Research Progress on the Environmental Behavior and Health Risks of Phthalates in Soil: Postprint

Authors: Yang Shan, Lu Shenghong, Wang Jun, Liu Kun, Chen Gangcai, Yong Zhang, Zhang Sheng, Teng Ying

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Abstract

Phthalate esters (PAEs), also known as phthalates, are environmental endocrine-disrupting organic compounds that are typically added as plasticizers in amounts ranging from 20% to 60% in plastic, resin, and rubber products. The primary sources of PAEs in soil include agricultural chemicals, sewage irrigation, and atmospheric deposition. PAEs exhibit strong enrichment in soil and can enter various environmental media through a series of environmental geochemical processes, thereby causing environmental pollution and posing risks to human health. By integrating relevant research findings on soil PAEs from domestic and international sources, this review summarizes the contamination status of PAEs in Chinese soils, analyzes the environmental behavior of PAEs at the soil-atmosphere interface (volatilization and deposition), in the soil-plant system (plant uptake and phytoremediation), and at the soil-water interface (adsorption-desorption), as well as the environmental health risks associated with soil PAEs contamination, and identifies existing deficiencies in domestic soil PAEs research. The results demonstrate that China' s soil environment has generally been subjected to varying degrees of PAEs contamination; furthermore, soil PAEs face high eco-environmental health risks through migration and transformation processes across different interfaces. It is proposed that future soil PAEs research should prioritize regional soil contamination and environmental behavior, conduct in-depth investigations into the spatiotemporal transport and evolution patterns of soil PAEs, multi-media migration and transformation mechanisms, and risk reduction and remediation measures, thereby providing a theoretical basis for safeguarding soil ecological environment and health.

Full Text

Environmental Fate and Health Risks of Phthalate Acid Esters in Soils: A Review

YANG Shan[1], LYU Shenghong[1], WANG Jun[1,2], LIU Kun[1], CHEN Gangcai[1], ZHANG Yong[1], ZHANG Sheng[1], TENG Ying[2]

[1]Chongqing Research Academy of Environmental Sciences, Chongqing 401147, China
[2]Key Laboratory of Soil Environment and Pollution Remediation, Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China

Abstract

Phthalic acid esters (PAEs), also known as phthalates, are environmental hormone-like organic compounds widely used as plasticizers in plastics, resins, and rubber products, typically accounting for 20%-60% of additive content. The primary anthropogenic sources of PAEs in soils include agricultural chemicals, sewage irrigation, and atmospheric deposition. PAEs exhibit strong accumulation in soils and can enter various environmental media through a series of environmental geochemical processes, causing environmental pollution and human health risks.

This review synthesizes research findings on soil PAEs from China and abroad, summarizing the current contamination status of PAEs in Chinese soils and analyzing their environmental behavior at the soil-atmosphere interface (volatilization and deposition), within the soil-plant system (plant uptake and phytoremediation), and at the soil-water interface (adsorption-desorption), as well as the associated environmental health risks. The review also identifies existing gaps in domestic research on soil PAEs. Results indicate that Chinese soil environments have generally suffered varying degrees of PAEs contamination, and soil PAEs pose high ecological and health risks through migration and transformation processes across different interfaces. Future research should focus on regional soil pollution and environmental behavior, with in-depth investigations into the spatiotemporal transport and evolution patterns of soil PAEs, multi-media migration and transformation mechanisms, and risk reduction and remediation strategies, thereby providing a theoretical basis for safeguarding soil ecological environments and human health.

Keywords: Phthalate acid esters; Environmental fate; Health risk; Soil environment; Migration and transformation

1. Sources and Characteristics of Soil PAEs Contamination

1.1 Sources of PAEs in Soils

The main sources of PAEs in soils are agricultural chemicals, sewage irrigation, and atmospheric deposition. Agricultural films [14,29], fertilizers and pesticides [30–32], and sewage sludge compost [31,33] are important sources of PAEs in Chinese farmland soils. PAEs have poor stability in agricultural films and easily leach from the matrix [2,34], with strong correlations observed between soil PAEs accumulation and film material, color, thickness, usage intensity, and mulching patterns [11,14,30].

Studies show that the average PAEs content in Chinese fertilizers is 0.25 mg · kg^{-1} [35], and application can increase soil PAEs contamination levels by 1–2 times [36]. Various sewage sludge composting methods can significantly increase soil PAEs concentrations [9,31,37]. Long-term sewage irrigation causes PAEs in wastewater to bind with soil organic matter, resulting in substantial accumulation in soils and exacerbating PAEs pollution [36]. PAEs attached to atmospheric particulate matter also enter soils through deposition, representing another important cause of PAEs contamination in suburban and industrial areas [31,38–39]. Thus, sources of soil PAEs are complex and widespread, and identifying these sources and their respective contributions is crucial for studying soil PAEs pollution and developing control measures.

1.2 Contamination Characteristics of Soil PAEs

Chinese soil environments have generally experienced varying degrees of PAEs contamination, with total concentrations reaching dozens of milligrams per kilogram [11,23,40]. Soil PAEs contamination exhibits high spatiotemporal variability and distinct regional patterns, closely related to land use type, cultivation practices, and proximity to pollution sources [41–42].

Vertically, soil PAEs are primarily distributed in the 0–20 cm soil layer, with concentrations decreasing with depth [36,43]. Horizontally, relatively high PAEs concentrations are found in economically developed, densely populated cities (such as Guangdong, Beijing, and Shanghai) [11,15], industrial zones, and sewage irrigation areas [7,33,44]. Affected by agricultural film use, farmland soils in northern China also show elevated PAEs levels [15,23]. Influenced by climatic conditions, soil PAEs concentrations tend to be higher in winter and lower in summer [45].

Compared with other regions worldwide, PAEs contamination levels in Chinese soils are several to dozens of times higher than those in Western developed countries [11,23]. For farmland soils, concentrations are 10–100 times higher than in the Netherlands [23]. Furthermore, according to US soil PAEs control standards, most regions in China exceed the limits [11,23], with some areas exceeding standards by 37.6%–610.0% [29], though most remain below remediation thresholds [44]. This is primarily due to unscientific agricultural film use and unreasonable

cultivation practices in China, leading to substantial PAEs accumulation and constraining productivity in farmland ecosystems [42].

1.3 Component Characteristics of Soil PAEs

Approximately 14 types of PAEs are commercially used [1,42], but research has focused mainly on six compounds listed as priority pollutants by the US Environmental Protection Agency (USEPA), European Union (EU), and China: dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), butyl benzyl phthalate (BBP), di(2-ethylhexyl) phthalate (DEHP), and di-n-octyl phthalate (DnOP) [21,23,42,46]. The detection of PAEs compounds follows certain patterns, with high-molecular-weight DEHP and DnBP typically showing the highest detection rates and concentrations [11,14,22–23,30,47] (Table 1), as these are the primary plasticizers used in Chinese plastic production [19,23,41], accounting for 65.3%–75.4% of total soil PAEs [11,23]. The spatial distribution patterns of PAEs components are consistent with regional variations in ΣPAEs [8,15]. Compared with southern regions, agricultural soils in northern China contain 3–4 times higher DEHP and DnBP levels, likely related to lower temperatures, longer surface mulching periods, and slower degradation of residual films [23].

Globally, soil PAEs component characteristics are broadly similar, with DEHP and DnBP being the dominant pollutants [6,22,55] and other PAEs compounds present at relatively low concentrations [31]. Studies show that DEHP concentrations in some Chinese farmland soils reach up to 29.37 $mg \cdot kg^{-1}$ [38], far exceeding levels in the Netherlands (0.031–0.041 $mg \cdot kg^{-1}$) [55], Denmark (0.012–1.900 $mg \cdot kg^{-1}$) [31], and the Czech Republic (0.030–0.730 $mg \cdot kg^{-1}$) [36]. This is closely related to the long-term extensive use of low-standard agricultural films in China, resulting in large amounts of film fragments remaining in soils [14,29].

2. Environmental Behavior of PAEs in Soils

Soil serves as an important medium for PAEs accumulation, migration, and transformation. The environmental behavior of PAEs in soils refers to the dynamic equilibrium processes between soil and other environmental media, including volatilization [50], leaching [2], adsorption [56], biodegradation [46,50], abiotic degradation (photolysis and oxidation) [1,31], and plant uptake [18,24]. Through these processes, PAEs either remain in soils/plants [14,18,57] or transfer into the atmosphere and water bodies (Figure 1 [Figure 1: see original paper]) [13]. Numerous factors govern PAEs environmental behavior in soils with complex interactions, and differences in soil type [33], soil physicochemical properties [11,30,50,58], and environmental interface conditions [59] across various habitats exert substantial influences. Studies indicate that soil organic matter strongly adsorbs PAEs, with higher organic matter content leading to greater adsorption [38,41]. Variations in soil organic matter content inevitably affect

PAEs residue and transformation processes [30,41], while changes in soil structure and moisture content also influence PAEs migration and transformation [31,37].

2.1 Soil–Atmosphere Interface Processes

Exchange processes of PAEs at the soil–atmosphere interface are critical links in their soil environmental behavior, affecting pollutant transport, distribution, and fate across different regional scales [12,60–61] and potentially altering PAEs exposure pathways [12,62]. The main exchange pathways include dry/wet deposition from air to soil [11,13,47,60] and volatilization from soil to air [12,22,62], though volatilization rates are slow and deposition from the air phase to the soil phase dominates [59].

PAEs are readily adsorbed by atmospheric aerosols and particulate matter, reaching the soil surface through dry/wet deposition and showing significant correlation with particulate deposition amounts [13,60,63]. Due to their strong hydrophobicity and adsorption capacity, PAEs attached to atmospheric particles accumulate substantially in the soil surface after adsorption and are not easily leached to deeper soil layers, with accumulated concentrations decreasing with soil depth [12,62]. Compared with dry deposition, wet deposition can deliver approximately double the amount of PAEs to soils [13,60]. PAEs deposited in soils may return to the atmosphere through volatilization via water evaporation/diffusion and soil disturbance [13], forming a dynamic equilibrium of various processes at the soil–atmosphere interface under certain emission conditions.

The migration and partitioning of PAEs between soil and atmosphere are typically described using fugacity (f) models to estimate fluxes and directions [55,59,61–62]. By calculating the ratio of soil fugacity coefficient (fS) to air fugacity coefficient (fA), when $fS/fA = 1$, the system is at equilibrium with no net migration; when $fS/fA \neq 1$, the system is non-equilibrium, and PAEs migrate from the medium with higher fugacity to that with lower fugacity [12,61–62], causing soils to exhibit dual characteristics as both "sinks" and "sources" [42,47]. Generally, PAEs with high vapor pressure (PV), low octanol–water partition coefficient (Kow), and high water concentration (Cw), such as DMP, tend to migrate from soil to air; conversely, PAEs with low PV, high Kow, and low Cw, such as DEHP, tend to migrate from air to soil [12,62].

Local pollution levels [12,44], soil physicochemical properties [59,61], environmental conditions (wind speed, temperature, vegetation cover) [13,60–61], farming activities [60], and degradation [12] can all disrupt the dynamic equilibrium of PAEs between soil and atmosphere, leading to spatial redistribution. Therefore, clarifying PAEs environmental behavior at the soil–atmosphere interface requires strengthened research on the distribution and influencing factors of PAEs in regional soils and atmospheres.

2.2 Transformation in the Soil-Plant System

Plant uptake is the source of PAEs transfer and accumulation in food chains [17,39,42]. Generally, low-molecular-weight PAEs with high Kow (logK $\geq$ 5), such as DEP/DEHP, are more readily absorbed by plants [36,39], with uptake amounts proportional to soil contamination levels [24,57,64]. Plants absorb soil PAEs through two pathways: (1) direct root uptake of PAEs from soil solution, followed by upward translocation to aboveground stems and leaves driven by transpiration flow through the xylem, and accumulation in plant organic components [25,57], as observed in soybean (*Glycine max*) and maize (*Zea mays*) [1,18,24]; and (2) absorption of PAEs from surface air by aboveground plant parts (leaves, stems) and accumulation in plant organic components, as seen in flowering cabbage (*Brassica campestris*) [64]. Which transport pathway dominates depends on crop species, environmental conditions, and PAEs properties. Under the same soil conditions, the same plant shows different absorption capacities for various PAEs compounds. PAEs compounds with logKow > 3.5, such as DEHP, have low degradation rates and strong lipophilicity, strongly adsorbing to root surfaces with only small amounts transferred to leaves [24,65]; whereas PAEs compounds with 1 < logKow < 3.5, such as DEP, show more pronounced translocation from roots to plant tissues after absorption [24]. However, plant uptake rates are influenced by plant species [1,64], trait indicators (leaf shape, root type, etc.) [36,64], and cultivation methods [39,66]. Generally, PAEs content shows a positive correlation with leaf surface area [64], and more developed root systems exhibit stronger PAEs absorption capacity [18]. Some PAEs compounds with low water solubility, such as DnBP/DEHP, are difficult for plants to degrade or metabolize, accumulating in roots or stems and leaves [64], interfering with normal plant physiological metabolic activities [1,65], reducing vegetable vitamin C content, and severely harming plant growth, development, and quality [39,67].

Phytoremediation utilizes inherent plant physiological processes or integrated soil-plant-microorganism systems to absorb, transform, and transfer pollutants in situ [25,28,66], thereby reducing contamination. Studies show that soil PAEs phytoremediation mechanisms mainly include: (1) planting remediation plants such as alfalfa (*Medicago sativa*) [28,39] to absorb soil PAEs, which then accumulate non-phytotoxic metabolites in plant tissues—a direct absorption removal method [18,39]; however, plant absorption is limited, removing less than 2% of initial contamination [20,25,28]. (2) Within suitable ranges, plant root exudates or enzymes can increase available carbon sources and energy for microbial utilization, altering rhizosphere microbial community structure and abundance and enhancing microbial activity [18]; or inducing secretion of peroxidases, laccases, and other PAEs-degrading enzymes that co-metabolize with PAEs [65]—this is the primary phytoremediation pathway [25]. However, as exudate content increases, they may become competitive carbon sources for microbial PAEs degradation, potentially inhibiting PAEs attenuation, particularly for DEHP [18,65]. (3) Inoculation with arbuscular mycorrhizal (AM) fungi, whose hyphae

play important roles in accelerating PAEs degradation and reducing plant PAEs residues [18,27,57]; compared with non-inoculated soils, AM inoculation reduces soil PAEs content by 21.7%–66.4% [27,57]. Additionally, pollutant bioavailability [25,65] and selection of remediation plant combinations [66] also affect phytoremediation efficacy. Therefore, research should be strengthened on interactions between root exudates and soil PAEs, plant–microorganism combined remediation of PAEs, and plant uptake and transformation mechanisms to provide scientific support for clarifying PAEs transport and potential risks in the soil–plant system.

2.3 Soil–Water Interface Processes

Soil PAEs can enter water bodies through leaching, runoff, and infiltration, subsequently accumulating in sediments through precipitation, adsorption, and exchange processes [68–69], thereby affecting PAEs distribution and migration in soils [23,32]. After entering water bodies, the release of PAEs from soil particles into water increases with flooding duration [70]. The entire process consists of two stages: a rapid release phase and a slow release phase, with the slow release accounting for 99.12% of total release [70]. As the release process continues, PAEs concentrations in water reach a maximum, after which excess PAEs are readsorbed by sediments [69]. Eventually, PAEs content between soil and water reaches equilibrium, achieving migration and exchange among soil, water, and sediments. Changes in water conditions such as ionic strength and organic matter content [70], environmental conditions like temperature and light [71], and microbial activity [70] can disrupt the equilibrium of PAEs between soil and water.

Some PAEs compounds with high Kow, low solubility, and long side chains, such as DEHP, tend to transfer from the water phase to organic-rich sediments [1,26,69] at concentrations of 0.1–331.7 $mg \cdot kg^{-1}$ [32,69,72–73], making sediments the ultimate reservoir for PAEs [33,55,72]. Similar to soil PAEs distribution patterns, PAEs concentrations in river and lake sediments near industrial and commercial areas with high population density are higher than in farmland areas [32]. DEHP is the most contaminated PAEs compound in sediments, accounting for 49.26%–98.1% of the total [32,69,73], and can accumulate in deep sediments [45]. Different PAEs compounds show varying adsorption rates and capacities; when DnBP approaches equilibrium between sediment and water phases, DMP and DEP tend to migrate from sediment to water, while DEHP migrates from water to sediment. Under water flow and aquatic organism disturbance, sediments may become suspended in water, affecting PAEs migration and release at the soil–water interface [47,74]. Therefore, research should focus on the influence of aquatic environmental conditions on PAEs transformation processes in sediments and the mechanisms of PAEs action and transformation on sediment surfaces.

3. Environmental Health Risks of Soil PAEs

The environmental health risk of soil PAEs characterizes the potential for adverse effects on humans or other receptors exposed to environmental hazards [14]. Based on different risk receptors, risks are categorized as ecological or health risks [15,75]. For potential hazards of soil PAEs to ecosystems, ecological risk assessment can quantitatively predict the acceptability of risks to ecosystem structure and function [76]. Currently, ecological risk assessments for PAEs based on aquatic ecosystems are common in China [32,72,77], while those for soil systems are rarely reported. Results show that ecological risks of PAEs in some aquatic ecosystems exceed relevant foreign standards and China' s surface water environmental quality standard (GHZB 1—1999) [32,77], indicating significant negative ecological effects. PAEs in sediments or water bodies can enter soils through fertilization or irrigation, exposing soil environments to high ecological risks.

Soil PAEs health risks refer to hazard effects on humans under various exposure pathways including respiration, drinking water, dermal contact, skin absorption, and food ingestion [3,7,41], with risk levels determined by comparing exposure doses to critical risk values [11,39,67]. Soil PAEs health risks include carcinogenic and non-carcinogenic risks, predominantly carcinogenic risks [11,67]. Studies indicate that carcinogenic and non-carcinogenic risks of PAEs compounds in most Chinese farmland soils are below acceptable health risk thresholds, representing relatively safe conditions [3,41]. However, some farmland soils in Nanjing, Xinjiang, and Guangdong show elevated carcinogenic risks from DEHP, with risk values of 7.37$\times10^{\{-6\}-3.94\times10}\{-5\}$ (>1$\times10^\{-6\}$) [11,67], related to highly intensive agriculture and widespread agricultural film use in these regions [8,67]. DEHP has become the most potentially hazardous PAEs compound [15]. Furthermore, dietary intake is considered the primary exposure pathway for Chinese adult populations, accounting for over 90% of total intake [15,17,47]. Health risks from different exposure pathways vary with PAEs physicochemical properties: for low-molecular-weight PAEs, carcinogenic risk follows the order dermal contact > respiratory intake > oral intake, while for high-molecular-weight PAEs, the order is dermal contact > oral intake > respiratory intake [67]. In summary, rational use of agricultural products and protection against exposure pathways are effective measures for controlling soil PAEs environmental health risks.

PAEs have become a global soil environmental organic pollutant, and extensive research has been conducted worldwide on soil PAEs pollution behavior. However, in-depth investigations of PAEs migration and transformation processes and mechanisms in soil environments are still lacking. From the perspective of environmental safety and human health protection, the following research areas urgently need strengthening:

1) **Strengthen research on spatiotemporal evolution characteristics of soil PAEs pollution.** Understanding pollution characteristics of soil

PAEs at different regional scales is fundamental for accurate risk assessment. Current soil PAEs surveys have focused primarily on eastern regions, urban, or suburban soils [38,41,50], with relatively few reports from central and western regions and rural areas. Regarding land use, studies have concentrated on facility vegetable fields [58,67], with less research on grain crop farmland. For compounds, research has been limited to one or a few PAEs compounds [33,64,66,68–78], which cannot represent the overall regional soil PAEs contamination status.

2) **Strengthen research on multi-media migration and transformation mechanisms of soil PAEs.** Studying PAEs environmental behavior across different media can effectively track PAEs metabolic processes. Currently, research on single environmental behaviors among multiple media is insufficient, mostly remaining at the descriptive level or speculative about mechanisms, lacking in-depth mechanistic investigations. The transformation mechanisms and joint toxic effects of PAEs with other organic pollutants in various environmental media remain unclear. Research should focus on PAEs residue dynamics in soils and their transport mechanisms and limiting factors throughout entire ecosystems, quantitatively estimating PAEs partitioning behavior and contribution rates in different environmental media.

3) **Strengthen research on soil PAEs pollution and human health risks.** Risk assessment is crucial for environmental risk management and ensuring environmental safety and human health. Currently, soil PAEs risk assessment objects remain limited to either ecological environments [32,78] or human health [11,23], rarely considering the combined environmental health risk effects. Furthermore, existing assessments mostly identify single pollution sources, while risks from multiple or historical pollution sources are unclear, potentially underestimating actual risks. Additionally, China has not yet established relevant standards for soil PAEs pollution assessment and remediation, with most studies referring to foreign standards [11,23], which may introduce bias in defining PAEs pollution levels.

4) **Strengthen research on soil PAEs reduction and remediation technologies.** Soil reduction and remediation technologies can effectively reduce or eliminate hazards to the environment and human health, representing fundamental measures for soil environmental protection. Compared with single phytoremediation or microbial remediation technologies, microbial-plant combined remediation can effectively improve PAEs remediation efficiency [25,28], but such studies remain limited.

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Note: Figure translations are in progress. See original paper for figures.