A comparative study on the efficacy of conventional and green chelating agents for soil heavy metal remediation

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Abstract. Soil heavy metal contamination poses significant risks to ecological balance, vegetation, agricultural productivity, and human health due to the toxicity, persistence, and resistance to degradation of heavy metal ions. Chelation presents a widely employed method for heavy metal ion removal. While conventional chelating agents exhibit high removal efficiencies, they are associated with ecological hazards. In contrast, green chelating agents have gained prominence for their environmentally friendly attributes. This paper meticulously examines the heavy metal removal efficiencies, mechanisms, application advantages and limitations, as well as optimal pH ranges of both the traditional chelating agent Ethylenediaminetetraacetic acid (EDTA) and the green chelating agents Tetrasodium glutamate diacetate (GLDA), Methylglycine diacetic acid (MGDA), and [S, S]-ethylenediaminedisuccinic acid (EDDS). A comprehensive comparison reveals that EDDS exhibits superior performance followed by MGDA, GLDA, and EDTA, with MGDA displaying the broadest pH range spanning from 2 to 13.5. EDDS emerges as the most biodegradable option. Moreover, in terms of elimination rates, both MGDA and EDDS demonstrate comparable efficacy to EDTA. In conclusion, the green chelating agents scrutinized herein hold promise to potentially supplant conventional EDTA in large-scale practical applications.

Keywords: EDTA, GLDA, MGDA, EDDS, Heavy Metal Chelating Agents.

1. Introduction

Soil ecosystem contamination by heavy metals represents a significant global challenge. The risk to ecological balance, ecotoxicology, and human health posed by heavy metals, particularly those found in road dust containing polluted soil, has been observed to be most pronounced in Asia, followed by Europe, Australia, America, and Africa [1]. Heavy metals in soil exhibit characteristics such as limited mobility, prolonged retention times, and resistance to microbial degradation, ultimately posing threats to human health through water and plant pathways

Numerous sources contribute to heavy metal pollution in soil. These metals have naturally occurred in the Earth's crust since its formation. However, anthropogenic activities, including mining, road construction, smelting, foundries, landfills, waste disposal, runoff, and metal-based industries, predominantly drive heavy metal pollution [2]. Atmospheric deposition is a significant route through which heavy metals enter the soil, originating primarily from emissions related to energy production, transportation, =metallurgical activities, and construction material manufacturing. Initially, heavy metals

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enter the atmosphere as aerosols before settling in the soil through natural sedimentation and precipitation. Natural processes such as volcanic eruptions, metal corrosion, metal evaporation from soil and water, sediment resuspension, soil erosion, and geological weathering also contribute to increased heavy metal concentrations. Wastewater serves as another source of heavy metals in soil; untreated discharge or irrigation with industrial and mining wastewater can result in heavy metal deposition. Solid waste, including various forms of mining and industrial waste, poses a severe threat of heavy metal pollution. Exposure to sunlight, rain, and water washing during waste stacking or processing facilitates the migration of heavy metals into surrounding soil and water.

Certain heavy metals pose serious health risks to humans, such as the historical occurrences of Itai-Itai and Minamata diseases in Japan, while others directly inhibit plant growth [3]. The chemical forms of heavy metals in soil are highly complex, existing in various states including exchangeable, carbonate-bound, manganese-iron-bound, organic-bound, and residual states [4-5].

Mainstream approaches to remediate heavy metal contamination in soil typically involve stability and solidification methods, as well as strategies involving the absorption and elimination of heavy metals by certain plant parts. Various substances, including those containing silicon (e.g., zeolites, diatomaceous earth, silicates), calcium (e.g., quicklime, limestone, hydrated lime), phosphorus (e.g., sodium hydrogen phosphate, calcium phosphate), and metal oxides (e.g., red mud, slag), are utilized to precipitate, adsorb, or immobilize heavy metals. Chelating agents such as MGDA, GLDA, EDTA, and DTC form stable complexes with heavy metals [6-7].

Chelation-induced remediation technology enhances the absorption or enrichment of heavy metals by plants by releasing solid-phase-bound heavy metals into the soil solution in dissolved or readily soluble forms [8]. Certain biodegradable green chelating agents offer energy-efficient manufacturing processes, environmental friendliness, low ecological footprints, and excellent solubility across a broad pH spectrum [9]. This article focuses on the use of chelating chemicals to solidify and stabilize heavy metals.

The objective of this article is to comprehensively analyze the performance, advantages, and limitations of various chelating agents, with a specific focus on conventional agents like EDTA and environmentally friendly alternatives such as GLDA, MGDA, and EDDS. It includes detailed assessments of the heavy metal removal rates of EDTA and GLDA in different environmental contexts, comparisons of their ecological and environmental impacts, elucidation of specific reaction mechanisms, and identification of optimal pH ranges.

2. EDTA

2.1. Advantages and Mechanisms

Ethylenediaminetetraacetic acid (EDTA), a prominent chelating agent, excels in the remediation of heavy metals due to its high solubility with metals in aquatic environments and its capability to mobilize heavy metal ions such as Pb, Cu, Zn, and Cd. Its hexadentate ligand structure, featuring four carboxyl and two amine groups, facilitates the formation of chelates with various metal ions including Mn(II), Cu(II), Fe(III), and Co(II), enhancing its versatility in metal sequestration [10-11].

The fundamental mechanisms driving its effectiveness include redox reactions and metal ion exchange processes [12]:

(i) Metal ion exchange involves the substitution between an EDTA-bound metal ion (M_1) and a free heavy metal ion (M_2), following the general reaction:

$$M_1EDTA^{n+/-} + M_2^{n+/-} \rightarrow M_2EDTA^{n+/-} + M_1^{n+/-} \quad (1)$$

(ii) Dissolution of minerals plays a critical role, wherein EDTA facilitates the exchange of metal ions (M_3 and M_4) with those in soil minerals:

$$M_3EDTA+M_{4\,x}O_y \rightarrow M_4EDTA+M_3 \quad (2)$$

(iii) The complexing capacity of EDTA enables the extraction of metal ions as EDTA salts, which can then be separated in subsequent processes, illustrating metal remobilization [13]:

$$M_1EDTA_{dissolved} + M_{2absorbed} \rightarrow M_2EDTA_{dissolved} + M_{1absorbed} \tag{3}$$

(iv) Redox reactions occur with the complexed metals.

(v) Absorption reactions take place on ternary surface complexes.

The interplay between the concentration of EDTA and the heavy metal content in soil impacts the overall efficiency of the remediation process.

Heavy metals can be mitigated in soil through the removal of plants that have absorbed high concentrations of these contaminants. In a similar vein, Ethylenediaminetetraacetic acid (EDTA) enhances the uptake of metal ions by plants, thereby increasing the bioavailability of heavy metals within the rhizosphere and significantly boosting the plants' capacity to accumulate them, with a notable effect on lead (Pb) [14]. This process effectively lowers the concentration of free heavy metal cations. EDTA, known chemically as $EDTA^{4-}$, and its conjugate acid, H_4EDTA, exhibit pronounced efficacy in complexing with divalent and trivalent metal cations due to its nature as a polybasic weak acid [15-17]. Additionally, while EDTA has the potential to act as a buffering agent, this attribute remains relatively underexplored.

2.2. *Stability of EDTA*

The stability of metal-EDTA complexes is known to be influenced by fluctuations in pH [18-19]. Studies have indicated that heavy metal removal efficiencies decrease with rising pH levels [20-21]. EDTA exhibits the highest removal rates for Cd, Cu, and Ni under acidic conditions. However, excessive acidity during extraction can alter sediment structure and diminish the long-term viability of sediment utilization [22]. The metal affinity for EDTA is contingent upon soil pH, given constant cation molarity. Consequently, metals with higher affinity for EDTA become dominant in metal-EDTA coordination when pH levels are equivalent [23]. To optimize EDTA usage, it is advisable to apply it under buffered solutions to mitigate potential pH-related efficacy fluctuations [24]. Additionally, the optimal pH for EDTA application is reported to be 4 [22]. Table 1 summarizes the removal efficiencies of metals by EDTA under various conditions.

Table 1. Performance of EDTA under various conditions

Heavy metals	Removal performance with EDTA
Pb	High clay and silt content: 64.2% [25]
	Biotic soil: 58.4% [26]
	Biomass: 99% [27]
	Smelting/battery recycling site: 58% [28]
	Natural calcareous soil: 16%-19% [23]
Zn	Biotic soil (by soil leaching): 25% [26]
	Sand-loam soil (after PV): 63.5% [29]
Cd	Biotic soil (by soil leaching): 68% [30]
	Natural calcareous soil: 24%-57% [23]
	Bottom mud in minging water: 78% [22]
Cu	High clay and silt content: 38.8% [30]
	Natural calcareous soil: 15%-22% [23]
	Bottom mud in mining water: 63% [22]

Despite EDTA's effectiveness in heavy metal remediation, its environmental impact raises concerns. The agent's poor biodegradability and prolonged soil residence time contribute to significant ecological disruptions once metals are extracted [31]. Notably, EDTA usage increases soil phosphorus levels and facilitates base ion leaching [31-32]. Although EDTA plays a critical role in mitigating heavy metal pollution and protecting ecosystems, its adverse effects on soil ecology cannot be overlooked. Thus, a comprehensive assessment of EDTA's impact necessitates consideration of both its remediation efficiency and the broader environmental implications.

3. Green Chelating Agents

3.1. GLDA

3.1.1. Advantages and Mechanisms. GLDA, recognized as a green chelating agent, is gaining traction in practical applications due to its environmentally benign characteristics [33]. As a biodegradable chelator, GLDA exhibits a minimal ecological footprint and is produced from biological carbon sources, aligning with sustainable practices [34]. Notably, GLDA achieves over 60% degradation within 28 days, highlighting its environmental compatibility [35]. This agent is increasingly employed to reclaim hazardous elements and rare metals from diverse solid waste forms, presenting a viable alternative to mitigate ecological risks associated with conventional chelators like EDTA [34]. The Swedish Society for Nature Conservation (SSNC) has endorsed GLDA, confirming that 86% of its composition is sourced from sustainable materials, earning the Bra Miljöval label. GLDA distinguishes itself among biodegradable chelators with its superior efficacy in situ-washing treatments for toxic content removal [36]. In certain environments, such as mining and agricultural soils, GLDA's metal extraction performance closely mirrors that of EDTA, showcasing comparable rates of Cd and Ni removal under optimal conditions of elution and pH [37]. Furthermore, GLDA demonstrates remarkable proficiency in eliminating Cu, Ni, Zn, and Cd from soils [9]. Similar to EDTA, the effectiveness of GLDA is influenced by soil pH levels and the dosed concentration during the remediation process, necessitating careful consideration of these factors in its application.

The chelating mechanism of GLDA encompasses several steps, beginning with an equilibrium reaction between the ligand (L) and metal ions (M), indicative of metal-ligand interaction dynamics [38]:

$$\mathrm{M}^{\mathrm{m}+} + \mathrm{L}^{\mathrm{n}-} \leftrightarrow \mathrm{ML}^{(\mathrm{m}-\mathrm{n})} \tag{4}$$

where m represents the charge on the metal ion, and n the charge on the ligand containing GLDA.

The subsequent reactions facilitate the effective removal of heavy metals from soil:

(i)

$$\mathrm{H_3GLDA^-} + \mathrm{M^{2+}} = [\mathrm{MH_3(GLDA)}]^+ \tag{5}$$

(ii)

$$\mathrm{H_2GLDA^{2-}} + \mathrm{M^{2+}} = [\mathrm{MH_2(GLDA)}] \tag{6}$$

(iii)

$$\mathrm{HGLDA^{3-}} + \mathrm{M^{2+}} = [\mathrm{MH(GLDA)}]^- \tag{7}$$

(iv)

$$\mathrm{GLDA^{4-}} + \mathrm{M^{2+}} = [\mathrm{M(GLDA)}]^{2-} \tag{8}$$

Firstly, in assessing the impact of pH value on the efficacy of GLDA, the stability constant in logarithmic form is considered. Secondly, drawing from a previous study, the stability constant (log KML, where M represents the metal ion and L denotes the ligand, GLDA) for 8 representative heavy metals was examined [35]. Cu exhibited the highest value, indicating GLDA's strongest chelating affinity for this ion, followed by Fe (III), Ni, Zn, Cd, Fe (II), Mg, and Ca, respectively. Additionally, when evaluating the efficiency of heavy metal ion extraction by GLDA, calculations incorporate factors such as the volume of the extraction solution (mL), the concentration of dissolved heavy metal ions in the extraction solution (μg L-1), the mass of the sludge sample (g), and the concentration of the metal in the sludge sample (mg kg-1).

3.1.2. Performance of GLDA. Through the application of the outlined process and the formulation of the extraction efficiency of heavy metal ions using the biodegradable chelator GLDA, the removal percentages were summarized in Table 2.

Table 2. Removal performance of different metals by GLDA

Heavy metals	Removal performance with GLDA
Pb	pH=3.56, 67.75% [39]
	pH=4, 54% [33]
	pH=5.5, 87.9% [37]
Zn	pH=3, 32% [9]
	pH=4, 62% [33]
	pH=5.5, 91.9% [37]
Cd	pH=3, 76% [9]
	pH=3.56, 81.04% [39]
	pH=4, 84% [33]
	pH=5, 10% [9]
	pH=5.5, 93.8% [37]
Cu	pH=3, 49% [9]
	pH=3.56, 77.35% [39]
	pH=4, 94% [33]
Ni	pH=3, 47% [9]
	pH=3.56, 75.78% [39]
	pH=4, 39% [33]

Table 2 highlights the exceptional performance of GLDA in extracting heavy metals such as Pb, Zn, Cd, Cu, and Ni from soil with pH values around 3 and 4. This suggests that GLDA is most effective under acidic conditions. At pH levels of 3.56 and 4, both Cu and Cd exhibited high extraction percentages using GLDA, with removal ratios of 81.04% and 94%, respectively. Moreover, the variation in removal efficiency is notably influenced by changes in pH, with percentages generally decreasing as pH values increase, consistent with findings from EDTA studies [20-21]. However, as an exception, the figures for Cd and Cu showed a significant increase, especially for Cd, rising from 10% at pH 5 to 93.8% at pH 5.5. It has been inferred that variations in GLDA concentration and the ratio between GLDA and heavy metal ions must be considered.

The extraction efficiency of GLDA appears heavily reliant on the pH of the surrounding environment and the concentration of GLDA. Therefore, careful consideration must be given to the ratio setting of this chelating agent prior to application. Consequently, while GLDA exhibits considerable promise, it is not a universal solution and requires cautious application in specific scenarios. Consequently, sample analysis is imperative before GLDA utilization, with the ideal GLDA concentration determined based on the results. Additionally, a study revealed that leaching with GLDA increased sample pH from 6.1 to 7.3 [40]. Apart from elevating phosphorus concentration, GLDA altered sandy soil texture to sandy loam, thereby modifying the original soil composition and structure [32].

In conclusion, the new biodegradable chelator, GLDA, performs admirably in many respects, particularly in extraction efficiency and environmental preservation. However, challenges persist, necessitating further investigation and careful application.

3.2. MGDA

MGDA stands out as a phosphorus-free chelator, distinguishing it from traditional heavy metal chelating agents. Unlike EDTA, MGDA does not contribute to soil and water eutrophication by leaving excessive phosphorus residues in the soil post-extraction. Furthermore, MGDA's production process is considerably safer than EDTA's, having passed the OECD 301D test with a biodegradation rate exceeding 80% in 28 days. Notably, MGDA does not carry the hazardous label associated with EDTA and NTA. However, it's worth noting that GLDA exhibits a higher degradation capacity in the OECD 301D test compared to MGDA [41-42]. MGDA demonstrates remarkable pH tolerance, ranging from pH 2 to pH 13.5, although it may not achieve the highest removal rates across all pH levels [43-44]. Additionally, MGDA boasts a high stability constant (log K), indicative of its strong affinity for specific metal ions [40]. Given its advantages in production safety and removal efficiency, there is a discernible shift towards replacing NTA or EDTA with MGDA [45].

3.3. EDDS

EDDS, classified as another biodegradable chelator, demonstrates extraction efficiency almost on par with EDTA and MGDA, surpassing that of CA and SCLC. Notably, EDDS boasts the highest biodegradability rate (99%) within 28 days, outstripping CA, MGDA, and EDTA [42]. Moreover, EDDS exhibits versatility across a wide pH range while maintaining robust stability [46]. Additionally, EDDS displays a notable selectivity for specific metals, particularly transition metals such as Cu and Fe over Ca and Mg. However, a drawback lies in EDDS's impact on soil pH, organic matter, and nutrient levels [47-48]. Presently, EDDS finds predominant application as a soil conditioner, chelating or complexing agent for agricultural trace elements, and surface treatment for transition metal removal.

4. Comparison and Further Trends

4.1. Comparison

Drawing from the data and concepts elucidated in preceding sections, the distinctive characteristics of EDTA, GLDA, MGDA, and EDDS are summarized in Table 3 [49].

Table 3. Characteristics of EDTA, GLDA, MGDA, and EDDS.

Chelating agent	Biodegradability	Short ecological footprint	Safety	Strong chelator
EDTA	×	×	×	√
GLDA	√	√	√	√
MGDA	√	√	√	√
EDDS	√	√	√	√

* × indicates that there is no such feature and √ represents the existence of such feature.

Further detailed information regarding these four chelators is provided in Table 4.

Table 4. Comparison of EDTA, GLDA, MGDA, and EDDS.

Chelating Agent	Advantage	Disadvantage
EDTA	Ability to chelate a variety of metal ions High capacity of the complexing effect Promoting the absorption of metal ions by plants High removal ratio for Cd, Cu, and Ni under acidic conditions	Long ecological footprint Environmental unfriendliness Poor biodegradability Destroy of soil structure and contents Increase of the phosphorus concentration
GLDA	Biodegradable Short ecological footprint Relatively high extraction ability among green chelators Best performance in Cu, Fe (III), Ni, Zn, Cd, Fe (II), Mg, and Ca High degradation ability in OECD 301D test	Significantly affected by pH Unsuitable for every soil condition Instead of the phosphorus concentration Transition of sandy soil texture to a sandy loam

Table 4. (continued).

MGDA	No excessive phosphorus leaching in soil A biodegradation rate over 80% in 28 days Non-hazardous Widest scope of pH values	Not the most significant removal rate at any pH level
EDDS	High affinity for specific metal ions Highest general extraction efficiency Highest biodegradability (99%) within 28 days Wide range of pH values High stability	Effect on the soil pH Impacts on organic matter and other nutrients

4.2. Further Trends

While the paramount consideration remains the protection of soil environment and quality through heavy metal elimination, attention must also be directed towards the ecological impact of chelating agents themselves. Leveraging the advantages of novel green chelating agents like MGDA, GLDA, and EDDS in terms of heavy metal removal efficiency and eco-friendliness, their potential large-scale substitution for traditional chelating agents such as EDTA warrants future consideration. Moreover, the combined application of multiple chelating agents holds promise. Some chelating compounds exhibit high sensitivity to pH fluctuations, while others boast minimal ecological footprints but less effective removal rates. By combining different chelating agents, synergistic effects may be harnessed to optimize environmental friendliness and cost-effectiveness while achieving superior heavy metal removal rates. For instance, the combination of CA and GLDA in specific ratios has shown promise for heavy metal extraction [50].

5. Conclusion

Based on the analysis presented in this paper, the effectiveness of the four chelating agents is ranked as follows: EDDS > MGDA > GLDA > EDTA, considering their overall heavy metal removal rates, versatility in application, and environmental friendliness. Factors influencing the efficiency of chelators, such as pH levels, ratios of chelating agents to heavy metal ions, and soil conditions, have been comprehensively summarized. MGDA exhibits remarkable resilience to pH variations and can be utilized across a broad pH range from 2 to 13.5. Similarly, EDDS demonstrates applicability over a relatively wide pH spectrum. However, EDTA's efficacy is influenced by fluctuations in pH, while GLDA proves most suitable for acidic conditions, particularly between pH 3.56 and 4. Given that green chelating agents pose minimal risks of low biodegradation rates or soil eutrophication on a large scale, there is a compelling case for expanding their application scope and substituting hazardous conventional chelating agents. To facilitate further in-depth research on this matter, future studies could explore the operational intricacies and optimal ratios involved in the combined use of multiple chelating agents, taking into account their respective complementary benefits and drawbacks. Such investigations would contribute significantly to advancing our understanding of environmentally sustainable heavy metal remediation techniques.

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