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Authors

Zeng, Saiqi

Dai, Zhongmin

Ma, Bin

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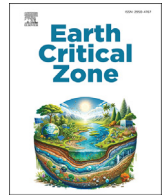
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Environmental interactions and remediation strategies for co-occurring pollutants in soil



Saiqi Zeng^{a,b}, Zhongmin Dai^{a,b}, Bin Ma^{a,b}, Randy A. Dahlgren^{a,c}, Jianming Xu^{a,b,*}

^a Institute of Soil and Water Resources and Environmental Science, College of Environmental and Resource Sciences, Zhejiang University, Hangzhou, China

^b Zhejiang Provincial Key Laboratory of Agricultural Resources and Environment, Zhejiang University, Hangzhou, China

^c Department of Land, Air and Water Resources, University of California, Davis, CA, 95616, USA

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ABSTRACT

With the development of industrialization, urbanization and modern agriculture, a myriad of pollutants have accumulated in soils, deteriorating soil health and food safety, and causing risks to human and ecosystem health. Environmental interactions and remediation strategies to address a single pollutant in soil have been widely documented, whereas information concerning remediation of co-occurring pollutants in soil, which is more prevalent and important for agricultural management, has received far less attention. The paucity of information regarding remediation strategies for multiple pollutants can be ascribed to the complicated interactions between physiochemical and biological processes affecting co-occurring pollutants in soil. Research on soil co-contaminants has received greater attention in recent years as advancements in single pollutant remediation strategies have progressed, thereby building a fundamental foundation to investigate more complex co-contamination issues. This review summarizes the current understanding of environmental interactions and remediation strategies for individual pollutants in soil, and then focuses on how this information can be used to develop effective remediation strategies for co-occurring pollutants in soil. Based on this synthesis, we identify important knowledge gaps and opportunities for future work to advance the science of pollutant remediation in soil systems.

1. Introduction

With the rapid development of industry, urbanization and agriculture in China during recent decades, the release of a myriad of pollutants into the environment has severely deteriorated environmental quality, causing widespread soil pollution (Zhao et al., 2015). Among critical environmental issues, contamination of arable land threatens food safety, and thus is of great concern for human health. A nation-wide survey of Chinese soils identified that 16.1% of all soils and 19.4% of agricultural soils are contaminated by various pollutants, including potentially toxic elements (PTEs; such as Cd, As, Cr, Hg and Pb) and organic pollutants (OPs; including hexachlorocyclohexane (HCH), dichlorodiphenyltrichloroethane (DDT) and polycyclic aromatic hydrocarbons (PAH)). The deleterious effects and toxicity mechanisms of pollutants on soil ecosystems have been widely reported, such as Cd²⁺ inhibiting photo-activation of photosystem II by competitive binding to essential Ca²⁺ sites (Faller et al., 2005); the presence of Cu²⁺, Cd²⁺ and Zn²⁺ adversely

affecting soil microbial biomass and bacterial community structure (Song et al., 2018); and exposure to hexabromocyclododecane (HBCD) altering the growth, locomotion behavior, reactive oxygen species (ROS) formation, lipofuscin accumulation, and cell apoptosis of nematodes (Wang et al., 2018a). Scientists have also investigated the complex interactions among pollutants in soil systems that affect their fate and transport in the environment, including sorption on soil colloids (Jiang et al., 2020a; Van Groeningen et al., 2021; Xu et al., 2019), biosorption by microbes (Jin et al., 2020), uptake, accumulation and transport by plants (Liu et al., 2019; Xu et al., 2021) and degradation of organic pollutants (Muskus et al., 2020). This fundamental understanding of pollutant behavior in soil systems provides essential guidance to design remediation strategies, such as addition of amendments (Qiu et al., 2020; Xue et al., 2018), phytoremediation (Ma et al., 2001), and bio-degradation (Qin et al., 2016; Sun et al., 2019).

While previous studies greatly improved our understanding of soil pollution and remediation strategies, they primarily focused on a single

* Corresponding author. Institute of Soil and Water Resources and Environmental Science, College of Environmental and Resource Sciences, Zhejiang University, Hangzhou, China.

E-mail address: jmxu@zju.edu.cn (J. Xu).

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pollutant. Conversely, the ubiquitous occurrence of multiple pollutants requires a multifaceted remediation approach based on an understanding of the numerous soil-pollutant interactions that occur within the soil biogeochemical system (Alvarez et al., 2017; Anae et al., 2021; Ye et al., 2017a, 2017b). Pollutants in co-contaminated soils involve various combinations of inorganic and organic pollutants, including PTE-PTE (Wu et al., 2020), OP-OP (Yang et al., 2006) and PTE-OP (Cao et al., 2013). The co-occurrence of pollutants with different physiochemical properties leads to a wide variety of soil interactions (physical, chemical and biological), toxicities and ecological perturbations, which require enhanced remediation strategies. For example, the effects of PTEs and PAHs on urease activity change at different PTE:PAH ratios and incubation times (Shen et al., 2006). These interactions may result from competition of metals (Cd^{2+} , Pb^{2+} and Zn^{2+}) for adsorption sites that decreases remediation efficiency (Fan et al., 2008), alteration of soil microbial communities (Zou et al., 2021), and/or inhibition of bacteria activities that decrease the degradation of OPs (Biswas et al., 2015; Van Zwieten et al., 2003). As toxicity and environmental interactions among co-contaminants are generally not a simple additive effect (e.g., synergistic vs. antagonistic), remediation strategies for each individual pollutant may not be effective under co-contamination scenarios. Nevertheless, previous studies on pollutant fate and transport, as well as remediation strategies for single soil pollutants, provide necessary fundamental information for further investigation of interactions between multiple pollutants. This information allows for development of new remediation strategies for soils with multiple pollutant impairments based on the distinct behavior of pollutants under co-contamination scenarios.

This review aims to summarize the biogeochemical processes and interactions between various pollutants affecting their fate and transport in soil systems; and the remediation strategies to address co-contaminated soils where pollutants may show different behaviors compared to a single-pollutant scenario. Finally, we identify important knowledge gaps and opportunities for future work to advance the science of pollutant remediation in co-contaminated soil systems.

2. Environmental processes and pollutant interactions in co-contaminated soil systems

Multiple pollutants in a given soil may react with each other directly and/or indirectly, resulting in changes to their environmental reactivity, speciation, toxicity and bioavailability through various pollutant interactions. These interactions are categorized as direct reactions between pollutants and disturbances of environmental processes by co-occurring pollutants in this synthesis. Fig. 1 briefly depicts interactions between pollutants discussed in this synthesis.

2.1. Molecular interactions

Some ubiquitous molecular interactions between pollutants, such as hydrogen bonding, electrostatic attraction, etc., are not discussed here as they are rigorously addressed in previous texts and reviews. This section focuses on cation- π and π - π interactions, which affect pollutant behavior in co-contamination scenarios (anion- π interaction is not included here due to its rarity in soil). Fig. 2 shows typical conformations of cation (anion)- π and π - π interactions between pollutants.

Cation- π interaction: In soils co-contaminated with cationic PTEs and polarizable aromatic OPs, especially PAHs, cation- π interactions often occur. This interaction favors soil remediation through application of amendments, the sorbed cations (or PAHs) serve as new sorption sites for PAHs (or cations) through cation- π interaction, increasing the number of potential sorption sites that enhance the sorption of pollutants by soil and amendments. For example, Liang et al. (2016) and Saeedi et al. (2018) showed that the addition of heavy metals, such as Ni, Pb and Zn, increased the sorption of PAHs (acenaphthene, fluorene and fluoanthene) to various clay minerals, wherein cation- π interactions between sorbed metal cations and PAHs are a prominent mechanism. Li et al. (2024) investigated the effect of Cu on adsorption of phenanthrene in 20 types of soil and revealed a universal increase in phenanthrene adsorption with the presence of Cu, whereby cation- π interactions between Cu and phenanthrene were a key mechanism. The sorbed PAHs also enhance the sorption of cationic pollutants through cation- π interaction, as reported in Zhang et al. (2010). This work also indicates that

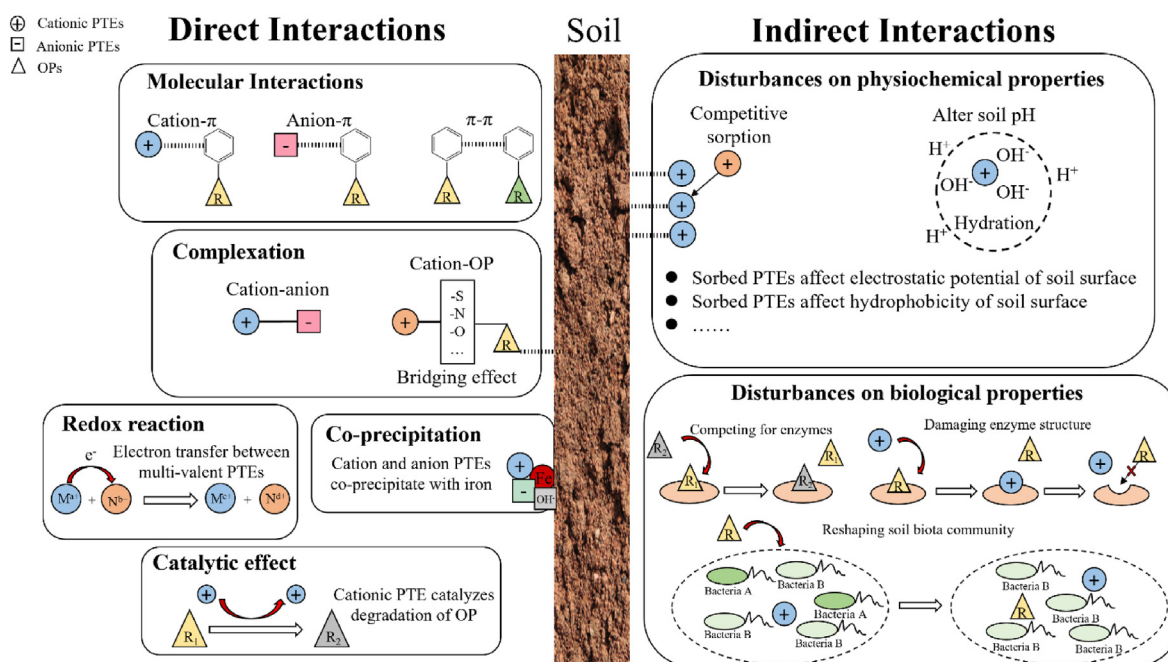


Fig. 1. Common interactions between co-occurring pollutants in soil. Different colors refer to different potentially toxic elements (PTEs) and organic pollutants (OPs). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

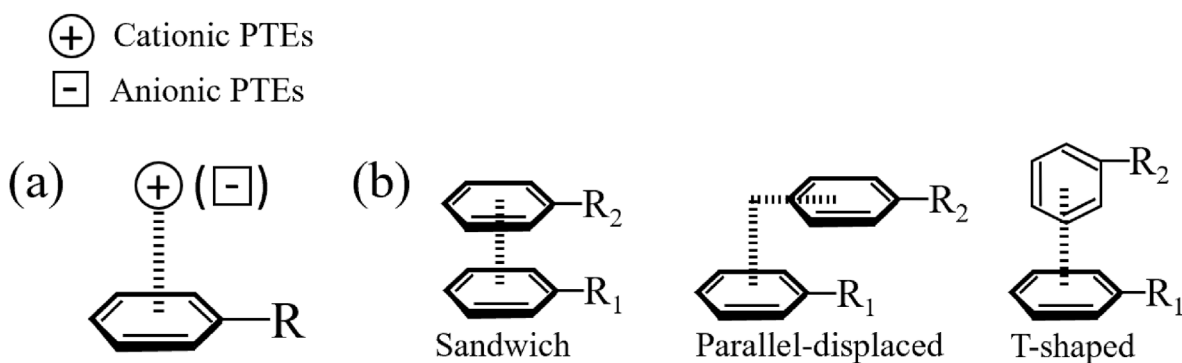


Fig. 2. Typical conformations of (a) a cation/anion- π interaction and (b) three representative π - π interactions.

the effect of cation- π interaction between pollutants on soil sorption properties is concentration-dependent. A low concentration of phenanthrene slightly suppressed Pb^{2+} sorption, whereas Pb^{2+} sorption was enhanced at higher phenanthrene concentrations. This divergent concentration-induced response is attributed to different cation- π bonding mechanisms at different phenanthrene loadings. At low concentration, phenanthrene is adsorbed mainly through cation- π bonds with other cations accumulating on the soil surface, which effectively competes with Pb^{2+} for cation exchange sites. Conversely, at high concentration, phenanthrene is adsorbed mainly through hydrophobic forces to form cation- π bonds with Pb^{2+} in solution, thereby enhancing Pb^{2+} sorption.

π - π interaction: The π - π interaction occurs in aromatic ring-containing OP-OP co-contaminated soil (Pei et al., 2012). Similar to cation- π interaction, π - π interactions potentially enhances sorption of pollutants. Among the aromatic ring-containing OPs, microplastics (MPs) are considered as a prominent soil contaminant and have received increasing attention in recent years. The sorption capacity of MPs for OPs was investigated and π - π interactions play a prominent role. For example, Liu et al. (2016) revealed that π - π interactions were responsible for the high sorption affinity of polystyrene nanoparticles to PAHs. Xu et al. (2018) investigated the sorption of tetracycline (TC) to different MPs, including polyethylene (PE), polypropylene (PP) and polystyrene (PS). The PS microplastics showed a greater sorption capacity for TC than PE and PP microplastics, which was attributed to polar and π - π interactions between PS and TC. These examples indicate that co-occurring MPs may serve as carriers for OPs, thereby significantly affecting their environmental transport. More detailed discussions about potential π - π interactions between OPs and MPs, along with other sorption mechanisms such as electrostatic interactions and hydrogen bonds can be found in a recent review of Agboola and Benson (2021).

2.1.1. Complexation

Complexation between pollutants can be found in soil co-contaminated with cationic PTE-anionic PTE, but is more prevalent in cationic PTE-OP co-contamination. Complexation reactions appreciably change the behavior of pollutants including their sorption, bioavailability, toxicity, etc., that generally result in a decrease of the pollutant's toxicity and bioavailability.

Complexation in cationic PTE-anionic PTE co-contamination: Li et al. (2019) investigated Cd detoxification by Se in the nematode *Caenorhabditis elegans*. They showed that Cd-Se interactions, mediated by multiple thiols including glutathione and phytochelatin, result in the formation of less toxic CdSe/CdS nanoparticles. These nanoparticles are mainly distributed in the pharynx and intestine of the nematode and are continuously excreted from the body to benefit its survival. Guo et al. (2021) reported that the toxicity of Se and Cd in rice was effectively alleviated by the elemental interaction. The possible formation of insoluble Cd-Se complexes in root tissues was supported by the Cd:Se molar ratio, which attenuated the absorption and transport of Cd and Se to the

shoots. In co-contaminated soil, one of the pollutants could serve as a cross-linking agent between the pollutant and binding site to form ternary complexes, which is known as the "bridging effect". The sorbed pollutants provide new binding sites for other pollutants through this "bridging effect", thereby enhancing the sorption of other pollutants. For example, in the work of Jiang et al. (2013), the enhanced sorption of Cd with co-occurring As was attributed to electrostatic interactions, as well as the formation of a Cd-As(V)-goethite complex, as inferred from EXAFS results.

Complexation in cationic PTE-OP co-contamination: Compared to cationic PTE-anionic PTE co-contamination, the "bridging effect" is more prevalent in cationic PTE-OP co-contaminated soils, especially OPs with O-, S-, and N-containing functional groups. For example, the bridging effect of Cu^{2+} cross-linked sulfamethoxazole (SMX) and soil particles significantly increased the sorption of SMX (Morel et al., 2014). Moreover, Wu et al. (2014) reported that Cu (or ofloxacin, OFL) complexed with adsorbed OFL (or Cu), increased the sorption of both pollutants, which was evidenced by fluorescence quenching of OFL by Cu (Pan et al., 2012). It can be inferred from the work of Wu et al. (2014) that specific sorption of pollutants is probably a prerequisite for the "bridging effect". They revealed that the sorption of the co-adsorbate Cu/OFL decreased as the concentration of the primary adsorbate OFL/Cu increased. This phenomenon was attributed to competition for common binding sites on Cu and OFL, indicating the "bridging effect" did not occur in the case of a nonspecific-sorption scenario. If a nonspecific-sorbed pollutant could support the "bridging effect", the increased sorption of both pollutants should be observed at any concentration.

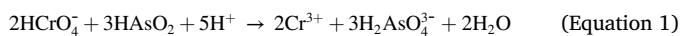
2.1.2. (Co-)Precipitation

The (co-)precipitation of pollutants mainly happens with cationic PTE-anionic PTE co-contamination, often with the assist of iron-bearing soil components/amendments. The pollutants become unavailable once (co-)precipitates are formed. In the work of Jiang et al. (2013) and Li et al. (2018), there was no co-precipitation between As and Cd/Pb under aqueous conditions (at pH = 6/6.5). However, in the presence of goethite, the formation of Cd-As(V) co-precipitates greatly decreased the mobility of both Cd and As (Jiang et al., 2013). The co-precipitated compounds have the potential to dissolve during desorption/dissolution processes, with As being more readily released from co-precipitates than from the As(V)-goethite complex. Other iron bearing amendments, for example, nano-scale zero valent iron (NZVI) amendments, lead to the formation of $\text{Cd}_3(\text{AsO}_4)_2$, $\text{PbFe}_2(\text{AsO}_4)_2(\text{OH})_2$ and As^0 in both acidic and alkaline soils (pH varied from 5.12 to 7.93), resulting in immobilization of pollutants (Li et al., 2020a). Some other soil components, such as hydroxides and sulfides, could also result in co-precipitation of co-occurring PTEs (Xue et al., 2018; Wu et al., 2020).

2.1.3. Redox reactions

In soil co-contaminated with several multi-valence PTEs, redox reactions between them could alter their mobility and bioavailability. For

example, Dong et al. (2021) documented competition between the reduction of Fe (III) and As (V) in the presence of Mn (IV) under flooded soil conditions. Further, MnO₂ and Mn (II) oxidize Cr (III) to Cr (VI) under alkaline (pH 9–11), oxic conditions (Liu et al., 2020a). Similarly, Park (2020) found that the adsorption of Cr (III) on Mn (IV) oxides provided a local surface environment that coupled Cr (III) oxidation and Mn (IV) reduction to form Cr (VI) and Mn (II), which resulted in higher toxicity to lettuce. However, this reaction requires a high oxidative capacity to oxidize soluble Cr (III). Specifically, a high content of Mn (IV) and low content of organic matter facilitated Cr (III) oxidation, which does not occur frequently in many arable soils (Kožuh et al., 2000). Moreover, Dong et al. (2014) found that As (III) was oxidized by Cr (VI) in ice in the absence of DOM, as described by Equation (1). These data provide new insights into Cr and As redox couples and their associated effects on speciation and toxicity in cold regions.



Microbially mediated redox reactions between multi-valence PTEs and OPs are prominent in soils. Jiang et al. (2020b) reviewed the coupled intracellular oxidation of organic matter, including organic contaminants such as benzene, phenol, etc., and extracellular reduction of oxidized metal ions including Fe(III) and Mn(VI) by *Shewanella* and *Geobacter* spp. Whether this interaction between pollutants is beneficial for soil remediation depends on the properties of pollutants, as well as remediation strategies. For example, the oxidation of As(III) to As(V) could significantly decrease the availability of arsenic, which is preferred for immobilization strategies, but would decrease phytoextraction of As through hyperaccumulators (Ma et al., 2001).

2.1.4. Catalytic effects

In cationic PTE-OP co-contaminated soil, some cationic-PTEs (e.g., Fe, Cu) could serve as catalysts in (bio-)chemical processes, enhancing the degradation of OPs. For example, Seger and Maciel (2006) found that partially hydrated Cu(II)- and Al-montmorillonites facilitated the highest decomposition efficiency for methyl parathion, whereas Ca-montmorillonite and kaolin were least effective. This is attributed to the ability of Cu²⁺ in clay minerals to catalyze the oxidation of methyl parathion. Similarly, Lien et al. (2007) found that Cu²⁺ associated with iron nanoparticles enhanced carbon tetrachloride dechlorination, producing more benign intermediate breakdown products. In contrast, when Pb²⁺ was associated with iron nanoparticles, the carbon tetrachloride reduction rate was only slightly increased; however, the intermediate breakdown products are more toxic, such as dichloromethane. Scanning electron microscopy-energy dispersive X-ray (SEM-EDX) analysis indicated that Cu²⁺ was deposited on the surface of the iron nanoparticles, forming a bimetallic surface structure. This leads to galvanic corrosion taking place at the surface, thereby facilitating electron transfer for the surface-mediated dechlorination of carbon tetrachloride.

2.2. Soil disturbance effects by environmental processes associated with co-occurring pollutants

Owing to the complexity of soil biogeochemical systems, various interactions between pollutants and soil components occur, which potentially change the soil environment and affect the behavior of co-occurring pollutants. Compared to direct interactions between pollutants, the indirect effects of co-occurring pollutants through disturbances on soil environment are more ubiquitous.

2.2.1. Competitive sorption

In all combinations of co-occurring pollutants, especially pollutants of similar properties (cationic PTEs, OPs of similar structure/functional groups, etc.), competitive sorption would occur. The competition for physiochemical sorption sites in soil results in increased mobility of pollutants (Li et al., 2018; Davari et al., 2015; Wang et al., 2006) and

reduced remediation efficiency of soil amendments. These interactions have been extensively investigated in soils, thus we will not discuss the details in this synthesis. However, it should be noted that a sufficiently high concentration of a pollutant is required to invoke an appreciable competitive effect. When pollutant concentrations are low, there is little competitive sorption as there are excess binding sites for sorption of additional pollutants, hence low sorbent saturation. As the concentration of a pollutant increases, sorbent saturation increases, limiting the number of sorption sites for other pollutants due to competitive sorption for the remaining binding sites.

2.2.2. Perturbations on soil environment

Pollutants in soil undergo various environmental processes which affect soil properties and influence the final equilibrium state among soil constituents and pollutants. These perturbations can be broadly categorized as perturbations on soil physiochemical and biological environments.

Perturbations on the soil physiochemical environment: Co-occurring pollutants interact with soil components and potentially change the physiochemical properties of the soil, such as electrostatic potential, pH, Eh, SOM/DOM content, hydrophobicity/hydrophilicity, thereby affecting the behaviors of other pollutants. For example, Wu et al. (2018) reported that the presence of As(III) enhanced the sorption of Cd(II) by a novel biochar, and the change in electrostatic potential was an important mechanism responsible for the enhanced sorption of Cd. The solubilization capabilities of saponin for organic contaminants (e.g., PAHs) increase in the presence of Cd²⁺ and Zn²⁺, which can be ascribed to decreased soil pH and complexation of Cd²⁺/Zn²⁺ with saponin that led to the increased mobility of PAHs (Zhou et al., 2011). Furthermore, Shen et al. (2022) found that the addition of tetracycline increased soil pH and DOC concentration, which decreased soil Eh during a 30-day incubation under flooded conditions and resulted in As release from soil colloids. Moreover, Gao et al. (2006) reported the transformation of dissolved organic matter (DOM) to soil organic matter (SOM) through “bridging effects” by heavy metals, with the SOM having a stronger affinity for PAH retention. The change of soil hydrophobicity/hydrophilicity and its influence co-occurring pollutants was reported in Duan et al. (2022).

Perturbations on the soil biological environment: Co-occurring pollutants in soil could interact with soil biota and reshape the soil biota community. For example, Shen et al. (2022) demonstrated that the presence of tetracycline not only increased DOC concentration and decreased soil Eh, but also increased the abundance of *arsC* and *arrA* genes in the soil, leading to enhanced As reduction. These findings were attributed to the co-selection of tetracycline resistance genes and As reduction genes. Due to the synergistic toxicity of co-occurring pollutants, the reshaping of the soil biota community is very common in polluted soils (Zhang et al., 2023a, 2024).

Co-occurring pollutants can also interfere with key biological processes in soils with existing pollutants. The presence of Sb increased the toxicity of As in *Ipomoea aquatica*, as Sb competes with As detoxification processes, including Sb binding to phytochelatin, methylation and sequestration in the vacuoles, making *I. aquatica* more susceptible to As toxicity (Egodawatta et al., 2020). Similarly, Cd²⁺ and Zn²⁺ enter plants through Zn²⁺ transporters in the roots due to their similarity in physiochemical properties, thereby contributing to Cd uptake and potential toxicity (Tan et al., 2020). The added toxicity induced by co-occurring pollutants negatively affects the activity and diversity of soil biota, and generally leads to reduced remediation efficiency. However, some researchers reported enhanced remediation efficiency in co-contaminated soil. Liu et al. (2023a) reported that Cu upregulates the P transporter *PvPht1;3* and As(III) antiporters of As hyperaccumulator *Pteris vittata* promoted As(V) uptake, As translocation and sequestration, which benefits phytoextraction of As. In PAHs co-contaminated soil, low-molecular-weight (LMW) PAHs stimulate the degradation of high-molecular-weight (HMW) PAHs. Zhang et al. (2013) showed that

anthracene, fluoranthene and pyrene become biodegradable by *Micrococcus* sp. PHE3 and *Sphingobium* sp. PHE9 after addition of naphthalene or phenanthrene. This was attributed to LMW-PAHs serving as a growth substrate to increase microbial activity, the so-called priming effect. Similarly, Wang et al. (2014) found that pyrene addition promoted benzo[a]pyrene degradation. In contrast, competitive degradation between PAHs of similar molecular weight was noted. Kashyap et al. (2020) observed strong competitive degradation between phenanthrene and pyrene by the yeast *Candida tropicalis*, and revealed multiple parallel metabolic pathways by GC-MS analysis.

3. Remediation strategies for co-contaminated soils

Remediation strategies for soil pollutants can be broadly categorized as immobilization, extraction and degradation. Immobilization reduces the bioavailability of pollutants in soil, with the addition of amendments being the most common practice (Anaee et al., 2021; Liu et al., 2020b; Zhai et al., 2020). Extraction refers to removal of pollutants from soil, with phytoextraction using hyperaccumulating plants most prominent (Ma et al., 2001; Qiao et al., 2020; Zhou et al., 2020). Degradation of OPs occurs via transformations by soil biota, especially soil microbes, which

convert toxic organic pollutants to benign products, although more toxic intermediate degradation products can sometimes be formed (Sun et al., 2021; Matjašić et al., 2021). The use of extraction and degradation are limited in most co-contaminated soil due to the specificity of the biological processes involved. A summary of typical remediation strategies for co-contaminated soils is found in Table 1.

3.1. Immobilization

Addition of amendments to immobilize soil pollutants is a widely-used remediation strategy. The amendments provide extra binding sites to immobilize various soil pollutants via the physiochemical processes discussed above, and are selected based on their higher affinity for pollutants than provided by the ambient soil alone. Hence, the reactions between pollutants and amendments out-compete those between pollutants and soil, thereby retaining the pollutant in a relatively non-bioaccessible/bioavailable form. Amendments, such as biochar (Bandara et al., 2020; He et al., 2019) and nanoscale zero-valent iron (NZVI) (Qiu et al., 2020; Li et al., 2018), are effective for immobilizing multiple pollutants simultaneously, especially multiple cationic PTEs, as the binding sites are not strictly specific to certain pollutants. However, the

Table 1
Typical strategies utilized for remediation of co-contaminated soils.

Pollutant	Remediation strategies	Remediation Effects	Mechanisms	Ref
As, Cd	Immobilization by calcium-based magnetic biochar	CaCl ₂ -extractable Cd decreased by 23–68%, NH ₄ H ₂ PO ₄ -extractable As decreased by 12%.	Application of biochar elevates soil pH and cation exchange capacity, immobilizes pollutants through complexation and adsorption.	Wu et al. (2020)
Zn, Cd, Cu, As, PAHs	Immobilization by hardwood-derived biochar	C _{Zn} and C _{Cd} decreased; C _{Cu} and C _{As} increased; C _{4-/5-ringed} and C _{2-/3-ringed} PAHs reduced by over 50% and 40%, respectively.	Application of biochar increases soil pH, dissolved organic carbon content, and sorption of pollutants.	Beesley et al. (2010)
As, Cd, Zn	Immobilization by hardwood-derived biochar	Concentrations of Cd and Zn reduced by 300- and 45-fold, As also retained on biochar surface.	The sorption of pollutants on biochar surface is the main mechanism.	Beesley and Marmiroli (2011)
As, Cd, Cr and Pb, PCBs	Immobilization by nano-metallic Ca/CaO dispersion mixture	About 95–99% of heavy metal was immobilized and up to 97% of PCB was hydro-dechlorinated by nano-metallic Ca/CaO dispersion mixture.	Heavy metals were immobilized through adsorption, precipitation and encapsulation; mechanisms for hydro-dechlorination of PCBs were not elucidated.	Mallampati et al. (2014)
Cd, Pb	Immobilization by alkaline lignin	Concentrations of Cd and Pb were reduced after addition of alkaline lignin in acid soil.	Application of alkaline lignin increases soil P content, which possibly precipitates Cd/Pb in acid soil, thus reducing Cd and Pb toxicity.	He et al. (2021)
Cu, Cd	Extraction by periphyton	39.2%–68.8% of Cu and 22.5%–49.9% of Cd were removed.	Periphyton increases solubility of Cu and Cd and retain/accumulate them from water.	Yang et al. (2016)
Cd, endosulfan	Extraction and degradation by <i>Pleurotus eryngii</i> and <i>Coprinus comatus</i>	Accumulation of Cd and dissipation of endosulfan were promoted by co-cultivation of two fungi.	Co-cultivation increases the biomass of mushroom, laccase activity and bacterial counts, enhancing Cd extraction and endosulfan dissipation.	Wang et al. (2018b)
Cd, Cu, PCB	<i>Sedum plumbizincicola</i> extracts Cd and stimulates degradation of PCBs	Concentrations of soil Cd and PCBs were significantly decreased after remediation in both pot and field experiments.	<i>S. plumbizincicola</i> hyperaccumulates Cd, takes up PCBs, along with promoting rhizobacteria activities thereby promoting PCB degradation.	Wu et al. (2012)
Cd, PAHs	<i>Juncus subsecundus</i> extracts Cd and stimulates degradation of PCBs	<i>J. subsecundus</i> takes up and translocates Cd and PAHs; presence of PAHs lessens Cd toxicity.	<i>J. subsecundus</i> takes up and accumulates Cd and increases the abundance of PAH-degrading bacteria, in the rhizosphere.	Zhang et al. (2012)
Cd, PCP	<i>Zea mays</i> L. extracts Cd and stimulates biodegradation of PCP	PCP removal of 86–91% with maize and 74–80% without maize after a 10-week culture period; Cd was mobilized and extracted by maize.	Maize mobilizes and accumulates Cd; produces root exudates and facilitates microbial activities in the rhizosphere, thereby enhancing PCP dissipation.	Hechmi et al. (2013)
Cd, PAHs	<i>Festuca</i> L. and <i>Echinacea purpurea</i> extracts Cd and <i>Mycobacterium</i> strain N12 degrades PAHs	PAH degradation and accumulation in belowground biomass of <i>Festuca</i> L. inoculated with N12 increased from 68.3% to 76.3%.	Removal of Cd relies on plant uptake; inoculation of N12 enhance dissipation of PAHs, but was plant dependent, <i>Festuca</i> L. show better efficiency than <i>E. purpurea</i> .	Li et al. (2021)
Cd, carbendazim	<i>Sedum alfredii</i> extracts Cd and bacteria strains degrade carbendazim	Cd accumulation by <i>S. alfredii</i> was 8.76% and 4.69% for low and high contaminant levels; carbendazim degradation by bacteria increased by 32.1–42.5%.	Enhanced microbial biomass, microbial diversities and dehydrogenase activities increased removal of Cd and carbendazim.	Xiao et al. (2013)
Cd, Pb, Zn, decabromodiphenyl ether (BDE-209)	<i>Sedum alfredii</i> and <i>Festuca arundinaceae</i> extracts PTEs and <i>Bacillus cereus</i> strain JP12 degrade BDE-209	Co-planting and inoculation of JP12 enhanced heavy metal accumulation and BDE-209 dissipation.	Synergistic effects of plants and microbes enhance plant growth and microbial activities, improving metal removal and BDE-209 dissipation.	Lu and Zhang (2014)
Phenanthrene, pyrene	<i>Medicago sativa</i> L., <i>Glomus mosseae</i> and <i>G. etunicatum</i> synergistically degrade PAHs	More than 98.6% and 88.1% of phenanthrene and pyrene were degraded after 70 days.	Colonization of AM in plants enhances soil microbial activity, changes root exudation, and form extraradical mycelium, thereby enhancing PAH degradation.	Gao et al. (2011)

overall immobilization efficiency for multiple pollutants is generally lower than for single-pollutant remediation scenarios due to the competitive processes discussed above. Moreover, for soil co-contaminated with pollutants of contrasting properties (e.g., cationic and anionic PTEs), amendments may fail to simultaneously immobilize these pollutants efficiently. As shown in Liu et al. (2020c), the application of carbide slag and biochar significantly decreased the concentration of CaCl_2 -extractable Cd, while carbide slag, biochar and lodestone had no significant effects on NaH_2PO_4 -extractable As in As–Cd co-contaminated soil.

Novel amendments are synthesized by incorporating new components, especially iron, into the existing amendments to simultaneously immobilize cationic and anionic pollutants. Biochar received great attention as a support for new components (e.g., iron incorporation), as well as its benefits to soil quality and nutrient availability. Fig. 3 illustrates typical sorption mechanisms of biochar and iron-bearing biochar to cationic and anionic PTEs. Wu et al. (2020) synthesized a calcium-based magnetic biochar (Ca-MBC) by pyrolyzing rice straw mixed with Fe oxide and CaCO_3 that decreased the bioavailability of both Cd (II) and As (III). In contrast, rice straw biochar derived from the same procedure without Fe oxide and CaCO_3 decreased Cd (II) bioavailability, but increased that of As (III). Similarly, the biochar-supported NZVI exhibited satisfactory remediation of Cd (II) and As (III) via electrostatic adsorption, complexation, oxidation, (co-)precipitation and formation of type-B ternary surface complexes (Yang et al., 2020a). Some novel amendments based on minerals also exhibited good remediation capacity. The synthesis of zeolite-supported NZVI overcomes the intrinsic limitation of zeolite for AsO_2^- adsorption, and also efficiently immobilizes Pb^{2+} and Cd^{2+} (Li et al., 2018). The mechanisms including type-B ternary

complexation, heterogeneous co-precipitation and concurrent redox reactions, especially the formation of $\text{Cd}_3(\text{AsO}_4)_2$, $\text{PbFe}_2(\text{AsO}_4)_2(\text{OH})_2$ and As^0 , were identified to explain the divergent immobilization and speciation distribution under different soil conditions (Li et al., 2020a). Jiang et al. (2023) modified montmorillonite with ferrihydrite and the new amendment effectively immobilize As, Pb and Sb. Materials from industrial waste such as coal gasification slag also displayed a strong efficacy to simultaneously immobilize Cd and As after modification with iron (Zhou et al., 2023).

Some research has revealed the potential of amendments incorporating microbes with biosorption capacity to remediate cationic-anionic PTEs co-contamination. For example, the magnetic biochar synthesized by Wang et al. (2021) exhibited similar results for Cd (II) and As (III) immobilization, but with *Bacillus* sp. K1 loaded onto the magnetic biochar, the new composite significantly enhanced Cd removal by 230%. This large improvement in Cd retention was ascribed to new biosorption sites, such as amine and hydroxyl groups provided by *Bacillus* sp. K1. Apart from providing extra sorption sites, the formation of type-B ternary complexes formed on the surface of biochar is responsible for enhanced Cd (II) and As (III) immobilization.

3.2. Combined remediation strategies for co-contaminated soils

The immobilization of pollutants through amendments efficiently remediates soils co-contaminated with multiple PTEs. For co-contamination involving OPs (PTE-OP and OP-OP), degradation is the first choice. As previously discussed, interferences (mostly suppression) of soil biota metabolism may arise from the presence of co-occurring pollutants (Chigbo et al., 2013; Hechmi et al., 2014), which may substantially limit the remediation efficiency of extraction and degradation strategies.

3.2.1. Combination of extraction and degradation

Combining remediation strategies that target each respective pollutant in co-contaminated soil is the most simplistic approach. As an example, the Cd-hyperaccumulating plant *S. alfredii* shows significant bioremediation capacity for Cd-DDT co-contaminated soil when inoculated with *Pseudomonas* sp. DDT-1, a DDT-degrading microbe (Zhu et al., 2012). Inoculation of *Pseudomonas* sp. DDT-1 increases the root biomass of *S. alfredii*, which in turn benefits the metabolic activities of *Pseudomonas* sp. DDT-1, thereby improving the removal efficiency of Cd-DDT co-contaminants. The effect of enhanced bioremediation efficiency by mutual beneficial interactions between plants and microbes is widely documented and termed “bioaugmentation”. As another example, an integrated remediation strategy involving bacteria (*Bacillus thuringiensis* FQ1) and a fungal mushroom (*Pleurotus cornucopiae*) effectively remediated Cd-phenanthrene co-contaminated soils (Jiang et al., 2015). The bacteria increase the biomass and Cd-accumulation capacity of the mushroom by alleviating its oxidative stress as indicated by decreased lipid peroxidation and enzyme activities (e.g., superoxide dismutase, catalase and peroxidase). Moreover, the inoculation of *Piriformospora indica* elevated the chlorophyll content of *Medicago sativa*, as well as soil enzyme activities (e.g., urease, invertase and polyphenol oxidase) in the rhizosphere (Li et al., 2020b). These alterations increased photosynthetic production and the tolerance of *M. sativa* in soil co-contaminated with Cd and phenanthrene, thereby enhancing phytoextraction efficiency. Yet another example involves the rhizobacteria *Burkholderia cepacian*, which enhances metal tolerance of *S. alfredii*, increasing phytoextraction efficiency for Cd^{2+} and Pb^{2+} , as well as phenanthrene removal (Li and Wong, 2012). The inoculation of *B. cepacian* increases translocation and metal tolerance of *S. alfredii*, but does not enhance plant growth and metal uptake. Improved soil nitrogen and phosphorous nutrition provides a plausible explanation for the increased translocation and metal tolerance, as phosphorous is directly involved in metal detoxification in plants.

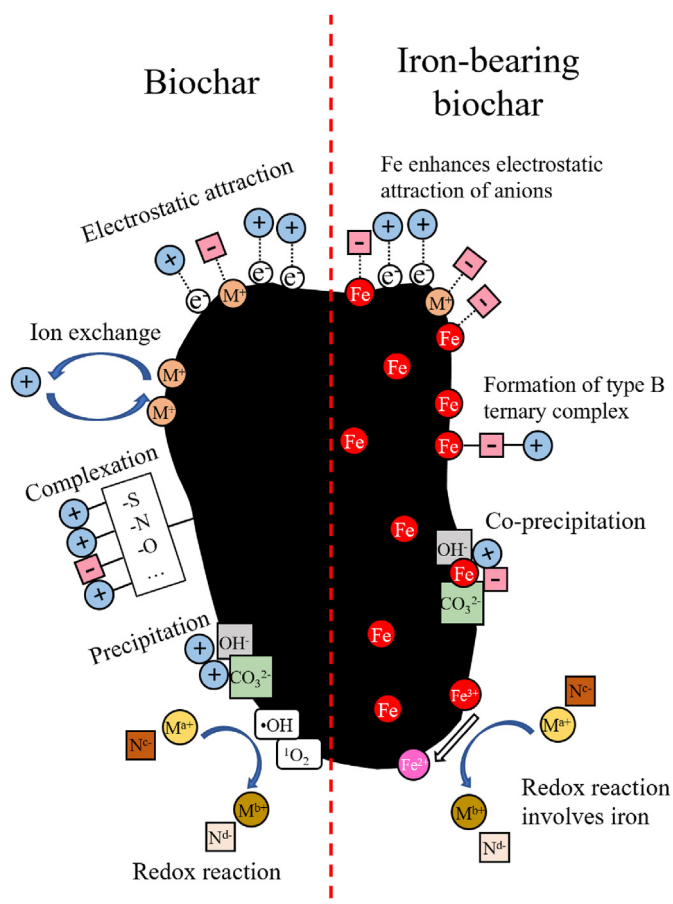


Fig. 3. Typical sorption mechanisms of biochar and iron-bearing biochar to cationic and anionic PTEs.

The abovementioned publications focused on the effects of reducing the concentrations of pollutants in soil, while [Chen et al. \(2016\)](#) further verified the safety of agricultural products grown on co-contaminated soil remediated through an extraction-degradation strategy. They conducted a 2-year greenhouse experiment to remediate a soil co-contaminated by PAHs and heavy metals (Zn and Cd) through interplanting ryegrass with *S. alfredii* and regular inoculation with *Microbacterium* sp. KL5 and *Candida tropicalis* C10. Plant shoots were harvested at a 4-month interval and microbes were inoculated every 2 months due to the natural attenuation of exogenous microbes. The harvested ryegrass was deemed safe as an animal feed based on Chinese national standards. Notably, the Cd/Zn hyperaccumulating plant *S. alfredii* outcompeted the ryegrass for heavy metal uptake, thereby greatly reducing metal accumulation in ryegrass. The PAH degradation capacity of the inoculated bacteria strains, *Microbacterium* sp. KL5 and *C. tropicalis* C10, has been previously reported. By comparing the results of different treatments in their experimental design, they demonstrated different roles for the bacteria strains. The *Microbacterium* sp. KL5 enhances soil enzyme activity, plant growth, PAH removal and metal phytoextraction, whereas *C. tropicalis* C10 promotes soil enzyme activity and removal of high-molecular-weight PAHs. This work corroborates the feasibility of simultaneous remediation of co-contaminated soils while retaining safe agricultural production via a combination of remediation strategies.

3.2.2. Combination of immobilization and degradation

The extraction-degradation strategy achieved simultaneous remediation of PTEs and OPs in soil. However, agricultural production may be adversely affected by the remediation processes, as the hyperaccumulating plants generally have little or no economic value. Thus, combining immobilization and degradation, i.e., a microbe-amendment consortium, could potentially be a better solution for remediation of PTE-OP co-contaminated soil while ensuring the economics of agricultural production. The enhanced remediation capacity of PTE/OP contaminated soil by microbe-amendment consortiums (microbe-biochar consortiums in most cases) has been widely investigated, and typical pollution alleviation mechanisms are depicted in [Fig. 4](#). More detailed information is summarized in review papers by [Xiang et al. \(2022\)](#) and [Bolan et al. \(2023\)](#).

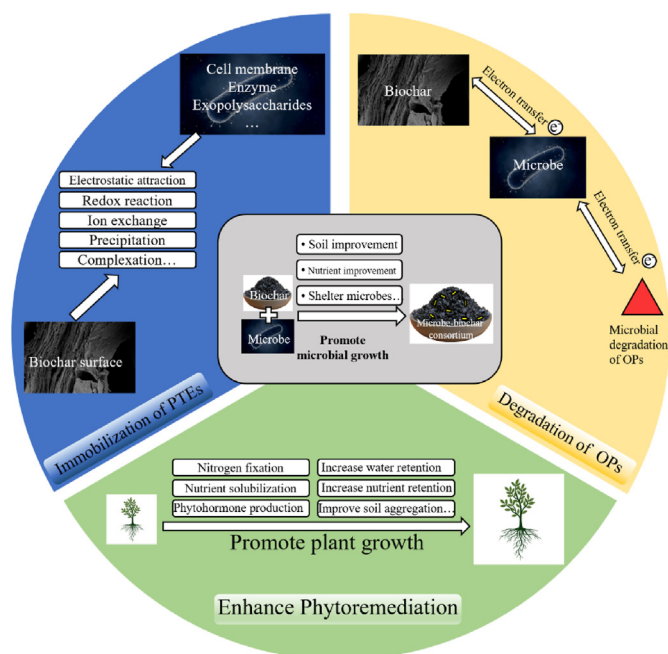


Fig. 4. Typical mechanisms for enhanced remediation capacity of PTE/OP contaminated soil by microbe-biochar consortiums.

Microbe-amendment consortiums also provide options for the remediation of PTE-OP co-contaminated soil. In the work of [Zhang et al. \(2023b\)](#), biochar inoculated with *Sphingobium abikonense* was introduced to remediate soil co-contaminated with Cu and phenanthrene. The results showed enhanced and sustained remediation capacity by the microbe-amendment consortium. More than 96% of phenanthrene was degraded with the bacteria-immobilized biochar in the first- (0–6 days) and second-remediation (6–12 days) stages, comparing to 88% and 36.7% degradation efficiencies for first- and second-remediation stages by free *S. abikonense*. The immobilization of Cu also increased from approximately 40 to 58 mg kg⁻¹ when compared to the biochar-only treatment. They attributed the enhanced Cu remediation capacity of the microbe-biochar consortium to extra biosorption sites from the immobilized microbes on biochar. The biochar promoted soil quality and nutrient availability, protection of immobilized bacteria and a reshaping of the soil microbe community, which further facilitated the degradation of OPs.

Conversely, there are reports of decreased degradation efficiency in the presence of soil amendments. [Yang et al. \(2020b\)](#) reported inhibited pyrene degradation by *Escherichia* sp. In the presence of Cd in both solution and soil, and lower pyrene degradation efficiency by immobilized *Escherichia* sp. on biochar than free cells in Cd-pyrene co-contaminated soil. These findings were ascribed to augmented Cd toxicity due to the strong adsorption affinity of biochar. Thus, ensuring the activity of immobilized OP-degrading bacteria is a high priority for effective remediation of PTE-OP co-contaminated soil using microbe-amendment consortiums. This can be achieved either through inoculation of OP-degrading microbes with heavy metal tolerance to the microbe-biochar consortiums, or through alleviation of heavy metal stress on the immobilized OP-degrading microbes. Mechanisms for heavy metal tolerance of soil microbes such as biosorption, bio-oxidation, active transport (efflux), etc. are summarized in the review paper of [Joshi et al. \(2023\)](#). Among these mechanisms, the biosorption of PTEs through exopolysaccharide (EPS, also known as extracellular polymeric substances) is of great importance. [Fig. 5](#) illustrates possible beneficial effects of EPS on bacteria survival and growth in microbe-biochar consortiums.

EPS can decrease mobility of various PTEs through biosorption, complexation, etc. ([Liu et al., 2023b](#); [Yue et al., 2015](#)), and some microbes secrete more EPS under PTE stress to survive ([Mohite et al., 2018](#)). Moreover, EPS can be secreted by various bacteria genera capable of OP degradation, which can be potentially incorporated into microbe-biochar consortiums to remediate PTE-OP co-contaminated soils. [Table 2](#) summarizes recent publications on biodegradation of OPs using EPS-secreting bacteria. Apart from immobilization of PTEs, EPS also forms capsules or slimy coatings around cells. The net-like structure of EPS not only shields microbes from direct exposure to hazardous contaminants, but also retains water and nutrients and enhances attachment of cells to adsorbates, thereby providing beneficial niches for bacteria ([Rehm, 2010](#)) ([Fig. 5](#)). Thus, microbe-biochar consortiums synthesized through immobilizing the coculture of EPS-secreting bacteria and OP-degrading bacteria to biochar could potentially enhance the remediation of PTE-OP co-contaminated soils. However, the effectiveness of these strategies requires further validation for various PTE-OP pairings.

4. Future research perspective

Co-contaminated soils are of increasing concern and warrant further research to understand the interactions among co-occurring pollutants and to develop effective remediation strategies. The soil environment is complex and interacts with co-occurring pollutants through direct (e.g., complexation, redox reactions) and indirect interactions (e.g., competition for substrates/nutrients, changing soil environmental conditions). Additionally, co-occurring pollutants interact among themselves and may serve as catalyst for soil interactions with other pollutants.

As for remediation of co-contaminated soils, the abiotic remediation

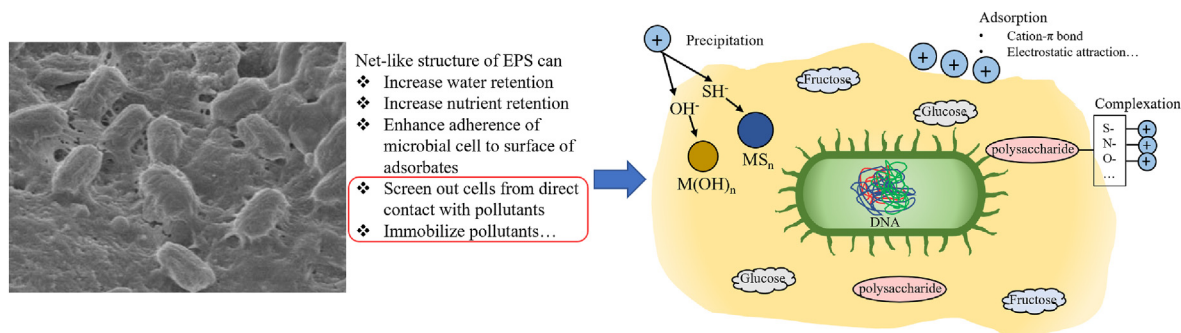


Fig. 5. Beneficial effects of EPS on bacteria survival and growth in a microbe-biochar consortium.

Table 2

Degradation of organic pollutants via EPS-secreting bacteria.

Microorganisms	Pollutants	Remediation Effects	Reference
<i>Pseudomonas furukawaii</i> PPS-19	Petroleum hydrocarbons	<i>P. furukawaii</i> PPS-19 and its EPS complex degrades and emulsifies 89.52% of crude oil within 5 days.	Vandana (2021)
<i>Micrococcus</i> sp. PHE9 and <i>Mycobacterium</i> sp. NJS-P	Phenanthrene, pyrene	EPS increases degradation rate of phenanthrene and pyrene from 24.38 to 3.24 mg L ⁻¹ day ⁻¹ (66% and 58%) to 35.18 and 5.08 mg L ⁻¹ day ⁻¹ (>90%), respectively.	Zhang et al. (2015)
<i>Enterobacter cloacae</i> strain TU	<i>n</i> -hexadecane	<i>E. cloacae</i> TU degraded <i>n</i> -hexadecane up to 75% through biodegradation and emulsification.	Hua et al. (2010)
<i>Alcaligenes faecalis</i> NBRI OSS2-5	Chlorpyrifos (organophosphate)	NBRI OSS2-5 degraded 94% of chlorpyrifos within 10 days.	Yadav et al. (2020)
<i>Kosakonia oryzae</i> strain – VITPSCQ3	Profenofos, Quinalphos (organophosphate)	Up to 82% and 92% of Profenofos and Quinalphos were degraded within 48 h.	Dash and Osborne (2020)
<i>Bacillus</i> sp. E5 and <i>Sphingomonas</i> sp. 224	Toclofos-methyl (organophosphate)	The degradation rate of Toclofos-methyl was approximately 90% within 48 h.	Kwak et al. (2013)
<i>Pichia kudriavzevii</i> Atz-EN-01	Atrazine	The removal efficiency of atrazine by Atz-EN-01 ranged from 94% to 100%.	Abigail and Das (2015)

strategies for single and multiple pollutants may be similar, employing sorption, complexation, precipitation and redox reactions between pollutants and amendments. Conversely, biotic remediation strategies for single and multiple pollutants are often different due to the specificity of degradation enzymes. Although some plants and microbes show excellent tolerance and remediation capacity for multiple pollutants in soil, the inhibition effects of co-occurring pollutants on remediating soil biota are prevalent. Thus, combining plants and microbes with capacities to remediate different pollutants in co-contaminated soil is challenging. While investigations of soil interactions and remediation strategies for co-contaminated soils have achieved good progress, most of the research has been based on laboratory studies, often under optimized environmental conditions. Thus, there is a critical need for future research aimed at translating the fundamental scientific findings from laboratory studies to the practical real-world of co-contaminated, complex soil systems at

the field scale.

4.1. Scientific aspects

The mechanisms and specific processes of microbial-plant combinations for remediation of co-contaminated soils are not fully understood. Additional information is required concerning why some microbial-plant combinations are capable of remediating multiple pollutants, whereas some pollutants inhibit biota metabolism and remediation success. Several studies indicate that the interactions, toxicity and remediation efficiency for multiple pollutants are dose-dependent, but investigations directly addressing dosage effects are lacking, especially with regard to synergistic and antagonistic interactions among pollutants. Understanding the dosage effects and potential toxicity interactions among pollutants are crucial for the success of practical field applications.

Investigations on the effects of soil physiochemical properties on interactions between pollutants and remediation strategies are not well elucidated. The effects of interactions with humic substances, biochar, clay minerals and Al/Fe (hydr)oxides, oxic vs anoxic conditions (redox), pH and soil organic matter quantity/quality are relevant to design effective remediation strategies. The effects of fertilizers and other soil amendments (e.g., biochar, lime, gypsum and animal wastes) on soil processes and remediation of co-contaminated soil require intensive investigation, as these common agricultural practices have a strong influence on plant-microbe communities and the physicochemical environment of the soil biogeochemical system.

Most studies of co-contaminated soils examine only 2–3 pollutants under laboratory conditions, whereas agricultural fields may contain a variety of pollutants and environmental conditions leading to unexpected outcomes. Thus, future studies need to address a wider range of interactions and remediation strategies in the presence of several minor pollutants/soil components. The factors affecting laboratory conditions are highly constrained, whereas under field conditions, the environmental factors are continuously shifting in time and space. Thus, it is important to investigate soil-pollutant interactions and remediation strategies under a variety of common real-world conditions, such as wet-dry cycles from rainfall/irrigation, diurnal/seasonal temperature fluctuations, cropping vs non-cropping seasons, and pre/post soil amendment additions.

4.2. Practical aspects

Most plants utilized for phytoremediation are not food crops. Thus, remediation strategies for contaminated arable land that can be applied without interrupting crop production (e.g., cover crops and polyculture) are of great importance to ensure both food safety and food security. The effects of remediation strategies on soil physiochemical properties and possible legacy effects from remediation activities on the economics of agricultural production need to be fully evaluated.

The potential ecological risks of bioremediation are not well elucidated. Changes in the soil biota community structure and/or abundance

might cause long-term negative outcomes, and hence should be scrutinized before application at the larger field scale. The application rate (i.e., soil amendments) and frequency of remediation strategies and their effects on agricultural practices require additional investigation. The development of economically-viable remediation strategies is crucial to large scale deployment of remediation efforts. Thus, a cost-benefit analysis of remediation strategies is required to evaluate the associated costs required for safe and sustainable agricultural production.

CRedit authorship contribution statement

Saiqi Zeng: Conceptualization, Writing – original draft. **Zhongmin Dai:** Writing – review & editing. **Bin Ma:** Writing – review & editing. **Randy A. Dahlgren:** Writing – review & editing. **Jianming Xu:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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