Published in final edited form as:

Chemosphere. 2022 November ; 307(Pt 4): 135989. doi:10.1016/j.chemosphere.2022.135989.

Composting and green technologies for remediation of phthalate (PAE)-contaminated soil: Current status and future perspectives

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Abstract

Phthalate esters (PAEs) are hazardous organic compounds that are widely added to plastics to enhance their flexibility, temperature, and acidic tolerance. The increase in global consumption and the corresponding environmental pollution of PAEs has caused broad public concerns. As most PAEs accumulate in soil due to their high hydrophobicity, composting is a robust remediation technology for PAE-contaminated soil (efficiency 25%–100%), where microbial activity plays an important role. This review summarized the roles of the microbial community, biodegradation pathways, and specific enzymes involved in the PAE degradation. Also, other green technologies, including biochar adsorption, bioaugmentation, and phytoremediation, for PAE degradation were also presented, compared, and discussed. Composting combined with these technologies significantly enhanced removal efficiency; yet, the properties and roles of each bacterial strain in the degradation, upscaling, and economic feasibility should be clarified in future research.

Graphical Abstract

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Credit authorship contribution statement

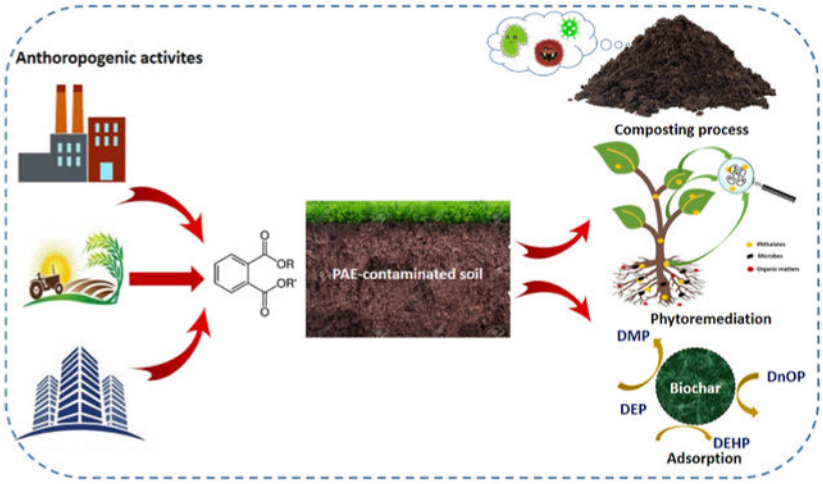
Huu Tuan Tran: Conceptualization, Formal analysis, Writing – original draft. **Minh Ky Nguyen**: Data curation Methodology, Writing – original draft. **Hong Giang Hoang**: Data curation, Review & editing. **Justin M. Hutchison**: Supervision, Review & editing. **Chi Thanh Vu**: Methodology, Review & 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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.135989.



Keywords

Plastic wastes; PAE-degrading strains; Biochar; Organic wastes; Microbial community

1. Introduction

In plastic manufacturing, phthalate esters (PAEs) are often used to enhance the flexibility and durability of plastics. PAEs are widely detected in many industrial, residential, and even agricultural areas worldwide because of the ubiquitous use of plastics (Abdel daiem et al., 2012; Ferreira and Morita, 2012; Kaewlaoyoong et al., 2018; Nguyen et al., 2022b). As the global production of plastics is over 150 million tons year^{-1}, the annual consumption of PAEs is around 6–8 million tons (Gao et al., 2018; Lönnstedt and Eklöv, 2016; Net et al., 2015). With this widespread use and subsequent environmental contamination, PAE has been found in every environmental media, including soil, sediment, water, and even air (Dargnat et al., 2009; Lee et al., 2019b; Lin et al., 2009). Yet, with high hydrophobicity, PAE contamination mainly occurs in soil (Lee et al., 2019a; Wei et al., 2020). In fact, PAEs are one of the most frequently detected persistent organic pollutants (POPs) in soil with half-lives varying from 3 to 2000 years under natural conditions (Gao and Wen, 2016).

PAEs are endocrine-disrupting chemicals (EDCs) and potential carcinogens. The impart on normal physiological hormones function has been previously reported (Diamanti-Kandarakis et al., 2009; Katsikantami et al., 2016; Mankidy et al., 2013), and exposure to PAEs is linked to an increased risk of breast cancer (Hsieh et al., 2012; López-Carrillo et al., 2010). Additionally, PAEs have shown adverse effects on the reproductive system (Schettler, 2006; Wang et al., 2019c). Due to their high toxicity, specific PAEs, di-*n*-butyl phthalate (DnBP), diethyl phthalate (DEP), diethylhexyl phthalate (DEHP), diisononyl phthalate (DINP), are listed as EDCs by WHO/IPCS (2002) and WHO-UNEP (2013) (Kay et al., 2014; Kay et al., 2013; WHO-UNEP, 2013). Therefore, in recent years, the remediation of PAE-contaminated soil has been a focus of environmental engineering, especially when the “hot” topic of micropollutants is attracting broad public interest.

Several treatment technologies are recommended for the remediation of PAE-contaminated soil, including biochar adsorption (Zhang et al., 2016), electrokinetic and nano oxidation processes (Yang et al., 2015), chemical oxidation (Wang et al., 2014), and composting (Tran et al., 2021b). Among them, composting is a promising biodegradation technology due to its high biodegradation efficiency, low environmental impacts, and cost feasibility (Tran et al., 2018). There are two main composting techniques, aerobic and anaerobic. Aerobic composting has shown higher removal efficiency and shorter incubation time than anaerobic composting (Tran et al., 2020). For instance, Yuan et al. (2011) reported 91%–96% DEHP removal (initial concentration 50–250 mg kg^{-1}) in soil after 30 days of aerobic incubation, whereas only 55%–69% DEHP (initial concentration 100 mg kg^{-1}) was removed after 112 days of anaerobic composting (He et al., 2018). In the composting biodegradation process, microbial activity plays a critical role in determining the removal efficiency and rate (Hoang et al., 2022). Among the microorganisms in the compost mixture, bacteria are key to biodegradation since their enzymes (e.g., hydrolase, esterase, protocatechuate) vastly accelerate biodegradation.

Numerous studies have evaluated bioremediation strategies for PAE-contaminated soil (Cai et al., 2008b; Das et al., 2021; Xiang et al., 2020). The role of the microbial community and the overall mechanism of PAE biodegradation have also been reported. However, a knowledge gap exists for the microbial community's structure and dynamic response during the composting process. Also, the role of bacterial strains and their specific enzymes in PAE degradation remains ambiguous. Therefore, this study aims to comprehensively discuss composting degradation of PAE-contaminated soil. Current knowledge on the variation of microbial community structure, the role of bacteria and their specific enzymes, and biodegradation pathways were also provided. Further, other green technologies, including biochar adsorption, phytoremediation, and bioaugmentation were summarized, compared, and discussed. Finally, research gaps were identified and recommendations for future research were accordingly provided.

2. Methodology

The database of this review was created using Google Scholar, NCBI, PubMed, and Web of Science (Table S1 and Fig. S1). We paid more attention to the publications between 2010 and 2022 since they provided up-to-date information. Reflecting the concern of PAE contamination, the number of publications has increased more than 10 times from 2010 – to 2020 (Fig. 1). Meanwhile, the number of publications on the composting remediation of PAE-contaminated soil has displayed a similar increasing trend.

3. Overview of PAEs

3.1. Physicochemical properties of PAEs

The fate and transport of PAEs in the environment depend much on their physicochemical properties, especially K_{OW} (octanol-water partition coefficient), K_{OA} (octanol-air partition coefficient), K_{AW} (air-water partition coefficient), K_{OC} (organic carbon partition coefficient) and vapor pressure (Kashyap and Agarwal, 2018) (Table 1). For example, Cousins et al. (2003) reported that high K_{OW} values mean easy sorption onto surfaces and organic matters,

and the Log (K_{OW}) increases with increasing alkyl chain length, which also means greater hydrophobicity. Vapor pressure (at 25 °C, in mm Hg) decreases with increasing molar volume (g mol^{-1}) or alkyl chain length. In general, low molecular weight (LMW-) PAEs (e.g., DEP and DBP) are easier to biodegrade than high molecular weight (HMW-) PAEs (e.g., DEHP and DnOP) (Tuan Tran et al., 2022).

3.2. Main sources of PAEs

Anthropogenic (residential, agricultural, and especially industrial) activities are the main PAE sources (Fig. 2). Currently, there are about 60 different PAE categories used for various types of industries such as personal care products (e.g., cosmetics, hair sprays, gels), material packaging, plastic manufacturing, lubricants, insecticides, paint additives, and adhesives (Eichler et al., 2019). Moreover, agricultural activities (e.g., irrigation, biosolid fertilization, and sewage sludge discharge) with the use of agricultural film mulching, pesticides, and fertilizers contribute significantly to PAE-contamination in soil (Cai et al., 2007; Guo and Kannan, 2012; Lü et al., 2018; Net et al., 2015; Tran et al., 2022a; Wang et al., 2013; Weschler et al., 2008; Yang et al., 2013b; Zhu et al., 2010). Residential activities can generate urban dust that contributes to soil contamination through deposition. Dry and wet deposition contributes to PAE contamination in soil in highly industrialized areas (Wu et al., 2015). Lan et al. (2012) found that both wastewater sludge and urban dust increased PAE levels in soil.

3.3. Current status of worldwide PAE-contaminated soil

PAE contamination in soil has been found worldwide, including Asia (e.g., China and Taiwan), Europe (e.g., Netherlands, Scotland, United Kingdom, and France), and Africa (e.g., South Africa) (Fig. 3) (Brodskiy et al., 2019; Gibson et al., 2005; Hu et al., 2003; Peijnenburg and Struijs, 2006; Rhind et al., 2013; Zeng et al., 2008). As a result of urbanization and industrialization, PAE concentrations are higher in urban areas (Zheng et al., 2014) than concentration in rural areas (Huo et al., 2016; Lü et al., 2018; Zhang et al., 2015). High PAE levels are often found in urban areas. For instance, Tran et al. (2015) reported that the total PAE concentrations in French urban and rural soils were 1.089 and 0.154 μg g^{-1}, respectively. In rural areas, high PAE contamination was found in agricultural fields (Xu et al., 2008; Zeng et al., 2008). Also, due to rapid industrialization and urbanization, PAE concentrations in China were higher than those in other countries (Lü et al., 2018). Among PAEs, DEHP and dimethyl phthalate (DMP) are the most frequently detected in the soil since they are the main constituents of plastic products (e.g., houseware, household appliances, and agricultural equipment).

3.4. Toxicity and risk assessments

PAEs are regarded as hazardous organic contaminants due to their adverse effects on the endocrine system, erythrocytic functions, and reproductive system of organisms (Li et al., 2018; škrbić et al., 2016; Tan et al., 2017). He et al. (2015) also reported that PAEs were highly toxic to microbes in soil. PAEs' toxicity is determined by their physicochemical properties (Giuliani et al., 2020). HMW-PAEs (e.g., DEHP and DnBP) can cause hormonally mediated diseases and are considered potential carcinogenic agents (Adeniyi et al., 2008; Fukuwatari et al., 2002; McKee et al., 2004). Thus, the United

States Environmental Protection Agency (USEPA) has listed DEHP as a high-priority pollutant (US.EPA, 2019). PAE exposure negatively impacts human health via pathways like inhalation of contaminated aerosols and consumption of contaminated food. Daily vegetable consumption is the main intake source of PAEs (Cheng et al., 2015; Niu et al., 2014; Wang et al., 2015a, 2018; Xia et al., 2011) The risks of PAEs exposure are assessed via a reference dose, which is related to the tolerable daily intake (Giuliani et al., 2020) with elevated risks for sensitive subpopulations including fetuses and breastfeeding infants (Filardi et al., 2020).

4. Composting remediation of PAE-contaminated soil

4.1. Overview of composting remediation of PAEs in soil

Composting has been reported as a promising treatment for PAEs in soil (Amir et al., 2005; Pakou et al., 2009). Aerobic and anaerobic composting with different scales (e.g., pilot (10–110 L) (Solano et al., 2022) and field (1800–5000 L) (Chen et al., 2022) could remove 20%–100% PAEs in soil (Table 2). Various types of organic wastes (e.g., sewage sludge, manure, fruit, and vegetable waste) have been used as compost material. Bulking agents (e.g., sawdust, wood waste, rice straw) have been used to adjust the moisture of the compost mixture. Composting biodegradation can (1) decrease moisture content due to metabolism and volatilization mechanisms, (2) increase pH due to the release of organic acids, and (3) decrease C/N ratio due to the decrease of substrates via microbial activity (Chang et al., 2009; Lin et al., 2017). Thus, in order to achieve high PAE biodegradation, composting conditions are often maintained as follows: initial moisture content of 50%–60%, and C/N ratio of 20–30 (Lü et al., 2021; Tran et al., 2021a).

In general, aerobic composting (oxygen content >10%) results in higher PAE removal and reguires a shorter incubation time compared to anaerobic composting (Amir et al., 2005; Liang et al., 2008). Aerobic composting could remove DEHP at 91%–97% after 30 days, whereas anaerobic composting removed 55%–70% after 112 days (He et al., 2018; Yuan et al., 2011). During the composting process, the biodegradation rate at the thermophilic phase is significantly higher than that of other phases (mesophilic, cooling, and maturation) (Fu et al., 2013; Tran et al., 2021b). For instance, Tran et al. (2021b) showed that composting biodegradation removed was 98% of DOTP, with the majority of the degradation (76%) occurring in the thermophilic phase. The thermophilic phase also had the highest degradation rate (0.149 day^{-1}). Similarly, the thermophilic phase accounted for 60% of DEHP degradation compared to the total removal of 85% (Cheng et al., 2008). Thermophilic phase degradation is higher for two reasons. First, high temperatures (55–70 °C) accelerate microbial growth, especially PAE-degrading strains. Second, at higher temperatures, the viscosity of the PAEs significantly decreases, enhancing oxygen penetration and interaction between PAEs and the bacterial community.

4.2 Roles of microbial community

The microbial community is key to PAE degradation in composting (Liang et al., 2008; Wang et al., 2015b). Next-generation sequencing has been used to identify microbial strains and their function in PAE degradation (Tran et al., 2021b). High-throughput sequencing technologies have increasingly been used to study the succession and characteristics of

microbial communities during composting of PAE-contaminated soil, including bacteria, fungi, algae, and yeast (Fang et al., 2017; Wang et al., 2019b). Various bacteria (e.g., *Rhodococcus pyridinivorans* XB, *Bacillus subtilis* No.66, *Gordonia* sp. QH-11), fungi (e.g., *Aspergillus niger*, *Tranmetes versicolor*, *Pleurotus ostreatus*), algae (e.g., *Chlorella pyrenoidosa*, *Closterium lunula*) and yeast (e.g., *Rhodotorula rubra*, *Saccharomyces cerevisiae*) have been reported as PAE-degraders (Benjamin et al., 2015; Liang et al., 2008). Among these microorganisms, bacteria are the most abundant and play major roles in degrading PAEs (Ren et al., 2018). The characteristics and function of the bacterial community are affected by both internal (e.g., initial compost materials, substrates, pH, and C/N ratio) and external factors (e.g., operational conditions, including: aeration, moisture content, and temperature).

The richness and diversity of the bacterial community are expressed via parameters like OTUs, Chao 1, Shannon diversity and Shannon evenness indexes (Bai et al., 2020). The richness and diversity vary with composting phases. Often, without PAEs, the indices increase from the mesophilic phase to the maturation phase (Wang et al., 2017, 2019a). For instance, Wang et al. (2017) indicated that after 29 days of food waste composting, the Chao 1 and Shannon index increased rapidly from 229 to 411 and 3.31 to 3.85, respectively. However, with the presence of high levels of PAEs (e.g., DEHP and DOP), the richness and diversity decrease (Bai et al., 2020; Zhang et al., 2017). For instance, at DOP concentration of 1000 mg kg^{-1}, the bacterial diversity dramatically reduced from 2.6 (day 2) to 1.7 (day 12) (Zhang et al., 2017). Similarly, at a DEHP concentration of 40 mg kg^{-1}, the measured Chao 1 (1293) and Shannon index (5.83) were lower than those at a concentration of 10 mg kg^{-1} (1411 and 5.97, respectively) (Gao et al., 2020). The toxicity of PAEs can disrupt cell membrane fluidity and integrity, causing growth abnormalities and thus adverse effects on the structure of the bacterial community (Cartwright et al., 2000).

Distinct shifts in bacterial community structure were observed during the composting process (Bai et al., 2020; He et al., 2018). Firmicutes, Proteobacteria, Acidobacteria, and Bacteroidetes play a vital role in PAE biodegradation. Among them, Firmicutes were the most abundant at the phylum level (Bai et al., 2020; Zhang et al., 2017). The relative abundance of Firmicutes declined remarkably with increasing temperature during composting (Wang et al., 2019a). For example, on day 1 (mesophilic phase), Firmicutes were the most abundant (76.7%) but gradually declined to 60% on day 10 (thermophilic phase, > 55 °C) (Wei et al., 2018). The thermophilic phase may limit bacterial growth and even eliminate some mesophilic members. Lactobacillales (phylum Firmicutes) were reported to dominate during the thermophilic phase, while Bacillales dominated the bacterial community during the mesophilic phase (Graça et al., 2021).

During the thermophilic phase, Proteobacteria were also found in high abundance (60.8%) (Huang et al., 2021). *Pseudoxathonomas* sp. (phylum Proteobacteria) could use PAEs as the sole carbon and energy source for metabolism (Meng et al., 2015). Likewise, the relative abundance of *Bacteroidetes* dramatically increased (to 12.4%) during the thermophilic phase since they can break down macromolecules (e.g., cellulose, lipid, and protein) for energy production (Huang et al., 2021). In contrast, *Acidobacteria* dominated the maturation phase, indicating the maturity of compost. Some gram-negative (e.g., *Shingomonas yanoikuyae*,

Comamonas acidovorans and *Delfia* sp.) and gram-positive bacteria (e.g., *Arthobacter* sp., *Gordonia* sp., and *Rhodococcus* sp.) have been identified as key PAE-degraders.

4.3. Biodegradation pathway

Complete PAE biodegradation requires primary and secondary pathways (Fig. 4). The primary pathway includes beta-oxidation, de-esterification or dealkylation, and *trans*-esterification or demethylation. PAEs with a long alkyl chain are converted to a short alkyl chain (such as diethyl phthalate (DEP)) via beta-oxidation. Then, the shorter chain is oxidized to phthalic acid (PA) through either de-esterification for DEP or *trans*-esterification DMP. For DEP, the hydrolysis of each ethyl group occurs in the de-esterification, producing mono-ethyl phthalate (MEP) and finally PA. For DMP, *trans*-esterification produces mono-methyl phthalate and finally PA.

In the secondary degradation, PA is metabolized and mineralized under aerobic and anaerobic conditions. Under aerobic conditions, PA's ring cleavage occurs differently with gram-negative and gram-positive bacteria. Gram-negative bacteria produce dioxygenase to catalyze phthalate 4,5- dioxygenase into *cis*-4,5-dihydroxy-4,5-dihydrophthalate, whereas gram-positive bacteria convert PA via *cis*-3,4-dihydroxy-3,4-dihydrophthalate. Both pathway produce protocatechuate, which is further metabolized via either *ortho*- or - *meta* cleavage (by ring cleavage enzymes). In the *ortho*-cleavage, protocatechuate is cleaved in the ring to form beta-carboxy-cis, *cis*-muconic acid, and finally beta-ketoadipate. *Pseudomonas fluorescens* and *P. putida* are known to support this metabolism. In the *meta*–cleavage, protocatechuate is degraded into 4-carboxy-2-hydroxymuconic and semi-aldehyde, which is finally oxidized to pyruvate and oxaloacetate. These intermediate products are then oxidized and enter the tricarboxylic acid (TCA) cycle.

Under anaerobic conditions, PA is converted to benzoate via decarboxylation. The benzoate is further degraded to acetate and methane. During the decarboxylation pathway, intermediate products have been identified in some cases. Anaerobic degradation pathway is still ambiguous due to the lack of some information on PAEs-bacteria degrading. So far, *Clostridium* sp. and methanogenic consortia were reported as degrading phthalate anaerobically.

4.4. The specific enzymes involved in PAE biodegradation

A list of bacteria and their specific enzymes for PAE biodegradation was summarized in Table 3. Esterase and hydrolase play an important role in the primary pathway (conversion to PA), whereas protocatechuate 3,4 dioxygenase, catechol 1,2, dioxygenase, and phthalate dioxygenase are key to the secondary pathway of PAE biodegradation. Niazi et al. (2001) reported that the four isoesterases (Et1–4) from the cell-free extract of the bacterium *Bacillus* sp. had the ability to utilize DMP as a carbon source. Their results also indicated that isoesterases Et-1 and Et-4 showed a significantly higher preference for DMP hydrolysis compared to Et-2 and Et-3, which played a vital role in the de-esterification. Hydrolase, purified from cell extracts of *Gordonia* sp. strain P8219, was reported to effectively hydrolyze DEHP (Nishioka et al., 2006). Serine hydrolases were reported to be able to

perform meta-cleavage of intermediates of the PAE-metabolism process (Habe et al., 2003; Omori et al., 1986).

Rhodococcus sp. strain DK17 was reported to be able to oxidize and subsequently dehydrogenate PAEs to form protocatechuate through the genes encoding protocatechuate 3,4-dioxygenase (ring-cleavage enzyme) (Choi et al., 2005). A similar finding revealed that two bacterial strains *Acinetobacter* sp. and *Arthrobacter* sp. could take Butyl Benzyl Phthalate (BBP) as the carbon source and the degradation occurred via protocatechuate 3,4-dioxygenase that was produced by these bacteria (Yang et al., 2013b). Moreover, catechol 1,2-dioxygenase and catechol 2,3-dioxygenase could help in cleaving the benzene ring, and the activity of the former was reported to be higher than that of the latter (Sanakis et al., 2003). Chen et al. (2007) indicated that *Microbacterium* sp. strain CQ0110Y contained both catechol 1,2-dioxygenase and catechol 2,3-dioxygenase, which helped accelerate DEHP degradation via hydroxylation of the benzoic acid. Then, the oxidation occurred to produce catechol and muconic acid, which finally entered the TCA cycle.

5. Green technologies for remediation of PAE-contaminated soil

In recent years, various promising green technologies such as biochar adsorption, bioaugmentation, and phytoremediation have successfully been employed in PAE removal (Tables 4–6, respectively). Their advantages and disadvantages are discussed below.

5.1. Biochar adsorption

Biochar, with a large surface area and high porosity, has been examined for the removal of many organic contaminants, including PAEs (Hung et al., 2018; Lap et al., 2021; Tran et al., 2022a; Vu and Wu, 2019; Yang et al., 2013a). Like other carbonaceous adsorbents, hydrophobic interaction, **π-π** coordination, and hydrogen bonding are the main adsorption mechanisms of biochar for organic pollutants (Vu and Wu, 2022; Zhang et al., 2013). Functional groups on the surface of carbonaceous adsorbents like biochar might improve the adsorption capacity via ion exchange, complexation, co-precipitation, and electrostatic interaction (Chen et al., 2021; Vu and Wu, 2022; Wu et al., 2019). Freundlich isotherm is often used to describe the biochar adsorption of PAEs. Zhang et al. (2016) amended PAE-contaminated soil with 0.5% bamboo biochar and achieved relatively high N values (sorption intensity) of 0.67–0.8. Similarly, Zhang et al. (2014) added rice straw biochar with 0.1–0.5 (w/w) to remove DEP and achieved N values of 0.31–1.01.

The adsorption efficiency is affected by the physicochemical properties of contaminated soil (e.g., organic matter and soil texture) and biochar (Nguyen et al., 2022a; Zhang et al., 2016). High organic carbon soil (HS) may increase adsorptive removal more than low organic carbon soil (LS). For example, reported K_f values for LS and HS with the amendment of 0.5%–1.0% pig manure-derived biochars were 2.78 ± 0.18 and 4.11 ± 0.17 $mg^{1-N}L^{N}kg^{-1}$, respectively (Chen et al. (2021). HS contains more functional groups, which help bind PAEs via electrostatic, hydrophobic interactions, and hydrogen bonds (Zhang et al., 2013). Also, high mineral-humus complexes in HS increase its affinity for PAEs (Chen et al., 2019; Wu et al., 2019; Zhang et al., 2016). Xiang et al. (2020) investigated the effects of paddy soil on DBP sorption using biochar. The findings showed that the positive effects on DBP sorption

were brought about by high aerobic surface paddy soil, which further helped enhance DBP biodegradation via aerobic bacterial metabolism (Jin et al., 2014). On the other hand, Qin et al. (2018) investigated the DBP sorption by pig biochar (PB) and bamboo biochar (BB). The higher efficiency of PB was attributed to its higher surface area, surface alkalinity, and mineral content (Jin et al., 2014; Zhang et al., 2016).

Biochar can also reduce the bioavailability of PAEs and improve the properties of soil (Zhang et al., 2016). Chen et al. (2019) reported that biochar significantly reduced the bioavailability of DEHP and enhanced microbial activity, reducing DEHP uptake in contaminated soil. He et al. (2016) revealed that organic matter was key to reducing DEHP bioavailability in contaminated soil. Moreover, biochar plays an important role in soil amendment through immobilization and adsorption of organic contaminants, which significantly enhanced the properties of contaminated soil (Chen et al., 2021). In soil, biochar increases pH by allowing hydrogen ions to form due to its negatively charged functional groups (Zhang et al., 2013) and through the potential release of alkali salts (e.g., Ca, Mg and Na). For instance, Dai et al. (2014) indicated that adding 1% biochar derived from swine manure increased the soil pH by 9%–19%.

5.2. Bioaugmentation

Microbial inoculation (bioaugmentation) is the addition of specific bacterial strains to accelerate the degradation of PAE in contaminated soil. Several bacterial strains have been isolated from contaminated soil to enhance PAE biodegradation. Many bacterial strains such as *Gordonia* sp. QH-11 (Kong et al., 2019), *Rhodococcus ruber* YC-YT1 (Yang et al., 2018), and *Rhodococcus pyridinivorans* XB (Zhao et al., 2018) have successfully been employed to degrade PAEs with impressive efficiencies (80%–100%). For instance, Kong et al. (2019) showed that at a high initial DEP concentration of 400 mg kg^{-1}, the biodegradation efficiency reached 100% with the addition of *Gordonia* sp. QH-11. Similarly, the DEHP degrading efficiency of *Rhodococcus ruber YC-YT1* reached 93% after seven days of incubation (Yang et al., 2018).

Bacterial degradation often occurs as the hydrolysis of ester linkage between alkyl chains and the aromatic ring (under aerobic/anaerobic conditions) (Liang et al., 2008). Thus, the biodegradation efficiency decreased with increasing alkyl chain length. For example, Kong et al. (2019) reported that 100% of the DBP (short-chain) at a high initial concentration (400 mg kg^{-1}) was degraded within 15 days of incubation by *Gordonia* sp. However, at an initial concentration of 50 mg kg^{-1}, only 92% of DEHP (long-chain) was removed after 30 days of incubation with a similar *Gordonia* strain (Zhang et al., 2020). Also, operational conditions (e.g., moisture, pH, temperature, and salinity) significantly affect the growth and activity of bacterial strains during incubation. Jin et al. (2016) indicated that with the addition of *Gordonia* sp. QH-12, DBP biodegradation rate of varying initial concentration (100–750 mg kg^{-1}) reached its highest at pH 7.0 and temperature 30 °C. With the addition of *Gordonia alkanivorans* YC-RL2, DEHP biodegradation was significantly inhibited at high salinity (6%), which caused stress and negative effects on bacterial growth (Nahurira et al., 2017).

Among the PAE-degrading bacteria, *Gordonia* plays the most important role (Benjamin et al., 2015; Jin et al., 2016). Various strains of the genus *Gordonia* have been isolated

from contaminated soil, e.g., *Gordonia* QH-12, *Gordonia terrae* RL-JC02, *Gordonia terrae* RL-JC02, and *Gordonia* sp. QH-11 (Nahurira et al., 2017; Zhang et al., 2020). As the sole carbon and energy source, these bacteria metabolized PAEs into intermediate products (Benjamin et al., 2015). For instance, *Gordonia* sp. QH-11 rapidly converted PAEs into phthalic acid (PA), which then was degraded by protocatechuate 3,4-phthalate dioxygenase (Kong et al., 2019). *Gordonia terrae* RL-JC02 hydrolyzed DEHP into PA via mono (2-ethylhexyl) phthalate (MEHP), and then PA was quickly metabolized to protocatechuic acid (PCA) (Zhang et al., 2020). *Gordonia* sp. JDC2 is able to metabolize PAEs, but doesn't appear to form PA (Wu et al., 2010). At present, the mechanism of PAE biodegradation pathways with *Gordonia* remains incomplete, which should require future research to clarify.

5.3. Phytoremediation

Phytoremediation uses plants/trees to remove contaminants from environmental media (Nguyen et al., 2020b, 2021). Therefore, it is considered a green and environmentally friendly treatment technology (Bui et al., 2017, 2019; Nguyen et al., 2020a). With phytoremediation, PAEs are removed through the uptake and translocation mechanisms by various plant species, e.g., *Ipomoea aquatica*, *Chinese cabbage*, and *Medicago sativa* (Cai et al., 2006, 2008b; Ren et al., 2020; Zhao et al., 2015). Ma et al. (2012) conducted field-scale phytoremediation experiments of PAEs using alfalfa (*Medicago sativa*) and achieved 80% removal of six PAEs. Maize cultivar was reported to remove up to ~88% DEHP after 40 days (Li et al., 2014). Similarly, Mo et al. (2009) showed that the bioconcentration factor of uptake of PAEs by vegetables (*Brassica chinensis* var. *parachinensis, Ipomoea aquatica*) varied from <0.0001–0.61.

The mechanisms of phytoremediation of PAEs are proposed in Fig. 5. Many plants adsorb PAEs and reduce their toxicity through mechanisms like rhizobacteria degradation, phytostabilization, phytoextraction, and phytovolatilization (Li et al., 2014). Other plants convert PAEs to monophthalates through detoxification with the help of microbial communities residing in the root nodules (Antoniadis et al., 2017). In the stem and leaf, phytostabilization and phytoextraction are the main removal mechanisms, in which extraction and transformation occur (Liao et al., 2019). Garden lettuce (*Lactuca sativa* L. *var. longifolia*) showed significant DBP absorption, in which high concentrations were found in the stem, root, and leaf (Liao et al., 2019). Leaves and stems were reported to accumulate DEHP better than roots (Cai et al., 2015). Ren et al. (2020) reported that DnBP could be taken by roots and shoots of alfalfa (*Medicago sativa*). The results also indicated that DnBP accumulated mainly in roots and adsorption to root epidermis and was the primary uptake mechanism. Further, DnBP could be converted to MnBP and PA through de-esterification followed by accumulation in cell components and organelles. The contaminants were gradually transferred to soluble components, and finally removed through phytoextraction.

6. Recommendations for future research

Composting process is robust and efficient for the remediation of PAE-contaminated soil. Further, combining composting with other technologies, e.g., bioaugmentation (inoculation

of bacterial strains), could enhance the removal efficiency and thus reduce the total treatment cost. However, knowledge gaps remain and future research is needed.

- Contaminated soil often contains a high number of contaminants, e. g., heavy metals and persistent organic compounds (e.g., pesticides, polycyclic aromatic hydrocarbons, petroleum hydrocarbons) with varying concentrations and forms (Tran et al., 2022b). Therefore, future research should address the removal of PAEs under the impacts of co-existing contaminants and other inorganic/organic chemicals in soil.
- The physicochemical properties of soil (e.g., texture, particle size, soil organic matter (SOM), ion exchange capacity) play important roles in PAE degradation. Yet, they are only addressed in a few studies so future research should pay more attention to these properties.
- The microbial community is key to effective PAE biodegradation. The structure and dynamics of the bacterial community in composting degradation of PAEs have been presented and well discussed. Thus, in the future, the bio-physicochemical properties of important PAE-degrading strains should be evaluated to establish the optimal conditions for composting remediation of PAEs in soil.
- Metagenomic sequencing is an effective tool to gain a comprehensive understanding of microbial communities. It should be widely applied during composting to identify and evaluate PAE biodegrading bacterial strains, especially anaerobic strains.
- The role of anaerobic bacteria is very limited in PAE biodegradation, and information on the anaerobic degradation pathways remains incomplete. Studies clarifying this should be conducted.
- Emissions of volatile organic compounds (VOCs) and the discharge of leachate reduce the number of materials in the composting mixture. Therefore, future research should perform the calculation of mass balance to evaluate the removal efficiency.
- Combining composting with other technologies is very promising in terms of enhancing removal efficiency and speeding up the degradation. However, the upscaling and economic feasibility of this idea should be further evaluated.
- As a green technology, biocatalysis is widely known for its robustness and effectiveness in degrading emerging organic pollutants through enzyme catalysis. In spite of this, there are still limitations associated with the combination of composting and biocatalysts. Therefore, combining these techniques in future studies will contribute to the full filling of knowledge gaps in this area.

7. Conclusions

PAE contamination in soil is one of the most concerning global environmental issues. Composting is a promising green and environmentally friendly PAE-degradation technology

(removal efficiency 25%–100%). Aerobic composting shows a higher biodegradation rate efficiency than anaerobic composting. In PAE biodegradation pathways, long alkyl chains are converted into short chains as by-products and other non-toxic products via microbial activity, including de-esterification, beta-oxidation, and *trans*-esterification. High-throughput sequencing provided insight into the structure and characteristics of microbial communities during the composting process. During the PAE biodegradation, the richness and diversity are significantly changed, and bacteria are key players in PAE degradation since their specific enzymes (e.g., hydrolase, esterase, catechol 1,2 dioxygenase, and protocatechute 3,4 dioxygenase) accelerate the degradation. Notably, green technologies including biochar adsorption, bioaugmentation (inoculation of bacterial strains like *Rhodococcus* sp. and *Gordonia* sp.), and phytoremediation have shown their effectiveness in PAE degradation. Combining composting with these technologies, clarifying the roles of each bacterial strain, and assessing the upscaling and economic feasibility is welcomed in the future research.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgement

This work is partially supported by the KU Center for Chemical Biology of Infectious Diseases (P20GM113117) (JMH).

Data availability

Data will be made available on request.

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HIGH LIGHTS

- PAE-contaminated soil is an environmental concern worldwide.
- Composting is robust and effective for the remediation of PAE-contaminated soil.
- The richness and diversity of the microbial community changed during composting.
- Bacteria with their specific enzymes play key roles in PAE degradation.
- Green technologies can be integrated into composting for enhanced PAE removal.

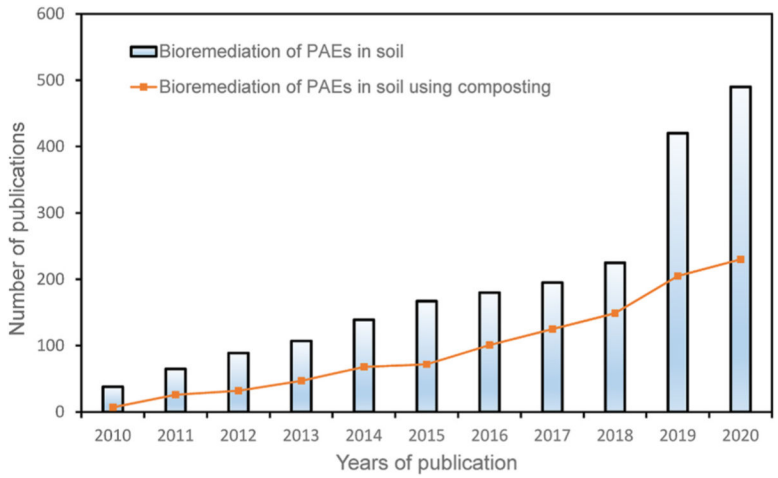


Fig. 1.

Growth in several publications of bioremediation of phthalates in soil and that of bioremediation of phthalates in soil using composting from 2010 to 2020.

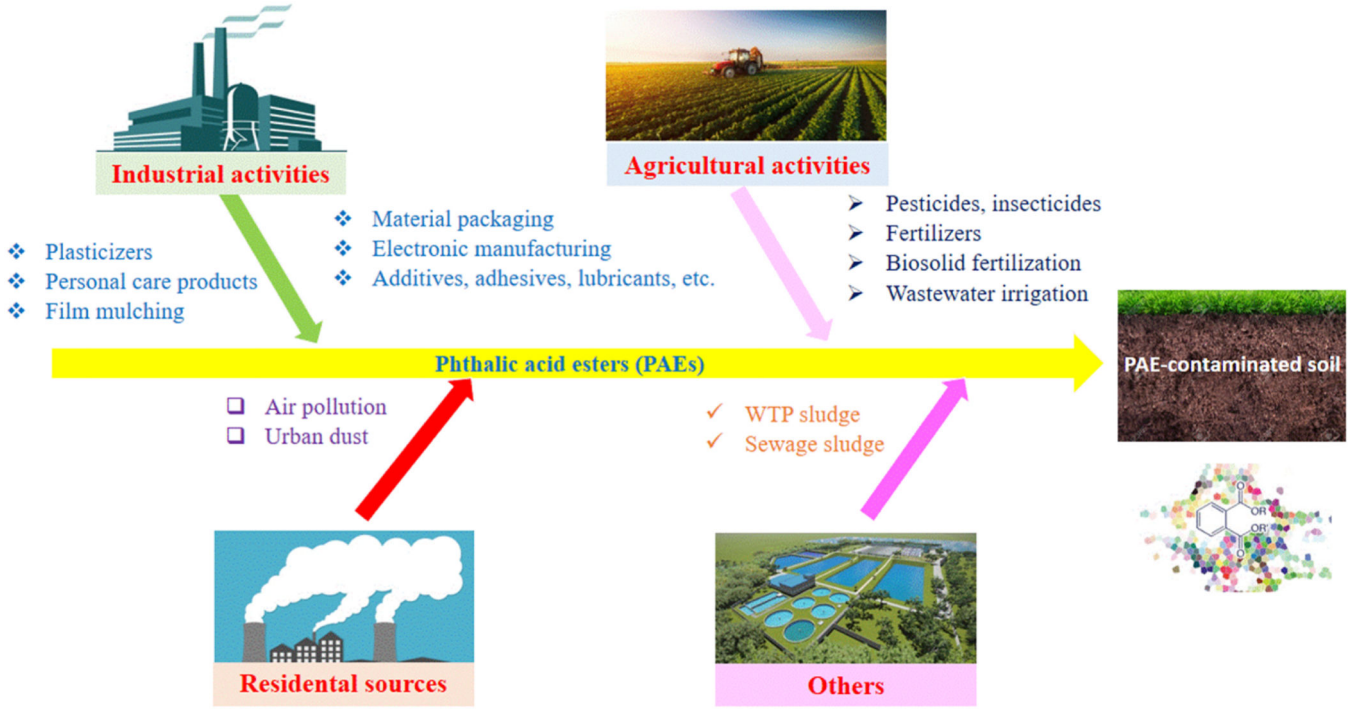


Fig. 2.
Main sources of PAEs in contaminated soils.

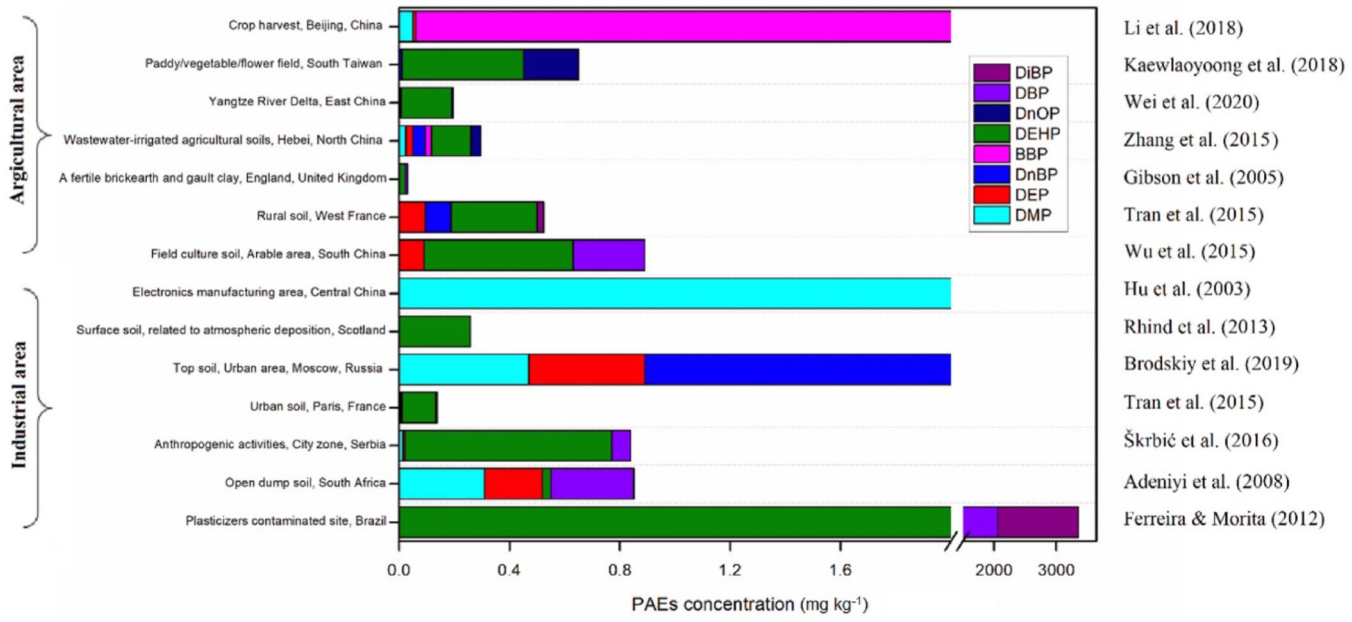


Fig. 3.
The situation of PAE-contaminated soil worldwide.

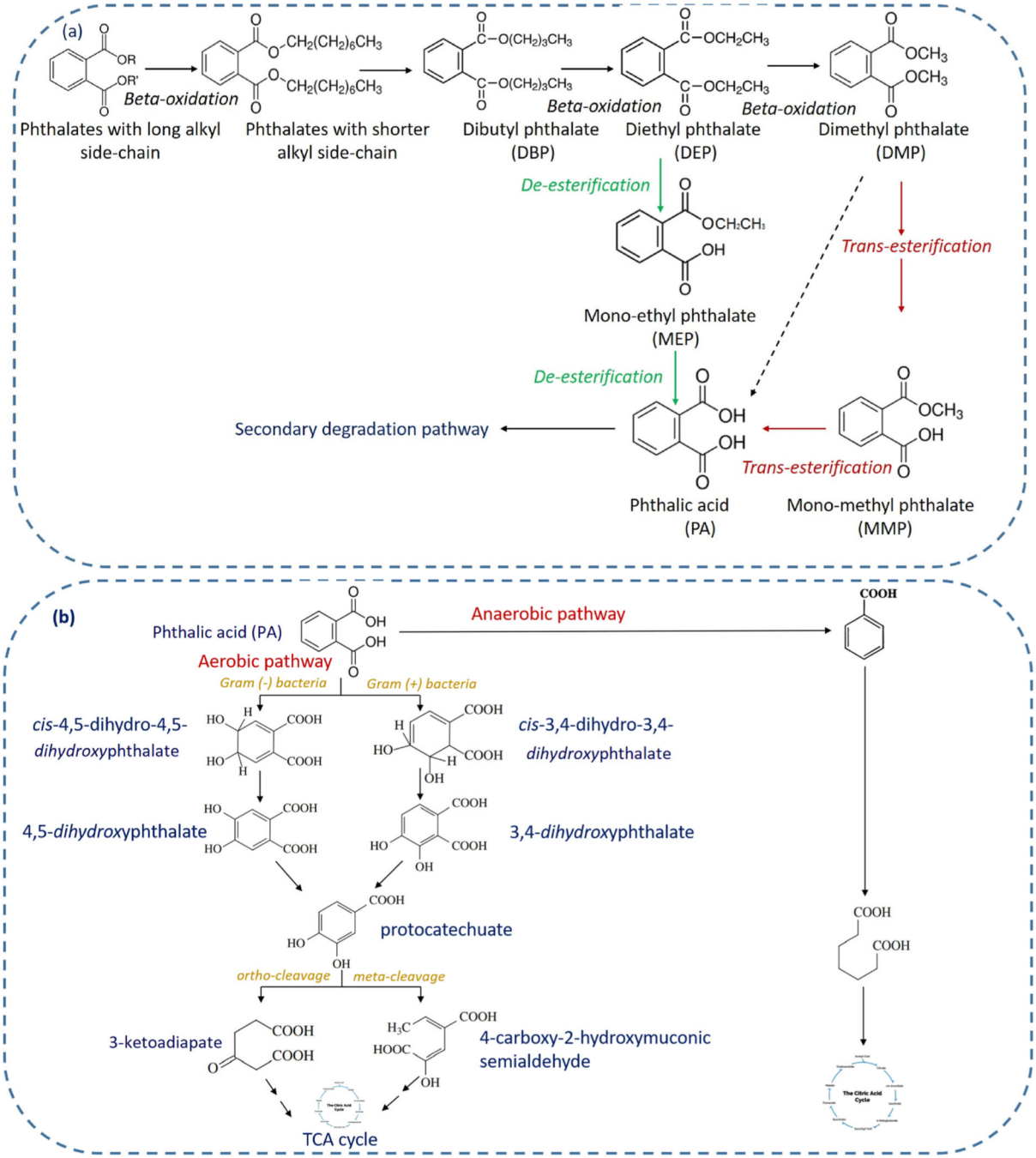


Fig. 4.

Proposed (a) primary and (b) secondary pathways for PAE biodegradation under aerobic and anaerobic conditions. Adapted from (Benjamin et al., 2015; Liang et al., 2008).

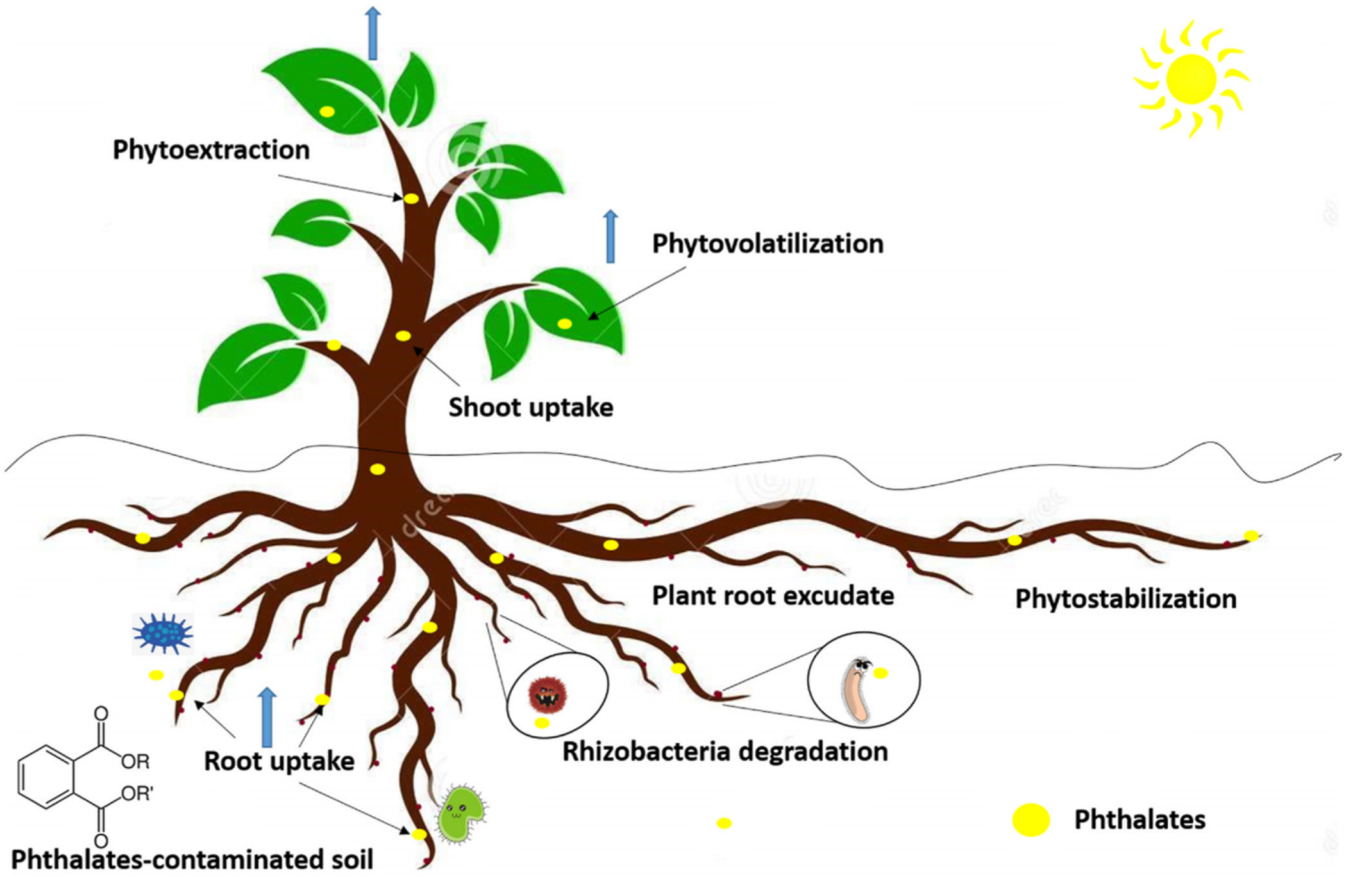
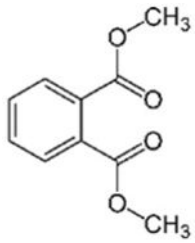
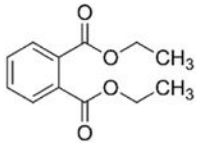
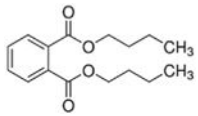
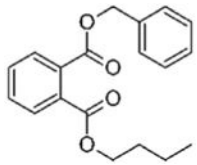
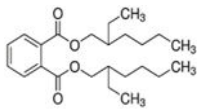
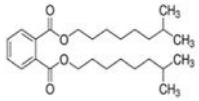
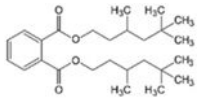
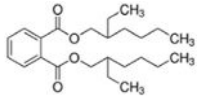
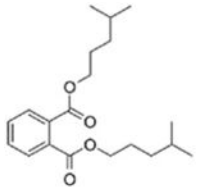
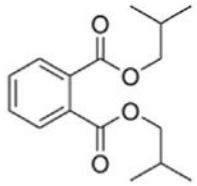


Fig. 5.
Proposed mechanism of phytoremediation of PAE-contaminated soil.

Table 1

Characteristics of various commercial PAEs (data were mostly obtained from the US National Library of Medicine).

Abbr.	Compounds	Formula	Structure	Log (K_{ow})	Log K_{oc} (Lkg^{-1})	Vapor pressure (mmHg at 25)	Solubility (mgL^{-1}, 25 °C)	Melting point (°C)	Boiling point (°C, at 760 mm Hg)
DMP	Dimethyl phthalate	$C10H10O_4$ M = 194.18 g mol^{-1}		1.64	39.81	9.12×10^{-3}	4000	5.5	283.7
DEP	Diethyl phthalate	$C12H14O_4$ M = 222.24 g mol^{-1}		2.50	69.18	1.67×10^{-3}	1080	− 40.5	295.0
DBP	Dibutyl phthalate	C16H22O4 M = 278.34 g mol^{-1}		4.50	1.38×10^{3}	2.01×10^{-5}	11.20	− 35.0	340.0
BBP	Butyl benzyl phthalate	$C_{19}H_{20}O_4$ M = 312.40 g mol^{-1}		4.73	5.25×10^{3}	8.25×10^{-6}	2.69	− 35.0	370.0
DEHP	Diethylhexyl phthalate	C24H38O4 M = 390.60 g mol^{-1}		7.60	8.71×10^{4}	1.42×10^{-7}	0.27	− 55.0	384.0

Abbr.	Compounds	Formula	Structure	Log (K_{ow})	Log K_{oc} (Lkg^{-1})	Vapor pressure (mmHg at 25)	Solubility (mgL^{-1}, 25 °C)	Melting point (°C)	Boiling point (°C, at 760 mm Hg)
DINP	Diisononyl phthalate	$C_{26}H_{42}O_4$ M = 418.60 g mol^{-1}		9.37	5.52	5.40×10^{-7}	0.20 (20 °C)	− 43.0	77.8
DiDP	Diisodecyl phthalate	$C_{28}H_{46}O_4$ M = 446.70 g mol^{-1}		10.36	6.04	5.28×10^{-7}	0.28	− 45.6	425.8
DnOP	Dinoctyl phthalate	C24H38O4 M = 390.60 g mol^{-1}		8.10	2.40×10^{4}	1.00×10^{-7}	0.02	25.0	220.0
DIHP	Diisohexyl phthalate	C20H30O4 M = 334.40 g mol^{-1}		6.30	5.25×10^{4}	5.00×10^{-6}	0.05	− 58.0	350.0
DIBP	Diisobutyl phthalate	C16H22O4 M = 278.34 g mol^{-1}		4.11	1.38×10^{3}	4.76×10^{-5}	0.01	− 64.0	296.0

Chemosphere. Author manuscript; available in PMC 2023 November 01.

Table 2

Summary of aerobic/anaerobic composting remediation of PAE-contaminated soil.

PAEs	Compost materials	Initial concentration (mg kg^{-1})	Conditions (aerobic/ anaerobic)	Incubation time (days)	Scale	Removal efficiency	Ref.
Total PAEs	Household waste and wood waste	14.1–19.4	Aerobic (minimum oxygen level of 12%)	56	Pilot scale (30 m^3, W × L × H = 6 × 2.4 × 2.2 m)	70.8%–97.4%	Graça et al., (2021)
DEHP	Dead pigs, bamboo, and composted sheep manure	100	Anaerobic	112	Lab-scale	55.5%–69.8%	He et al., (2018)
Total PAEs	Sludge, pig manure, and rice straw	25.5	Aerobic	60	Bioreactor (volume = 30 L)	74.7%–78.0%	Fu et al., (2013)
Total PAEs	Sewage sludge, sawdust, and wood chip	4.8–11.6	Aerobic (total aeration of 4 h day^{-1})	56	Full-scale compost pile (W × H × L = 1.5 × 2.0 × 2.0 m)	58.3%–90.6%	Cai et al., (2012)
DEHP	Sewage sludge	50–250	Aerobic	30	Lab-scale	91.3%–96.5%	Yuan et al., (2011)
DBP				10		92.1%–97.5%	
DEHP	Sewage sludge and yard and park waste	31	Aerobic	24	Full-scale compost pile (W × H × L = 4 × 2 × 80 m)	84%	Poulsen & Bester, (2010)
DEHP	Sewage sludge and sheep manure	31	Aerobic	62	Bioreactor (V = 28 L, D = 0.3 m)	97%	Pakou et al., (2009)
DBP	Spent mushroom compost and animal manure	500	Aerobic	20	Lab-scale	91%	Chang et al., (2009)
DEHP		500		20		88%	
DEHP	Sludge cake and sawdust	200–300	Aerobic, air pump (10 L min^{-1})	18	Bioreactor (V = 110 L)	85%–88%	Cheng et al., (2008)
DEHP and DBP	Sewage sludge and rice straw	8.1	Aerobic	56	Lab-scale	76%	Cai et al., (2008a)
DEHP	Sewage sludge and wheat straw	36.37	Anaerobic	143	Pilot scale (5 tons, W × L × H = 2 × 4 × 1.5 m)	60%	Gibson et al., (2007)
DEP	Fruit and vegetable waste and bark/peat	10–100,000	Aerobic	84	–	25%–100%	Kapanen et al., (2007)
DEHP	Lagoon sludge and straw	6.26	Aerobic	180	Composter (2.5 tons)	91%	Amir et al.,
	Lagoon sludge and grass	28.7		135		94%	(2005)
DEHP	Sewage sludge and	62–99	Anaerobic	270	Composter (40 kg)	>50%	Bagó et al., (2005)
DEHP	Sewage sludge and tree-bark and peat	77	Aerobic	85	Bioreactor (V = 220 L, D = 1.2 m) Rotary drum (V = 5 m^3)	62% 34%	Marttinen et al., (2004)
DEHP	Sewage sludge and garden park waste	38–160	Aerobic	25	Bioreactor (V = 10 L)	96%–99%	Moeller & Reeh, (2003)

PAEs	Compost materials	Initial concentration (mg kg^{-1})	Conditions (aerobic/ anaerobic)	Incubation time (days)	Scale	Removal efficiency	Ref.
DEHP	Sewage sludge and straw and green waste Municipal biowaste and shredded shrubbery	3–6 2.35	Aerobic	370	Bioreactor (V = 1.8 m^3, D = 1.5 m)	91% 86%	Hartlieb et al., (2001)

Table 3

Bacteria with their encoding specific enzymes for PAE biodegradation.

Phylum	Bacteria	Characteristics	Enzymes	Compounds	References
Firmicutes	*Bacillus* sp.	Gram positive, aerobes	Esterase	DMP	Niazi et al., (2001)
Actinobacteria	*Gordonia* sp. strain P8219	Gram positive, aerobes	Hydrolase	DEHP	Nishioka et al., (2006)
	Rhodococcus erythropolis	Gram positive, aerobes	Esterase	DAPs	Kurane, (1997)
	Micrococcus sp.YGJ1	Gram positive, aerobes	Esterase	DAPs, MAPs	Akita et al., (2001)
	Arthobacter sp.	Gram positive, aerobes	Protocatechuate 3,4 dioxygenase	BBP	Yang et al., (2013b)
	Rhodococcus sp.strain DK17	Gram positive, aerobes	Protocatechuate 3,4 dioxygenase	Terephthalate	Choi et al., (2005)
	Microbacterium sp. strain *CQ0110Y*	Gram positive, aerobes	Catechol 1,2,dioxygenase Catechol 2,3,dioxygenase	DEHP	Chen et al., (2007)
Proteobacteria	*Pseudomonas* sp. 054	Gram positive, aerobes	Esterase, Protocatechuate 4,5 dioxygenase	DMTP	Tserovska et al., (2006)
	Shingomonas yanoikuyae DOS01	Gram negative, aerobes	Esterase	DMP	Gu et al., (2009)
	Comamonas acidovorans D4	Gram negative, aerobes		DMTP	Patel et al., (1998)
	Agrobacterium sp.	Gram negative, aerobes	Esterase	DBP	Wu et al., (2011)
	Acinetobacter sp	Gram negative, aerobes	Esterase, Hydrolase	BBP	Hashizume et al., (2002)
	Delfia sp.TBKNP05	Gram negative, aerobes	Esterase, Phthalate dioxygenase	DBP	Patil et al., (2006)

Table 4

Remediation PAE-contaminated soil by biochars.

Biochar components	Production temperature (°C)	Added biochars rate (%, w/w)	PAEs types	Best adsorption parameters (Freundlich equation)			Important factors on sorption capacity	Ref.
				Kf ($mg^{1-N}L^{N}kg^{-1}$	N	R^2		
Bamboo and rice straw	350 650	0.1, 0.5	DEP	HS: 590.30 ± 13.63 LS: 22.25 ± 1.50	1.01 0.31	0.97 0.96	Feedstock materials, pyrolysis temperature, pH of biochar, and soil organic carbon levels	(Zhang et al. (2014)
Wood biochar, grass biochar, and animal waste biochar	450	N/A	DBP	288.40	0.34	0.97	Presence of Cd^{2+} could increase the sorption of low concentrations of DBP	(Jin et al. (2014)
Bamboo	820	0.5	DEP	LS: 45.02 ± 0.35 HS: 21.88 ± 0.11	0.67 0.80	0.99 0.98	Soil organic carbon levels, and aging processes of biochar	(Zhang et al. (2016)
Bamboo sawdust and rice straw	500	2.0	DEHP	N/A	N/A	N/A	OC contents	(He et al. (2016)
Pig and bamboo biochar	650 750	0.5	DEHP	N/A	N/A	N/A	Organic amendments, soil pH, and the organic carbon contents	(He et al. (2018)
Straw-derived biochar	600	1.0, 5.0	DBP	0.61 ± 0.02	0.84	0.78	OM contents, the pore size of the soil	(Xiang et al. (2020)
Wheat straw biochar	450	0.01, 0.02, 0.05	DMP	2.43	1.25	0.99	Biochar addition	(Yan and Quan (2020)
Pig biochar	650	0.5, 1.0	DEP	LS: 2.78 ± 0.18 HS: 4.11 ± 0.17	0.65 0.92	0.92 0.94	Biochar type, application dose, and soil organic carbon Content	(Chen et al. (2021)

Notes: HS – high organic carbon; LS – low organic carbon; K_f – Freundlich sorption coefficient, N (dimensionless) – nonlinearity index; N/A – Not Available.

Table 5

Bioaugmentation of specific bacterial strains for remediation of PAE-contaminated soil.

Bacterial strains	PAEs	Initial concentration (mg kg^{-1})	Incubation time (days)	Removal efficiency	Ref.
Rhodococcus pyridinivorans XB	DEHP	25–100	50	67.6%–85.4%	Zhao et al., (2018)
Pseudomonas sp. *DNB-S1*	DBP	5	7	36.5%–40.42%	Li et al., (2020)
Microbial consortium (CM9)	DEHP	100	42	87.5%	(Bai et al., 2020) (biochar)
Gordonia terrae RL-JC02	DEHP	50	30	91.8%	Zhang et al., (2020)
Gordonia sp. *QH-11*	DBP	400	15	100.0%	Kong et al., (2019)
Rhodococcus sp. *WJ4*	DEHP	1000	21	57.5%	Wang et al., (2015b)
Agromyces sp. *MT-O*	DEHP	100	12	82.1%	Zhao et al., (2016)
Mycobacterium sp. *YC-RL4*	DEHP	100	10	83.3%	Ren et al., (2016)
Gordonia sp. *Lff*	DEHP	100	35	90.0%	Wang et al., (2019b)
Rhodococcus ruber YC-YT1	DEHP	100	7	79.7%–92.9%	Yang et al., (2018)

Table 6

Phytoremediation of PAE-contaminated soil.

Plants species	PAEs	PAE concentrations (*) (**) (***)	Bioconcentration factors (BCFs)	Experimental duration (days)	Remarks	References
Water spinach (*Ipomoea Aquatica*)	DnOP, BBP, DBP, DEHP, etc.	4.5, 10.3, 22.5 mg kg^{-1} (***)	0.014–0.744	39	Plant uptake PAEs by roots, shoots	(Cai et al. (2008b)
Elven vegetable species (*Brassica parachinensis, Brassica Chinensis, Ipomoea Aquatica, Brassica juncea*, etc.)	DnOP, DMP, BBP, DBP, DEHP, etc.	0.073–11.2 mg kg^{-1} (*)	<0.0001–0.61	N/A	The highest PAEs were found in *B. juncea* and *B. parachinensis*	(Mo et al. (2009)
Alfalfa *(Medicago sativa*)	DEHP	117.4 ± 5.2 mg kg^{-1} (***)	<0.1	40	Can remove 76.7% DEHP	(Li et al. (2014)
Twenty cultivars of rice (*Oryza sativa* L.)	DEHP	19.68 ± 0.23 mg kg^{-1} (***)	Uptake 0.40–7.58 mg kg^{-1} (in shoots) Uptake 0.26–11.8 mg kg^{-1} (in roots)	Growth stages	DEHP was highly accumulated in stems and leaves	(Cai et al. (2015)
Edible plants (lettuce, strawberry, and carrot)	DnBP DEHP, etc.	500 μg kg^{-1} (**)	0.16 ± 0.01–4.78 ± 0.59	28	Vegetable plants could take up PAEs	(Sun et al. (2015)
Chinese flowering cabbage (*Brassica parachinensis* L.)	DBP, DEHP	DBP: 81.35 mg kg^{-1} (**) DEHP: 85.92 mg kg^{-1} (**)	DBP: 0.139 ± 0.028–0.170 ± 0.029 DEHP: 0.126 ± 0.029–0.164 ± 0.029	45	PAEs were accumulated by Chinese cabbage	(Zhao et al. (2015)
Vegetable samples in the diet (*Brassica rapasubsp. Chinensis, Cucumis sativus, Solanum melongena,Phaseolus vulgaris*, etc.)	DMP, DEP, DnBP, DEHP	0.95–8.09 mg kg^{-1} (*)	N/A	N/A	PAE exposure through the vegetable intake	(Chen et al. (2017)
Garden lettuce *(Lactuca sativa* L. *var.longifolia*)	DBP	5 mg L^{-1} (**)	N/A	21	A promising plant for DBP phytoremediation	(Liao et al. (2019)
Alfalfa *(Medicago sativa)*	DnBP	8.0 mg L^{-1} (**)	Uptake 36.5 μg kg^{-1} (in roots) Uptake 4.1 μg kg^{-1} (in shoots	0.5–10	DnBP was accumulated mainly in roots	(Ren et al. (2020)
Sunflower (Helianthus annuus)	DEHP	1000 mg kg^{-1} (**)	Uptake 20 mg kg^{-1} (in shoots) Uptake 25 mg kg^{-1} (in roots	25	Uptake of DEHP in contaminated soil Maximum DEHP accumulated in their roots	(Mustafa et al. (2021)
Chinese cabbage (Brassica rapa var. Chinensis)	DnBP, DEHP, etc.	100 μg mL^{-1} (**)	N/A	5	PAE uptake by root Theoretical evidence for uptake, accumulation, and metabolism of PAEs in plants	(Cheng et al. (2021)

Notes:

(*)- PAEs levels in plants/vegetables;

(**) - Added PAE levels;

(***) - PAE levels in soil.