Scientific Paper

Enhance electrokinetic remediation of potentially toxic elements in the dredged sediment of Camorim Lagoon

Remediação eletrocinética de elementos potencialmente tóxicos de sedimentos da lagoa de Camorim

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ABSTRACT

Electrokinetic remediation, effective for low-permeability sediments, can remove both organic and inorganic contaminants simultaneously. This study investigated its application to dredged sediments from Camorim Lagoon (Jacarepaguá lagoon complex, Rio de Janeiro), known to be contaminated with potentially toxic metals (chromium, copper, lead, zinc, and nickel). A buffer solution controlled pH changes, while the complexing agent ethylenediaminetetraacetic acid (EDTA) and humic substances aided metal desorption and removal. Treatments utilizing EDTA (EK2 and EK3) achieved lower final concentrations of zinc and lead. Notably, EK1 (freshwater only) achieved the lowest nickel concentration observed in the anodic section. EK1 also achieved the lowest copper concentration; however, none of the treatments reached regulatory limits for copper. Chromium, which is the least mobile metal, is concentrated in the middle section for all treatments, suggesting the need for further electrolyte optimization to promote efficient chromium transport.

Keywords: electrokinetic remediation; potentially toxic elements; dredged sediment; pH control; circular economy.

RESUMO

Aremediação eletrocinética é eficaz para sedimentos de baixa permeabilidade, permite a remoção simultânea de contaminantes orgânicos e inorgânicos. Neste estudo, foi investigada a aplicação da remediação eletrocinética em sedimentos dragados da lagoa de Camorim (complexo lagunar de Jacarepaguá, Rio de Janeiro), contaminada por metais potencialmente tóxicos, como cromo, cobre, chumbo, zinco e níquel. Utilizou-se uma solução tampão para controlar as variações de pH, enquanto o agente complexante EDTA e substâncias húmicas auxiliaram na dessorção e na remoção dos metais. Os tratamentos com EDTA (EK2 e EK3) resultaram em menores concentrações finais de zinco e chumbo. De forma notável, o tratamento EK1 (apenas água doce) alcançou a menor concentração de níquel na seção anódica. Embora o EK1 também tenha apresentado a menor concentração de cobre, nenhum dos tratamentos atingiu os limites regulatórios para esse metal. O cromo, metal menos móvel, concentrou-se na seção média em todos os tratamentos, indicando a necessidade de otimizar o eletrólito para promover sua remoção mais eficiente.

Palavras-chave: remediação eletrocinética; elementos potencialmente tóxicos; sedimentos dragados; controle de pH; economia circular.

INTRODUCTION

Dredging activities in lagoons, rivers, and ports are crucial for maintaining water quality, depth, and navigation channels (Çevikbilen *et al.*, 2020; Kim *et al.*, 2011). However, this process generates vast volumes of sediment annually, posing a management challenge, while international conventions (London

(1972), Barcelona (1976), Helsinki (1992), and OSPAR (1992)) promote considering dredged sediment as a resource (Amar *et al.*, 2021).

According to Crocetti *et al.* (2022), "a linear sediment treatment strategy that only focuses on pollutant removal and toxicity reduction is often not sustainable." Therefore, Spadaro and Rosenthal (2020) and Crocetti *et al.* (2022)

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suggest transitioning toward a circular economy as an alternative. Dredged sediments can be beneficially reused in the civil construction industry, for example, as pavement or landscaping materials. Another alternative is recycling, where the sediment is transformed into a raw material for mortar production. By promoting a circular economy, industrial symbiosis can be fostered in areas with established industrial growth (Rossa *et al.*, 2017). However, contaminated sediments require treatment before reuse.

Electrokinetic remediation emerges as a promising alternative for treating low-permeability, contaminated sediments and can remove both organic and inorganic contaminants simultaneously (Acar; Alshawabkeh, 1993). This technique utilizes a low-intensity direct current (DC) applied through electrodes inserted into the sediment (Asadollahfardi *et al.*, 2021; Bauddh; Singh; Korstad, 2017; Osman, 2018; Ribeiro; Mateus; Couto, 2016; Sharma *et al.*, 2018). The generated electric field mobilizes and transports pollutants, nutrients, and microorganisms through mechanisms such as electro-osmosis, electromigration, electrophoresis, and diffusion (Osman, 2018; Sharma *et al.*, 2018). Notably, pH changes play a critical role in this process, impacting various aspects such as contaminant behavior and complex formation (Reddy; Cameselle, 2009).

The Jacarepaguá lagoon complex in Rio de Janeiro, Brazil, exemplifies the need for innovative sediment remediation strategies. Decades of industrial and urban pollution have resulted in silting, reduced water surface area, and poor water quality (Masterplan, 2015). Untreated domestic and industrial waste has contributed to the presence of potentially toxic elements (Almeida; Borma; Barbosa, 2001; Araruna Junior *et al.*, 2012; Fernandes *et al.*, 1994) in the dredged sediments, particularly chromium (Cr), copper (Cu), lead (Pb), zinc (Zn), and nickel (Ni), exceeding regulatory limits set by CONAMA resolution No. 454 of 2012.

Electrokinetic remediation has not been explored as an alternative to remediate the dredged sediment in the Camorim lagoon. Therefore, this research proposes to study the application of electrokinetic remediation to dredged sediments from Camorim Lagoon. It will utilize a buffer solution to control pH changes and employ the complexation agent ethylenediaminetetraacetic acid (EDTA) and humic substances to enhance the desorption and removal of potentially toxic metals.

While dredging is essential for maintaining waterways, the management of contaminated dredged sediments remains a challenge. Electrokinetic remediation shows promise for treating low-permeability sediments contaminated with heavy metals, but its application to dredged sediments from the Jacarepaguá lagoon complex has not been explored. This research aims to investigate the effectiveness of electrokinetic remediation for decontaminating dredged sediments from Camorim Lagoon, a particularly impacted area within the complex. The study will employ a buffer solution to control pH changes and utilize the complexation agent EDTA and humic substances to enhance the desorption and removal of potentially toxic metals.

METHOD

Sediment sampling

Sediment samples were collected in July 2022 from Camorim Lagoon, Rio de Janeiro, Brazil (22° 58.533' S, 43° 21.511' W), designated as sampling site CP1. A Van Veen grab sampler was used to collect approximately 47 kg of sediment, stored in plastic bags within an ice chest for transport to the laboratory. Upon arrival, the sediment was homogenized using mechanical mixing and refrigerated at 4°C. Following air-drying for 48 h, the sediment was analyzed for particle size distribution (ABNT NBR 7181, 2016a), moisture content (ASTM D2216, 2019), liquid limit, plasticity limit, and plasticity index (ABNT NBR [6459 (2016a), 7180 (2016b)]), and grain density (ABNT NBR 6457, 2016c). The concentration of potentially toxic metals was determined using US-EPA 3051A. The pH and electrical conductivity were determined according to ASTM D4972 (2019) and ISO 11265 (1994), respectively.

Electrokinetic device

The electrokinetic cell consisted of two electrodes fixed on an acrylic plate, spaced 88.5 mm apart from the sediment sample. The electrodes were connected to a DC power supply to facilitate the application of an electric field. For solution adjustment and gas exhaust, two 50-mL pipettes, each marked with 0.1-mL graduations, were connected to the electrode compartments. This design is consistent with the approach described by da Rocha *et al.* (2009).

Electrokinetic test

The sediment sample underwent preparation for the electrokinetic treatment. First, the reconstituted sediment was loaded into the designated sample chamber. To ensure proper compaction, a press was used to consolidate the sediment with a vertical stress of 34.5 kPa. After achieving the desired dimensions, the sample was trimmed. To prevent fine sediment particles from migrating into the electrolyte chambers during the experiment, filter papers were placed at each end of the sample chamber. Finally, specific electrolyte solutions, detailed in Table 1, were added to their respective chambers.

Throughout the treatment process, aliquots of anolyte and catholyte were collected using a syringe with a 2-mm-diameter hose to monitor the pH and EC. The removed electrolytes were replaced with fresh solution. Additionally, the electric current was monitored. After the test, the samples were sectioned into three roughly equal portions (anodic, middle, and cathodic) for analysis of pH, EC, and potentially toxic elements concentration. Table 2 presents the nomenclature used for electrolytes and sediment samples in test EK1, which is similar to other tests. Figure 1 shows the electrokinetic device used in this investigation.

Table 1 - Experimental conditions of electrokinetic tests.

Treatments	Anolyte	Catholyte	Electric potential (V cm ⁻¹)	Duration (days)
EK1	Freshwater	Freshwater	1.0	7
EK2	0.01 M NaOH	Buffer solution + 0.1M EDTA	1.0	9
EK3	Buffer solution	Buffer solution + 0.1 M EDTA	1.0	11
EK4	HS 5 g L ⁻¹	HS 5 g L⁻¹	1.0	11

Analytical method

The electrical conductivity (EC) of the sediment samples was measured using a 1:5 sediment-to-water ratio (w:v) ratio. A separate 1:2.5 sediment-to-water ratio (w:v) was used for pH measurements. The EC and pH of the electrolyte

Table 2 - Nomenclature of electrolytic samples and sediment samples.

Treatment	Nomenclature	Description	
EKI	EK1-C	Catholyte for experiment EK1	
	EK1-A	Anolyte for experiment EK1	
	EK1-C-C	Control catholyte for experiment EK1	
	EK1-A-C	Control anolyte for experiment EK1	
	EK1-CS	Cathodic sediment for experiment EK1	
	EK1-MS	Middle sediment for experiment EK1	
	EK1-AS	Anodic sediment for experiment EK1	
	EK1-CS-C	Control cathodic sediment for experiment EK1	
	EK1-MS-C	Control middle sediment for experiment EK1	
	EK1-AS-C	Control anodic sediment for experiment EK1	

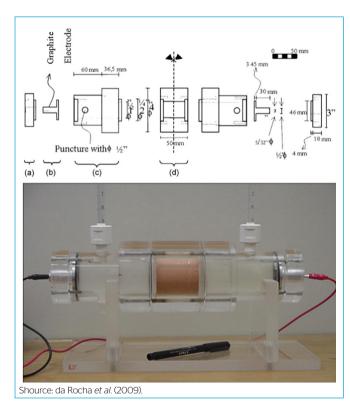


Figure 1 - Electrokinetic cell.

solutions were measured directly. A Hanna Instruments pH meter (model HI11310) and a Hanna Instruments conductivity electrode (model HI763100) were used for all measurements.

Microwave digestion following the EPA 3051A method was employed to prepare the sediment samples for analysis of their elemental composition (Zn, Ni, Cr, and Cu). Briefly, this method involves digesting 1.0 g of ground sediment with a mixture of concentrated nitric acid (9 mL) and concentrated hydrochloric acid (3 mL) within a microwave digester. The resulting digest was then analyzed using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) Optima 7300 DV (Perkin Elmer Instruments) to determine the concentrations of the target elements.

RESULTS AND DISCUSSION

Characterization of sediment

The physicochemical properties and initial concentration of potentially toxic sediments in sediment are summarized in Tables 3 and 4.

Electrolytes pH

The solutions EK1-C and EK4-C, containing freshwater and humic acid, respectively, exhibited a rapid rise in pH (above 12 within 24 h) due to their inherent lack of buffering capacity. This increase can be attributed to the generation of hydroxyl ions (OH $^-$) during water electrolysis, as reported in the literature (Beyrami, 2021; Estabragh *et al.*, 2019). Kanbar *et al.* (2023) also observed an increase in pH after 24 h of treatment despite using citric acid in the catholyte. Conversely, treatments EK2-C and EK3-C, which included a buffer solution,

Table 3 - Physical and chemical characterization of sediments.

Parameter	Testing method	Results	
		Medium sand	1.5%
Particle size distribution	ABNT NBR 7181(2016a)	Fine sand	6.7%
Particle size distribution	ADINT INDR / IOI(2010d)	Silt	72.4%
		Clay	19.4%
Moisture content	ASTM D2216 (2019).	318.5%	
Liquid limit	ABNT NBR 6459 (2016b)	141.6%	
Plasticity limit	ABNT NBR 7180 (2016c)	56.0%	
Plasticity index	ABNT NBR 7180 (2016c)	85.6%	
Grain density	ABNT NBR 6457 (2016c)	2.54	
рН	ASTM D4972 (2019)	7:11	
Electrical conductivity	ISO 11265 (1994)	13.37 mS cm ⁻¹	

Table 4 - Initial concentration of potentially toxic metals in sediments.

Parameter	Testing method	Results	Limit quantitation	Limit detection
Zn	ICP-OES EPA 3051A	287.33 mg kg ⁻¹	0.005	0.0016
Pb		46.00 mg kg ⁻¹	0.034	0.0104
Ni		27.33 mg kg ⁻¹	0.007	0.0022
Cu		76.00 mg kg ⁻¹	0.00002	0.0001
Cr		40.67 mg kg ⁻¹	0.005	0.0016

composed of acetic acid and NaOH, effectively controlled the pH rise, maintaining it below 6.5, as can be seen in Figure 2.

All treatments displayed a rapid decrease in anolyte pH (<2 within 2 days) as a result of water electrolysis generating hydrogen ions (H^+). Notably, neither the buffer solutions nor NaOH alone was successful in preventing this decrease, as can be seen in Figure 3.

Control treatments generally maintained stable pH, except EK1-C-C and EK1-A-C, with a slight initial drop (7.33–6), possibly due to freshwater-sediment interaction. EK2-C-C catholyte remained constant (around 4) due to buffer capacity, while the anolyte decreased (Chen *et al.*, 2023). Both EK3 controls had a constant pH (around 4), likely from the buffer. EK4 controls exhibited a stable range (9.86–8.37), likely due to humic acid composition and sediment interaction.

Sediment pH

Treatments EK1 and EK4, which lacked inherent buffering capacity in their electrolytes, caused significant pH changes in the sediment. Water electrolysis at the cathode generated hydroxyl ions (OH⁻), resulting in a notable pH increase (9.1 and 8.4 for EK1 and EK4, respectively), while hydrogen ion (H*) production at the anode led to a marked pH decrease (2.5 and 1.9 for EK1 and EK4, respectively), as can be seen in Figure 4. Similar findings were reported by Fu

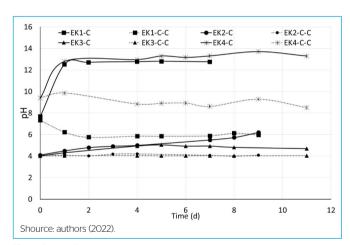


Figure 2 - pH of catholyte solutions.

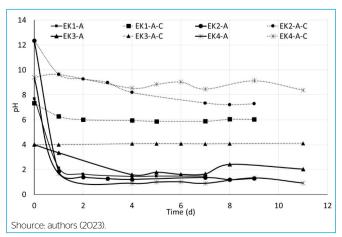


Figure 3 - pH of anolyte solutions.

et al. (2017), Kanbar et al. (2023), and Mohamadi, Saeedi, and Mollahosseini (2019), where soil near the anode exhibited a low pH and soil near the cathode showed a higher pH.

The presence of a buffer solution and EDTA in the catholyte of treatments EK2 and EK3 effectively mitigated the pH rise near the cathode, resulting in lower pH values (5.9 and 4.6 for EK2 and EK3, respectively) compared to treatments lacking buffer control. However, the use of NaOH as an anolyte in EK3 was not as effective in controlling pH near the anode, leading to decreased pH values in anodic sediment, as can be seen in Figure 4. This suggests the anolyte composition plays a crucial role in overall pH management during electrokinetic remediation.

The pH of the control treatments displayed minimal changes compared to the electrokinetic treatments. The most significant change was observed in the cathodic zone of the EK2 control (pH=5.4). This is likely due to the starting pH of the catholyte (pH=4.0) used in this control.

Electrolytes EC

Figure 5 presents the initial EC of each treatment, reflecting the varying electrolytes used. Both anolytes and catholytes exhibited a rapid EC increase in the initial days, followed by a gradual decrease toward a stable state. Estabragh *et al.* (2019) observed similar behavior, where the EC increased over time. However, the

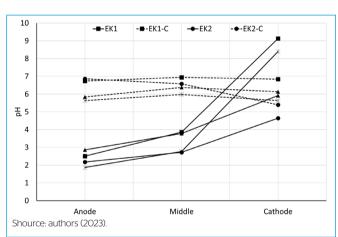


Figure 4 - pH of sediment.

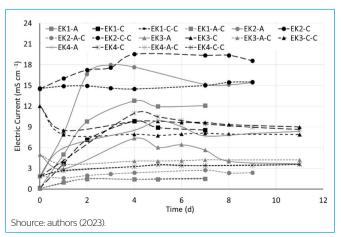


Figure 5 - EC of electrolytes.

EC in the electrolytes of EK3 and its control initially decreased, which might be attributed to the precipitation of buffer components from the electrolytes, as suggested by Kanbar, Ammami, and Benamar (2024).

In the EK1 treatment, the anolytes exhibited higher EC than the catholytes. Thisbehavior aligns with observations by Kanbar *et al.* (2023), who attributed it to the higher ionic mobility of H⁺ compared to OH⁻ ions. Conversely, EK2, EK3, and EK4 displayed the opposite behavior, likely due to the specific electrolytes used. An unexpected observation was the higher initial EC value of the catholyte solution in EK2-C and EK2-C-C (14.57 mS cm⁻¹) compared to EK3-C and EK3-C-C (12.04 mS cm⁻¹). This variation may be due to differences in preparation times and slight variations in pH (EK2: 4.1 vs. EK3: 4.0).

Control EC remained constant due to the absence of an electric field. A slight initial rise, possibly due to electrolyte-sediment interaction and ion dissolution, was observed, most prominently in EK3-C-C (12.04 to 7.91 mS cm $^{-1}$), before stabilizing.

Sediment EC

Figure 6 illustrates the EC of sediment samples collected from the electrokinetic treatment zones (anodic, middle, and cathodic) following treatment completion. Interestingly, the sediment near the anode and cathode displayed higher EC values compared to the central regions. This observation can be explained by the proximity of these zones to the electrolytes, which possessed high initial EC values (in the order of mS cm⁻¹) and the electric field-driven transport of ions driven by the applied electric field during electrokinetic treatment (Kanbar *et al.*, 2023; Tang *et al.*, 2020; Tang *et al.*, 2021). Kanbar *et al.* (2023) observed similar results and attributed this behavior to the ions, mean H⁺ and OH⁻, produced during the water electrolysis and the ions released from the sediment.

EK2 and EK3 control treatments exhibited higher EC readings in the cathodic section, likely due to the presence of high concentrations of ions in the catholytes. Conversely, EK1 and EK4 controls showed minimal changes in EC.

Electrical current response

The electrical current measured during the experiment differed between the treatments (Figure 7). EK1 and EK4 started with low current readings, followed by a rapid rise to peak values (7.5 and 8.6 mA, respectively). The initial increase in conductivity is probably due to ions desorbing from sediment, as noted by

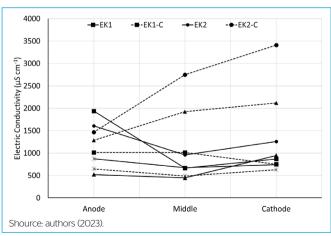


Figure 6 - EC of sediment.

Song *et al.* (2018). Similar findings were reported by Kanbar, Ammami, and Benamar (2024) and Garcia-Blas, Jimenez-Relinque, and Castellote (2022), as well as Song *et al.* (2016), who also linked this rise to sediment ion desorption.

Afterward, the current decreased and stabilized, suggesting a depletion of available ions, possibly due to the precipitation of electrically resistive compounds (Falciglia *et al.*, 2017). This decrease aligns with the concept of electromigration and electro-osmosis, which transport ions away from the electrodes.

In contrast, EK2 and EK3 exhibited a distinct "U-shaped" pattern in the first few hours. These treatments started with low current values, which then dipped further before rising again. This initial decrease might be attributed to factors specific to the electrolytes used, potentially affecting the initial availability of ions. Masi, Iannelli, and Losito (2016) observed a similar trend when using EDTA as the electrolyte. Following this initial phase, their current behavior resembled that of EK1 and EK4, suggesting a similar process of ion depletion and current stabilization. As. Maqbool and Jiang (2023) noted, the electric current profile is indeed related to ionic migration.

Potentially toxic elements

Figures 8–12 illustrate the potentially toxic elements' concentration (mg kg $^{-1}$) in sediment at the end of electrokinetic treatment, control tests, initial concentration, and levels 1 and 2 set by CONAMA Resolution N° 454 of 2012. All sections (anodic, middle, and cathodic) were analyzed for each treatment. Zinc concentration: EK1 and EK4 treatments exhibited the highest zinc concentrations in the cathode section (311 and 537 mg kg $^{-1}$, respectively), suggesting electro-osmosis and electromigration as primary transport mechanisms, potentially influenced by the lower pH at the anode aiding zinc desorption. This finding is consistent with Bahemmat, Farahbakhsh, and Kianirad (2016), who

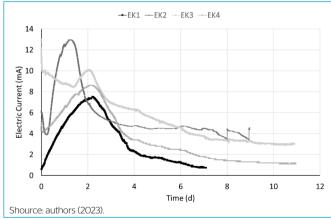


Figure 7 - Electric current variation in electrolytes during treatment.

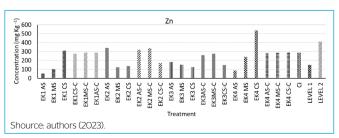


Figure 8 - Zinc concentration at the end of electrokinetic treatments.

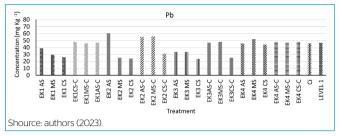


Figure 9 - Lead concentration at the end of electrokinetic treatments.

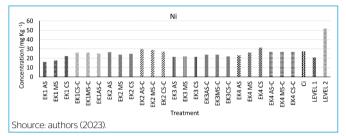


Figure 10 - Nickel concentration at the end of electrokinetic treatments.

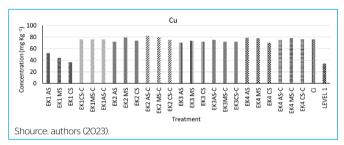


Figure 11 - Copper concentration at the end of electrokinetic treatments.

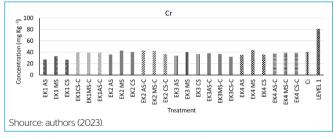


Figure 12 - Chromium concentration at the end of electrokinetic treatments.

found the highest concentrations there (340.5 and 183 mg kg $^{-1}$, respectively), indicating electromigration as the dominant mechanism, likely due to EDTA promoting zinc desorption and forming negatively charged complexes (EDTA-lead complexes). Song *et al.* (2016) achieved favorable results (~35% removal efficiency) using EDTA as both an anolyte and catholyte solution, suggesting its potential for enhancing zinc removal. Although EK3 had the lowest overall zinc concentration, only the cathode section met the regulatory limit (150 mg kg $^{-1}$), suggesting a need to extend the treatment duration for full compliance.

Controls showed minimal zinc removal except for a potential EDTA-driven decrease in the cathode section.

Lead concentration: In EK1, the lead migrated toward the anode, with the highest concentration (38.67 $\rm mg\,kg^{-1}$) in this region. This behavior is likely due to the formation of soluble metal hydroxides at the high pH near the anode. However, all sections achieved lead concentrations below the regulatory limit established in CONAMA Resolution N° 454 of 2012 (46.7 $\rm mg\,kg^{-1}$).

In EK2, lead moved toward the anode, reaching its highest concentration there $(60.5 \text{ mg kg}^{-1})$, likely due to electromigration facilitated by EDTA promoting lead desorption into negatively charged complexes. The middle and cathodic sections stayed below the regulatory limit.

EK3 achieved the lowest overall lead concentration across all sections compared to the initial value, with all sections meeting regulatory limits; the cathode section had the lowest concentration (23.67 mg kg $^{-1}$). EDTA likely aided lead desorption and complexation for transport toward the anode. Hahladakis, Latsos, and Gidarakos (2016) observed similar results with zinc, nickel, and lead, suggesting that potentially toxic metals may have formed complexes with organic matter, facilitating transport toward the anode. Wen *et al.* (2023) and Song *et al.* (2016) also found EDTA effective for lead removal.

Unlike other treatments, EK4 showed no clear direction of lead transport. Both the anode and cathode sections had lower concentrations compared to the middle section. While humic acid may have partially formed complexes with lead (Bahemmat; Farahbakhsh; Kianirad, 2016), it likely accumulated in the middle section rather than reaching the anode. Nonetheless, all sections remained below regulatory limits.

Controls displayed minimal lead removal, with some cathode sections showing a possible EDTA-driven decrease. Humic acid in the controls showed no significant lead desorption.

Nickel concentration: In EK1, nickel increased toward the cathode section, peaking there (22.33 mg kg $^{-1}$), suggesting electromigration or electro-osmosis. Masi, Iannelli, and Losito (2016) found similar results with freshwater electrolytes. Notably, two out of three sections achieved concentrations below the level 1 limit (20.9 mg kg $^{-1}$).

In EK2, nickel showed no clear transport direction, with uniform concentrations (around 25 mg kg⁻¹). All sections exceeded the level 1 limit, indicating EDTA's ineffectiveness in promoting nickel desorption.

EK3 mirrored EK2, with relatively constant nickel concentrations (around 21 mg kg $^{-1}$), exceeding the level 1 limit but lower than the initial value (26 mg kg $^{-1}$), possibly due to a buffering effect aiding desorption. EDTA remained ineffective. Song *et al.* (2016) identified nickel as the second most mobile metal with the use of EDTA.

Similar to EK1, EK4 saw nickel increase toward the cathode (31.33 mg kg⁻¹), hinting at electromigration or electro-osmosis. Despite exceeding the level 1 limit, Ge *et al.* (2022) noted a 29% Ni removal with fulvic acid, supported by Bahemmat, Farahbakhsh, and Kianirad (2016), indicating its promise in enhancing nickel removal.

Controls (water and humic acid) showed minimal nickel removal, while the EDTA control (EK2/EK3) exhibited a negligible decrease in the cathode section, suggesting limited EDTA effectiveness for nickel mobilization in this case. Copper concentration: In EK1, copper migrated toward the anode, peaking there (52.33 mg kg $^{-1}$), likely due to soluble metal hydroxides forming at the high pH near the anode, facilitating transport by electromigration. However, none of the sections met the regulatory limit (34 mg kg $^{-1}$).

In EK2, copper transport lacked a clear direction, with uniform concentrations (around 75 mg $\rm kg^{-1}$) exceeding the regulatory limit, suggesting EDTA's ineffectiveness in promoting copper desorption.

EK3 mirrored EK2, with relatively stable copper concentrations (around 72 mg ${\rm kg}^{-1}$), exceeding the regulatory limit but lower than the initial value (76 mg ${\rm kg}^{-1}$), possibly due to a buffering effect aiding desorption. However, EDTA

remained ineffective. Ayyanar and Thatikonda (2021) found that EDTA can also lead to the dissolution of Fe, Al, and Ca, which can compete with the target potentially toxic metals.

In EK4, copper increased toward the anode (highest concentration: 78.67 mg kg⁻¹), indicating electromigration or electro-osmosis. Despite all sections exceeding the regulatory limit, the chosen electrolyte (humic and fulvic acids) showed inefficiency in copper removal. This contrasts with Ge *et al.* (2022), who achieved 28% Cu removal using fulvic acid. Song *et al.* (2016) also found EDTA ineffective for copper removal.

Similar to nickel, controls (water and humic acid) showed minimal copper removal. The EDTA control (EK2/EK3) exhibited a negligible decrease in the cathode section, suggesting limited effectiveness for copper mobilization. Chromium concentration: In the EK1 treatment, chromium distribution showed no clear direction of transport. The middle section exhibited the highest concentration (33.00 mg kg $^{-1}$), followed by the anodic and cathodic sections. All sections were below the initial concentration (40.67 mg kg $^{-1}$) and the regulatory limit (81 mg kg $^{-1}$). The lower pH in the anodic section likely facilitated chromium desorption, but the transport mechanism remains unclear.

Similar to EK1, chromium distribution in EK2 lacked a clear direction of transport. The middle section exhibited the highest concentration (42.67 mg kg $^{-1}$). All sections remained below the initial concentration but approached this value compared to EK1.

Similar to EK1 and EK2, chromium distribution in EK3 displayed no clear direction of transport, with the middle section again having the highest concentration (40.33 mg kg $^{-1}$). All sections remained below the initial concentration and the regulatory limit.

In EK4, chromium mobility was limited, with concentrations decreasing below the initial level in the anodic and cathodic sections, and the middle section showing the highest concentration (43.67 mg kg⁻¹). Humic acid control tests supported this, suggesting chromium immobilization within the sediment. This aligns with Tsang and Hartley (2014) who found synthetic

chelating agents more effective for metal extraction compared to natural humic substances.

Control tests were conducted across treatments, which showed minimal removal of chromium. The EDTA controls (EK2/EK3) exhibited negligible cathode decreases, implying limited EDTA effectiveness for chromium mobilization.

CONCLUSIONS

- This study investigated the effectiveness of electrokinetic remediation with various electrolyte compositions for decontaminating sediments from Camorim Lagoon, Rio de Janeiro. While complexing agents (EDTA and humic acid) were expected to enhance the removal of zinc, lead, copper, and chromium, their effectiveness varied across the tested metals.
- The EDTA used in treatments EK2 and EK3 promoted the removal of zinc
 and lead from the sediment. However, using EDTA as the anolyte also avoids
 the accumulation of these elements in the anode section. Thus, treatment
 EK3 resulted in a more uniform reduction in the potentially toxic elements
 concentration across the entire sediment sample. This suggests that EDTA
 effectively mobilized these metals under the tested conditions.
- Interestingly, treatment EK1 (distilled water) exhibited the lowest overall
 nickel concentration compared to other treatments, despite a tendency for
 nickel accumulation in the cathodic section. Likely due to soluble metal
 hydroxides forming at the low pH near the anode, facilitating transport by
 electromigration.

Under the employed conditions, complexing agents (EDTA and humic acid) were not effective in removing copper and chromium from the sediment. Distilled water (EK1) resulted in the lowest concentration of these metals, suggesting that the low pH environment near the anode likely induces the formation of soluble metal hydroxides. These charged species can then be transported more readily through the sediment by electromigration, a process driven by the electric field.

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