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Challenges and Future Prospects of Using Biochar for Soil Remediation

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ABSTRACT: Biochar gained significant attention as an eco-friendly and effective solution for remediating contaminated soils, particularly those impacted by pharmaceutical persistent pollutants (PPPs). These pollutants, known for their resistance to natural degradation and tendency to accumulate in soil, posed serious risks to both human health and ecosystems. To address this issue, researchers proposed the use of biochar as a remediation technology to remove PPPs through adsorption. As an efficient sorbent, biochar demonstrated the ability to immobilize pharmaceuticals in contaminated soils, thereby reducing their bioavailability and mobility, and ultimately mitigating their environmental impact. This review aimed to provide a comprehensive overview of the current understanding of PPPs contamination and the potential of biochar for remediation. It first summarized the occurrence of pharmaceutical pollutants in various countries and identified their primary sources. It then examined the environmental fate of these pollutants and outlined the key challenges associated with their management. The mechanisms by which biochar adsorbed pharmaceutical compounds were discussed in detail, followed by a case study that illustrated the effectiveness of this technology in practical applications. This review also evaluated the advantages and disadvantages of using biochar for remediation, along with the practical challenges encountered during its implementation. Future directions highlighted included developing methods for extracting toxic residues and enhancing the performance of biochar through chemical or structural modifications.

KEYWORDS: Pharmaceutical; biochar; remediation; soil contamination; pollution

1. Introduction

The pharmaceutical industry was a thriving sector that contributed significantly to the advancement of healthcare by addressing the medicinal treatment needs of humanity. Pharmaceutical products were defined as biologically active substances used in both human and veterinary medicine. They were primarily utilized for therapeutic and preventive purposes in the industry, albeit not exclusively; they were also widely employed as growth stimulants in food production and animal husbandry [1]. The production of pharmaceutical products aimed not only to promote health but also to provide protection against diseases for both humans and animals [2, 3]. By the time of reporting, several hundred types of pharmaceutical items were utilized by humans [4]. Some of the commonly found compounds in pharmaceuticals included caffeine, enrofloxacin, ibuprofen, salicylic acid, sulfamethoxazole, and tetracycline [2].

Moreover, pharmaceutical production and consumption steadily rose due to factors such as population growth, the discovery of new medications, and the development of new chemicals [5]. Within 15 years, worldwide sales by the U.S. pharmaceutical industry grew from US$390.2 billion to at least US$1105 billion [6]. Nevertheless, the waste generated from pharmaceutical manufacturing, along with ingredients from pharmaceuticals and personal care products (PPCPs), were identified as some of the most distinctive and diverse classes of emerging contaminants [7]. These toxic substances were continuously introduced into soil and water systems, leading to their bioaccumulation and persistence in the environment [6]. For example, antibiotics—among the most abundant pharmaceuticals—were detected not only in water bodies but also in air, soil, and bottom sediments. Their accumulation within the food chain ultimately posed significant health risks to both animals and humans [8].

On the other hand, biochar was a carbon-rich material produced through the pyrolysis of organic biomass—such as agricultural and forestry waste—under limited oxygen conditions [9–11]. Although other techniques such as hydrothermal carbonization (HTC), gasification, and torrefaction also converted biomass into char, the products generated through these methods typically did not meet the criteria outlined by the European Biochar Certificate (EBC) for biochar [12]. According to previous studies, biochar possessed a high pH value and cation exchange capacity, making it particularly effective in enhancing soil fertility [13, 14]. One of its emerging applications was in long-term carbon sequestration, due to the predominantly aromatic structure of its carbon content, which rendered biochar highly recalcitrant and resistant to environmental decomposition [15]. Furthermore, biochar's high adsorption capacity, versatility, and sustainability positioned it as an attractive option for adsorbing pollutants in soils [2, 16]. Consequently, there was growing interest in applying biochar for pollutant removal from wastewater and for remediating polluted soils [17]. The aim of this review was to assess and discuss the potential of biochar as a sustainable remediation strategy for soils contaminated by PPPs.

2. Overview of the Pollutant

An overview of PPPs was presented in this section to provide insights into their status, occurrence, and fate in the environment. Some of the challenges encountered in addressing issues caused by these pollutants were also discussed. When PPPs were not metabolized prior to discharge, they posed significant adverse effects to both human health and the environment upon entering the ecological system. Figure 1 illustrated the pathway by which pharmaceutical wastes were transferred within the aquatic environment.

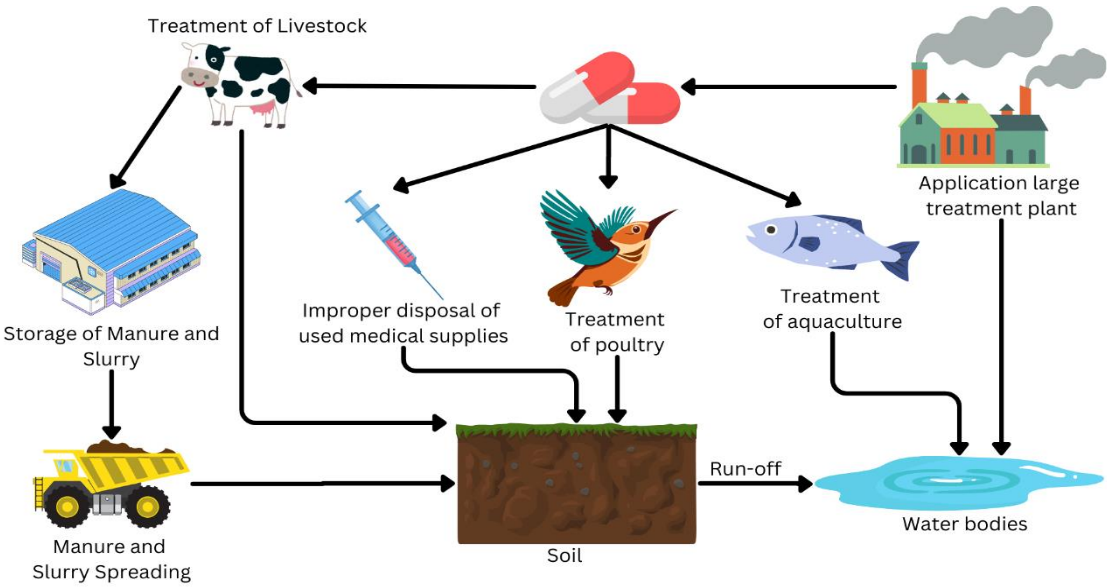

Figure 1. Sources and transfer pathways of pharmaceutical wastes in aquatic environment. Icon from Flaticon Basic License CC3.0 (Creative Commons)].

There were several adverse effects when PPPs entered the environment. When these contaminants accumulated in soil, they could eventually contaminate groundwater as they were carried by seepage into deeper soil layers [18]. Additionally, PPPs could be transported via runoff into surface water bodies. Due to their low degradability, PPPs tended to persist in the environment, thereby degrading water quality and threatening aquatic life [8]. In fact, a study by Brausch et al. showed that certain pharmaceuticals, such as thioridazine, dextropropoxyphene, and diphenhydramine, acted as acute pollutants for fish, invertebrate, and algae populations [19]. According to Pérez-Pereira et al., the European Medicines Agency (EMA) established a regulatory framework for environmental risk assessment (ERA) of pharmaceuticals, while the United Nations Educational, Scientific, and Cultural Organization (UNESCO) listed PPPs as emerging contaminants [20]. In recent years, extensive investigations into the occurrence and dispersion of PPPs in soil–plant systems and agricultural contexts were conducted, resulting in the detection of pharmaceutical contaminants in multiple countries [21]. For example, in urban biosolids intended for agricultural use in Ontario, Canada, over 80 types of PPPs and PPCPs were identified [8]. Table 1 presents some of the PPPs detected in various countries, along with their concentrations.

Trimethoprim was one of the pharmaceuticals found in the highest concentrations among various compounds, with levels exceeding 60 μg/kg in both Malaysia and the USA. In contrast, carbamazepine appeared to have the lowest concentrations detected in these countries, ranging from 0.02 to 0.06 μg/kg in Hebei, China, and was not detected at all in the USA. However, notably higher concentrations of carbamazepine, ranging from 2.6 to 7.5 μg/kg, were reported in Mexico. Other pharmaceuticals with significantly high concentrations were observed in Denmark, where oxytetracycline levels ranged from 33 to 2000 mg/g in pig waste and from 2.5 to 50 μg/g in soil [27]. In terms of the total mass of pharmaceutical pollutants, it was reported that soils in the Slovak Republic accumulated several hundred kilograms of medications annually. Nevertheless, the quantities remained within the maximum annual mass estimates for specific compounds such as verapamil (29 kg/year) and fexofenadine (120 kg/year) [28].

Table 1. Concentration of PPPs detected in soil across different countries.

Country/region	Number of samples	Types of pharmaceutical	Range of Concentration (μg/kg)	References
Hebei, China	18	Carbamazepine	0.02 – 0.06	[22]
		Diclofenac	0.35 – 1.16	
		Ibuprofen	1.51 – 5.03	
		Sulfadiazine	1.15 – 3.82	
		Trimethoprim	0.64 – 2.15	
Malaysia	10	Sulfadiazine	Not detectable	[23]
		Trimethoprim	3.1 – 60.1	
Mexico	4	Carbamazepine	2.6 – 7.5	[24]
		Diclofenac	Not detectable	
		Ibuprofen	Not detectable – 0.1	
		Triclosan	Not detectable – 16.7	
USA (a)	3	Carbamazepine	Not detectable	[25]
		Trimethoprim	Not detectable – 0.64	
USA (b)	1	Carbamazepine	0.7 – 1.4	[26]
		Triclosan	Not detectable	

2.1.Point source and non-point source.

The sources of PPPs were generally categorized into point source pollution and diffuse pollution. According to Lapworth et al., point source pollution referred to a single, identifiable origin that could be quantified using mathematical modelling. Primary sources of PPPs in the soil zone and water resources included sewage treatment plants, hospital effluents, and discharges from the pharmaceutical industry [30]. In contrast, diffuse pollution occurred across broad geographical areas and was difficult to trace to a specific location [29]. Examples of diffuse pollution included urban runoff from household waste, agricultural runoff from animal waste and manure, and leakage from facilities and waste treatment systems [31]. Notably, non-point sources had a greater potential for natural attenuation in the soil and subsurface, often resulting in less severe environmental impacts compared to point sources [32].

2.2. Fate of pharmaceutical persistent pollutants.

Most research on the fate and transport of pharmaceutical waste in the environment focused on understanding their behavior across various environmental matrices such as sludge, agricultural soils, aquatic ecosystems, and wastewater treatment systems [33]. Generally, pharmaceuticals were designed to be non-bioaccumulative and were intended to be rapidly metabolized and excreted from the bodies of humans and animals after consumption [34]. However, in many cases, pharmaceutical compounds were excreted unmetabolized and released into the environment without consideration of their potential negative impacts [6]. As a result, the pharmaceutical industry was identified as a major source of PPPs due to discharges during production processes [1]. Additionally, sewage sludge, biosolids, and animal feces containing antibiotics were found to be other significant contributors [35].

Numerous studies showed that, except for facilities employing tertiary treatment, conventional wastewater treatment plants (WWTPs) that met legal standards were only moderately effective in removing PPPs [36, 37]. Consequently, more than 25 types of pharmaceutical waste were detected in both effluent and sludge from WWTPs [30]. WWTPs were thus identified as major contributors to environmental pharmaceutical contamination [38, 39]. For instance, pharmaceuticals such as norfloxacin, ciprofloxacin, and other fluoroquinolones were commonly found in sewage sludge [40]. In Turkey, treated sludge was reported to contain approximately 1.002 kg of pharmaceuticals daily, amounting to an annual release of up to 71.6 kg into the environment [41].

These pollutants contributed to widespread environmental contamination. PPPs in agricultural soils could remain in the upper soil layers or migrate downward, eventually reaching groundwater aquifers. Upon entering the soil, these compounds interacted with microbial communities, potentially disrupting microbial activity and soil health. Moreover, PPPs could undergo partial biodegradation or be absorbed by plants and vegetables, raising additional concerns about their entry into the food chain and their effects on both ecosystems and human health [8].

2.3. Major challenges.

Several consequences arose from the negligent and indiscriminate disposal of pharmaceutical products, leading to environmental contamination [42]. The presence of PPPs in soil had, in fact, emerged as a significant environmental concern. Some of the anthropogenic wastes generated by the pharmaceutical industry included birth control pills, antibiotics, tetracycline, painkillers, and more posing a global issue that required urgent attention and solutions [2]. Moreover, PPPs were recognized as some of the most prominent contaminants in recent years, alongside microplastics [43]. Due to their high hydrophilicity and chemical stability in water [44, 45], PPPs tended to accumulate over time in aquatic ecosystems [46–49]. Their persistence in water bodies led to their classification as potential toxicological pollutants, posing serious threats to both aquatic ecosystems and human health. Therefore, preventing these pollutants from entering the environment became increasingly important. At an industrial scale, traditional chemical treatment methods—such as coagulation, sedimentation, photocatalytic treatment, phototransformation, and advanced oxidation techniques like ozonation—were found to be ineffective due to limitations in ease of use, cost-efficiency, the need for secondary treatments, and the potential release of hazardous by-products [2]. Additionally, the use of conventional wastewater treatment plants to remove PPPs and PPCPs was also shown to be insufficient [50, 51]. As a result, studies reported the continued presence of these contaminants in both biosolids and effluents from wastewater treatment plants, indicating that existing treatment processes were inadequate for the complete removal of PPPs and PPCPs from waste streams [34].

3. Remediation Technology

Several remediation methods were employed to address persistent organic contaminants, including the use of plants, compost, microorganisms, biochar, and organic manures [52, 53]. Nevertheless, over the past 20 years, biochar had gained increasing attention due to its versatile applications, particularly its potential to offer a practical and cost-effective method for decomposing pollutants and reducing the risk of soil contamination [42]. The mechanism by which biochar remediated PPPs in soil primarily involved adsorption, as biochar possessed a large, negatively charged surface area [17]. The adsorption of organic substances, including PPPs, onto biochar was significantly influenced by electrostatic interactions [54, 55]. Ionic attraction and repulsion, largely attributed to non-pyrolyzed organic matter within the biochar, played a key role in bonding charged pharmaceutical molecules to its surface. Biochar was capable of adsorbing both hydrophobic and hydrophilic organic molecules due to the presence of electronic interactions [56]. However, the sorption process involved more than just electrostatic forces. The carbonized organic matter on biochar's surface facilitated π–π interactions with pharmaceuticals in both neutral and ionized forms. The high adsorption capacity for pharmaceuticals such as tetracycline was further enhanced by additional mechanisms, including cation–π bonding, π–π electron-donor-acceptor interactions, and van der Waals forces. These mechanisms made the sorption process highly effective by allowing PPPs to bind to the graphene-like structures found in biochar [57].

3.1. Case study: removal of tetracycline using biochar.

In a study conducted by Monisha et al., the treatment of tetracycline, sulfa compounds, quinolone compounds, and anti-inflammatory drugs was investigated through adsorption and degradation using biochar [2]. Table 2 presents the results of using biochar to treat tetracyclines under different preparation methods and conditions.

Table 2. Treatment of tetracyclines in aqueous solutions using class 1, 2, and 3 biochar.

Class	Feedstock	Activation	Contaminant	Preparation and Condition	Efficiency/Uptake	References
1	Camellia oleifera shell	Phosphate	Tetracycline	Pyrolysis; 600 °C, 1 h	99.50%	[59, 60]
	Eucommia ulmoides	H_2SO_3	Tetracycline, Hydrochloride	Pyrolysis; 700 °C, 1 h	1163 mg/g	[61]
	Maize straw	Not available	Oxytetracycline	Pyrolysis; 350 °C, 1.5 h	63%	[62]
	Maple leaf	Not available	Tetracycline	Pyrolysis; 750 °C, 2 h	4017.3 mg/g	[63]
	Pinus taeda	NaOH	Tetracycline	Pyrolysis; 300 °C, 15 min	274.8 mg/g	[64]
	Rice straw	Hydrogen Peroxide	Tetracycline	Pyrolysis; 500 °C, 2 h	97%	[65]
	Rice straw	H_3PO_4	Tetracycline	Pyrolysis; 700 °C, limited O_2 supply	552 mg/g	[66]
	Sawdust	Not available	Tetracycline, Cu(II)	Pyrolysis; 600 °C, 2 h	94.1%	[67]
	Soybean residue	KOH, Ball Milling, HCl	Tetracycline	Pyrolysis; 800 °C, 2 h	84.15%	[68]
2	Chicken bone feather	$KMnO_4$	Tetracycline, Rhodamine B	Pyrolysis; 500 °C, 2 h	93.20%	[69]
	Chicken feather	Multilayered graphene phase	Tetracycline	Pyrolysis; 450 °C, 1 h	99.72%	[70, 71]
	Crayfish shell	Ball milled biochar	Tetracycline Hydrochloride	Pyrolysis; 800 °C, 2 h	60.7 mg/g	[72, 73]
	Spirulina species	Not available	Tetracycline	Pyrolysis; 750 °C	132.8 mg/g	[74]
	Swine manure	H_3PO_4	Tetracycline	Pyrolysis; 700 °C, limited O_2 supply	365.4 mg/g	[66]
3	Pharmaceutical sludge	Sodium hydroxide	Tetracycline	Pyrolysis; 600 °C, 2 h	379.78 mg/g	[59]
	Municipal solid waste	Clay composite	Tetracycline	Slow pyrolysis; 500 °C, 30 min	26 mg/g	[75]

In the study, three classes of biochar were proposed based on the type of feedstock used for production. Class 1 biochar was derived from feedstocks such as crops, trees, and plants, making it the most extensively used category [2]. Class 2 biochar was produced from sources such as animals, microorganisms, and marine organisms, while Class 3 biochar was manufactured from domestic and industrial discharges. One of the most widely used antibiotics for human treatment and as an animal feed additive was the tetracycline group, which was known for its environmental persistence, with half-lives ranging from 30 to 180 days, and its strong adsorption affinity for manure in soils [58]. In fact, the global usage of tetracyclines was ranked second among antibiotics [2]. A total of 4017.3 mg of tetracycline was removed per gram of biochar produced from maple leaves when pyrolyzed at 750 °C for 2 hours. The highest removal rate was observed when the biochar was produced from chicken feathers using pyrolysis at 450 °C for 1 hour, activated with a multilayered graphene phase, resulting in a 99.72% removal of tetracycline. In contrast, biochar produced from maize straw under pyrolysis at 350 °C for 1.5 hours achieved only a 63% removal of oxytetracycline.

3.2. Pros and cons using biochar as a remediation technology.

There are several advantages and disadvantages to utilizing biochar as a remediation technology for removing PPPs from contaminated soil. One of the main benefits of using biochar is its ability to enhance soil fertility, which promotes plant growth by efficiently addressing carbon sequestration and nutrient delivery [12, 76, 77]. This is because biochar contains a large number of inorganic components, which act as soil fertilizers to support plant growth [78]. Additionally, since biochar is produced from organic wastes, such as forestry residues, municipal wastes, and agricultural by-products [12], it has the added advantage of being an environmentally friendly and cost-effective bio-based adsorbent [2]. Biochar's potential for reuse as both an adsorbent and catalyst is one of its key advantages. Furthermore, biochar has been reported to be highly effective in removing both organic and inorganic pollutants due to its stable structure, high cation/anion exchange capacity, large surface area, and rich carbon content [79]. It has also been found to decrease the bioavailability of organic contaminants in soil, reducing the uptake of these pollutants by plants and microbes [80, 81]. Additionally, biochar application has been shown to reduce the toxicity and depletion of insecticides and pesticides, further remediating PPPs in soil [82]. This suggests that biochar may help limit the bioavailability of harmful substances, reducing their negative effects. There is also a wide variety of biochar types that can be used for contaminant adsorption. For instance, biochar produced from Camellia oleifera shells with acid treatment has a sorption capacity nearly 17 times greater than that of raw biochar and is adaptable to a wider pH range (1–9) [59, 60]. Ultimately, the main benefits of biochar over other sorbents are its abundance of feedstocks, cost-effectiveness, reusability, unique functional groups, and environmentally friendly, relatively simple production processes [2].

However, biochar also has some potential disadvantages, as it contains naturally occurring hazardous chemicals that may have adverse environmental effects [2]. Metalloids, intermediate organic molecules, heavy metals, and polycyclic aromatic hydrocarbons can be present in biochar that has been treated with pharmaceuticals, such as tetracyclines, sulfonamides, quinolones, and anti-inflammatory drugs [2]. Furthermore, biochar can slow down the aging process of soil, and for optimal nutrient cycling and soil-water balance, fresh biomass may need to be periodically added [83]. Consequently, aged biochar in soil can have negative effects, such as inhibiting the growth of earthworms [84]. It has also been shown that aged biochar reduces the biomass of subterranean roots in *Solanum lycopersicum* (tomato) and *Oryza sativa* (rice), which may impact plant growth. Additionally, because biochar has low thermal diffusivity by nature, it has been shown to reduce soil thermal diffusivity [85]. Moreover, the use of biochar can decrease soil nutrient absorption and carbon mineralization [86]. The presence of toxins and highly volatile chemicals in biochar has also been linked to a decrease in crop productivity [87], as biochar made from pyrolyzing forest waste can contain toxic contaminants [42]. Another significant finding by Zhu et al. is that the effectiveness of biochar in soil remediation is highly dependent on the type of soil, suggesting that biochar may not have a universally positive effect on all soil types [88].

4. Challenges and Prospects

Despite the advantages biochar application offers for remediating PPPs in soil, there are several limitations and challenges that need to be addressed for this technology to be widely adopted. A lack of fundamental information in this area and uncertainty surrounding the safety of biochar use are two major obstacles [17]. The presence of toxins in spent biochar poses a significant challenge, as the concentration of these toxins can vary depending on several factors, including the types of feedstocks used, pyrolysis temperature, activation agents, and the composition of adsorbed pollutants. It is crucial to thoroughly evaluate the levels of toxins or hazardous compounds in spent biochar before large-scale commercial or industrial use, even if these levels are typically low and within allowable limits [2]. Therefore, additional studies are needed to address the challenge of removing these toxic chemicals from biochar, such as improving biochar pyrolysis and reaction conditions to mitigate environmental issues [42].

Moreover, before biochar can be practically applied, future research should thoroughly investigate changes in pH levels and the production of secondary byproducts [89]. Since there are different types of biochar, those produced from various biomass sources may require different dosages. Accordingly, Kang et al. have recommended that a suitable biochar application strategy be developed based on the properties of soil, compost, and organic waste [89]. Furthermore, Zhang et al. have suggested conducting large-scale field trials to assess biochar's effectiveness in removing PPPs, as only laboratory, greenhouse, and small plot trials have been conducted thus far. As mentioned previously, the aging of biochar in soil can pose several risks. Therefore, Zhang et al. emphasized the need for further studies to better understand the aging process and improve remediation efficiency [16].

In addition, several studies have highlighted the benefits of using composite biochar to overcome its limitations [90]. For instance, rice-based biochar made from solid digestate was used to create three distinct composite forms—Copper (Cu), Iron (Fe), and Cu-Fe composites—after nitrogen purging. The Cu-modified biochar demonstrated a removal effectiveness of 97% for PPPs [2]. Further research is also required to assess the risks associated with biochar, which has been found to potentially accelerate the dissipation of certain organic contaminants in soil [16]. Finally, it is suggested to conduct thorough research on the implementation of biochar in wastewater treatment plants. Biochar could serve as a pretreatment method for removing toxic compounds, allowing it to adsorb harmful contaminants before the wastewater undergoes biological treatment [82]. Challenge and prospect of biochar application in PPPs remediation is summarized in Figure 2.

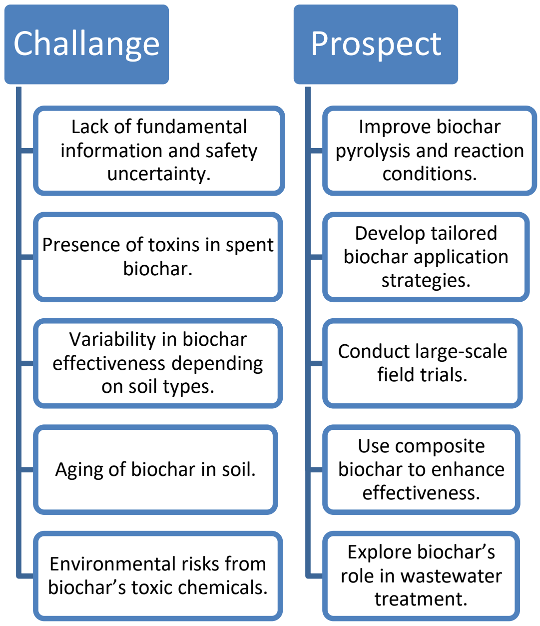

Figure 2. Challenge and prospect of biochar application in PPPs remediation.

5. Conclusions

This paper has provided a comprehensive overview of the impacts caused by the contamination of PPPs in soil, as well as explored the use of biochar as a remediation technology. PPPs have raised significant environmental concerns due to their widespread occurrence and persistence in various environmental matrices, which in turn pose risks to human health and the environment. The global extent of the problem is highlighted through the occurrence of these pollutants in different countries, including China, Mexico, and the USA. Biochar has emerged as a promising method for remediating soils contaminated with PPPs. It is often favored over other conventional sorbents due to its large surface area and its ability to adsorb a wide range of contaminants. However, the risk of releasing toxins from spent biochar remains a challenge, alongside its variable effectiveness on different soil types and its potential to accelerate the dissipation of certain organic contaminants in soil. Despite these challenges, biochar still holds significant potential as a sustainable and cost-effective tool for remediating soil contaminated by PPPs. Further research is needed to address concerns regarding its environmental safety and its potential for commercial and industrial-scale applications. Additionally, more studies are required to investigate the possible negative effects associated with the implementation of this technology.

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Author Contribution

Audrey Primus, Alexandru Marculescu: drafting of the manuscript, writing and revision, conceptualization, data analysis; Linh Thi Thuy Cao, Gina Nadifah, Daniel Twum-Ampofo:

methodology and interpretation of the results; Md Abu Hanifa Jannat, Jovale Vincent Tongco: research design, data collection, interpretation, writing and revision.

Competing Interest

The authors declare no competing interests.

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