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Assessing the influence of particle size and dissolved organic carbon on heavy metal leaching from construction and demolition waste modified with carbon-rich materials

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ABSTRACT

Construction and demolition waste (CDW) represents a substantial proportion of the overall waste generated in both developed and developing countries, attracting attention due to its large volume, reuse potential, and environmental risks. This study aims to investigate the impact of biochar and activated carbon on the leaching characteristics of copper (Cu) and nickel (Ni) from CDW towards groundwater resources. This research investigates, for the first time, the impact of dissolved organic carbon (DOC) on the mobilization and release of Cu and Ni in biochar-amended CDW. The leaching behavior of Cu and Ni from CDW was assessed using the column leaching test. Activated carbon and wood-derived biochar, produced at temperatures up to 750°C, were incorporated into the columns and applied to CDW at two different rates. Column leachates collected at different intervals were analyzed for Cu and Ni using inductively coupled plasma-mass spectrometry (ICP-MS). During the experiment, the concentrations of Cu and Ni in the effluents of the unamended column decreased by 94.67% and 73.68%, respectively. Applying 2% activated carbon to CDW reduced Cu and Ni leachate release by 30.82% and 33.33%, respectively, compared to 2% pulverized biochar. The results indicated high initial mobilization followed by a rapid decline in leached concentrations of the heavy metals in all treatments. The application of pulverized biochar and activated carbon significantly reduced leaching and cumulative release of heavy metals from CDW into water, whereas crushed biochar elevated mobilization and leached concentrations of Cu and Ni. A positive correlation was obtained between the leached concentrations of Cu and Ni from CDW with DOC. The results indicated that biochar could serve as a promising and cost-effective carbon-rich amendment for immobilizing Cu and Ni within CDW; however, biochar particle size played a critical role in controlling the mobilization and release of heavy metals in CDW.

1. Introduction

Heavy metals have been recognized as a serious threat to the environment and food security due to the rapid growth of industrial and agricultural activities, overpopulation, and the disruption of natural ecosystems [1]. It is reported that more

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than 10 million sites with an area of more than 20 million ha are contaminated with various contaminants worldwide, with over 50% of the sites contaminated with heavy metal(loid)s [2, 3]. Main causes of soil contamination with heavy metals include, but are not limited to, metal plating and finishing operations, manure, biosolids, sewage sludge, industrial wastewater, metal mining and smelting, fossil fuel combustion, pesticides, herbicides, fertilizers, and waste dumping [4]. CDW is being increasingly recognized as a significant contributor to groundwater contamination when not appropriately managed, with a heightened potential risk in upcoming years. A diverse array of components is present in CDW. Among these, the most commonly found materials include rocks, bricks, tiles, concrete, stones, gypsum plaster, bituminous materials, and other mineral composite materials. Heavy metals found in CDW frequently originate from paints, plumbing, and electrical components, whereas organic carbon may come from decomposed wood and other organic construction materials. These constituents represent notable environmental hazards, especially regarding groundwater contamination [5, 6].

CDW stands out as the primary component of solid waste generated in both developed and developing countries, with a more pronounced growth rate observed in developing nations [6]. For instance, Iran has experienced escalating urbanization, population expansion, and construction activities in recent decades, leading to a surge in CDW generation. In Tehran, the capital of Iran, around 50,000 tons of CDW are generated daily, with a substantial amount directed to unregulated landfills and dumping sites. Moreover, 5,000 to 10,000 tons undergo processing daily for potential reutilization [7], underscoring significant opportunities for CDW reuse in the upcoming years. However, it's crucial to prevent adverse environmental impacts on subsoil and groundwater resources when a reuse strategy is opted for CDW. For instance, the release of elevated concentrations of contaminants from CDW into groundwater resources must be strictly controlled. Until now, certain studies have concentrated on the environmental hazards linked to the discharge of heavy metals from soil [8], while scant consideration has been given to the risks associated with the release of heavy metals from CDW, and only limited data is accessible regarding the leaching and release of heavy metals from CDW.

One of the primary mechanisms for controlling the migration of contaminants from the solid phase to groundwater is their sorption onto solid phase materials. Typically, inorganic contaminants such as heavy metals exhibit less affinity to the organic fraction of soil in contrast to organic compounds, primarily due to their lower hydrophobicity and higher bioavailability in the solid phase [9]. For example, in a study conducted by Wang et al. (2024), over 50% of Cu and Pb were mobilized and released from contaminated soil into the aqueous phase [10]. In contrast, the leaching of persistent organic pollutants from soil was found to be much lower [11], implying serious groundwater risk associated with heavy metals present in a solid matrix. The inclusion of carbonaceous materials like biochar and activated carbon could modify the affinity of heavy metals within a solid matrix by altering their bioavailability, consequently impacting their leaching characteristics, which will be addressed in this study. Biochar is a carbon-rich product created by heating organic biomass under oxygen-limited conditions. While it is frequently used as a soil additive to improve fertility [12], biochar has recently garnered attention for its environmental remediation capabilities. The literature has reported a decrease in the bioavailability of heavy metals in both soil [13] and sediment [14] with the addition of activated carbon. However, according to the reviewed literature, the application of activated carbon, particularly wood-derived biochar, to mitigate the mobility and leaching of heavy metals in CDW has been limitedly investigated and is the focus of this study. The use of organic amendments to stabilize a solid matrix may also influence the leaching behavior of DOC, a factor often overlooked. Greater values of leached concentrations of heavy metals associated with higher amounts of mobilized DOC in soil have been reported in the literature [15]. The main aims of this study include: (i) investigating the impact of biochar application, including its particle size, on the leaching and release of selected heavy metals from construction and demolition waste; (ii) comparing the performance of activated carbon and biochar in the modification of the leaching behavior of heavy metals within the CDW matrix, and (iii) assessing the impact of dissolved organic carbon on the mobilization of heavy metals in CDW across different amendment conditions.

2. Materials and Methods

2.1. Construction and demolition waste, biochar, and activated carbon

The CDW used in this study was prepared by the Federal Institute for Materials Research and Testing in Berlin, Germany. The CDW exhibited significant variability in the quantities of its various constituents, reflecting its heterogeneous nature. The materials composing the CDW consisted primarily of asphalt (1.1% w/w), brick (17.8% w/w), concrete (63.2% w/w), solid rock (13.1% w/w), gravel (4.6% w/w), and miscellaneous materials, such as wood, plastics, paper, rubber, and textiles (0.2% w/w). A reference CDW sample was sieved using a 2 mm sieve to eliminate large debris. The forest wood-derived biochar utilized in this study was derived from forest wood and contained 78.3% organic carbon with a bulk density of 1.48 g cm^{-3}. The wood chips were air-dried and placed in open crucibles. Then, they were weighed and completely covered with aluminum foil to create an oxygen-limited environment. The biochar was produced at temperatures of up to 750°C, with a temperature gradient of approximately 10°C min^{-1}, until reaching the desired pyrolysis temperature of 750 ± 5°C in a muffle furnace operating at atmospheric pressure, with a residence time of 42 minutes. The resulting chips were left in the furnace overnight to cool to room temperature.

The obtained biochar chips were originally in granular form with a wide range of particle sizes. In order to investigate the impact of biochar particle size on the leaching behavior of heavy metals, biochar was applied to CDW in two forms: crushed and pulverized. The produced biochar chips were air-dried over a week and ground using a ceramic mortar and pestle. The obtained biochar was further milled using a zirconium oxide planet ball mill and sieved to 63-75 μm diameter to yield fine-graded biochar to be used as pulverized wood-derived biochar in this study. Some other biochar chips were only ground and sieved to 1-2 mm diameter and mixed thoroughly to gain homogenous crushed wood-derived biochar and did not undergo the milling process before applying to CDW. An elemental analyzer was employed to assess the carbon (C), hydrogen (H), and nitrogen (N) composition of the produced biochar. A suspension of the <0.25 mm fraction of the biochar and deionized water, with a solid to liquid ratio of 1:5, was agitated for 24 hours [16]. Subsequently, the pH value of the resulting slurry was measured using a calibrated pH meter. Some physico-chemical characteristics of the produced wood-derived biochar are presented in Table 1. Filtrasorb TL 830 granular activated carbon (GAC) purchased from Chemviron carbon containing 890 mg/g (89%) organic carbon was used in this research.

Table 1. Physico-chemical characteristics of the wood-derived biochar.

Biochar Source	Max. Pyrolysis Temp. (°C)	Pyrolysis Time (min)	Bulk Density (g cm^{-3})	pH	N (%)	H (%)	C (%)	Ash Content (%)
Forest Wood chips	750 ± 5	42	1.48	9.3	0.6	3.43	78.3	3.1

2.2. Experimental setup for column leaching tests

Columns were filled with CDW and its mixture with organic amendments in the following order: Column S1: 100% CDW, Column S2: 98% CDW and 2% pulverized biochar (w/w %), Column S3: 96% CDW and 4% pulverized biochar (w/w %), Column S4: 98% CDW and 2% crushed biochar (w/w %), Column S5: 96% CDW and 4% crushed biochar (w/w %), Column S6: 98% CDW and 2% activated carbon (w/w %), and Column S7: 96% CDW and 4% activated carbon (w/w %). A separate column filled with clean quartz sand alone was also prepared and operated as a control to ensure the absence of background contamination with heavy metals. Leaching studies were conducted using small glass columns with an inner diameter of 6 cm. These columns were filled with CDW, as well as a mixture of CDW and biochar, arranged in five sub-layers up to a height of 12 cm. The experimental setup for the column tests is detailed in Table 2. A layer of one centimeter of clean quartz sand was placed at the top and bottom of the columns to facilitate uniform water flow and prevent the loss of soil and biochar particles. The leaching tests were conducted following the guidelines outlined in DIN 19528 (2009) [17]. The German standard DIN 19528 is considered the most advanced leaching test method [18].

The experiments were conducted in a room with a constant temperature of 20°C. Given that the study aimed to investigate groundwater contamination risk under a worst-case scenario, the columns were operated under saturated conditions with upward flow. This approach ensured continuous saturation throughout the leaching process and mitigated the risk of air bubble entrapment. MilliQ water, stored in a 50-liter vessel, was used as the leaching agent to avoid interference with potential non-target ions and compounds in the water. Water was pumped into the columns using a multi-channel peristaltic pump at a constant flow rate (Table 2). The columns were initially saturated with deionized water at a higher flow rate. The tests commenced no later than one hour after the completion of saturation.

Table 2. Leaching test experimental set-up.

Parameter	Description
LS^{-1} ratio	Increasing from 0.25 to 4 L kg^{-1}
Column inner diameter	6 cm
Filling height	12 cm
Flow rate	0.4 mL min^{-1}, up-flow percolation
Leachant	MilliQwater (Millipoer, MA, USA)
Ambient temperature	20±1°C
Contact time	5 h
Leaching test procedure	DIN 19528 (2009) [17]

2.3. Analysis of column percolates for heavy metals and other parameters

Column percolates were collected in dark glass bottles by connecting the columns to the bottles using a stainless-steel tube. Collection bottles containing eluate fractions were changed at specified liquid-to-solid (LS) ratios up to 4 L kg^{-1}. Column effluents were analyzed for the selected heavy metals, i.e., Cr, Cu, and Ni, in this study. Leachate samples were characterized according to the Standard Method for the Examination of Water and Wastewater. 100 ml of column percolates at certain LS ratios were filtered, digested with nitric acid, and then analyzed for the concentrations of Cr, Cu, and Ni using inductively coupled plasma-mass spectrometry (ICP-MS). Measurements were performed in triplicate, and the average values were reported [19, 20]. 30 mL of the collected eluates were collected in 50 mL dark glass bottles to be analyzed for other parameters, i.e., pH dissolved organic carbon. Results of the tests were expressed as a function of LS ratio in terms of mg or µg of a given compound measured per liter of leachate. Dissolved organic carbon (DOC) values were measured within 24 hours after sampling. Each sample was filtered through 0.45 µm filtration membranes (MILEXHA) before being analyzed for DOC by a HighTOC analyzer (Elementar).

3. Results and Discussion

3.1. Leaching and release behavior of Cu and Ni from Construction and Demolition Waste across different leaching scenarios

Redirecting CDW away from landfill sites and shifting towards reuse and recycling strategies can alleviate the demand for landfill space, conserve finances, and preserve natural resources [21]. However, it's imperative to assess their potential ramifications, especially on groundwater resources, and minimize any potential adverse impacts. This study examines the leaching and release patterns of copper (Cu) and nickel (Ni), the prevalent heavy metals in the composition of CDW. Figure 1 illustrates the concentrations of Cu and Ni in the effluent from Column S1, filled solely with CDW. The concentrations of heavy metals are depicted at various liquid-to-solid ratios. The LS ratio is primarily employed in column leaching test studies to enable comparisons of findings across different leaching tests. This approach facilitates comparison across different leaching tests, such as batch and column experiments, as well as tests conducted at different scales. The LS ratio represents the volume of water passed through the column after a certain time relative to the dry weight of the solids in the column and can be calculated as the quantity of percolated water in relation to the solids' dry weight [17, 22].

High initial concentrations of Cu and Ni were observed in the unamended CDW for Column S1. However, a rapid decrease in heavy metal concentrations in the demolition waste matrix was followed by an extended tailing period for the studied heavy metals, namely copper and nickel. The decrease in Ni concentration in the effluent from Column S1 was less pronounced compared to Cu. Throughout the experiment, the concentrations of Cu and Ni in the effluents of Column S1 decreased by 94.67% and 73.68%, respectively (Figure 1), implying a higher mobilization of Cu in CDW compared to Ni.

It is commonly understood that water solubility plays a crucial role in controlling the mobilization of compounds in the solid phase. However, other factors, such as complexation with DOC and colloidal matter within the solid matrix, as well as fluctuations in pH during the leaching process, may influence leaching behavior and impact the release kinetics of a particular compound. The results of this research revealed high initial mobility and leaching of Cu and Ni. Susset and Grathwohl (2011) proposed that contaminants displaying a rapid initial decrease in leached concentrations could be classified as "finite" [22]. This aspect should be considered in groundwater risk assessment, as the elevated concentrations may only be a temporary phenomenon [23]. The significant initial mobilization of heavy metals within CDW, followed by a rapid decline in their leached concentrations, suggests a rapid depletion of heavy metals within the construction and demolition waste matrix. Higher concentrations of leached Cu were observed throughout the leaching process compared to the leached amounts of Ni, which might also be attributed to the higher content of Cu in the solid phase.

The leaching behavior of Cu and Ni from organically amended CDW is also depicted in Figure 1. The results indicated that the leaching of heavy metals followed a similar trend in both unamended and amended CDW. However, the addition of 2% pulverized biochar to CDW slightly increased the initial mobilization of Ni compared to the unamended treatment. Furthermore, the application of 4% pulverized biochar resulted in increased initial leached concentrations of both studied heavy metals compared to Column S1. The increase in pulverized biochar content was associated with a higher initial mobilization of Cu and Ni within CDW. The initial concentrations of Cu and Ni in effluents from Columns S2 and S3 showed rapid declines over time, with prolonged tailing observed at higher LS ratios.

The comparable effectiveness of higher levels of pulverized biochar in reducing the leaching of heavy metals may be due to the saturation of biochar's adsorption capacity at these higher levels, resulting in diminished additional benefits [24]. The addition of pulverized biochar to CDW led to a reduction in the leaching of Cu during the later stages of the leaching process (at LS ratios exceeding 0.5 L kg^{-1}) compared to the unamended treatment (Figure 1). These findings suggest that the decrease in Ni concentration in leachates from pulverized biochar-amended CDW was not as rapid as the values obtained for Cu. Previous studies have reported enhanced immobilization of heavy metals in contaminated soils using organic amendments [13]. The Ni concentration decreased by 75% and 87.5%, respectively, throughout the experiment following the incorporation of 2% and 4% biochar (Figure 1).

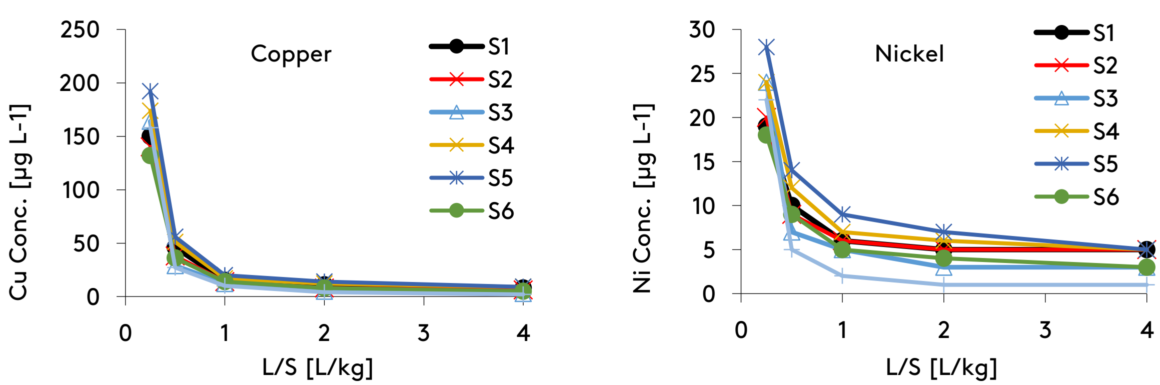


Fig. 1. Leaching of copper (Cu) and nickel (Ni) from CDW across different leaching scenarios.

The lack of reduction in the cumulative leaching of Ni from Column S2, which was amended with 2% biochar compared to S1 (Figure 2), could be due to several probable factors. A 2% application rate might be insufficient to significantly influence the leaching of Ni. Higher concentrations of biochar might be required to observe a noticeable impact, as observed in the presence of 4% biochar. Nickel and copper both form complexes with solid-phase organic matter, but Ni typically has a higher coordination number, allowing it to form more complex and stable interactions. With its lower coordination number, Cu tends to form fewer and less stable complexes. These differences affect their mobility and bioavailability in the solid phase, with Ni often binding more strongly to organic matter that potentially reduces its leachability compared to Cu in the presence of lower amounts of biochar in the solid phase, as observed in this study. The CDW physical and chemical characteristics, such as pH, organic matter content, and existing metal-binding sites, could also influence how effectively biochar interacts with Ni at lower applied rates. In addition, the quality and type of biochar used might affect its performance. Not all biochars have the same adsorption capacities or efficiencies for different heavy metals.

The comparison of cumulative release of Cu and Ni across S1, S2, and S3 treatments suggested that amending CDW with pulverized biochar could effectively reduce their mobilization and release into the aqueous phase (Figure 2), with a greater impact observed when a higher percentage of pulverized biochar was utilized. With the addition of 4% pulverized biochar to CDW, there was a 40% reduction in the cumulative leaching of Cu; the addition of 2% pulverized biochar to CDW resulted in a 16.6% decrease in the cumulative leaching of Cu. Overall, incorporating pulverized biochar into CDW had the greatest impact on the cumulative release of Cu, followed by Ni. The effectiveness of biochar in reducing heavy metal leaching can vary depending on factors such as biochar feedstock, pyrolysis temperature, application rate, and solid phase properties [25]. These findings suggested that pulverized biochar showed promise as an effective amendment for mitigating the release of the studied heavy metals in contaminated porous media, such as CDW.

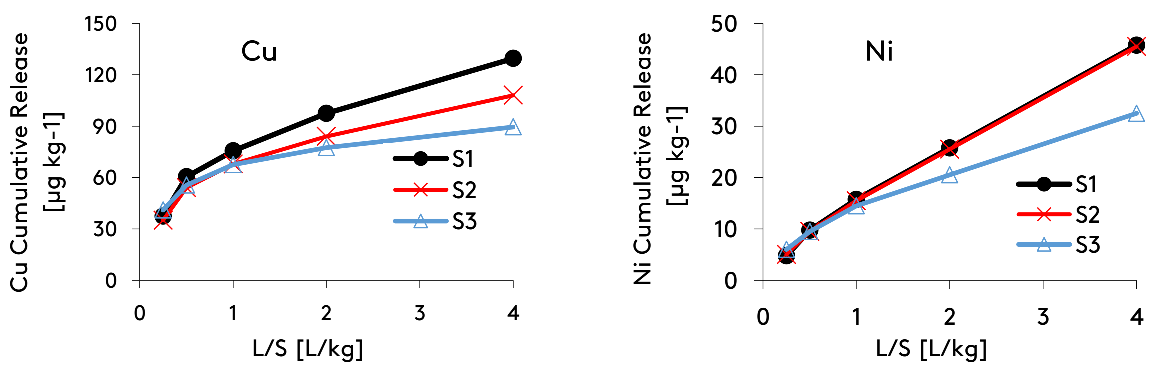


Fig. 2. Accumulative leaching of copper (Cu) and nickel (Ni) from CDW (S1) and CDW amended with 2% (S2) and 4% (S3) pulverized biochar.

3.2. Effect of biochar particle size on mobilization and leaching of Cu and Ni from CWB

A significant difference was observed between the performance of crushed and pulverized biochar in immobilizing Cu and Ni in CDW, highlighting the crucial role of biochar particle size in controlling the fate of heavy metals in CDW. Results of the present study revealed a significant influence of biochar particle size on the mobilization and leaching of both the studied heavy metals in CDW. For instance, the results demonstrated that reducing biochar particle size enhanced its performance significantly in stabilizing Cu in CDW and reduced its cumulative release into water by 35.18% and 44.06% at biochar application rates of 2% and 4%, respectively (Figure 3). The mobilization and release behavior of Cu in the presence of both crushed and pulverized biochar was comparable to that of Ni. In other words, the addition of crushed biochar increased the cumulative release of all three heavy metals, whereas the use of pulverized biochar decreased their mobilization and release throughout the leaching process.

Analysis of the data suggests that the cumulative concentration of Ni increased by 24.59%, compared to unamended treatment, when 4% crushed biochar was introduced at an LS ratio of 4 L kg $^{-1}$. Conversely, the addition of 4% pulverized biochar to CDW resulted in a reduction of cumulative Ni concentration by 28.96% compared to unamended CDW. This trend was consistent across similar observations for cumulative concentrations of Cu in the study. Results of the present study showed higher effectiveness of the higher dosage of pulverized biochar in promoting immobilization of heavy metals in CDW both for Cu and Ni (Figure 3). The heterogeneous distribution of crushed biochar in the columns and the insufficient contact between crushed biochar particles and heavy metals likely contributed to the lower immobilization of heavy metals in CDW amended with crushed biochar, compared to CDW amended with pulverized biochar. This resulted in higher solute concentrations in the aqueous phase when crushed biochar was used. Pulverizing the biochar to reduce its particle size could lead to a more homogeneous distribution within the CDW along the length of the column, thereby providing more effective contact and a more stable impact on the leaching concentrations of heavy metals. Studies have shown that decreasing the particle size of materials such as zeolites, clay minerals, and iron-based sorbents can lead to increased sorption capacities for heavy metals like lead, cadmium, and zinc. This enhanced sorption performance is attributed to the larger surface area and higher reactivity of finer particles, which provide more active sites for heavy metal sorption [26]. Additionally, reducing biochar particle size has been shown to enhance its sorption capacity for heavy metals like zinc in contaminated soil, attributed to increased surface area and reactivity [27].

Biochar was found to be able to attenuate the leaching of heavy metals by adsorbing them onto its surface, thereby diminishing their mobility and availability for leaching [25, 28]. This observation is consistent with the findings of the present study when pulverized biochar was utilized (Columns S2 and S3). However, the incorporation of crushed biochar in CDW notably enhanced the leaching of Cu and Ni compared to unamended CDW. This highlights the significance of considering biochar particle size as a crucial factor influencing its efficacy in mobilizing heavy metals within the CDW. Ensuring adequate contact between the biochar and heavy metals in the CDW appears necessary for achieving efficient adsorption onto solid particles, thereby reducing their mobility and leaching potential. Further investigation is recommended into the impact of biochar particle size and its distribution in the solid phase on the behavior and fate of heavy metals in construction and demolition waste. The results also indicated that using pulverized biochar resulted in markedly lower peak levels of DOC concentration in the column effluents, whereas the incorporation of crushed biochar resulted in an elevation of DOC concentration compared to the unamended treatment. The application of biochar to soil has been reported to influence DOC content by modifying organic matter decomposition rates and the structure of the solid phase. This can enhance soil carbon sequestration by stabilizing organic matter, thus potentially increasing DOC levels. Conversely, biochar can also adsorb and immobilize DOC through surface interactions, leading to a reduction in DOC content [29]. The influence of biochar on the DOC content of CDW may vary depending on biochar particle size and its distribution within the CDW. Further studies are suggested to explore the influence of biochar particle size and its distribution within construction and demolition waste on the mobilization and leaching behavior of heavy metals, as well as dissolved organic carbon, to provide deeper insights into these interactions and their real-world implications.

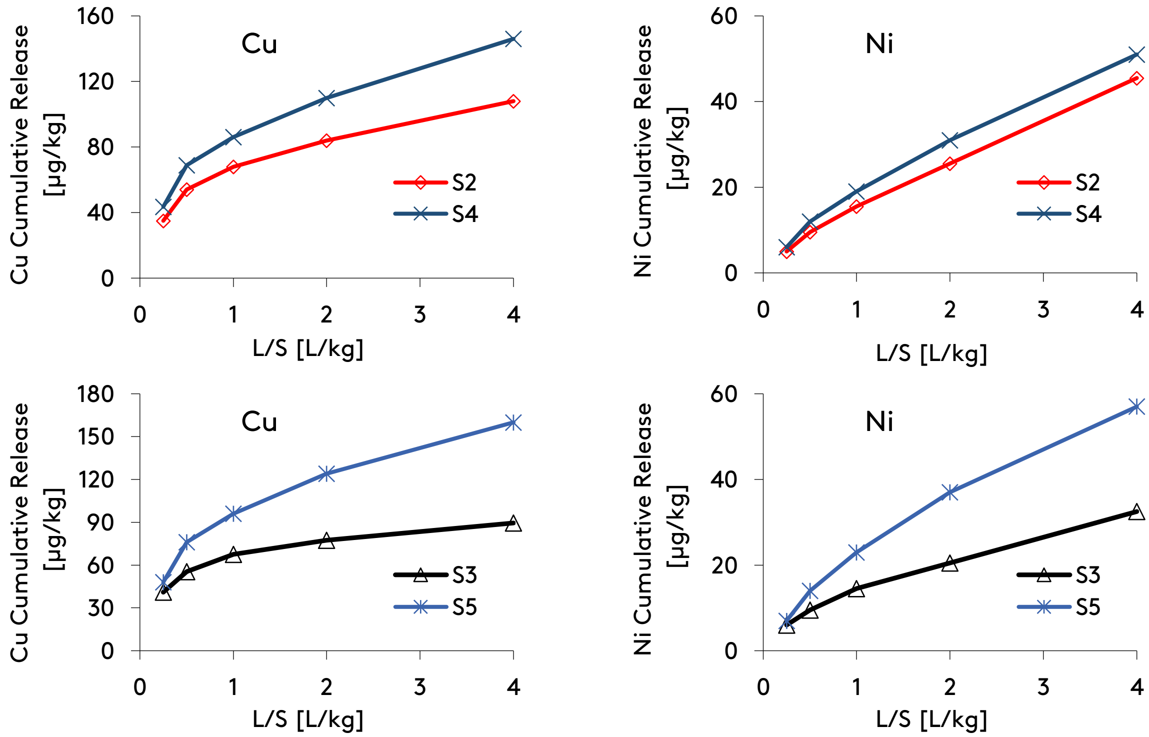


Fig. 3. Influence of biochar particle size on cumulative release of copper (Co) and nickel (Ni) from CDW (S2: CDW amended with 2% pulverized biochar, S3: CDW amended with 4% pulverized biochar, S4: CDW amended with 2% crushed biochar, and S5: CDW amended with 4% crushed biochar).

3.3. Comparison of performance of activated carbon and biochar in modification of leaching characteristics of Cu and Ni in construction and demolition waste

Activated carbon has been extensively used for environmental remediation purposes due to its large specific surface area, rich pore structure, high stability, and reasonable price [13]. Adding 2% granular activated carbon to CDW reduced leached concentrations of Cu and Ni compared to the unamended treatment across all the LS ratios examined. A similar trend was found for the treatment that received 4% granular activated carbon, except leached concentrations of Cu and Ni at an LS ratio of 0.25 L kg $^{-1}$, which was higher than those of unamended CDW (Figure 1). Greater immobilization of Cu and Ni in CDW was observed in the presence of 4% activated carbon compared to 2% activated carbon. For instance, the application of 4% activated carbon reduced Cu and Ni concentrations in the water phase by 60% and 66.67%, respectively, compared to the treatment amended with 2% activated carbon at an LS ratio of 4 L kg $^{-1}$.

The organic carbon content of an organic amendment significantly influences the immobilization of heavy metals in soil. For instance, incorporating municipal waste compost with 54% organic carbon content resulted in the release of approximately 4% of copper and more than 58.3% of zinc from the soil into groundwater [30], which was not favorable in the context of environmental remediation and poses a threat to groundwater quality. Conversely, the effectiveness of activated carbon in altering the leaching characteristics of heavy metals in sediments was demonstrated by Ting et al. (2018) [31]. Their research revealed that incorporating activated carbon decreased the leaching of mercury (Hg) significantly, showing the highest reduction rate when 3% activated carbon was added to the soil. This underscores the potential of activated carbon for applications in soil remediation.

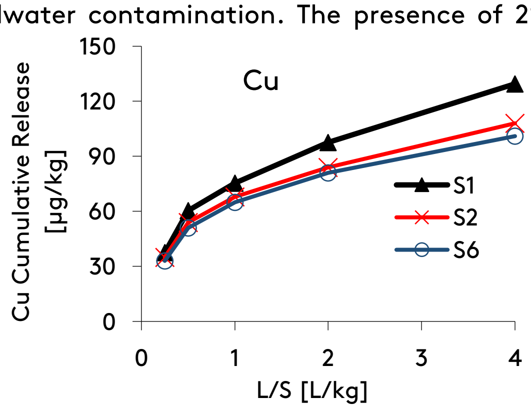
Figure 4 illustrates the enhanced immobilization of the studied heavy metals within CDW when both pulverized biochar and activated carbon are present, with activated carbon demonstrating a slightly stronger effect at both 2% and 4%

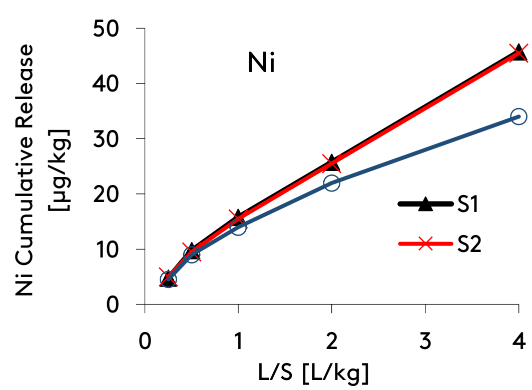
application rates. The immobilization effect of crushed biochar was below activated carbon; therefore, only the effectiveness of pulverized biochar and activated carbon has been compared in Figure 4. The immobilization effect of crushed biochar was far less than that of pulverized biochar and activated carbon. Thus, Figure 4 specifically compares the effectiveness of pulverized biochar and activated carbon to each other and in relation to unamended CDW. The results of the present investigation indicated that a higher dosage of activated carbon was more effective in reducing the mobilization and leaching of heavy metals from CDW, yielding a comparable result to the effects observed with pulverized biochar. Activated carbon demonstrated higher efficiency in reducing the cumulative release of Cu and Ni in CDW compared to pulverized biochar-amended CDW (Figure 4). Activated carbon likely exhibited greater efficiency due to its higher surface area, well-defined pore structure, superior adsorption capacity compared to biochar, and specific chemical properties of activated carbon, such as its surface chemistry and functional groups [32, 33]. For instance, it could be inferred from Figure 4 that applying 2% activated carbon to CDW resulted in a 30.82% and 33.33% reduction in the cumulative release of Cu and Ni into leachates, respectively, compared to applying 2% pulverized biochar over the course of the leaching process.

Applying carbon-rich materials like biochar and activated carbon can improve the immobilization of contaminants within the contaminated solid matrix such as CDW. This process reduces contaminants' bioavailability, favorably stabilizing them in the solid phase and mitigating the risk of groundwater contamination. The presence of 2% activated carbon in CDW resulted in a decrease of 22.01% in the accumulated release of Cu and 25.68% in Ni into the water phase during the leaching process, compared to unamended CDW. Furthermore, incorporating higher levels of activated carbon, i.e., 4% in CDW, significantly lowered the cumulative release of Cu and Ni by 38.99% and 65.03%, respectively, compared to S1 (Figure 4). Previous studies have documented the effectiveness of activated carbon in reducing the mobility of heavy metals in soil [34]. Overall, the present research findings suggested that pulverized wood-derived biochar could serve as a cost-effective alternative to activated carbon for immobilizing heavy metals within construction and demolition waste.

3.3. Contribution of dissolved organic carbon on mobilization of Cu and Ni in construction and demolition waste

DOC measurements were performed on percolates from both amended and unamended columns at specified LS ratios to investigate the leaching behavior of DOC from construction and demolition waste, as well as its possible role in the mobilization and leaching of heavy metals. The interaction between heavy metals and DOC during leaching from porous media has received notable attention in recent years. DOC can act as a carrier for compounds present in the solid phase, potentially increasing their mobilization and transport towards groundwater [35]. The highest levels of DOC were measured during the initial stages of the leaching process, i.e., at LS ratios of 0.25 L kg $^{-1}$, followed by LS ratio of 0.5 L kg $^{-1}$ in Column S1, which was solely filled with CDW (Figure 5).





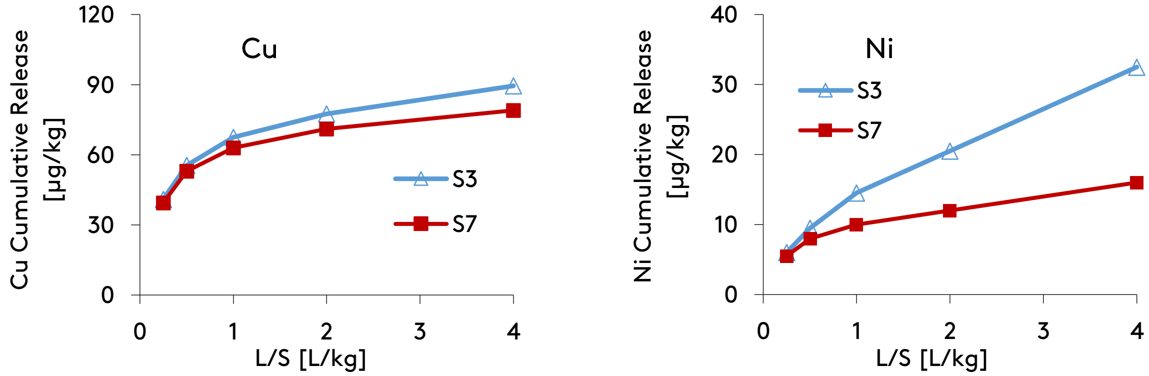


Fig. 4. Comparison of cumulative leaching and release of copper (Co) and nickel (Ni) from CDW in presence of pulverized biochar and activated carbon in CDW (S2: CDW amended with 2% pulverized biochar, S3: CDW amended with 4% pulverized biochar, S6: CDW amended with 2% activated carbon, and S7: CDW amended with 4% activated carbon).

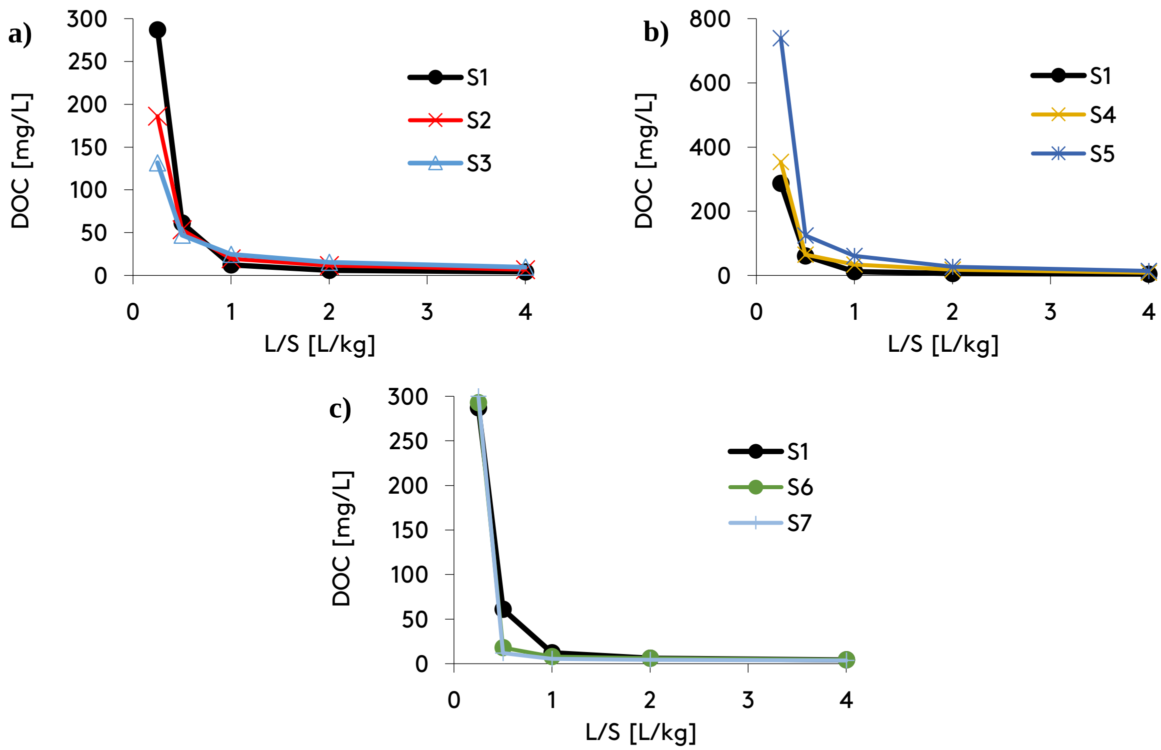


Fig. 5. Leaching behavior of Dissolved Organic Carbon from CDW in presence of 2% and 4% pulverized biochar (a), crushed biochar (b) and activated carbon (c).

Applying 2% pulverized biochar to CDW reduced the initial mobilization of DOC compared to unamended CDW. Further reductions were achieved with higher levels of pulverized biochar. Specifically, adding 4% pulverized biochar to CDW decreased the initial leaching of DOC from 287 mg L-1 to 131.7 mg L-1 at an LS ratio of 0.25 L kg -1, marking a reduction of 54.11%. However, higher levels of DOC were detected at higher LS ratios in pulverized biochar-amended CDW compared to Column S1 (Figure 5), implying the influence of the higher solid-phase content of the organic fraction in biochar-amended CDW.

A rapid initial decline in DOC content was observed in the percolates of both amended and unamended CDW, followed by a prolonged tailing phase, as depicted in Figure 5. The depletion of highly soluble organic carbon resources in CDW likely contributed to the rapid reduction in DOC concentrations in the percolates. However, adding pulverized biochar to CDW led to an increase in detected DOC content in the effluents of the columns at higher LS ratios. For instance, adding 2% and 4% pulverized biochar to CDW resulted in increases in the DOC contents of the leachates in later stages of the leaching process, specifically by 62.47% and 128.03%, respectively, at an LS ratio of 4 L kg^{-1}. The obtained results suggested that the mobilization of the studied heavy metals through the leaching of dissolved organic colloids was more likely to occur during the initial stages of a leaching process, regardless of the presence or absence of organic amendments in the CDW.

Most metal cations exhibit limited solubility in water primarily due to their sorption onto mineral grains and organic matter. The presence of DOC can enhance the solubility and mobility of heavy metals by forming organic-metal complexes, thereby influencing their transport and fate [18]. Therefore, the rapid decrease in copper and nickel concentrations may be correlated with the decline in DOC concentration. Moreover, DOC can impact the sorption and desorption processes of heavy metals on solid phase particles, affecting their availability for leaching. Adding crushed biochar to CDW increased the leaching of DOC at all tested LS ratios in this experiment, compared to unamended CDW (Figure 5b). For instance, the incorporation of 2% and 4% crushed biochar into CDW increased the leached amounts of DOC by 103.09% and 234.92%, respectively, at an LS ratio of 4 L kg^{-1}, suggesting the contribution of DOC-facilitated transport in the higher leached amounts of Cu and Ni in crushed biochar-amended treatments compared to the unamended CDW.

As time passed, DOC leaching from the amended soil with crushed biochar declined significantly from an LS ratio of 0.25 L kg^{-1} to 4 L kg^{-1}. Throughout the percolation process, the leaching of DOC from CDW decreased by 97.58% and 98.09% in the presence of 2% and 4% crushed biochar, respectively. The obtained results indicated that the increase in leaching of the studied heavy metals in the presence of crushed biochar in CDW was likely associated with the leaching of dissolved organic compounds throughout the leaching process, particularly at the initial stages, which was comparable with the trend observed in the presence of pulverized biochar. However, the application of crushed biochar to CDW may exert an inhibitory effect on the mobilization of DOC and its associated heavy metals at higher LS ratios. This could be attributed to the stabilization of the remaining dissolved carbon and its incorporation into the CDW matrix, thereby immobilizing the associated heavy metals. Dissolved organic matter (DOM) can impact the leaching of heavy metals in the soil through various mechanisms, including complexation, chelation, and formation of soluble complexes, affecting their mobility and bioavailability in the environment [36]. It has been suggested that DOM may elevate the solubility of heavy metals, facilitating their leaching from a solid matrix into groundwater resources, thereby posing risks to ecosystems and human health [37].

At LS ratios higher than 0.5 L kg^{-1}, the DOC content of leachates experienced a notable decrease when activated carbon was added to CDW compared to the unamended treatment. This indicated the strong sorption capacity of activated carbon, which inhibited the mobilization and leaching of dissolved organic matter in construction and demolition waste. This phenomenon was likely responsible for reducing the DOC-mediated transport of heavy metals in CDW. The use of pulverized biochar significantly reduced the leaching of DOC from CDW compared to unamended CDW. The addition of activated carbon and biochar, in crushed form, might contribute to an increased amount of initially leached heavy metals associated with DOC. However, activated carbon caused lower leaching of DOC from CDW compared to both crushed and pulverized biochar, especially at LS ratios higher than 0.5 L kg^{-1}. In this study, the decline in mobilization and release of Cu and Ni from CDW was found after adding pulverized biochar and activated carbon, while the application of crushed biochar elevated the mobilization and leaching of the heavy metals throughout the leaching process.

Additionally, the application of biochar in both forms slightly reduced the pH values in leached water from CDW, which may affect the solubility of heavy metals and alter their leaching behavior. The pH reduction was more notable with increasing concentrations of the organic amendment applied to the CDW in this study. Specifically, the effect of activated carbon exceeded that of pulverized biochar, which in turn surpassed the impact observed with crushed biochar. The rise in pH can alter the speciation of heavy metals, potentially decreasing their solubility and mobility in the solid phase, such as CDW, thereby reducing their leaching into groundwater [38, 39]. However, the precise impact of a particular biochar on heavy metal leaching depends on factors such as biochar type, application rate, characteristics of the solid phase, and properties of the heavy metals involved [40].

3.5. Correlation between Dissolved Organic Carbon (DOC) and heavy metal leaching from Construction and Demolition Waste

Figures 6 and 7 depict the correlation between the leached concentrations of DOC and the leached amounts of copper (Cu) and nickel (Ni) across various treatments investigated in this study. Although directly quantifying DOC associated with the release of heavy metals poses challenges, analyzing correlations and coefficients of determination (R^2) can offer indirect insights into the potential relationship between fluctuations in leached metal concentrations and DOC. DOC refers to organic molecules within aquatic systems with a molecular size smaller than 0.45 µm [18]. Some metals can form complexes with DOC, potentially changing the solubility of these compounds in its presence. As a result, these metals can be mobilized and released from CDW into water, bound to the leached DOC.

A positive correlation between the leached concentrations of Cu and Ni from CDW with DOC at corresponding LS ratios was consistently observed, suggesting that higher values of DOC might be linked to a greater likelihood of heavy metal mobilization in CDW. Pearson correlation coefficients (R) approaching +1 signify a robust positive correlation as a statistical measure indicating the strength of a linear relationship between paired data. In both unamended CDW and CDW amended with various levels of pulverized biochar, Pearson correlation coefficients (R) for Cu and Ni were found to be more than 0.99, indicating a highly significant statistical correlation [41], highlighting a remarkable dependency of Cu and Ni mobilization and leaching within CDW on leached concentrations of DOC. It can be inferred from Figures 6 and 7 that the Pearson correlation coefficient (R), used as an indicator of the correlation between leaching of DOC and leaching of Cu and Ni, remained relatively stable following the addition of 2% and 4% of biochar and activated carbon to CDW.

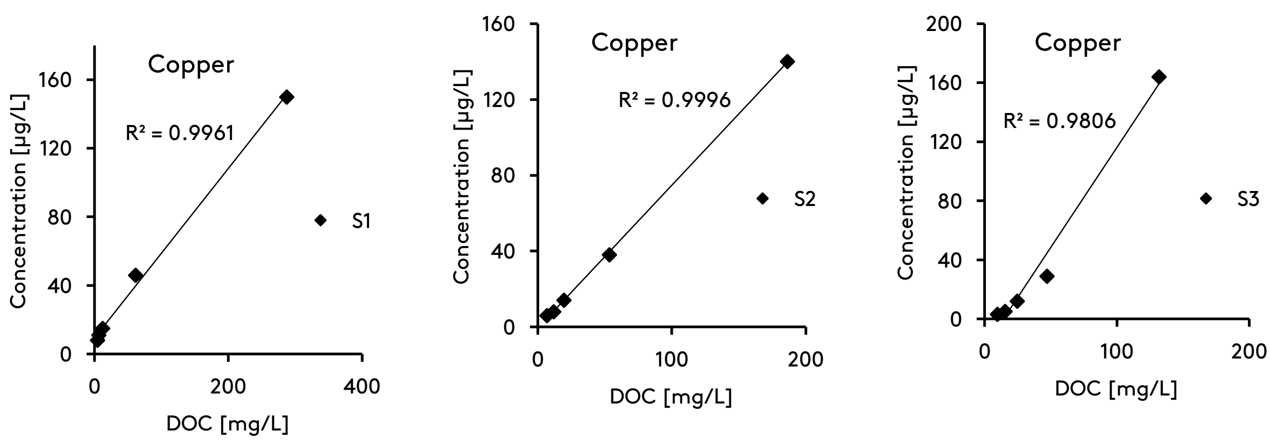


Fig. 6. Linear correlations between the leached quantities of copper (Cu) and dissolved organic carbon (DOC) from construction and demolition waste across various leaching scenarios.

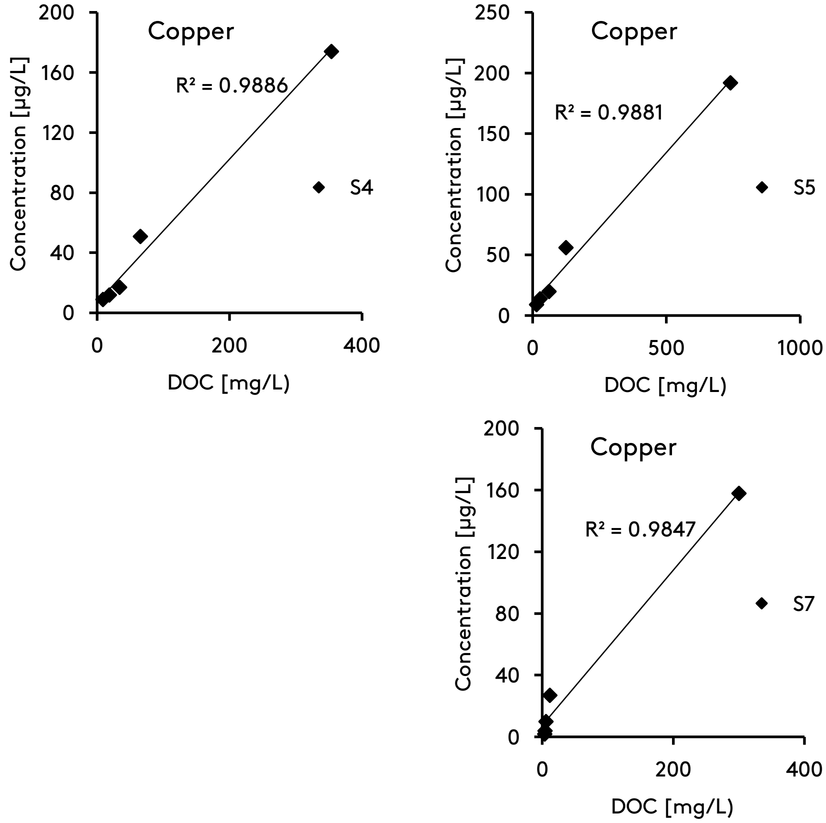


Fig. 6. (Continiued) Linear correlations between the leached quantities of copper (Cu) and dissolved organic carbon (DOC) from construction and demolition waste across various leaching scenarios.

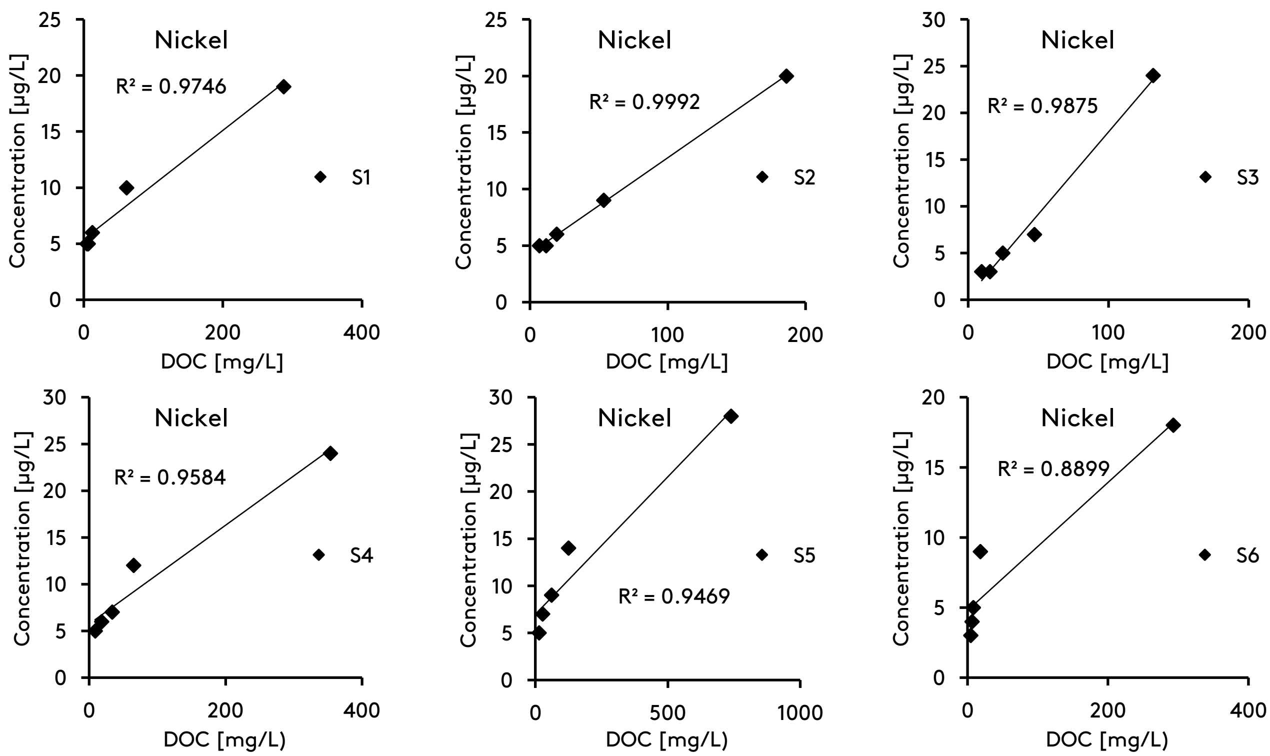


Fig. 7. Linear correlations between the leached quantities of nickel (Ni) and dissolved organic carbon (DOC) from construction and demolition waste across various leaching scenarios.

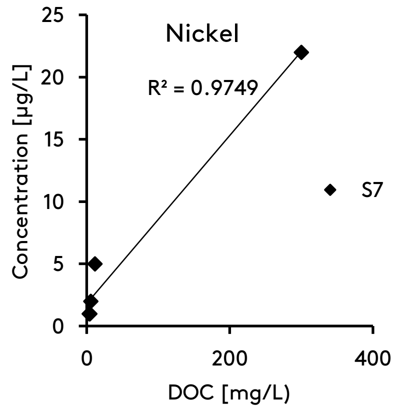


Fig. 7. (Continued) Linear correlations between the leached quantities of nickel (Ni) and dissolved organic carbon (DOC) from construction and demolition waste across various leaching scenarios.

In this study, the reduction in pulverized biochar content from 4% to 2% in CDW did not appear to markedly alter the role of DOC in the leaching of heavy metals, and the transport facilitated by DOC remained a significant mechanism for the mobilization and leaching of heavy metals. A positive correlation between the leached concentrations of Cu and Ni with DOC at corresponding LS ratios was also observed in CDW amended with crushed biochar and activated carbon, with slightly higher R^2 values obtained for Cu. Adding crushed biochar to CDW increased the leaching and cumulative release of Cu and Ni, with a more pronounced effect observed for 4% compared to 2%. However, the incorporation of crushed biochar to CDW did not change R^2 values compared to unamended treatment in terms of the correlation between leached concentrations of Cu and Ni with DOC (Figures 6 and 7). Increasing the crushed biochar content of CDW also resulted in a significant rise in the leaching of DOC, particularly with the higher percentage of biochar. Calculated R values remained fairly constant following the increase in crushed biochar content of CDW from 2% to 4%. A strong correlation between the leaching of the heavy metals and the leaching of DOC in the presence of 0, 2, and 4 percent crushed biochar in CDW was found in this study. Variations in biochar properties at different concentrations, such as surface chemistry, might influence the relative importance of DOC and colloids in heavy metal transport [42].

Initial leaching of DOC from CDW in the presence of 2% and 4% activated carbon increased slightly at an LS ratio of 0.25 L kg $^{-1}$, followed by reduced leaching of DOC at higher LS ratios. The application of 2% and 4% activated carbon decreased the mobilization and leaching of all studied heavy metals compared to unamended soil, with a greater impact observed for 4% activated carbon than 2%. Calculated R values remained fairly constant following the increase in activated carbon content of CDW from 2% to 4%. Adding activated carbon to CDW did not change R^2 values notably compared to the unamended treatment in terms of correlation between leached concentrations of Cu and Ni with DOC, except for a slight decrease observed for Ni in the S6 treatment. The R values calculated for Cu and Ni in activated carbon-amended CDW fell in the range of 0.94-0.99, suggesting significant dependence of mobilization and leaching of the heavy metals on the leaching of DOC in S6 and S7 treatments. Overall, a robust correlation between the leaching of Cu and Ni and the leaching of DOC was observed in both unamended CDW and CDW amended with biochar and activated carbon.

4. Conclusions

This study investigated the mobilization and leaching behavior of copper and nickel within construction and demolition waste using column leaching tests under various conditions. The influence of incorporating pulverized wood-derived biochar, crushed wood-derived biochar, and activated carbon on the leaching of heavy metals from CDW was also investigated in this research. The effect of biochar particle size on the leaching and release characteristics of heavy metals within CDW was also discovered. Additionally, the potential role of Dissolved Organic Carbon in the mobilization and release of these heavy metals from CDW was examined across different treatments. Unamended CDW in Column S1 exhibited high initial concentrations of heavy metals, followed by a rapid decrease with extended tailing, mirroring the trend observed in CDW amended with biochar. Column S1, filled solely with unamended CDW, showed initially high concentrations of leached heavy metals, followed by a rapid decline with prolonged tailing, similar to the trend observed in CDW amended with pulverized biochar (Columns S2 and S3), crushed biochar (Columns S4 and S5), and activated carbon (Columns S6 and S7). The application of biochar in two forms elevated the initial leached concentrations of both Cu and Ni compared to the unamended column. A comparison of the cumulative release of Cu and Ni among treatments indicated that amending CDW with activated carbon and pulverized biochar significantly reduced their mobilization and release into the aqueous phase. The results demonstrated that wood-derived biochar could serve as a promising and cost-effective alternative to activated carbon for immobilizing Cu and Ni within CDW, thereby effectively reducing their leaching into the aqueous phase when applied in pulverized form. The efficacy of biochar in reducing heavy metal leaching may vary based on various factors, including solid phase properties, feedstock, pyrolysis temperature, biochar particle size, and application rate. Excessive amounts of organic amendments may alter soil properties, further influencing the contaminants' fate and transport. Balancing the application of organic amendments in CDW to optimize remediation effectiveness while mitigating adverse effects on solid phase processes is crucial and should be further investigated in prospective research. The results also underscore the importance of ensuring adequate contact between the biochar particles and heavy metals within CDW to achieve effective immobilization and reduced leaching of heavy metals from the solid phase towards the aqueous phase, highlighting the critical role of opting for the proper particle size of biochar for stabilizing heavy metals in CDW. The results showed a higher effectiveness of the higher dosage of pulverized biochar in promoting the immobilization of heavy metals in CDW. In contrast, the application of 4% crushed biochar resulted in greater mobilization and release of both Cu and Ni compared to the application of 2% crushed biochar. Rapid declines in DOC content were observed in the percolates of both amended and unamended columns, followed by the extended tailing of DOC. Adding crushed biochar to CDW increased the leaching of DOC at all tested LS ratios compared to unamended CDW. A consistent positive correlation was found between the concentrations of leached Cu and Ni from CDW and DOC at corresponding LS ratios, suggesting that higher levels of DOC might enhance the mobilization of heavy metals in CDW. While construction and demolition waste offer remarkable potential for reuse, especially in road construction projects, it is essential to carefully manage its utilization to minimize the risk of contaminating groundwater.

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Declaration of competing interest

The author declares that he has no known competing financial interests or personal relationships that would influence this paper.

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