use of solidified land may be limited, so immobilization technology is the last selection for soil remediation. Chemical leaching is to remove contaminants from soil by applying a chemical solvent through the soil. The chemical solvent can promote the dissolution or migration of pollutants in the soil environment, which can be recovered and eventually disposed of later. Soil flushing is technically simple, however, installing solution collection wells or underground drains can be challenging and expensive. The measure is suitable for uniform and rough texture soils with high permeability.

Biological remediation is the process of removing heavy metals from the soil by using microorganisms and plants. Phytoremediation is the process of planting green plants in contaminated soils to remove heavy metals by extraction, volatilization, root filtration, degradation, stabilization. Some special plants with strong tolerance and enrichment capacity for heavy metals, and break down pollutants into less toxic elements and absorb them in the stems and leaves. For instance, Sebertia acuminate could accumulate Ni in its latex up to 26% dry weight [19]. This plant-based measure has the advantage of low cost and easy control of secondary pollution, especially, after vegetation planting, it has the effect of protecting topsoil and reducing erosion and soil erosion. Bioremediation is to decontaminate soil by using microorganisms to detoxify metals through valence transformation, biosorption, extracellular chemical precipitation, and volatilization. Vaxevanidou et al. (2008) found that the iron-reducing bacterium Desulfituromonas palmatites greatly contributed to the release of As in calcareous soil [20]. Generally, bioremediation is assisted with other measures to facilitate the solubilization of heavy metals before extraction. For the soil polluted by various heavy metals, the effect of microbial remediation is not significant. But it's more effective than using phytoremediation alone.
Table 1. The advantages and disadvantages of different remediation techniques and their application status

| Method                  | Remediation technique | Advantages                        | Disadvantages                         |
|-------------------------|-----------------------|-----------------------------------|---------------------------------------|
| Physical remediation    | Soil replacement      | Complete amelioration of heavy metals | Laborious, time-consuming, not economically |
|                         | Encapsulation         | High security                     | High cost                              |
|                         | Vitrification         | High efficiency                   | High cost, loss of soil environmental functions |
|                         | Electrokinetic        | Less disturbance to the soil       | Time-consuming, low efficiency         |
| Chemical remediation    | Immobilization /stabilization | Economical, easy to implement, quick results | Temporary effectiveness                |
|                         | Soil flushing         | Low cost, simple implementation    | High requirement for soil structure, potential groundwater pollution |
| Biological remediation  | Phytoremediation      | Low cost, simple implementation    | Limited to low contamination areas, low efficiency |
|                         | Bioremediation        | Low cost                          | Low efficiency                         |

3. The method of risk assessment of soil contamination with heavy metals

Various assessment methods are applied to assess risk in the heavy metals pollution land. Two groups are divided: the methods to evaluate human health risk and ecological risk, respectively.

3.1. Health risk assessment

Health risk assessment is to establish a corresponding relationship between environmental pollution and human dose-effect and quantitatively describe the risks caused by pollutants to the human body. For human health assessment who are exposed to heavy metals, a model developed by the USEPA (Environmental Protection Agency of the United States) was used in most of the studies to evaluate the risk of carcinogenic and non-carcinogenic [21]. There are three ways in which hazardous pollutants are exposed to individuals: oral intake, breath intake, and skin contact. The average daily intake (ADI) of the three exposure pathways for potentially toxic metals in the human body is calculated as follows [22] [23]:

\[
ADI_{\text{oral}} = \frac{C \times IR_{\text{oral}} \times EF \times ED}{BW \times AT} \quad (1)
\]

\[
ADI_{\text{breath}} = \frac{C \times IR_{\text{breath}} \times EF \times ED}{PEF \times BW \times AT} \quad (2)
\]

\[
ADI_{\text{skin}} = \frac{C \times SA \times SAF \times ABS \times EF \times ED}{BW \times AT} \quad (3)
\]

where \(ADI_{\text{oral}}\), \(ADI_{\text{breath}}\) and \(ADI_{\text{skin}}\) are the average daily intake by oral intake (mg kg\(^{-1}\) d\(^{-1}\)), breath intake (mg kg\(^{-1}\) d\(^{-1}\)) and skin contact (mg kg\(^{-1}\) d\(^{-1}\)), respectively. C is the concentration of the contaminant (mg kg\(^{-1}\)). The exposure parameters for the three models are based on the Technical guidelines for risk assessment of soil contamination of land for construction (2019) (Table 2).
Table 2. Recommend parameters for the risk assessment model.

| Parameter     | Meaning                                               | Adult | Children | Dimension |
|---------------|-------------------------------------------------------|-------|----------|-----------|
| $\text{IR}_{\text{oral}}$ | oral intake rate of soils                          | 100   | 200      | mg d$^{-1}$ |
| $\text{IR}_{\text{breath}}$ | breath rate                                           | 14.7  | 7.63     | m$^3$d$^{-1}$ |
| EF            | exposure frequency                                     | 350   | 350      | d a$^{-1}$  |
| ED            | exposure duration                                      | 24    | 6        | a          |
| BW            | average body weight                                    | 61.8  | 19.2     | kg         |
| AT            | average time for non-carcinogenic effect               | 2190  | 9125     | d          |
| SA            | exposure skin surface area                            | 4530  | 1600     | cm$^2$     |
| ABS           | dermal absorption factor                              | 0.001 | 0.001    | dimensionless |
| PEF           | particle emission factor                              | 1.32×10$^9$ | 1.32×10$^9$ | m$^3$kg$^{-1}$ |

The non-carcinogenic risk refers to the level at which a population is exposed to a non-carcinogenic pollutant. It can be evaluated by the ratio of the average daily dose and a reference dose (RfD) (mg kg$^{-1}$ day$^{-1}$). The calculation of the hazard quotient (HQ) is as follows:

$$HQ = \frac{ADI}{RfD}$$

where RfD is the maximum allowable concentration of a heavy metal that has no effects on human health (Table 3).

For a variety of heavy metals pollution, the sum of HQ for every heavy metal is calculated to examine the entirety of non-carcinogenic risk. The calculation equation of hazard index (HI) is as follows:

$$HI = \sum HQ_i = \sum \frac{ADI_i}{RfD_i}$$

If the HI value is much less than 1, the exposed adult or children is not likely to undergo bad health effects.

Carcinogenic risks (CR) refer to the probability that an adult or child's exposure to potential carcinogen pollutants in their lifetime resulting carcinogenic diseases, which are calculated as the product of the average daily intake and SF (mg kg$^{-1}$ day$^{-1}$). The carcinogenicity slope factor converts the average daily intake of potentially toxic metals to the incremental risk of adult or children developing cancer [24]. For multiple carcinogenic pollutants, the carcinogenic risks are sum.

$$CR = ADI \times SF$$

$$TCR = \sum CR_i$$

The carcinogenic risk value between $1 \times 10^{-4}$ and $1 \times 10^{-6}$ is acceptable [25]. The carcinogenic risks of Cr, Ni, and As were assessed because of the absence of carcinogenic slope factors for other heavy metals. The parameter values of RfD and SF through three exposure pathways of the eight heavy metals are resulting from the research analyzed in China (Table 3) [26] [27].

Table 3. The reference dose and cancer slope factor of heavy metals.

| Metals | RfD_{oral} | RfD_{breath} | RfD_{skin} | SF_{oral} | SF_{breath} | SF_{skin} |
|--------|------------|-------------|------------|-----------|-------------|-----------|
| Cr     | 3.00×10$^{-3}$ | 2.86×10$^{-5}$ | 6.00×10$^{-5}$ | 0.50      | 4.20        | 2.00      |
| Ni     | 2.00×10$^{-2}$ | 2.06×10$^{-2}$ | 5.40×10$^{-4}$ | 1.70      | 0.90        | 4.25      |
| As     | 3.00×10$^{-4}$ | 3.10×10$^{-4}$ | 1.20×10$^{-4}$ | 1.50      | 1.51        | 3.66      |
| Cu     | 4.00×10$^{-2}$ | 4.02×10$^{-2}$ | 1.20×10$^{-2}$ |           |             |           |
| Hg     | 3.00×10$^{-4}$ | 8.57×10$^{-5}$ | 2.10×10$^{-3}$ |           |             |           |
| Cd     | 1.00×10$^{-3}$ | 1.00×10$^{-3}$ | 1.00×10$^{-3}$ |           |             |           |
| Zn     | 3.00×10$^{-1}$ | 3.00×10$^{-1}$ | 6.00×10$^{-2}$ |           |             |           |
| Pb     | 3.50×10$^{-3}$ | 3.52×10$^{-3}$ | 5.30×10$^{-4}$ |           |             |           |
3.2. Ecological risk assessment

Ecological risk assessment is to evaluate the possibility of adverse ecological consequences after the ecosystem is affected by stress factors. For ecological risk assessment of heavy metal pollution in soils, toxic metal is the important stress factor. Various methods are applied to evaluate the ecological risk and danger of soil contaminated with heavy metals.

3.2.1. Single-factor pollution index method and Nemerow Comprehensive Pollution Index Method. The single-factor pollution index ($P_i$) method is based on the soil environmental quality standard to evaluate the pollution level in the soil, which is the most basic evaluation method. Generally, $P_i$ is the ratio of the heavy metal concentration $C_i$ to the $S_i$, which are the second-grade standard values according to Soil environmental quality-Risk control standard for soil contamination of development or agriculture land (GB36600-2018 or GB15618-2018):

$$P_i = \frac{C_i}{S_i} \quad (8)$$

If $P_i < 1$, there is no heavy metal contamination; conversely, it means pollution happens.

The single factor pollution index can assess the main pollutants and their harm level. However, it is limited to show the overall situation of heavy metal contamination in the region. The Nemerow Comprehensive Pollution Index ($P_c$) Method considers not only the maximum value of $P_i$, but also the average value of all considered metals, which can more objectively evaluate the pollution status of soil heavy metals [28]. The formula is as follows:

$$P_c = \sqrt{P_{i,\text{max}}^2 + \frac{1}{n} \sum_{i=1}^{n} P_i^2} \quad (9)$$

It is common to use both the $P_i$ method and the $P_c$ method to assess soil heavy metal contamination. These two methods are used to study the pollution degree of four kinds of heavy metals Cd, Pb, Zn, and Cu in the soil of the Dabaoshan mine in Guangdong, the results showed that there was a great difference in the concentration of heavy metals of different soil, but they were all significantly higher than the background value of the heavy metal of soil in Guangdong province [29]. Lin et al. (2014) used GIS and geostatistics to systematically assess the heavy metal contamination of cultivated soil in Jiangxi Province by applying the $P_i$ method and $P_c$ method [30].

3.2.2. Potential ecological risk index method. The potential ecological risk index (RI) could assess the pollution level and ecological risk of heavy metals based on their properties and environmental behavior, the calculation formula is as follows [31]:

$$RI = \sum_{i=1}^{n} E_i = \sum_{i=1}^{n} T_i \times \frac{C_i}{B_i} \quad (10)$$

where $E_i$ is the potential ecological risk index of heavy metal “i”. $T_i$, $C_i$, and $B_i$ is the toxicity response coefficient, the measured concentration (mg kg-1), and the background values (mg kg-1) of heavy metals “i”, respectively. $n$ is the number of heavy metal elements.

RI method takes into account the addition of toxicity of various elements, eliminates the influence of regional differences, and can be used to assess the pollution of heavy metals relatively comprehensively. However, it only considers common heavy metals and lacks the toxicity coefficient of some specific heavy metals. This method was initially applied to the study of heavy metal contamination in sediments, and then gradually introduced into the ecological harm evaluation of heavy metal contamination in soils and has been widely used. Hu et al. (2013) used the RI method to analyze the ecological risk degree of different heavy metals in the surface soil of the Pearl River Delta [32]. Islam et al. (2015) used the Hakanson index method to evaluate 6 harmful elements (Cr, Ni, Cu, As, Cd, Pb) in urban soil under 12 different land-use conditions in Bangladesh [33]. Rafique et al. (2016) evaluated the potential ecological risk of heavy metals in the soil under the cotton-wheat planting mode in Ocala, Pakistan [34]. Also, some scholars have improved the RI method. Wang et al. (2017) made an
an in-depth study of Hakanson’s principle, calculated the toxicity coefficient of Sb, and extended the application scope of the RI method [35]. Zhuang et al. (2018) improved the toxicity coefficient of Be, Sb, and Ti in the ecological risk assessment of Nansi Lake [36].

3.2.3. Geoaccumulation index method. German scientist Muller first founded the geoaccumulation index ($I_{geo}$) method, and it has been widely used to research the pollution degree of a certain heavy metal in soil [37]. This method assesses the natural variation characteristics of heavy metal distribution and discriminating against the impact of human activities on heavy metal pollution [38]. The following formulation is provided to computing the $I_{geo}$ for the soils of the pollution areas.

$$I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right)$$

Where $C_n$ and $B_n$ are the measured value and the geochemical background concentration of each heavy metal in the pollution areas (mg kg$^{-1}$), respectively. The latter value is different in different provinces. The constant 1.5 is applied because of potential changes in the baseline data [39] [40]. The $I_{geo}$ contain 7 classes (Table 4).

| Class | Value | Soil quality               |
|-------|-------|---------------------------|
| 0     | $I_{geo} \leq 0$ | Uncontaminated             |
| 1     | $0 < I_{geo} \leq 1$ | Uncontaminated to moderately contaminated |
| 2     | $1 < I_{geo} \leq 2$ | Moderately contaminated    |
| 3     | $2 < I_{geo} \leq 3$ | Moderately to heavily contaminated |
| 4     | $3 < I_{geo} \leq 4$ | Heavily contaminated       |
| 5     | $4 < I_{geo} \leq 5$ | Heavily to extremely contaminated |
| 6     | $I_{geo} > 5$ | Extremely contaminated     |

Geoaccumulation index methods not only consider the impact of background values processes but also pays attention to the influence of human activities on heavy metal pollution. In recent years, $I_{geo}$ method has been widely applied to assess soil heavy metal pollution. Fang et al. (2019) used $I_{geo}$ method for the analysis of heavy metal pollution in some parks in China [41]. Wang et al. (2020) evaluate the urban roadside soil heavy metals pollution levels [42]. Li et al. (2018) analyzed and evaluated heavy metal accumulation in typical areas of Chongming and Fengxian in Shanghai according to $I_{geo}$ evaluation and grading standards [43].

There are some limitations and deficiencies in the practical application of the above methods. In the evaluation process, it is necessary to use a combination of various methods to achieve the expected effect.

4. Conclusion
This review summarizes the sources, remediation methods, and risk assessment methods of soil heavy metal pollution. Although the concentration of heavy metals in the soil is low in China, its pollution in the soil is a serious problem associated with rapid social development. The heavy metals in soil were mainly from anthropogenic activities and atmospheric deposition, such as the burning of coal and petroleum, mining activities, application of fertilizer and pesticides, and sewage irrigation. Various remedial methods have been adopted to minimize soil heavy metals and to limit their mobility into the food chain. These techniques involve physical, chemical, and biological to rectify soil contamination with heavy metals. It leads to soil replacement, encapsulation, vitrification, electrokinetic, immobilization, soil flushing, phytoremediation, and bioremediation.

As a result of soil heavy metal contamination, green plants and agricultural grain products are increasingly polluted, resulting in public and ecological health risks. Some scientific methods are proposed to assess the levels of pollution and contamination risk degrees. The exposure risk of soil
heavy metals for human health risk assessment can be evaluated by using the model developed by the USEPA. The hazard quotient (HQ) is used to evaluate potential non-cancer risk, which is calculated by dividing the dose of the average daily intake (ADI) of potentially toxic metals for adults and children from three pathways including the oral intake, breath intake, and skin contact, by the reference dose (RfD) of a specific heavy metal. Meanwhile, as for carcinogens, the degree of cancer risk is calculated by multiplying the dose and the corresponding slope factor (SF). Moreover, there are some useful methods to assess the degree and risk of heavy metal contamination, including the Single-factor pollution index method, the Nemerow Comprehensive Pollution Index Method, the Potential ecological risk index method, and the Geoaccumulation index method.

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