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The Impact of Different Fertilization Methods on Soil Heavy Metal Pollution and Restoration Methods

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Abstract. Soil heavy metal contamination, exacerbated by the long-term use of phosphate and organic fertilizers, poses a significant threat to agriculture and global ecosystems. Accumulation of heavy metals such as arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) in soils can endanger human health and ecosystem services. This paper evaluates the content of heavy metals in various fertilizers and reviews existing remediation technologies. Among these, plant-microbe collaborative remediation shows promise for mitigating heavy metal pollution, although its effectiveness in multi-pollutant environments remains underexplored. Current research reveals a gap between laboratory results and practical applications, with limited studies on long-term ecological impacts. Future research should prioritize addressing complex pollution scenarios and developing efficient, eco-friendly remediation strategies to promote sustainable agriculture and environmental protection.

Keywords: Heavy metal, Soil, Fertilizer, Remediation.

1. Introduction

Soil contamination by heavy metals has become a significant global environmental concern, primarily due to the widespread use of fertilizers in contemporary agriculture. Although fertilizers are essential for boosting crop production, their prolonged use, particularly of phosphate and organic fertilizers, has resulted in the buildup of toxic heavy metals such as arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) in the soil. This buildup not only deteriorates soil health but also threatens human health and ecosystems, as these metals can be absorbed by crops and enter the food chain.

China, as a leading agricultural nation, faces severe challenges related to soil heavy metal contamination. Overuse of fertilizers has been identified as a key contributor to heavy metal accumulation in soils, which in turn threatens food security and undermines ecological stability. Understanding the sources of soil heavy metal contamination and developing effective remediation strategies are therefore critical for ensuring sustainable agricultural practices and safeguarding ecological health.

This study examines the content of heavy metals in different fertilizers and evaluates existing remediation technologies. Additionally, it highlights the urgent need for research into the remediation of composite pollution and the development of sustainable solutions to mitigate the long-term effects of soil contamination.

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2. Analysis of Heavy Metal Content in Fertilizers and Their Contribution to Soil Contamination

Heavy metal contamination in fertilizers, particularly phosphorus fertilizers, has been identified as a significant source of soil pollution. According to the national standard GB 38400-2019 [1], the permissible limits for heavy metals such as arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) in inorganic fertilizers are set to ensure soil safety. However, research by Duan Lulu [2] reveals that among 400 fertilizer samples tested, 11 samples exceeded these limits, with phosphorus fertilizers showing the highest exceedance rate of 26.92% (Table 1)..

The overall analysis of heavy metal content in fertilizers (Table 2) shows that the average values of six heavy metals—As, Cd, Pb, Cr, Hg, and Tl—are all less than 100 mg/kg. Cd has the lowest average value at 0.04 mg/kg, while Cr has the highest at 67.77 mg/kg. The content ranges for these metals vary significantly: As (0.1–72 mg/kg), Cd (0.04–110 mg/kg), Pb (0.29–323 mg/kg), Cr (0.07–212 mg/kg), Hg (0.01–53 mg/kg), and Tl (0.024–88 mg/kg). Such wide ranges highlight the potential for substantial variability in contamination levels across different fertilizer types. Phosphorus fertilizers exhibit the highest concentrations of heavy metals, with significant exceedance rates for As, Cd, Pb, and Hg, compared to the national safety limits set by GB 38400-2019. Compound fertilizers also show elevated Tl levels. In contrast, nitrogen fertilizers generally have the lowest average content of these metals.

Table 1. The heavy metals exceeding standard rate of different fertilizers

Fertilizer variety	Sample size	Over standard rate(%)						Total (%)
		As	Cd	Pb	Cr	Hg	Tl	
Nitrogenous fertilizer	35	0	0	0	0	0	0	0
Phosphate fertilizer	26	4	4	4	0	27	0	26.92
Potassium fertilizer	52	0	0	0	0	0	0	0
Compound fertilizer	203	1	0	0	0	0	1	0.97
Blended fertilizer	18	0	0	0	0	0	0	0
Other fertilizers	66	0	0	0	0	0	2	1.52
Total	400	0.75	0.25	0.25	0	1.75	1.00	2.75

Table 2. Heavy metal contents in different fertilizers

Element	Fertilizer type	Range	Average	Medium value	SD	CV
As		0.1~2	0.36	0.30	0.31	0.88
	Nitrogenous fertilizer	0.3~62	19.01	15.50	15.91	0.84
	Phosphate fertilizer	0.22~4.28	0.63	0.30	0.82	1.29
	Potassium fertilizer					
	Compound fertilizer	0.3~72	11.34	9.00	9.85	0.87
	Blended fertilizer					
	Other fertilizers	0.29~26	8.64	5.59	8.18	0.95
		0.3~37	3.20	0.74	6.00	1.87

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Table 2. (continued)

		0.04~0.06	0.04	0.04	0.004	0.09
	Nitrogenous fertilizer	0.04~110	5.49	0.35	21.40	3.90
	Phosphate fertilizer	0.04~0.35	0.05	0.04	0.04	0.91
Cd	**Potassium fertilizer**					
	Compound fertilizer	0.04~7	0.31	0.04	0.69	2.23
	Blended fertilizer					
	Other fertilizers	0.04~1	0.30	0.04	0.23	1.74
		0.0028~1	0.12	0.05	0.15	1.32
		0.4~4	0.50	0.40	0.61	1.21
	Nitrogenous fertilizer	0.4~323	50.58	15.50	70.57	1.40
	Phosphate fertilizer	0.4~7.13	0.79	0.40	1.08	1.36
Pb	**Potassium fertilizer**					
	Compound fertilizer	0.29~195	5.40	0.40	16.77	3.11
	Blended fertilizer					
	Other fertilizers	0.37~33	4.11	0.40	9.14	2.22
		0.27~67	3.49	0.40	10.99	3.15
		0.07~8	0.55	0.07	1.56	2.81
	Nitrogenous fertilizer	0.07~212	67.77	51.00	59.68	0.88
	Phosphate fertilizer	0.4~48.69	8.15	5.35	8.97	1.10
Cr	**Potassium fertilizer**					
	Compound fertilizer	0.07~46	18.97	14.00	17.16	0.90
	Blended fertilizer					
	Other fertilizers	3~24	12.34	10.28	6.18	0.50
		0.07~68	7.40	2.00	12.28	1.66
		0.01~0.08	0.07	0.08	0.03	0.41
	Nitrogenous fertilizer	0.08~53	7.66	1.50	13.26	1.73
	Phosphate fertilizer	0.01~0.1	0.06	0.08	0.03	0.49
Hg	**Potassium fertilizer**					
	Compound fertilizer	0.01~3	0.32	0.08	0.43	6.19
	Blended fertilizer					
	Other fertilizers	0.02~1	0.18	0.08	0.22	1.26
		0.028~0.32	0.08	0.08	0.03	0.37

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Table 2. (continued).

		0.03~0.8	0.08	0.03	0.18	2.18
	Nitrogenous fertilizer	0.03~1.6	0.30	0.03	0.48	1.59
	Phosphate fertilizer	0.03~1.2	0.13	0.03	0.29	2.24
Tl	**Potassium fertilizer**					
	Compound fertilizer	0.03~88	0.82	0.90	6.19	7.58
	Blended fertilizer					
	Other fertilizers	0.03~1.41	0.27	0.12	0.35	1.31
		0.024~13.7	0.33	0.03	1.69	5.15

3. Experimental Study of Soil Contamination Induced by Fertilizer Use

3.1. Experiment and result analysis

According to the research by Xia Wenjian et al. [3], Since 1984, the soil has been monitored for 35 years, and the soil is the middle-yellow mud field developed in the quaternary acidic red soil. The experimental planting pattern was a rotation of early rice-late rice-winter fallow, with a plot area of 33.3 m², separated by cement dikes, and independent irrigation and drainage. During both the early and late rice growing seasons, nitrogen, phosphorus, and potassium fertilizers were used, sourced from urea, calcium superphosphate, and potassium chloride. Organic fertilizers consisted of vetch for early rice and pig manure for late rice. The pig manure had a moisture content of 85% and nutrient content of N 0.45%, P2O5 0.19%, and K2O 0.60%; vetch had a moisture content of 88% and nutrient content of N 0.30%, P2O5 0.08%, and K2O 0.23%. Table 4 to 8 are parameters related to this experiment.

Table 3. Heavy metal content in local soil

Element	**Cu**	**Zn**	**Cr**	**Pb**	**Cd**	**As**	**Hg**	**Fe**	**Mn**
Content mg·kg^{-1}	47.0	115	72.0	43.0	0.171	11.0	0.30	29.9x10^3	475

Table 4. Total fertilizer consumption per year under different test treatments

Treatment method **Dosage**	**Pig manure /t·hm^{-2} (Organic fertilizer, late rice)**	**Chinese purple cloud root/ t·hm^{-2} (Organic fertilizer, early rice)**	**Superphosphate /kg·hm^{-2} (Phosphate fertilizer, containing P_2O_5 12%)**	**Urea /kg·hm^{-2}(Nitrogenous fertilizer)**	**Potassium chloride /kg·hm^{-2}(Potassium fertilizer, containingK_2O 60%)**
Not fertilize(CK)	0	0	0	0	0
Application of phosphorus and potassium fertilizers(PK)	0	0	1000	0	500
Application of nitrogen, phosphorus and potassium fertilizers(NPK)	0	0	1000	717	500
70%Chemical fertilizer + 30%organic fertilizer (M1NPK)	12.0	14.8	710	512	324

DOI: 10.54254/2753-8818/75/2024.LA19383

Table 4. (continued).

50%Chemical fertilizer+ 50%organic fertilizer (M2NPK)	20.0	24.8	520	358	206
30%Chemical fertilizer + 70%organic fertilizer (M3NPK)	28.0	34.6	337	218	119

Table 5. Content of heavy metals in different fertilizers

Content($mg \cdot kg^{-1}$)	Cu	Zn	Cr	Pb	Cd	As	Hg	Fe	Mn
Pig manure	110	280	2.35	1.16	0.27	0.48	0.09	391	93.2
Chinese purple cloud root	1.3	3.8	0.26	0.12	0.04	0.03	0.001	16.2	21.4
Urea	0.13	2.31	—	—	—	—	—	10.2	0.21
Potassium chloride	1.31	5.8	—	—	—	—	—	106	8.42
Superphosphate	5.80	22.8	1.87	0.36	0.33	0.01	0.01	431	23. 2

The detailed proportions of nitrogen, phosphate, and potassium fertilizers in different growth stages can be simplified, focusing only on the main types of fertilizers used (phosphate, nitrogen, potassium, and organic fertilizers) and their role as base fertilizers or topdressing. The practices of sowing, transplanting, irrigation, and pesticide application in all plots followed local management traditions. In red soil paddy fields, the total heavy metal content in soils treated with PK and NPK fertilizers showed minimal change compared to the control (CK). However, the bioavailable forms of Cr and As increased significantly, while other heavy metals displayed a slight upward trend in bioavailability. The proportion of bioavailable heavy metals to the total heavy metal content in soils treated with chemical fertilizers changed slightly. The PK treatment significantly increased the proportion of bioavailable Cu, Cr, As, and Fe, while the NPK treatment significantly increased the proportion of bioavailable Cr, As, and Hg. Generally, the long-term application of chemical fertilizers has little impact on the total heavy metal content in the soil[4], but there is still an accumulation risk. Ren Shunrong et al. [5] observed that after 25 years of continuous fertilizer use, levels of heavy metals such as Cu, Zn, Cd, Cr, Hg, and As in the soil exhibited a rising trend. While nitrogen fertilizers had minimal impact on heavy metal accumulation, phosphate fertilizers contributed to a significant increase in their content.

3.2. The availability of soil heavy metals and their correlation with soil chemical properties and total heavy metal content

Bioavailable forms of Cu, Zn, Cr, Cd, As, and Fe show positive correlations with soil organic matter (SOM), cation exchange capacity (CEC), available phosphorus (AP), and nitrate nitrogen (NO3--N). Bioavailable Zn and Cd positively correlate with soil pH, whereas bioavailable Pb negatively correlates with soil pH, SOM, CEC, and NO_3^--N content. The correlations between bioavailable Hg and Mn and soil chemical indicators are not significant, and there is no significant correlation between available potassium (AK) and ammonium nitrogen (NH_4^+-N) with the bioavailable forms of heavy metals.

Bioavailable Cu, Zn, and Cd are positively correlated with their total content, while Cr and Fe exhibit negative correlations. Other heavy metals show no significant correlation with their total content. Bioavailable Cu, Zn, Cd, As, and Fe correlate positively with total Cu, Zn, Cd, and Hg, while bioavailable Pb correlates negatively with total Cu, Zn, Cd, and As. Total Cr and Fe both show negative correlations with bioavailable As, Hg, and Fe.

This indicates that long-term fertilization affects the relationship between the bioavailable and total contents of different heavy metals in soil, and there are complex correlations between the bioavailable

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forms of different heavy metals and other elements. The underlying reasons for these correlations require further investigation.

3.3. Soil Heavy Metal Pollution from Organic Fertilizer Application

In red soil paddy fields, organic fertilizer treatments (M1NPK, M2NPK, and M3NPK) increased the soil content of Cu, Zn, and Cd, while As, Hg, Mn, and Pb remained unchanged, and Cr and Fe decreased compared to the control (CK).. According to the Chinese Farmland Soil Standard GB15618-2018, the soil copper content under the organic fertilizer treatments (M1NPK, M2NPK, and M3NPK, ranging from 56.5 to 82.0 $mg \cdot kg^{-1}$) slightly exceeded the standard limit, and the cadmium content under the high organic fertilizer treatment M3NPK (0.486 $mg \cdot kg^{-1}$) also slightly exceeded the limit. Other elements did not exceed the standard limits.

The organic fertilizer treatments M1NPK, M2NPK, and M3NPK notably elevated the bioavailable levels of Cu, Zn, Cr, Cd, As, and Fe in the soil. These treatments also significantly boosted the proportion of bioavailable Cu, Zn, Cr, As, and Fe. While the M3NPK treatment notably reduced bioavailable Pb, the M1NPK treatment significantly raised the proportion of bioavailable Hg. Over time, fertilization, especially with organic treatments, altered soil chemical properties by significantly increasing soil SOM, CEC, AP, AK, and NO_3-N.

According to the Soil Environmental Quality Standard (GB 15618-2018), the safe limit for copper in soil is 50 mg/kg, but under the high organic fertilizer treatment, the soil copper content reached 82.0 mg/kg, exceeding the safety threshold. This poses a risk of heavy metal contamination in crops grown in such soils. Long-term use of organic fertilizers increased both total and bioavailable Cu, Zn, and Cd, while also raising bioavailable Cr, As, and Fe. Studies confirm that frequent use of animal manure or sludge elevates soil Zn and Cu levels [6-13].

4. Effect of pollution

Research indicates that superphosphate application increases Cd accumulation in crops like lettuce, peas, and radishes [14], and that nitrogen, phosphorus, and potassium fertilizers enhance Cd uptake in carrots and oats on loam soils with pH values between 4.8 and 6.8 [15,16].

Cadmium damages plant cells, inhibits photosynthesis, and suppresses antioxidant capacity. Its accumulation in tissues displaces essential nutrients such as iron (Fe) and zinc (Zn), leading to deficiencies and reduced crop quality. Cadmium also stimulates soil microbial cells, cause mutagenic effects. It inhibits protein and nucleic acid synthesis by breaking DNA strands and binding to proteins with carboxyl and thiol groups, thus imparting toxicity to the microorganisms.

Studies suggest that organic fertilizers increase heavy metal content in crops. For example, Huang Yaxi et al. reported higher Cd levels in lettuce after sludge composting [17], while Wang Jin et al. observed increased Zn content in celery treated with pig manure, exceeding safety limits [18].

5. Remediation technology

5.1. Engineering remediation technology

Engineering remediation technologies use physical methods to address soil contamination, including surface soil removal, soil replacement, topsoil covering, and deep plowing. These methods prevent plant roots from contacting heavy metals, allowing healthy plant growth. Deep plowing is suitable for mild contamination, while soil replacement or covering is preferred for severe pollution. Studies have demonstrated that techniques like deep plowing can reduce Cd contamination [19], while topsoil covering is effective in diluting high levels of Cd and Cu [20]. However, these methods are costly, disrupt soil structure, and mainly relocate pollutants, without addressing the root cause of contamination.

5.2. Physical remediation technology

Physical remediation immobilizes heavy metals in soil to prevent their spread, using methods like microwave heating, pyrolysis, and steam extraction. While these techniques reduce heavy metal impacts,

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they require high financial investment and do not fully address pollution sources. For example, pyrolysis can remove up to 90% of Zn and Pb but has limitations like high energy consumption and untreated by-products [21].

5.3. Chemical remediation technology

Chemical remediation uses soil amendments to stabilize heavy metals, transforming them into less harmful compounds. Leaching is a common method, where solutions like acids and chelating agents extract heavy metals for recovery. Although effective, issues like high cost, soil fertility loss, and leachate disposal remain challenges [22-23].

5.4. Bioremediation technology

5.4.1. Phytoremediation technology

Phytoremediation relies on plants' ability to absorb heavy metals through their roots and accumulate them in their tissues. This technology is cost-effective and easy to maintain, offering economic and environmental benefits. However, it has limitations, such as slow remediation rates and susceptibility to factors like soil type and climate conditions.

5.4.2. Microbial remediation technology

Microbial remediation uses microorganisms such as bacteria, fungi, and actinomycetes to restore soil by degrading or stabilizing heavy metals. While effective, this technology is limited by high costs, which restrict its broader application in soil ecosystem restoration [24].

5.4.3. Plant - microbial combined repair technology

The combination of plants and microorganisms enhances soil remediation, with microorganisms promoting plant growth and stabilizing contaminants. For example, adding Fe-platanus carbon increased microbial diversity and improved heavy metal adsorption efficiency [25]. This approach is both cost-effective and environmentally friendly but requires specialized knowledge and a longer remediation cycle. Studies have shown that the combination of Kentucky bluegrass and microorganisms [26] Research on Leymus chinensis shows its strong ability to accumulate heavy metals, particularly chromium, indicating its potential for phytoremediation [27]. Additionally, marigold has demonstrated superior tolerance and accumulation of cadmium, making it a viable option for heavy metal-contaminated soils [28]. While phytoremediation is environmentally friendly, its longer remediation cycle and reduced effectiveness in highly contaminated areas remain challenges [29].

5.5. Summary of major findings

Heavy Metal Content in Chemical Fertilizers: A study of 400 fertilizer samples revealed phosphorus fertilizers as the leading source of heavy metal contamination, with an exceedance rate of 26.97%. Specifically, the exceedance rates for arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) were 4% or higher, with mercury reaching 27%. This highlights phosphorus fertilizers as a major contributor to heavy metal soil pollution.

Accumulation Effects of Heavy Metals in Soil: Long-term use of phosphorus and organic fertilizers results in heavy metal accumulation, such as cadmium (Cd) and chromium (Cr), which can infiltrate the food chain, threatening ecosystems and human health. Differences in the Impact of Various Fertilizers on Soil Heavy Metals: Although organic fertilizers enrich soil organic matter, they can also elevate copper (Cu), zinc (Zn), and cadmium (Cd) levels. In some cases, copper concentrations reached 82.0 mg/kg, exceeding the safety threshold of 50 mg/kg.

Application of Remediation Technologies: Remediation methods such as physical, chemical, and biological techniques exist, with combined phytoremediation and microbial remediation showing high potential for improving soil heavy metal detoxification. However, these techniques still show low

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efficiency when addressing complex pollution, and discrepancies often exist between laboratory results and practical applications.

Insufficient Research on Complex Pollution: Current research predominantly addresses single-metal pollution, with limited focus on multi-metal and organic pollutant co-contamination. This gap reduces the effectiveness of remediation technologies in complex contamination scenarios.

5.6. Research Gaps and Future Directions

Although research on heavy metal pollution in soil has made some progress, significant gaps remain. Most research focuses on single heavy metal pollution and its remediation, neglecting the complexity of coexisting pollutants in real-world environments. Interactions between pollutants can alter their behavior, but current remediation methods inadequately address this complexity. In real-world soils, complex pollution often occurs, where interactions between pollutants alter the mobility, bioavailability, and toxicity of heavy metals. Yet, remediation technologies lack sufficient research on these dynamics.

Secondly, remediation technologies developed in the laboratory, such as chemical remediation, phytoremediation, and microbial remediation, demonstrate good results under controlled conditions. However, their effectiveness often declines significantly in practical applications due to influences such as soil physicochemical properties, climatic conditions, and complex pollution scenarios. Thus, the gap between laboratory results and real-world applications is a pressing issue that needs to be addressed.

Lastly, there is a lack of research on the long-term ecological effects of remediation. Most existing studies focus on short-term control of heavy metal concentrations, with limited in-depth tracking and evaluation of the long-term impacts on soil ecological health, changes in microbial communities, and the safety of crops after remediation. This limits our understanding of the long-term stability and sustainability of remediation measures.

6. Conclusion

Heavy metal pollution presents a major threat to environmental and agricultural systems, exacerbated by the widespread use of chemical fertilizers. Fertilizers, particularly phosphorus and organic variants, contribute to the accumulation of toxic metals such as copper, zinc, cadmium, arsenic, and lead in soils, posing significant risks to human health through the food chain. Remediation technologies often show high efficiency in laboratories but perform poorly in real-world conditions due to the interactions among soil properties, climate, and pollutants. Current studies tend to overlook long-term ecological impacts and the complexity of multiple pollutants interacting together. This study emphasizes the urgent need for effective remediation technologies, especially as traditional methods often fail to address the complexity of multi-metal contamination in real-world agricultural settings.

Future research must focus on developing comprehensive remediation strategies that tackle the combined effects of multiple heavy metals and organic pollutants. Integrating chemical and biological techniques—such as combining phytoremediation with microbial remediation—offers potential for improving metal stabilization in soils and boosting plant uptake. Additionally, emerging technologies such as nanotechnology should be explored to optimize existing methods. Bridging the gap between laboratory efficacy and field performance is crucial, requiring the adaptation of technologies to diverse environmental conditions.

Addressing these challenges will improve soil pollution management and contribute to safer agricultural production. Effective long-term monitoring systems will help evaluate the ecological impact of remediation efforts and provide crucial data for policymakers. Ultimately, by advancing remediation technologies and adopting comprehensive pollution control measures, we can ensure the sustainability of agricultural ecosystems and protect environmental health for future generations.

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