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Review

Evaluation methods for assessing effectiveness of in situ remediation of soil and sediment contaminated with organic pollutants and heavy metals

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ABSTRACT

Soil and sediment contamination has become a critical issue worldwide due to its great harm to the ecological environment and public health. In recent years, many remediation technologies including physical, chemical, biological, and combined methods have been proposed and adopted for the purpose of solving the problems of soil and sediment contamination. However, current research on evaluation methods for assessing these remediation technologies is scattered and lacks valid and integrated evaluation methods for assessing the remediation effectiveness. This paper provides a comprehensive review with an environmental perspective on the evaluation methods for assessing the effectiveness of in situ remediation of soil and sediment contaminated with organic pollutants and heavy metals. The review systematically summarizes recent exploration and attempts of the remediation effectiveness assessment based on the content of pollutants, soil and sediment characteristics, and ecological risks. Moreover, limitations and future research needs of the practical assessment are discussed. These limitations are not conducive to the implementation of the abatement and control programs for soil and sediment contamination. Therefore, more attention should be paid to the evaluation methods for assessing the remediation effectiveness while developing new in situ remediation technologies in future research.

1. Introduction

Nowadays, a growing public concern has been aroused over the issue of soil and sediment contamination resulting from industrial and municipal waste discharge, mining activities, improper use of chemical fertilizer and pesticides, and wastewater irrigation (Zeng et al., 2013a; Awad et al., 2014; Teng et al., 2014; Tang et al., 2015). In USA, over 100,000 sites which suffered from soil pollution have been reported (He et al., 2015), while 3.33 million hectares of arable land in China are too contaminated to use according to the correspondence of Chen and Ye (2014). Organic pollutants and heavy metals are two main types of contaminants found in soil and sediment (Zhang et al., 2014; Zeng et al., 2016; Zhang et al., 2016b; Liu et al., 2017; Song et al., 2017). These contaminants are harmful, posing great threat to food safety, human health, and ecological environment. For example, exposure to polycyclic aromatic hydrocarbons (PAHs) was associated with the increase in breast cancer incidence (White et al., 2016). Consequently, the remediation of soil and sediment contaminated with organic pollutants and heavy metals is crucial (Tchounwou et al., 2012; Benami et al., 2013; Wang et al., 2013b; Zeng et al., 2015; Cheng et al., 2016b).

Much effort has been made to solve the problem of soil and sediment contamination. Many strategies and methods have been adopted for the pollution abatement and control (Gerhardt et al., 2009; Peng et al., 2009; Xu et al., 2012; Zhang et al., 2013a; Cheng et al., 2016a). Generally, the remediation technologies can be categorized into two main strategies: in situ remediation and ex situ remediation. In situ remediation is the treatment of the pollutant in original place. This strategy is aimed at the removal of pollutant from soil or sediment without moving the soil or sediment itself. Ex situ remediation is carried out by excavation and treating the contaminated soil or sediment somewhere else away from the site. Comparing these two strategies, in situ remediation offers a number of potential technical, economic, and environmental advantages (Bardos et al., 2000; Kuppusamy et al., 2016). In some cases, in situ remediation is the only means of pollutant removal when considering the scale of the contaminated area and the cost effectiveness. For a large scale of contaminated soil or sediment, in situ remediation is appealing because

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it causes less disturbance to the ecosystem, the operation is relatively simple, and the cost is more cheaper than the ex situ treatment (Carberry and Wik, 2001; Guiwei et al., 2008; Velimirovic et al., 2014).

After the implementation of pollution abatement and control programs, follow-up observations and evaluation methods are needed for assessment ensuring that the remediation goals are achieved and environmental criteria are met. Assessing the remediation effectiveness not only is a significant part of the whole remediation project, but also provides basis and guidance for the management and reuse of the soil and sediment after remediation (Apitz et al., 2005; Kuppusamy et al., 2016). Many in situ remediation methods, which can effectively reduce the pollutant concentration in soil and sediment, have been studied and applied extensively (Carberry and Wik, 2001; Peng et al., 2009; Kuppusamy et al., 2016). However, the evaluation methods for assessment of the in situ remediation need to be studied in greater detail. To our knowledge, current research on the remediation effectiveness assessment is scattered, and reviews which mainly elucidated the evaluation methods for assessment of in situ remediation of contaminated soil and sediment have not been reported previously.

The main purpose of this paper is to provide a comprehensive review with an environmental perspective on the evaluation methods for assessing the effectiveness of in situ remediation of soil and sediment contaminated with organic pollutants and heavy metals. This review systematically summarizes and discusses not only recent exploration and attempts for in situ remediation assessment, but also the gaps that limit their field application. The paper firstly introduces the methods employed by recent researchers for in situ remediation of soil and sediment contaminated with organic pollutants and heavy metals. Primary attention is given to the evaluation methods for assessing the remediation effectiveness based on the content of pollutants, soil and sediment characteristics, and ecological risks. Then, limitations and future research needs of the practical assessment are discussed. The structure of this review is illustrated in Fig. 1.

2. In situ remediation of contaminated soil and sediment

During the past decades, many technologies for in situ remediation of contaminated soil and sediment have developed rapidly. These technologies include physical, chemical, biological, and combined remediation methods (Fig. 2).

Physical remediation is the process of reversal or stopping of damage to the environment using physical technologies such as capping, flushing, thermal treatment, etc. In recent studies, in situ capping was more often used for controlling the sediment pollution and improving the quality of overlying water and aquatic habitat (Samuelsson et al., 2015; Zhu et al., 2016). In situ capping is the placement of a covering of clean or suitable isolating material over the contaminated soil or sediment. The primary functions of this method include physical isolation, stabilization of contaminated sites, and reduction of contaminant transport (Tomaszewski et al., 2006). For some sites contaminated with hydrophobic organic pollutants (HOCs), flushing can be a quick and effective method. In situ flushing is a technology for the cleaning of contaminants by pumping flushing agents into the bottom layer of soil or sediment. A mixture of water and surfactant is often applied to improve the aqueous solubility of contaminants during the flushing process (Begum et al., 2012; Lee et al., 2014; Tick et al., 2015). For volatile contaminants, thermal treatment is usually applied. In situ thermal treatment is to heat the subsurface for enhancing the mobilization, volatilization, and destruction of contaminants in soil or sediment. The heating methods mainly include conductive heating, electrical resistive heating, steam-based heating, and radio-frequency heating (Triplett Kingston et al., 2012; Peng et al., 2015; Samaksaman et al., 2016). Sometimes the heating process is assisted with air sparging to accelerate the volatilization of contaminants.

Chemical remediation is a kind of method that chemical reagent, reaction, and principles are used to remove contaminants. Remediation mechanisms include adsorption, catalysis, ion exchange, oxidation,

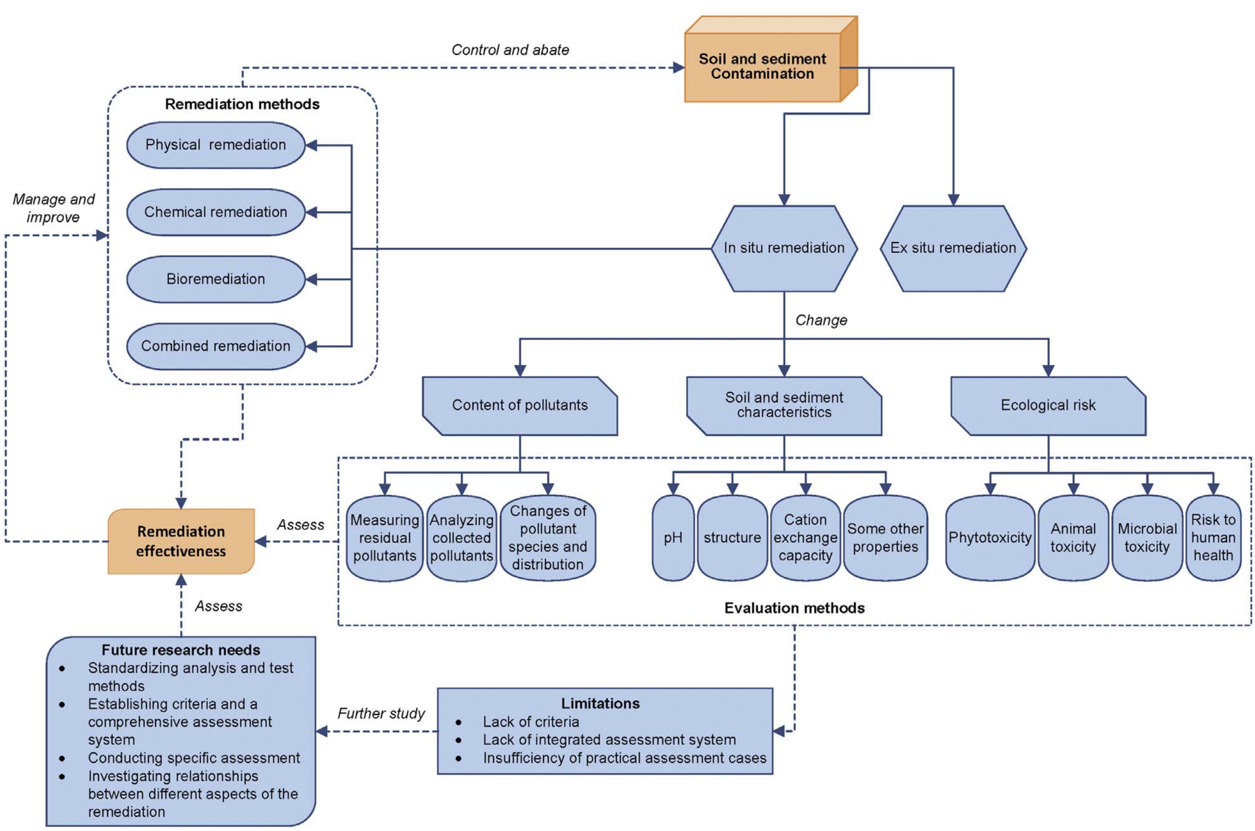

Fig. 1. Overview of the review structure.

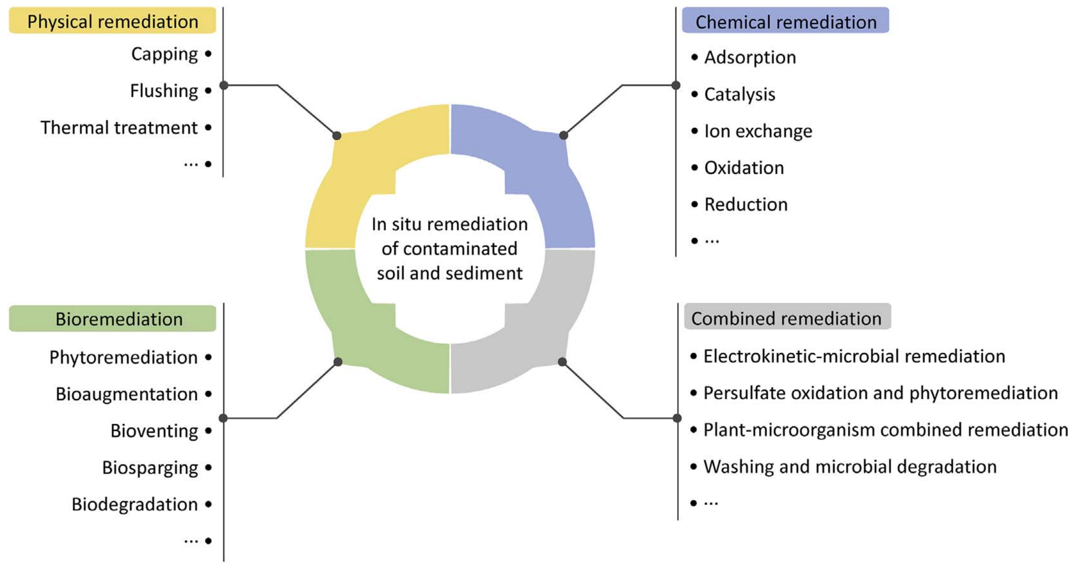

Fig. 2. Categorization of technologies for in situ remediation of soil and sediment contaminated with organic pollutants and heavy metals.

reduction, and so on (Gong et al., 2009; Feng et al., 2010; Hu et al., 2011; Usman et al., 2012; Kueper et al., 2014; Kang et al., 2015; Mao et al., 2015). Many researchers have been focusing their efforts on developing chemical remediation methods for in situ remediation of contaminated soil and sediment. Recently, using biochar and activated carbon for in situ remediation of contaminated sediment was adopted, which showed an effective reduction of both organic and inorganic pollutants in porewater (Gomez-Eyles et al., 2013; Choi et al., 2014). In a case of in situ stabilization of cadmium and lead in paddy soil using bentonite, multiple mechanisms including adsorption, ion exchange, complexation, and precipitation were probably involved in the remediation process (Sun et al., 2015). Latest research by Usman et al. (2016) indicated that Fenton oxidation showed great potential in treatment of recalcitrant organic contaminants, such as PAHs and polychlorinated biphenyls (PCBs). In order to improve the removal efficiency, Fenton oxidation coupling with other strategies like electrokinetic enhancement and magnetite catalysis has been studied widely (Usman et al., 2012; Wu et al., 2012; Bocos et al., 2015; Sandu et al., 2016).

Bioremediation is applied to clean up environmental pollutants by the aid of biological processes and reactions, which include absorption, transformation, and degradation caused by animals, plants, and microorganisms. Common bioremediation technologies include phytoremediation, bioaugmentation, bioventing, biosparging, and biodegradation. Recent research investigated the effectiveness of phytoremediation and bioaugmentation for bioremediation of soil contaminated by heavy metals and petroleum (Agnello et al., 2016; Cai et al., 2016; Vigliotta et al., 2016). Phytoremediation is recognized as a green technology with good public perception and is widely used for the removal of pollutants, particularly heavy metals. This technology uses green plants and their associated microbial communities to remove, degrade, or stabilize inorganic or organic contaminants and can be further divided into phytoextraction, phytofiltration, phytostabilization, phytovolatilization, phytodegradation, rhizodegradation, and phytodesalination (Ali et al., 2013; Ansari et al., 2015). Bioaugmentation is the practice of adding selected microorganisms into soil or sediment to enhance the biological activity of degradation and accelerate the removal of pollutants (Fotidis et al., 2014; Zhou et al., 2016). It is conceivable that a bioaugmentation-assisted phytoremediation was more helpful in treating contaminated sites compared to bioaugmentation or phytoremediation applied alone (Agnello et al., 2016). As for bioventing and biosparging, air or other gas is injected into the vadose zone and the saturated zone, respectively, to remove volatile pollutants or stimulate biodegradation. These two technologies were found to be more effective when paired with each other (Gillespie and Philp, 2013). Biodegradation plays a significant role in bioremediation. Biodegradation is the process through which organic substances are broken down by living organisms. Other bioremediation technologies are often accompanied with biodegradation when they are used for treating organic pollutants (Chen et al., 2015).

Combined remediation involves two or more different types of physical, chemical, or biological remediation technologies. The combination of diverse technologies can not only overcome the problems caused by using any one technology alone, but also take advantages of all and enhance the remediation efficiency. Therefore, it has also attracted much attention of researchers from all over the world (Huang et al., 2012; Chen et al., 2013; Khan et al., 2013; Gong et al., 2015; Chen et al., 2016; Lin et al., 2016).

3. Assessment of in situ remediation effectiveness

Whether a certain technology is effective for solving the problems of soil and sediment contamination, it requires scientific tests and evaluations. Here, the “effective” not only means the remarkable effect of the applied method on reducing the amount and toxicity of pollutants, but also refers to less disturbance to the natural environment. In actual remediation projects, the method is effective only when the remediation goals are achieved and environmental criteria are met. For assessment of in situ remediation effectiveness, recent studies mainly focus on three aspects: the content of pollutants, soil and sediment characteristics, and ecological risks.

3.1. The content of pollutants

The content of pollutants is a particularly reliable aspect during the remediation process when the hazardous substances are fully identified. Conventional evaluations of remediation effectiveness are based on the measurement of pollutant concentrations before and after remediation. Several evaluation methods concerning the content of pollutants are introduced as follows.

3.1.1. Measurement of residual pollutants

A part of pollutants can be removed through remediation treatment, while the residual fraction of pollutants is usually measured for calculating removal rate with the following equation:

$$\text{Removal rate (\%)} = (1 - A/B) \times 100 \quad (1)$$

where A is the residual fraction of pollutants after remediation, B is the total amount of pollutants before remediation.

For accurate measurement of the residual pollutants in soil and sediment, researchers have developed many methods (Zhang et al., 2007; Fan et al., 2008; Tang et al., 2008; Zhang et al., 2015; Zhang et al., 2016a). In general, samples need to be digested before detecting the content of heavy metals. Besides conventional acid digestion (Weng et al., 2014), microwave-assisted digestion (Hu et al., 2013) and ultrasonic-assisted digestion (Matong et al., 2016) are also used for heavy metal digestion. HF, HNO_3, $HClO_4$, HCl, H_2SO_4, and H_2O_2 are commonly used digesting agents. After the process of digestion, the content of heavy metals is quantitatively analyzed with precise detecting instruments. Generally, heavy metals can be detected by atomic absorption spectrometry (AAS), inductively coupled plasma-atomic emission spectrometry (ICP-AES), or inductively coupled plasma-mass spectrometry (ICP-MS). For organic pollutants, they need to be gathered by extractants before detecting them. The extraction is usually conducted by Soxhlet extraction (Schmidt et al., 2014), microwave extraction (Merdassa et al., 2013), ultrasonic extraction (Jurado-Sanchez et al., 2013), or pressurized fluid extraction (Fuller and Gautam, 2016) with *n*-hexane, dichloromethane, or acetone. High performance liquid chromatography (HPLC), liquid chromatography-mass spectrometry (LC-MS), gas chromatograph (GC), and gas chromatograph-mass spectrometer (GC–MS) are often used to detect the organic pollutants.

3.1.2. Analysis of collected pollutants

During the processes of some remediation treatments, the removed pollutants will be collected for the purpose of recycling or preventing secondary pollution. According to the conservation of mass, the more pollutants are collected, the fewer pollutants remained in soil or sediment. Thus, the collected fraction of pollutants can be used as an indirect evaluation parameter.

Phytoremediation through phytoextraction for remediating soil and sediment contaminated with heavy metals is a promising technology. The accumulated heavy metals can be recycled by combustion of the plant material after harvests (Willscher et al., 2013). Measuring the content of heavy metals in the ash provides useful information about the ability of the plant to accumulate heavy metals from soil and sediment, as well as the removal efficiency of the remediation method. Additionally, the metal uptake ratio, a quantitative parameter, can be calculated as the content of heavy metals in the whole plant to that in the corresponding growth soil or sediment (Hassan et al., 2013).

Thermal treatment of volatile pollutants in soil and sediment requires to collect the vapor. Collecting the vapor can prevent air pollution caused by pollutant volatilization. Moreover, the collected vapor can be used for recycling and monitoring the concentration change of pollutants in the vapor. Generally, a vapor cap is constructed or installed on the top of the remediation locality. Vapors are collected under vacuum created by extraction blowers (Kempa et al., 2013; Ma et al., 2015; Scholes et al., 2015).

In some circumstances, the concentration of pollutant is difficult to measure directly. For example, in a case of in situ stabilization, the digestion and extraction methods mentioned above are not suitable for the evaluation, because the stabilized pollutants still remain in the soil or sediment. It cannot be simply divided into a stabilized part and an unstabilized part. Therefore, more suitable methods are developed. Some researchers used polyethylene passive samplers (Choi et al., 2014; Pisanello et al., 2015) or Rhizon soil moisture samplers (Park et al., 2011; Sumon et al., 2012) to measure the reduction of contaminants in pore water. Pore water is the water existing in the pores between the soil or sediment particles. The pore water contains dissolved pollutants, and the concentration change of these pollutants can be a useful indicator of the remediation effectiveness in some ways. Under natural conditions, pollutants in sediment are likely to diffuse from sediment into overlying water through the sediment-water interface. Thus, determining the pollutant concentration in overlying water can be a feasible method to evaluation the potential risk and the remediation effectiveness (Zeng et al., 2013b; Xu et al., 2014; Vardy et al., 2015; Song et al., 2017). In some cases, the effectiveness of in situ remediation is estimated by measuring the pollutant concentrations in pore water or overlying water and then using equilibrium partitioning relationships to calculate the content of pollutants in soil and sediment (Gomez-Eyles et al., 2013; Han et al., 2015). However, one study showed that the remediation treatments have the potential to change characteristics of soil and sediment, and then affect the partitioning behavior of pollutants (Pan et al., 2012). The changes in pollutant partitioning behavior can affect the validity of assessing the remediation effectiveness with this method, which should be taken into account when choosing the method for assessment.

3.1.3. Changes of pollutant species and distribution

In situ immobilization or stabilization technology aims at increasing the stabilization of pollutants in soil and sediment by reducing their mobility, bioavailability, and toxicity. These properties strongly depend on the chemical speciation of pollutants. After treatment, determining the content of pollutants transformed into species with low-toxic or nontoxic is an effective way for the assessment. In sequential extraction methods, heavy metals are divided into exchangeable, carbonate bounded, Fe/Mn oxides bounded, organic matter bounded, and residual metal fractions (Islam et al., 2015). Among these defined chemical speciation, exchangeable fraction is considered as the most active and environmentally available part, while residual fraction is most stable and has little bioavailability. After remediation, the increase of residual fraction or/and the decrease of exchangeable fraction indicate a good remediation effectiveness of the adopted method (Jeon et al., 2015). For organic pollutants, the measurement of intermediate products during the degradation process can not only be useful for understanding the degradation mechanism, but also reflect the amount of the pollutants that have already been degraded.

Generally, the distribution characteristics of pollutants will change after remediation. Here, distribution change refers to the alteration of spatial position. In a study of in situ remediation of PAHs contaminated soils (Ma et al., 2013), different oxidation methods caused diverse distribution of PAHs in removing parts, soil residue parts, recycling parts, and supernatant. The results indicated different remediation ability of the chosen oxidants. In addition, the remediation activities sometimes cause secondary pollution through altering the distribution of pollutants (McGuire et al., 2014). Therefore, studying the change of pollutant distribution would provide a possible way to evaluate the remediation effectiveness.

3.2. Alterations of soil and sediment characteristics

The remediation treatments can change the soil and sediment characteristics when dealing with the pollutants (Table 1). The alterations of soil and sediment characteristics, such as pH, structure, cation exchange capacity (CEC), organic carbon and electrical conductivity, have significant impacts on the migration behavior of pollutants (Sassman and Lee, 2005; Rodriguez-Cruz et al., 2006; Uchimiya and Bannon, 2013; Zhang et al., 2016c). Thus, the subsequent remediation effectiveness will be influenced. Additionally, the change of soil and sediment characteristics also affects the reusing modes and directions after remediation.

3.2.1. pH

Acid-base property of soil or sediment is expressed as pH value, which is under the influence of organic matter, bed rock, mineral substances, and carbon dioxide in the soil or sediment. The pH value is a basic parameter when analyzing the characteristics of soil and

Table 1
Alterations of soil and sediment characteristics after different in situ remediation treatments.

Characteristic	Pollutants	Remediation treatment	Effect	Reference
pH	Cd, Zn, and Pb	Biochar-induced immobilization	Increased	Houben et al. (2013b)
	Cd	Immobilization by mining, industrial, and agricultural solid by-products	Increased	Wang et al. (2013a)
	Cd and Zn	Phytoextraction in combination with amendments based on elemental sulphur and $(NH_4)_2SO_4$	Decreased	Amoakwah et al. (2014)
	Cd	Immobilization by sepiolite and palygorskite	Increased	Liang et al. (2014)
	PCBs	Electrokinetic remediation in combination with activated persulfate	Decreased	Fan et al. (2016)
	Cr	Reduction by acidified hydrazine hydrate	Increased	Ma et al. (2016)
	Heavy metals	Phytoremediation in combination with ferrous sulfate	Decreased	Sigua et al. (2016)
Structure	Metals, As, and phenanthrene	Remediation with biochar and iron filing amendments	Deteriorated	Sneath et al. (2013)
	Metal and metalloid contaminants	Remediation with iron-based amendments	Improved	Gargiulo et al. (2014)
	Pb, Zn, and Cd	EDTA washing	Remained stable	Zupanc et al. (2014)
	Pb and Zn	Washing assisted with reagents and ultrasound	Destroyed	Wang et al. (2015)
	As	Oxalate-based washing	Improved	Lee et al. (2016)
	Pb and pyrene	Phytoremediation (rhizodeposition)	Improved	Li et al. (2016)
Cation exchange capacity	Cu and Cd	Remediation using compost with inorganic amendments	Increased	Gadepalle et al. (2009)
	Cu, Cd, and Ni	Immobilization by biochar	Increased	Uchimiya et al. (2010)
	Heavy metals	Stabilization by biochar	Increased	Fellet et al. (2011)
	Cd, Pb, and Zn	Phytoremediation	Increased	Houben et al. (2013a)
	Zn	Adsorption by manure- and lignocellulose-derived biochars	Decreased	Dai et al. (2016)
	Petroleum oil	Low-temperature thermal desorption treatment	Decreased	Yi et al. (2016)
Enzyme activity	Heavy metals	Stabilization by pig slurry and compost	Increased	Pardo et al. (2014)
Electrical conductivity	Cu and pyrene	Enhanced-electrokinetic remediation with oxidants and pH control	Increased	Cang et al. (2013)
Capacity of nutrients and water retention	Heavy metals	Stabilization by biochar	Enhanced	Fellet et al. (2011)
Organic matter content	Metals and As	Remediation with compost and biochar amendments	Increased	Beesley et al. (2014)

sediment. For in situ immobilization of heavy metals, alkaline amendments are often used to reduce the mobility, leachability, and bioavailability of heavy metals in soil and sediment. Higher pH favors metal precipitation and simultaneously decreases the metal solubility. Therefore, the pH value usually increased after such remediation (Houben et al., 2013b; Wang et al., 2013a; Liang et al., 2014). However, excessive change of the pH value is not allowed. To avoid alkalization of the environment, acidified amendments are applied (Ma et al., 2016), and some remediation methods intentionally lower pH to facilitate migration of pollutants (Cang et al., 2013; Mena et al., 2016). Amoakwah et al. (2014) used some pH lowering amendments (elemental sulphur and $(NH_4)_2SO_4$) to enhance solubilization of Cd and Zn so as to facilitate phytoextraction of the heavy metals. After applying 200 and 16 mmol elemental sulphur and $(NH_4)_2SO_4$), the soil pH decreased by 2.6, 2.1, and 1.8 units, respectively, when compared with the control. The production of H^+ during the oxidation processes of elemental sulphur and $NH_4{}^+$ is the reason behind the decrease of soil pH. Similar results can also be found in the research by Sigua et al. (2016). Obviously, in these examples, the amendment dosage is the key factor. It is important to choose an optimal addition amount to ensure that the phytoextraction of heavy metals is facilitated while the potential risk caused by the movement of solubilized metals into groundwater is not increased.

3.2.2. Structure

Soil or sediment structure is the arrangement of particles and associated pores in soil or sediment across the size range from nanometers to centimeters (Oades, 1993). It plays an important role in the function of soil and sediment. In situ remediation treatments have considerable influences on the structure of soil and sediment. Researchers have reported that iron filings as amendments negatively impacted the soil structure (Sneath et al., 2013). Iron amendments can cause soil particles to cement together and reduce the soil porosity. The reason behind this is the high adsorption ability of iron oxides towards silicate minerals, which can form bridges between iron oxides and other soil particles, contributing to the cementation (Colombo and Torrent, 1991). But Gargiulo et al. (2014) found that iron-based amendments can increase soil porosity in the range of transmission pores (50–500 μm) in a low-plastic shrink-swell soil thus improve the soil structure. In a washing process of the soil contaminated by heavy metals, ultrasonic-assisted desorption was used to enhance the removal rates of heavy metals (Wang et al., 2015). At the same time, however, soil structure would be destroyed by the ultrasonic cavitation. Since strong acids could destroy soil structure by mineral dissolution, Lee et al. (2016) used oxalate-based washing process to remove arsenic in contaminated soil. Their results showed that using oxalate as washing agent could achieve high arsenic removal rate and minimize the destruction of soil structure. Additionally, an increase of organic matter content by using organic amendments could result in the decrease of soil bulk density and the increase of soil aeration because of the redistribution of pore space. (Tejada et al., 2006).

Alterations of soil and sediment structure (density, porosity, and pore size distribution) strongly influence the contaminant transport parameters (Safadoust et al., 2012). Facilitated transport of contaminants can cause more severe groundwater pollution, while blocked transport can result in that contaminants remain in soil and sediment for a longer time, bringing about a lasting threat towards surroundings. In the growth process of plants, root distribution and their ability to take up nutrients and water are affected by soil structure. Favorable soil structure is important to maintaining soil fertility, reducing erodibility, and increasing agricultural productivity (Bronick and Lal, 2005). Hence, evaluating the structure change of amended soil should be necessary for agricultural reuse.

3.2.3. CEC

CEC is a measurement of the ability of soil or sediment to adsorb cations in a form that these cations can be easily desorbed by their competing ions (Bache, 1976). Cation exchange is caused by negatively charged colloidal clay and humus particles of the soil matrix. To ensure electric neutrality of the whole, positively charged ions are adsorbed by

Table 2
Applications of ecotoxicological tests for assessing in situ remediation effectiveness.

Experimental type	Test species	Remediation treatment	Assessment results	Reference
Phytotoxicity test	Mung bean (*Phaseolus radiatus* L.)	Bioremediation with Cd-resistant strains	The strains were able to reduce Cd accumulation in roots and shoots.	Rani et al. (2012)
	Garden cress (*Lepidium sativum* L.)	Remediation with biochar, activated carbon, and multi-walled carbon nanotubes	These materials reduced sediment toxicity (inhibition of seed germination and root growth).	Jośko et al. (2013)
	Edible rape (*Brassica campestris* L.) and Chinese cabbage (*Brassica rapa* ssp. *pekinensis*)	Immobilization by sodium carboxymethyl cellulose stabilized nanoscale zero-valent iron	The remediation exerted an inhibitory effect on plant growth.	Wang et al. (2014)
	Tomato (*Lycopersicon esculentum* L.)	Immobilization by biochar	Metal-induced toxicities in tomato plant were reduced.	Herath et al. (2015)
	Ryegrass (*Lolium perenne* L.)	Phytoremediation enhanced by Pb immobilization using nano-hydroxyapatite	Ryegrass biomass was significantly increased.	Jin et al. (2016)
	Lettuce (*Lactuca sativa* L.)	Fenton oxidation and remediation with nanoscale zero-valent iron	Both of the remediation treatments showed to be prejudicial for lettuce seeds.	Rede et al. (2016)
	Cabbage mustard (*Brassica alboglabra* Bailey)	Remediation with biochar-supported nano-hydroxyapatite	The upward translocation capacity of Pb in soil-plant system was effectively reduced.	Yang et al. (2016b)
Animal toxicity test	Benthic communities	Remediation with activated carbon	The remediation showed negative effects on benthic communities.	Kupryianchyk et al. (2012)
	Earthworm (*Eisenia andrei*)	Remediation with organic and inorganic amendments	The inorganic amendment did not significantly reduce toxicity, while the organic amendment made the metal-contaminated soil more toxic.	González et al. (2013)
	Springtail (*Folsomia candida*) and earthworm (*Aporectodea caliginosa*, *Eisenia fetida*)	Remediation with activated carbon, biochar, and ferric oxyhydroxide	These soil amendments had variable but never strongly deleterious effects on the invertebrates studied.	Hale et al. (2013)
	Earthworm (*Eisenia fetida*)	Remediation using pig slurry and compost in combination with hydrated lime	Amendments reduced the soil toxicity to invertebrates.	Pardo et al. (2014)
	Polychaete (*Hediste diversicolor*) and clam (*Abra nitida*)	Capping with activated carbon	Bioaccumulation of PCBs and PAHs in benthic fauna was reduced.	Samuelsson et al. (2015a)
	Earthworm (*Lumbricus terrestris*)	Remediation with two types of composted manure, composted biosolids, and biochar	Addition of aged dairy manure compost and presence of grasses was effective in reducing the accumulation of soil-derived Pb in earthworms.	Centofanti et al. (2016)
	Nematode (*Caenorhabditis elegans*)	Remediation with zero-valent ion nanoparticles	Zero-valent ion nanoparticles induced infertility risk in nematode.	Yang et al. (2016a)
Microbial toxicity test	Bacteria (*Photobacterium phosphoreum*)	Biostimulation in combination with modified Fenton oxidation	Microbial toxicity of the treated soil extract reduced greatly after the chemical oxidation process.	Gong (2012)
	Bacteria (*Vibrio fischeri*)	Remediation with activated carbon, biochar, and ferric oxyhydroxide	All amendments reduced the inhibition of luminescence.	Hale et al. (2013)
	Microbial communities	Remediation with iron grit	The remediation treatment increased microbial diversity and respiratory activity.	Kaplan et al. (2013)
	Bacteria, actinomycete, and fungi	Stabilization by natural sepiolite	Functional diversity of soil microbial communities was recovered.	Sun et al. (2013)
	Microbial communities	Bioremediation with biostimulant	Diversity of sulfate reducing bacteria was enhanced.	Subha et al. (2015)
	Bacteria (*Escherichia coli*)	Biodegradation by plant-growth promoting *Acinetobacter* sp. AVLB2	Toxicological studies revealed non-toxic nature of 4-NA biodegraded metabolites.	Silambarasan and Vangnai (2016)
	Microbial communities	Immobilization by sodium alginate modified nanoscale zero-valent iron	The treatment increased bacterial taxa and improved bacterial abundance.	Huang et al. (2016)

soil particles (Bache, 1976). In the environmental area, CEC is widely used in characterizing soil and sediment for assessment of their fertility, nutrients retention capacity, and ability to prevent groundwater from pollution by cation contaminants. Implementing in situ treatments brings about significant impacts on CEC of soil and sediment. Acidification of soil and sediment decreases CEC during the remediation process (Dai et al., 2016). In an acidic condition, the negative charges of soil or sediment decrease, subsequently causing nutrients loss and CEC reduction as cations are disproportionately replaced with H^+ (Sharma et al., 2015). Besides, Gadepalle et al. (2009) found that using compost for remediation of the soil contaminated by copper and cadmium could increase CEC with the enhancement of organic matter content. This may be because organic matter makes significant contributions to the soil CEC (Helling et al., 1964). Some studies have shown that the CEC of soil and sediment increased after using biochars as amendments because biochars can cause large cation exchange on themselves (Zhang et al., 2013a; Abujabhah et al., 2016). And the oxidation of surface functional groups on biochar also contributes to the CEC increase of the amended soil due to the increases of charge density per unit surface of biochar and surface area for cation adsorption after the oxidation process (Atkinson et al., 2010; Singh et al., 2014).

Higher CEC favors stabilization of heavy metals in soil and sediment. Heavy metals can exchange with K^+, Na^+, Ca^{2+}, Mg^{2+}, and other cations, finally being co-precipitated or forming complexes with humic matters. However, increased CEC of soil can reduce the uptake and accumulation of heavy metals by plants (Haghiri, 1974). Therefore, it is unfavorable for phytoremediation with hyperaccumulator. When combined biochar stabilization and phytoremediation for treating heavy metal contaminated sites, the antagonistic effect caused by CEC alteration should be taken into consideration.

3.2.4. Some other properties

Apart from the above-mentioned properties, in situ remediation can also influence the enzyme activity, electrical conductivity, nutrients and water retention, mechanical strength, and organic matter content of soil and sediment. For example, Pardo et al. (2014) measured the enzyme activities after using organic and inorganic amendments for remediation of a contaminated mine soil. Their results showed that this method had a positive effect on soil enzyme activities after remediation. The organic amendments increased cellulase, β-glucosidase, and urease activities of the soil, and the increase was observed to be higher in the soil treated by compost. Soil enzyme activity mainly results from cell secretion of animals, plant roots, and microorganisms in soil, as well as decomposition of organic debris. Moreover, soil enzyme activity is closely associated with soil fertility and structure. Therefore, determination of soil enzyme activity can also be useful for assessing the remediation effectiveness in some cases. Additionally, due to the transport of ions from electrolytes into the soil, electrical conductivity of soil increased after electrokinetic remediation (Cang et al., 2013). Applying biochar for in situ remediation could enhance the capacity of nutrients and water retention (Fellet et al., 2011) as well as the mechanical strength of the soil and sediment (Lu et al., 2014). Addition of organic amendments like compost might increase organic matter content of the soil (Beesley et al., 2014). All these changes are significant in characterizing soil and sediment for effectiveness evaluation and land reusing after remediation.

3.3. Ecological risks and ecotoxicological tests

Contaminants in soil and sediment are of growing concern for their potentials to cause adverse ecological effects. Ecological risk is the likelihood of adverse ecological effects that are occurring or may occur as a result of exposure to one or more stressors (U.S. EPA, 1992). Herein, the stressors refer to the contaminants in soil and sediment, as well as the adverse factors caused by remediation activities. For evaluating remediation effectiveness, the assessment focuses on the change of original ecological risks and probable new risks caused by the remediation activities. Ecotoxicological tests are an important part of ecological risk assessment. Due to their sensitivity and reflection of the overall toxicity, ecotoxicological tests can provide a comprehensive assessment of the environmental state, which allows them to be used for evaluating the remediation effectiveness (Pardo et al., 2014). Some of recent applications of ecotoxicological tests for assessing in situ remediation effectiveness are presented in Table 2.

3.3.1. Phytotoxicity test

Phytotoxicity is defined as detrimental effects on various physiological processes (e.g., seed germination, seedling growth, and water uptake) of plants caused by specific substances or growing conditions (Deepesh et al., 2016). In order to conduct valid phytotoxicity tests, seeds of the test plant should be uniform, sensitive, and amenable. Besides, it should be considered that the tested species can give reliable and reproducible results, and they meet the validity criteria of the test. Cabbage mustard, mung bean, garden cress, lettuce, and ryegrass are recently used in phytotoxicity tests for assessing remediation effectiveness of contaminated soil and sediment (Rani et al., 2012; Jośko et al., 2013; Jin et al., 2016; Rede et al., 2016; Yang et al., 2016b; Song et al., 2017). Use of phytotoxicity test for evaluation of the remediation effectiveness is based on the alteration of hazardous substances in soil and sediment as well as plant growth conditions. The content, distribution, and species of pollutants are key factors influencing seed germination and plantlet growth. Generally, the more chance that seed and plantlet contact with the pollutant the more toxic it is, and the pollutant exhibits stronger toxicity when it is easy to be absorbed by plant. Additionally, soil and sediment properties will change during the remediation. Properties including pH, soil or sediment structure, CEC, electrical conductivity, nutrients and water retention, and organic matter content significantly affect the plant growth. Alteration of these properties changes the phytotoxicity of the soil and sediment, which implies the ecological impact of the adopted remediation technology.

Reduction and stabilization of the pollutants in soil and sediment usually diminish the phytotoxicity. Yang et al. (2016b) conducted phytotoxicity assessment for in situ remediation of lead contaminated soil by biochar-supported nano-hydroxyapatite. Their results showed that the materials could effectively reduce the concentration of lead in the aerial part of cabbage mustard to 0.1 mg/kg, a level which is below the tolerance limit (0.3 mg/kg). Such evaluation can be helpful in restoring the soil quality for planting. Jin et al. (2016) also used nano-hydroxyapatite for immobilizing lead in soil. According to their experiments, this remediation method increased the ryegrass biomass, and the removal of lead from soil by ryegrass was enhanced. However, not all of the remediation treatments are conducive to the reduction of phytotoxicity. Rede et al. (2016) investigated the ecotoxicological impact of two soil remediation treatments on lettuce seeds. In their experiments, both Fenton oxidation and nanoremediation showed negative impact on the development of lettuce seeds. A decrease of seed germination up to 45% and a root elongation inhibition around 80% were observed. Basing on these results, the use nanoscale zero-valent iron particles for remediation might cause the deposition of iron species in the root, inhibiting the uptake of nutrients and water. As for Fenton oxidation, the production of free radicals appears to contribute to the increased phytotoxicity. Additionally, the deterioration of plant growth conditions (e.g., water availability, temperature, and properties of soil and sediment) could also increase the phytotoxicity after remediation (Jośko et al., 2013; Song et al., 2017).

3.3.2. Animal toxicity test

Animal toxicity test investigates the effect of unfavorable factors on morphological, physiological, behavioral, and ecological characteristics of soil or sediment fauna. For soil contamination, earthworms are widely used as test species due to their intimate contact with soil and their importance in terrestrial food webs (González et al., 2013; Pardo et al., 2014; Centofanti et al., 2016). Besides earthworms, other animal species such as enchytraeids, collembolan, mites, isopods, nematodes, and protozoans also show a good potential for soil ecotoxicity assessment (Haimi, 2000). As to the ecotoxicity assessment of contaminated sediment, benthic invertebrates are used most often. Generally, benthic invertebrate species are abundant in aquatic ecosystem. Many benthic faunas are sensitive to sediment contaminants, showing specific response to the change of pollutants (Rodriguez-Iruretagoiena et al., 2016). Moreover, benthic invertebrates play important roles in ecosystem, and reflect the biodiversity of benthic communities (De Jonge et al., 2012). For these reasons, benthic invertebrates are recommended species for testing ecotoxicity of sediment (Lim et al., 2013; Diepens et al., 2014). Additionally, fishes and their biomarkers have been also explored for assessing sediment remediation (Meier et al., 2013).

Good remediation strategies can improve habitats of animals in soil and sediment, resulting in an increase of biomass and biodiversity. Meier et al. (2013) conducted bioassessment for a sediment remediation. Their results showed that the remediation effectively reduced contaminant concentrations and exposure of fish to PAHs, and improved fish assemblages with a increase of 60% in the index of biotic integrity scores in remediated river sections. And their further studies indicated that monitoring exposure of fish and benthic macroinvertebrates to sediment contaminants before and after remediation can be used to evaluate the remediation effectiveness (Meier et al., 2015). Kupryianchyk et al. (2012) used activated carbon (AC) for sediment remediation. According to their research, AC showed a positive trend on species abundance after the remediation with AC for 3 months, whereas a negative trend was observed on Pisidiidae and Lumbriculidae after 15 months. Based on this problem, using AC for sediment remediation only suitable for high concentration system of HOCs. And the concentration of AC should not be too high to ensure that the positive effects of AC in reducing HOCs toxicity outweigh the negative effects of AC application on benthic communities. In a case of remediation of metal-contaminated soil, the organic amendments caused an adverse effect (González et al., 2013). The organic compost amendment at concentrations above 25% increased available metal concentrations and electrical conductivity in the amended soil, leading to the death of all earthworms in undiluted soil and a complete reproduction inhibition. These results demonstrated that animal toxicity test is effective as a tool for assessing in situ remediation effectiveness.

3.3.3. Microbial toxicity test

Microbial toxicity refers to the adverse effects on physiological processes and ecological functions or characteristics of microorganisms resulting from exposure to unfavorable factors. Compared with toxicity tests using plants and animals, microbial toxicity tests of soil and sediment attract more attention. On the one hand, microorganisms are generally highly abundant in soil and sediment, and closely associated with the soil fertility and the degradation of pollutants. On the other hand, the microbial toxicity test is simple, fast, and inexpensive, and only a small quantity of sample is needed to induce physiological changes (Beelen and Doelman, 1997; Huang et al., 2008; Song et al., 2016). Currently, available microbial tests can be classified into single-species tests, community-level assessments based on functionality, biomass and physiological processes, and molecular methods (Diepens et al., 2014). These methods are effective for determining the toxicity of soil and sediment. Remediation treatments reduce pollutant concentration and change properties of soil and sediment, and these alterations have significant impacts on soil or sediment toxicity to microorganisms, which makes it possible to use microbial properties for assessment of in situ remediation effectiveness. For a better interpretation of soil microbial properties as indicators of soil quality, Gómez-Sagasti et al. (2012) grouped microbial properties within a set of ecosystem health attributes of ecological relevance, including vigor, organization, resilience, suppressiveness, and redundancy (Fig. 3). Through linking the concepts together, different microbial indicators of soil quality can be understood from the perspective of ecosystem health attributes of ecological relevance.

In general, a reduction of pollutant amount enhances the soil and sediment health, with increased microbial biomass, microbial respiration intensity, and structural diversity of microbial communities, on account of that the stress and influence of the pollutant to microorganisms can be relieved by remediation treatments (Zhang et al., 2013b). Kaplan et al. (2013) used iron grit to conduct in situ remediation for controlling trace metal contamination. The microbial diversity and respiratory activity increased after the effective remediation, which demonstrated that monitoring soil microbial communities and their activity can be a useful tool for evaluating the effectiveness of in situ remediation of contaminated soil and sediment. Sun et al. (2013) assessed microbial communities in acidic soil after applying natural sepiolite for cadmium stabilization. The treatment significantly affected microbial populations. For example, at a cadmium level of 1.25 mg/kg, the bacteria count increased by 71.1 to 232.9% after the application of sepiolite. Moreover, their results showed that bacteria were the most sensitive species in the soil, followed by actinomycete and fungi. These microorganisms play an important role in the formation of soil aggregates and the production of plant growth promoters. After sepiolite treatment, the increased amount and biodiversity of microbial communities indicated that a certain functional recovery of the soil occurred. Gong (2012) adopted a combined method of biostimulation and modified Fenton oxidation for the remediation of weathered petroleum oil-contaminated soil. In his research, although there was a considerable reduction of the total petroleum hydrocarbons, the amount of microbes (called "hydrocarbon degraders" by the author) decreased sharply at week 6 after the chemical oxidation process due to the cell wall disintegration caused by H_2O_2 and radicals.

3.3.4. Risk to human health

One of the main purposes of the remediation activities is to reduce risk to human health. Generally, the remediation will diminish the risk of immediate exposure to hazardous substances. This exposure refers to the direct contact with the contaminants in soil and sediment. However, improper remediation of contaminated sites may induce potential health risks via food chain or other productive reuse of the contaminated sites (Wcisło et al., 2016). For example, after the remediation of an abandoned mine in Portugal, surface water in the mining-impacted area was found to contain several metals and arsenic, which leads to that the water cannot be used for human consumption or agriculture due to the carcinogenic health risks (Antunes et al., 2016). Investigation of health risks needs long-time observation and monitoring, and the time spans from a few months to even more than 10 years (Schoof et al., 2016). Compared with toxicity tests using plants, animals, and microorganisms, assessing risk to human health is likely to be more complicated and difficult. But even so, it is still necessary to conduct health risk assessment, which in turn reflects the remediation effectiveness after treatment.

3.4. Other evaluation methods

In addition to the evaluation methods introduced above, some other possible methods have also been explored for assessing the effectiveness of in situ remediation. Santini and Fey (2013) observed a spontaneous vegetation encroachment upon bauxite residue and applied it as an indicator of in situ remediation processes. In their assessment, bauxite residue in vegetated areas had lower pH, alkalinity, electrical conductivity, and total aluminum and sodium compared to unvegetated areas. Since the process of vegetation encroachment took a long time (over 30 years), the assessment is not suitable for common remediation cases, but helpful for long-term study of ecological impacts. Yang et al. (2016a) used a mathematical modelling method in a case of assessing the risk of zero-valent iron nanoparticles released from the in situ

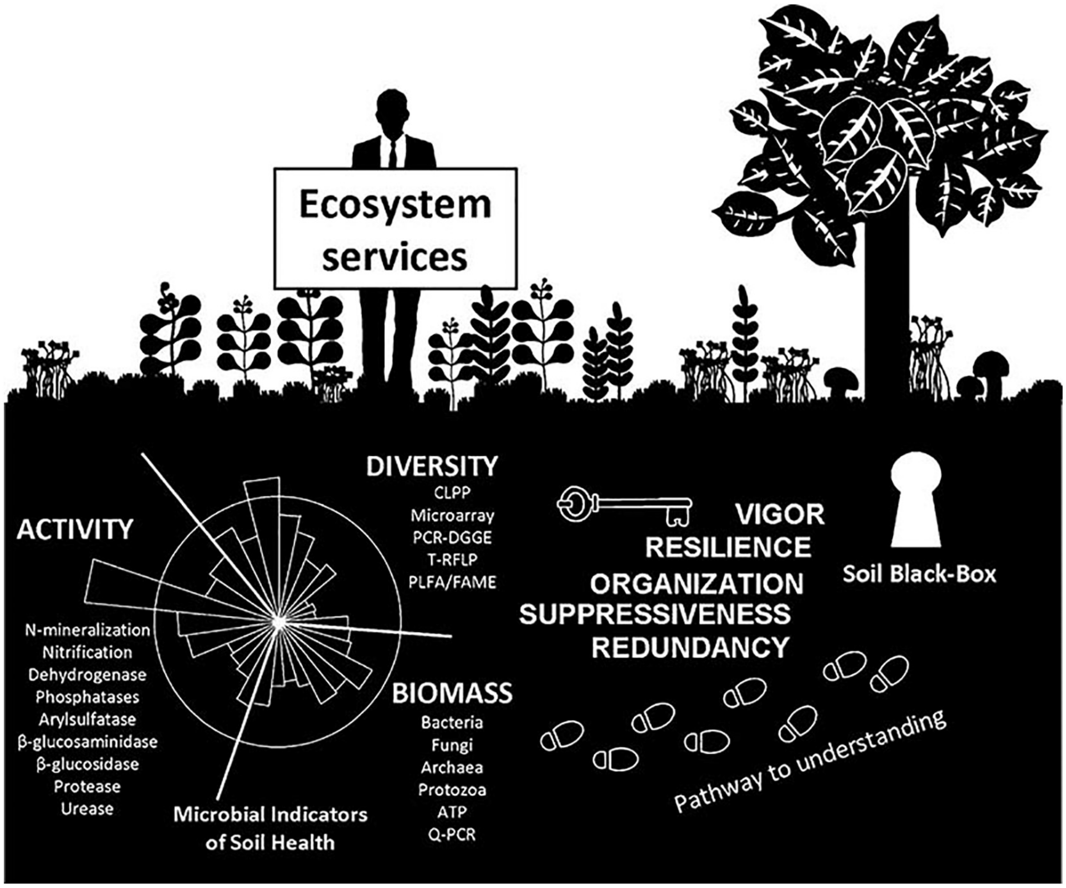

Fig. 3. An interpretation of soil microbial properties as indicators of soil quality.
Adopted from Gómez-Sagasti et al. (2012).

remediation. They developed a probabilistic risk assessment model based on *Caenorhabditis elegans* biomarker, and their model was validated to be useful for quantitatively assessing the potential environmental risks after remediation. Additionally, the combined evaluation which involves two or more different types of above-mentioned evaluation method has been being considered and explored for the assessment. However, there is still a lack of practical cases and some proposed evaluation methods are unfeasible at present. Therefore, it needs more development in the future.

3.5. *Validity of the evaluation methods*

A valid evaluation method can correctly reflect the changes of concentrations, toxicity, and bioavailability of the target pollutant. To date, relatively few studies that determined the validity of the evaluation methods have been reported. In most cases, researchers investigated only the changes of evaluation indicators after remediation. Song et al. (2017) investigated whether the phytotoxicity of sediment can be applied for assessing the remediation effectiveness. They used the changes of pollutant concentrations as a reference and studied the validity by comparing the Pearson correlation coefficients between evaluation indicators and associated parameters. According to their results, the phytotoxicity of sediment might inaccurately indicate the changes of pollutant concentrations after a long time remediation for the data showed both positive and negative correlations on the same group. In future, more research work and trials are need to verify the validity of the evaluation methods, especially for using soil or sediment characteristics and ecological risks in assessment.

4. Limitations and future research needs

Currently, there are some limitations when adopting the above-reviewed evaluation methods for assessment of in situ remediation of contaminated soil and sediment. Firstly, lack of criteria, including quality criteria for soil and sediment as well as technical criteria for remediation, makes it difficult for regulators to monitor the pollution and make decisions to remediation activities. Secondly, most of current assessments focus on a single aspect, lacking integrated assessment system. Researchers only partly studied the alteration of pollutant content, soil and sediment characteristics, or ecological risks. Thus, the results of assessment may not be convincing, and this is unfavorable for the project management and the enhancement of remediation effectiveness. Besides, the ecological assessment should be based on different level of ecosystem (single species, population, community, and the whole ecosystem), particularly when long-term impacts of the remediation need to be monitored (Rohr et al., 2016). Additionally, there are insufficient assessment cases in practical soil and sediment remediation. Actual situation is more complicated. The pollution of soil and sediment can also affect groundwater or atmosphere. And engineering difficulties, economic aspects, and social issues need to be considered.

Based on these gaps and challenges, future research is recommended as follows:

(1) Studies for establishing quality criteria of soil and sediment as well as technical criteria for remediation is encouraged. But before this, methods of pollutant analysis and ecotoxicological tests need to be standardized.

(2) There is a need to establish a comprehensive assessment system which is based on the quality criteria and technical criteria. Therefore, the relationship between various aspects of remediation assessment

needs to be explored.

(3) Specific assessment for in situ remediation of contaminated soil and sediment is strongly recommended. On the one hand, the assessment can be conducted according to the specific pollutant. On the other hand, different reuse purposes should adopt different assessment methods. For example, the health risks needs to be studied before the amended soil being reused for agricultural production.

(4) Coordination between assessment methods, engineering technologies, and economic aspects under field conditions need to be further investigated, ensuring that the assessment methods are indeed feasible.

5. Conclusions

This review presents the latest applications and knowledge on the evaluation methods for assessment of in situ remediation of contaminated soil and sediment from an environmental perspective. Physical, chemical, biological, and combined remediation methods have been being explored and adopted for solving the problems of soil and sediment contamination. Preliminary attempts and potential methods for assessing the remediation effectiveness are systematically summarized for the first time basing on the content of pollutants, soil and sediment characteristics, and ecological risks. When implementing abatement and control programs for soil and sediment contamination, the adopted method should not only have a remarkable effect on reducing the amount and toxicity of pollutants, but also cause less disturbance to the natural environment. In actual remediation projects, the ecological impact assessment needs to be involved in the establishment of remediation goals and environmental criteria. Therefore, more attention should be paid to the evaluation methods for assessing the remediation effectiveness while developing new remediation technologies in future research.

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