Nickel in soil and water: Sources, biogeochemistry, and remediation using biochar

Published in: *Journal of Hazardous Materials*

Citation for published version: El-Naggar, A., Ahmed, N., Mosa, A., Niazi, N.K., Yousaf, B., Sharma, A., Sarkar, B., Cai, Y., Chang, S.X., (2021) Nickel in soil and water: Sources, biogeochemistry, and remediation using biochar. *Journal of Hazardous Materials*. 419: 126421. doi: 10.1016/j.jhazmat.2021.126421.

Document version: Accepted peer-reviewed version.

Nickel in soil and water: Sources, biogeochemistry, and remediation using biochar

Ali El-Naggar[a,b], Naveed Ahmed[c], Ahmed Mosa[d], Nabeel Khan Niazi[e,f], Balal Yousaf [g], Anket Sharma[a, h], Binoy Sarkar[i], Yanjiang Cai[a], Scott X. Chang[a,j,*]

[a] *State Key Laboratory of Subtropical Silviculture, Zhejiang A&F University, Lin'an 311300, China*

[b] *Department of Soil Sciences, Faculty of Agriculture, Ain Shams University, Cairo 11241, Egypt*

[c] *U.S. Pakistan Center for Advanced Studies in Water, Mehran University of Engineering and Technology, Jamshoro, 76062, Sindh, Pakistan*

[d] *Soils Department, Faculty of Agriculture, Mansoura University, Mansoura 35516, Egypt*

[e] *Institute of Soil and Environmental Sciences, University of Agriculture Faisalabad, Faisalabad 38040, Pakistan*

[f] *School of Civil Engineering and Surveying, University of Southern Queensland, Toowoomba 4350 Queensland, Australia*

[g] *Department of Environmental Engineering, Middle East Technical University, Ankara 06800, Turkey*

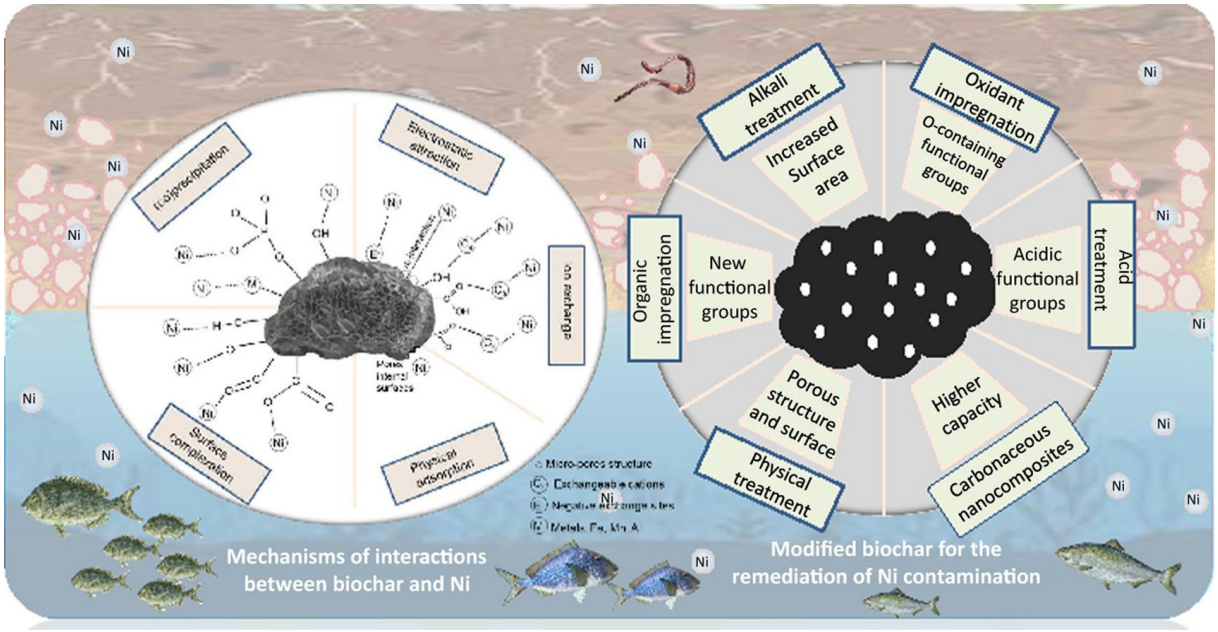
[h] *Department of plant science and land Architecture, University of Maryland, college park, USA*

[i] *Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK*

[j] *Department of Renewable Resources, University of Alberta, Edmonton, Alberta, T6G 2H1, Canada*

* Corresponding author: sxchang@ualberta.ca

Graphical abstract



Highlights

- Ni occurrence and biogeochemistry, and remediation using biochar are reviewed.
- Biochar affects redox-mediated transformations and reduces Ni availability.
- Negative-charged acidic functional groups act as electron donors, enhance Ni removal.
- Competitive adsorption on binding sites on biochar may impair Ni remediation.
- Biochars should be fabricated and designed for Ni remediation.

ABSTRACT

Nickel (Ni) is a potentially toxic element that contaminates soil and water, threatens food and water security, and hinders sustainable development on a global scale. Biochar has emerged as a promising novel material for remediating Ni-contaminated environments. However, the potential for pristine and functionalized biochars to immobilize/adsorb Ni in soil and water, and the mechanisms involved have not been systematically reviewed. Here, we critically review the different dimensions of Ni contamination and remediation in soil and water, including its occurrence and biogeochemical behavior under different environmental conditions and ecotoxicological hazards, and its remediation using biochar. Biochar is effective in immobilizing Ni in soil and water via ion exchange, electrostatic attraction, surface complexation, (co)precipitation, physical adsorption, and reduction due to the biogeochemistry of Ni and the interaction of Ni with surface functional groups and organic/inorganic compounds contained in biochar. The efficiency for Ni removal is consistently greater with functionalized than pristine biochars. Physical (e.g., ball milling) and chemical (e.g., alkali/acidic treatment) activation achieve higher surface area, porosity, and active surface groups on biochar that enhance Ni immobilization. This review highlights possible risks and challenges of biochar application in Ni remediation, suggests future research directions, and discusses implications for environmental agencies and decision-makers.

Keywords: Biowaste; Charcoal; Immobilization; Review; Soil contamination; Sorption; Toxic trace element; Wastewater

1. Introduction

Nickel (Ni) is a potentially toxic element that is commonly found in soil and water systems. It is a transition metal with a high density (8.9 g cm^{-3}), melting point (1455 °C), and boiling point (2730 °C) (Gonnelli and Renella, 2013; Tsadilas and Rinklebe, 2018). Nickel has an average atomic weight of 58.69, belongs to Group X of the periodic table and the iron family, along with Fe and Co (Kabata-Pendias, 2011). Nickel has four oxidation states (+1, +2, +3 and +4), while its main oxidation state is +2. The Ni(II) is readily available and more toxic in cationic form than its complexes (Albanese et al., 2015; Parades-Aguilar et al., 2021). Nickel is widely released into soil and water systems from various anthropogenic and natural sources. For example, Ni can be discharged into the environment from Ni alloy industries, pigment manufacturing processes, tannery industry wastewater, and mafic and ultramafic rocks and their derived soils as a result of weathering and pedogenesis (Albanese et al., 2015; Babaahmadifooladi et al., 2020; Panagopoulos et al., 2015). Improper disposal of industrial wastes and aerial deposition of contaminants can lead to elevated levels of Ni in soil and water systems. Generally, the levels of Ni is 5-40 ng m^{-3} in the air, 3-1000 mg kg^{-1} in agricultural soils, and less than 2 µg L^{-1} in fresh water and oceans (WHO, 2000). Nickel contamination can be transferred directly from air and soil to surface water bodies through deposition and runoff from soil, or indirectly to groundwater via leaching (Tsadilas and Rinklebe, 2018). The pollution of this metal is increasingly becoming a concern, particularly in developing countries, due to its non-degradability in the environment (Panagopoulos et al., 2015; Poznanović Spahić et al., 2019).

The presence of excessive amounts of Ni over permissible limit in soil (35 mg kg^{-1}) and water (0.02 mg L^{-1}) causes toxicity to all living organisms (Antoniadis et al., 2017; Hussain et al., 2017; WHO, 1996). Even though Ni is an essential micronutrient for plants with concentrations <10 mg kg^{-1} of plant biomass (Sugawara and Nikaido, 2014) and in the biochemistry of human microbiota at low consumption rate of 5 µg Ni kg^{-1} body weight day^{-}

[1] (Russell et al., 2001), a high rate of intake of Ni may cause serious harm to human health, such as causing allergy, cancer and reduced lung function (Zambelli et al., 2016). Therefore, high concentrations of Ni in drinking water and the soil, or the entry of Ni into the food chain via plant uptake poses a significant health threat for humans and animals, and threatens the ecological sustainability of the global system (Rinklebe et al., 2019; Shaheen et al., 2020).

Several techniques, including soil washing (Zou et al., 2019), adsorption (Cheng et al., 2019; Fiyadh et al., 2019), reduction (Di Palma et al., 2015), membrane process (Samantaray et al., 2019), electrokinetic remediation (Wang et al., 2019), photodegradation (Singh et al., 2018), phytoremediation (Antoniadis et al., 2017) and microbial remediation (Guan et al., 2019), have been investigated for the treatment of Ni contaminated soil and water systems. In-situ stabilization methods have been widely recommended as an eco-friendly and less destructive technique for the remediation of contaminated soils (Shaheen et al., 2018a). Those methods involve the application of immobilizing agents to the soil matrix, aiming to transform the soluble toxic element into immobile forms, making it unavailable for plant uptake or translocation in the soil. Among different immobilizing agents such as biosolids, lime, clays, compost, metal oxides, and animal waste (Palansooriya et al., 2020; Shaheen et al., 2018a), biochar garnered high attention for the remediation of Ni contaminated environments owing to its multiple environmental benefits (El-Naggar et al., 2018b).

The utilization of biochar for the remediation of Ni contaminated soil and water has been increasing over the last decade, as indicated by the steadily increasing number of publications in indexed journals that deal with this hot topic (Figure 1). Biochar exhibited a high immobilization efficiency for Ni in soil (Munir et al., 2020; Nawab et al., 2018; Rehman et al., 2016; Shaheen et al., 2015) and aquatic systems (Zhu et al., 2019). Biochar as a C-rich porous material with a high surface area, reactivity and adsorption capacity has the

potential to immobilize toxic elements in the soil (He et al., 2019). However, some studies have reported contradictory results where biochar did not affect or decreased Ni immobilization in contaminated environments (Mourgela et al., 2020; X. Yang et al., 2019). Therefore, a critical review on the effectiveness of biochar for the remediation of Ni contaminated soil and water is urgently needed.

Recently, a number of publications reviewed the potential of using biochar to remediate contaminated soil and water, with most of those reviews discussing either the immobilization efficiency of biochar for a group of toxic elements (Ahmad et al., 2014; Bandara et al., 2020; Beesley et al., 2011; Chemerys and Baltrėnaitė, 2018; Deng et al., 2020; Derakhshan Nejad et al., 2018; He et al., 2019; Rajapaksha et al., 2016; Rinklebe et al., 2019) or focusing on a specific element such as arsenic (As), cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn) (Li et al., 2019; Palansooriya et al., 2020; Shaaban et al., 2018; Zama et al., 2018; Zhang et al., 2020). However, these reviews did not provide specific and in-depth discussion for the remediation of Ni contamination in soil and water systems. The sources and biogeochemical behavior of metals and their immobilization/adsorption mechanisms on biochar can widely vary. Therefore, the most suitable method for biochar production and modification to achieve the best performance can also greatly vary for the remediation of the specific potentially toxic element, either in the soil or the water environment (Shaheen et al., 2018b). Therefore, a review is needed to synthesize the knowledge, draw holistic conclusions, and recommend future research directions specifically for the remediation of Ni contamination. Although a large number of publications (>750) have been focussed on the application of biochar (and sometimes referred to as charcoal) in treating Ni-contamination (according to the Scopus database), few have provided a synthesis of the current status or future perspectives in this area of research. In particular, the use of pristine (unmodified) and modified (functionalized or designed)

biochars as immobilizing agents for Ni and the mechanisms involved have not been systematically reviewed.

This paper synthesizes information on the different dimensions of Ni contamination, the occurrence of Ni in the environment, the biogeochemistry of Ni in soil and water, the potential for pristine and designer biochars to mitigate Ni contamination, and the mechanisms involved in Ni remediation using biochar. We also discuss the potential risks and challenges of biochar application for Ni remediation and future research perspectives. This review provides state-of-the-art knowledge on Ni contamination in soil and water, and the advances in biochar application as a remediation tool. We expect that this review will benefit not only researchers for designing future research projects but also environmental agencies and decision-makers for developing policies related to the management of Ni pollution.

2. Sources and occurrence of Ni contamination

Nickle contamination occurs via disparate sources in the ecosphere, which is composed of terrestrial (soil), freshwater, and marine ecosystems. Nickle is formed in the soil via various lithogenic/pedogenic and anthropogenic processes; Ni contained in the soil can then be transferred to ground and surface waters. The origin of Ni in a specific site plays a key role in determining its biogeochemical behaviour and the proper remediation method for its environmental management.

2.1. Lithogenic/pedogenic sources of Ni contamination

The type of parent material at a site affects the geochemical background of potentially toxic elements in the soil. Nickle can occur naturally in the parent material; those derived from pedogenic sources are affected by pedogenic soil formation processes that change their concentration and distribution (Evans and Barabash, 2010). In particular,

weathering of soil minerals leads to chemical alteration of surfaces of primary minerals and the release of more soluble forms of Ni (e.g., in association with chloride, sulphate, or nitrate), which might form secondary minerals such as alumino-silicates, carbonates, oxides, hydroxides, sulfates, and amorphous minerals (Massoura et al., 2006). For instance, Ni contaminations in southeast Mexico and in the Mitidja plain in Algeria, were from lithogenic sources such as volcanic plumes/ashes and the parent material, respectively, with pedogenic processes comprising a major portion of Ni in the soil solution, containing 39-318 mg Ni kg^{-1} (Hernández-Quiroz et al., 2012; Laribi et al., 2019). Nickle contamination has also been reported in soils in Balkan and Serbia due to the presence of ultramafic and mafic parent rocks having Ni in the range of 28-589 mg kg^{-1} (Albanese et al., 2015; Poznanović Spahić et al., 2019).

The spatial distribution of Ni in agricultural and grazed soils in Europe has been studied (Albanese et al., 2015). Many areas in north-western Italy contained >109-114 mg kg^{-1} of Ni (Figure 2) because of the dominance of Ni-rich sedimentary rocks, alluvial sediments and ophiolite complexes in that areas (Albanese et al., 2015). Sedimentary rocks contain up to 90 mg kg^{-1} of Ni (Kabata-Pendias, 2011). In general, the mineral composition of parent materials has a crucial role to play in enriching Ni concentrations to > 75 mg kg^{-1} in soils of many Mediterranean countries (Albanese et al., 2015). In a study on Czech serpentinites, which contain 2000 to 3000 mg kg^{-1} of Ni, Ni was highly mobile, mainly released from olivine and complexes of Ni sulphide-iron oxide minerals, influenced by weathering under the temperate and cold continental climates (Quantin et al., 2008). Contamination of soil and water with Ni from lithogenic/pedogenic sources is usually associated with parent materials that are ultramafic rocks, such as serpentinites, peridotites and pyroxenites, and ophiolite outcrops (Panagopoulos et al., 2015). However, potentially toxic elements in naturally contaminated soils are typically immobile and less toxic due to their occurrence in the solid phase as compared to those contained in anthropogenically

contaminated soils (Poznanović Spahić et al., 2019). Therefore, the transfer of Ni from such soils to water is limited as compared to that in anthropogenically contaminated soils. Nevertheless, lithogenically/pedogenically contaminated soils still pose significant potential environmental risks, in particular, under changing pH and alternating reducing-oxidizing conditions, in which more labile fractions of toxic elements can be formed.

2.2. Anthropogenic sources of Ni contamination

Soil contamination of Ni from anthropogenic sources is a major threat to the sustainability of our ecosystems. For instance, dust from cement and metal processing industries and fly ash from fossil fuel combustion are sources of Ni at range of 91-1200 mg kg^{-1} in the soil (Panagopoulos et al., 2015; Poznanović Spahić et al., 2019). Many industrial activities, such as the production of austenitic stainless steel, chemicals, and batteries, as well as exhaust fumes from automobiles and refineries, are sources of Ni (Albanese et al., 2015; Hernández-Quiroz et al., 2012). These industries often generate Ni-containing disposals, effluents, and wastewater; those biproducts, if inappropriately discharged, could pose a huge threat to soil and water contamination of Ni.

The Asopos river basin in Greece underwent major Ni contamination in the 1960s, with Ni concentration in the contaminated sites 2.5 times higher than the baseline in the soil and groundwater, mainly due to emissions from nearby activities of more than 400 industrial units (Panagopoulos et al., 2015). In agricultural soils, Ni can come from fossil fuel combustion, industrial discharge, mining, or smelting operations, as well as the application of Ni-containing materials such as insecticides, fungicides, herbicides, and fertilizers (El-Naggar et al., 2018c; Khan et al., 2017; Palansooriya et al., 2020). Therefore, soil and water contamination with Ni is usually driven by anthropogenic sources with high mobility and toxicity risks. Therefore, wastewater disposal was banned by law in the Asopos area

(Palansooriya et al., 2020; Panagopoulos et al., 2015), contaminants previously entered into soil and water systems pose constant risks for the environment and human health.

2.3. Stable isotope tracing of Ni contamination

The composition of the ^{60}Ni isotope in different terrestrial samples, including ultramafic soils, Fe/Mn crust, fresh water and sweater and river sediments, ranges between -1.5 and 2.5‰ (Cameron and Vance, 2014; Gall et al., 2013; Ratié et al., 2015a). The source of Ni in the soil can be traced using isotope analysis, since anthropogenic Ni has a higher ^{60}Ni isotope composition than naturally occurring Ni (Ratié et al., 2015b). For example, smelting and refining activities increased isotopic ^{60}Ni composition, which ranged between 0.01 and 0.20 ‰ for fly ash, and 0.11 and 0.27‰ for smelting slags, while it ranged between -0.19 and 0.10 ‰ in surrounding non-ultramafic natural soils in Niquelandia, Brazil (Ratié et al., 2015b). As another example, isotopic ^{60}Ni composition ranged between 0.03 and 0.22‰ in smelting and refining slags, while it ranged between -0.30 and 0.11 in non-ultramafic soils in Barro Alto, Brazil (Ratié et al., 2015b). The isotopic ^{60}Ni composition ranged between 0.56 and 1.00‰ in anthropogenic inputs (smelter slag and feeding material), while it ranged between 0.22-0.49‰ in natural background (bedrock) in northeast Norway (Šillerová et al., 2017). The isotopic composition of Ni can therefore be used to trace anthropogenic Ni contamination in the environment.

Isotopic exchange kinetics for Ni also enable us to distinguish different available pools of Ni via determining the rate of isotopic exchange over time (Kabata-Pendias, 2011; Tongtavee et al., 2005). In this approach, the relative contribution of each Ni-bearing mineral to the total Ni availability can be used to demonstrate the fractionation of available Ni. Isotopically exchangeable pool of Ni is a reliable indicator of the bioavailable pool of Ni in the mineral/water interface. Isotopic exchange kinetics were studied for typical Ni bearing minerals such as serpentines, chlorite, smectite, goethite, and hematite (Zelano et al., 2016).

The rates of isotopic exchange were 100 - 1000 times higher for Ni in outer-sphere complexes than that in inner-sphere complexes (Zelano et al., 2016). This phenomenon was responsible for the regulation of Ni availability from Ni bearing minerals in natural samples. For instance, the high abundance of the formation of outer-sphere complex sites (>80%) of smectite enhanced Ni availability in the soil quickly as compared to soils rich in iron oxides; whereas the formation of outer-sphere complexes was not the primary process that governs Ni isotopic exchange in chlorite and deweylite. The Ni availability of pure Ni-bearing minerals was successfully described via assessing the isotopic properties of ultramafic soils (Echevarria, 2021).

Isotopically exchangeable Ni measured over time refers to the Ni retained onto soil particles via sorption or surface complexation (Echevarria, 2021). The availability of Ni in ultramafic soils was almost solely controlled by the abundance of Ni-bearing minerals, with a limited effect by pH (Echevarria, 2021). Available Ni commonly originates from secondary 2:1 clay minerals (e.g., Fe-rich smectite) and amorphous Fe oxyhydroxides (Echevarria, 2021). Hence, Ni contamination is often associated with smectite-rich soils formed on serpentinite, and poorly weathered Cambisols developed on peridotite under temperate climates (Echevarria, 2021). The utilization of Ni isotopes for tracing the source of contamination in the environment is a promising, yet challenging, approach due to the scarcity of knowledge about the impact of different biogeochemical processes on the isotopic fractionation of Ni (Šillerová et al., 2017).

3. Biogeochemical behavior and environmental risks of Ni

Elevated Ni concentrations over a permissible limit in soil and water pose significant environmental risks, particularly if a significant portion of the element is in mobile or potentially mobile fractions. The mobility and phytoavailability of Ni are dynamically influenced by multiple environmental factors and processes such as pH, reducing-oxidizing

condition, precipitation, dissolution, complexation, ion exchange, and biological transformations (Rajapaksha et al., 2018; Shaheen et al., 2018b). Geochemical fractionation of Ni results in different forms of Ni in the soil, including exchangeable form and those bound to carbonates, iron and manganese oxides, organic matter, and other soil fractions (Tessier et al., 1979). Methods have been developed to further investigate sulfide-, crystalline iron oxide-, and amorphous iron oxide-bound fractions of toxic elements (El-Naggar et al., 2018c). Exchangeable and carbonate-bound fractions are considered mobile fractions, non-residual fractions are considered potentially mobile when soil biogeochemistry (e.g., soil pH, reduction-oxidation (redox) condition, and dissolved organic carbon concentration) changes (El-Naggar et al., 2018c; Shaheen et al., 2020). The mobility of toxic elements might facilitate their uptake by plants or transfer among environmental compartments via soil and water routes. Soil erosion and surface runoff may lead to the transfer of Ni from the soil to aquatic ecosystems and contribute to water pollution. Leaching of Ni may occur, in particular, in acidic and coarse-textured soils due to the weak binding between Ni and soil colloids (Palansooriya et al., 2020). Therefore, the biogeochemical behavior of Ni determines their speciation, mobility and phytotoxicity in the soil, bioaccumulation by plants, and transfer to the water system. Understanding the biogeochemical behavior of Ni is essential to properly assess the potential environmental and health risks of Ni pollution.

3.1. Nickel in soil

Nickel shows affinity with metallic Fe and S; hence, Ni-Fe and Ni-S minerals such as pentlandite $(Fe, Ni)_9S_8$, ullmannite (NiSbS), millerite (NiS), and gersdorffite (NiAsS) are formed (Albanese et al., 2015; Hooda, 2010) in the soil environment. Nickel also occurs in other minerals such as kullerudite ($NiSe_2$), niccolite (NiAs), and ferromagnesian minerals (Hooda, 2010).

Nickel can be mobilized during weathering of Ni-containing soil minerals via different soil forming processes, and become soluble in soil solutions, allowing its translocation along the soil profile (Kabata-Pendias, 2011). The mobility of Ni in soils might pose a health risk as it can be moved towards the rooting zone and be taken up by plants as $Ni(H_2O)_6^{2+}$ (Palansooriya et al., 2020). The maximum allowable concentration of Ni in agricultural soils range from 20 to 60 mg kg^{-1}, while the threshold for requiring remediation ranges from 75 to 150 mg kg^{-1} (Kabata-Pendias, 2011). These values are based on a range of studies and seem to be questionable due to their wide range; however, they are realistic considering the variation in soil properties and Ni forms. The biogeochemical behavior and forms of Ni are governed by various factors in the soil including clay type and content, organic matter content and soil pH, and can be indirectly influenced by soil E_H, and the content of Fe-Mn (hydr)oxides (El-Naggar et al., 2018b; Panagopoulos et al., 2015), with the mobility of Ni increases under oxidizing and acidic conditions (Albanese et al., 2015; El-Naggar et al., 2018c). Due to the high affinity of Ni with organic matter, the mobility of Ni is dependent on soil organic matter content and composition. On the one hand, Ni binding with organic ligands might diminish its mobility, as organic molecules can react with Ni to create less mobile Ni forms. On the other hand, the mobility of Ni might increase with the presence of fulvic and humic acids in organic matter-rich soils as those organic acids have a high chelation ability (Kabata-Pendias, 2011). Nickel can co-precipitate with clay minerals such as montmorillonite, which can be easily mobilized during weathering (Dähn et al., 2004). Nickel can be adsorbed/complexed with soil Fe (hydro)oxides and minerals at a range of 100-170 mg Ni kg^{-1}, and with soil Mn (hydro)oxides and minerals within 39-4900 mg Ni kg^{-1} (Kabata-Pendias, 2011). Extended X-ray absorption fine structure (EXAFS) analysis on samples collected from an industrial contaminated site in southern Italy showed that Ni was in a spinel-type geochemical form (trevorite; $NiFe^{3+}{}_2O_4$) associated with magnetite and hematite (Terzano et al., 2007). The EXAFS analysis on a clayey paddy soil

(Ultisols) collected from Taipei City showed that Ni was incorporated into the MnO_2 layer of lithiophorite in the soil (Manceau et al., 2005); this finding is in line with results from Manceau et al. (2002). They demonstrated that the incorporation mechanism of Ni into the MnO_2 layer might be dependent on the abundance of natural lithiophorite and the condition of its formation (Manceau et al., 2005). Nickel adsorption on Fe-Mn (hydr)oxides is pH-dependent, as the release of Ni into the soil solution increases with decreasing soil pH due to the effect of pH on the surface charge of adsorbents in the soil (El-Naggar et al., 2018b; Kabata-Pendias, 2011).

3.2. Nickel in water

Nickel is widely present in dissolved and particulate forms in natural water. The dissolved forms include the hydrated divalent Ni, inorganic Ni complexes (with OH, CO_3 and Cl), and organic Ni complexes (i.e., Ni-humic substance inclusions), while particulate forms are associated with colloids, forming larger particles (Donat et al., 1994). The presence of excessive Ca and Mg can trigger the formation of weak complexes of Ni ($CaMgNiH_4$) and the release of its bioavailable forms ($NiCO_3$, NiS, Ni_3S_2, NiO) that is highly toxic to freshwater and marine organisms (Mandal et al., 2002). In aqueous solutions, four main species of Ni, including Ni(II), $Ni(OH)^+$, $Ni(OH)_2$ and $Ni(OH)_3^-$, are formed with the absence of external oxidizing agents or complete chelation for Ni (Anoop Krishnan et al., 2011). The Ni(II) is the predominant form in highly acidic (pH = 0) to alkaline (pH 8.5) systems (Figure 3). With increasing pH, hydroxide complexation increases and forms various hydroxide species such as $Ni(OH)^+$, $Ni(OH)_2$ and $Ni(OH)_3^-$. Mainly, hydroxides of nickel get phagocytised when inside the body of organisms and the Ni(II) is the actual carcinogen that binds with DNA (Schaumlöffel, 2012). In aqueous solutions, Ni(II) is relatively stable due to the formation of complexes with inorganic and organic ligands and association with suspended soil colloids (Rinklebe and Shaheen, 2017).

In general, the formation of Ni-organic complexes depends mainly on the dissolved organic carbon content in soil solutions or aquatic systems (El-Naggar et al., 2018c). Both $Ni(OH)^+$ and $Ni(OH)_2$ are abundant when the pH ranges between 8.0 and 12.0, and $Ni(OH)_3^-$ occurs mainly in highly alkaline conditions (pH = 12.0-14.0) (Anoop Krishnan et al., 2011). Nickel can be rapidly sorbed on surfaces of solids when offered in the solution. For instance, Ni can be adsorbed through the formation of outer-sphere complexes, in the form of hydrated Ni species on the surfaces of solids. In outer-sphere complexation, the electrostatic forces or H-bonding facilitates adsorption of Ni ions; therefore, they could be easily released or exchanged with other ions (Rinklebe and Shaheen, 2017). On the other hand, Ni can form inner-sphere complexes on the surfaces of colloids, in the form of dehydrated Ni species, which are more stable (Shi et al., 2012). In water-soil systems such as wetlands, and floodplains, reduction or oxidation conditions can also affect the biogeochemical behavior of Ni, and its distribution among dissolved and colloidal phases (Rinklebe and Shaheen, 2017). Dissolved Ni concentrations were found to increase at high E_H values (oxic conditions) than at low E_H values (anoxic conditions) (El-Naggar et al., 2018c). Nickel tends to bound to insoluble Fe and Mn (hydr)oxides or dissolved organic matter (e.g., aromatic compounds) under oxidizing conditions (El-Naggar et al., 2018c; Pyle and Couture, 2011). Nickel also has an affinity to sulfates under oxic conditions, while it is commonly bound to insoluble sulfides under reduced conditions; in particular, E_H-induced transformation of sulfates (at high E_H values)-sulfides (at low E_H values) affected the release dynamics of Ni associated with them into dissolved and colloidal phases (El-Naggar et al., 2018c).

However, the redox chemistry of Ni and its interactions with the solid phase and colloids in aquatic systems is not sufficiently documented and requires further research to understand the factors governing the mobility and redistribution of Ni among different solid-water phases.

3. Nickel remediation strategies and biochar

A variety of remediation strategies has been developed to combat soil and water contamination with potentially toxic elements such as Ni. Those strategies are classified into physical (e.g., soil replacement, spading and washing), biological (e.g., microbial treatment and phytoremediation) and chemical (electrokinetic remediation, chemical reduction, chemical stabilization, and photocatalysis) technologies (Derakhshan Nejad et al., 2018; Li et al., 2019; Palansooriya et al., 2020). For instance, photocatalysis is deemed a green technology for the remediation of contaminated aquatic systems by converting photon energy from sun light into chemical energy, and at the same time transform toxic elements into nontoxic ones (Santhosh et al., 2018; Wu et al., 2020, 2018). However, the potential formation of undesirable secondary products, which could be more toxic than the original contaminant, via this method is a serious disadvantage (Vasilache et al., 2013). Some remediation strategies aimed to permanently overcome co-existing toxic elements in the soil such as soil replacement and phytoremediation. Soil replacement involves removing the contaminated soil and adding a large amount of uncontaminated soil to dilute the contaminant in the soil. This method is labor-demanding and has a high cost; however, it can be a suitable strategy for remediating a small area of heavily contaminated soils. Utilization of Ni hyperaccumulating plants such as canola (*Brassica napus* L.) for the phytoextraction and removal of Ni from soil has also been reported (Adiloğlu et al., 2016; Deng et al., 2018). Although phytoremediation technology can be a feasible strategy for removing Ni from contaminated soils (Adiloğlu et al., 2016), it is considered an expensive and time-consuming approach, and the improper management of the polluted plant can raise new environmental issues (Antoniadis et al., 2017; Li et al., 2019). However, phytoremediation of Ni contaminated soils is a technology that should be further explored.

Based on new risk-based regulatory requirements, more attention has been paid to combating the potential mobility and phytoavailability of toxic elements, rather than

targeting the reduction of their total concentrations (Beesley et al., 2011; Zama et al., 2018). Therefore, chemical stabilization technologies via the application of soil amendments to immobilize toxic elements and subsequently reduce their transfer to groundwater or uptake by plants have received growing interest. Remediation methods based on the in-situ stabilization of contaminants are often more cost-effective, have no/minimal negative impacts on soil fertility, and require simple operations (El-Naggar et al., 2020). Several amendments, including clay, Fe oxides, liming materials, nanoparticles, biosolids, manure, composts, coal fly ash, and biochar, have been extensively employed to immobilize toxic elements in soil and water (Abbas et al., 2020; Biswas et al., 2019; Hu et al., 2015). Recent studies have recommended biochar as a promising eco-friendly remediation material due to its unique surface characteristics and inorganic and organic compositions (Bolan et al., 2014; El-Naggar et al., 2019c, 2021; Elkhlifi et al., 2021; He et al., 2019; Man et al., 2021). In particular, biochar may diminish the potential mobility and phytoavailability of Ni in the soil, and play an important role in their removal from aquatic systems, as discussed in the sections below.

Compared with the application of other restoration materials or technologies, the application of biochar is superior due to its lower cost, reusability, relatively high stability once reacts with the contaminant, and adaptable in the environment. In addition, biochar provides various benefits such as improving soil fertility, enhancing soil water holding capacity, aeration, nutrient retention and carbon sequestration, and stimulating (commonly) microbial activities. Thus, biochar provides a win-win strategy for waste management and climate change mitigation. Although biochar is typically more expensive than other soil amendments such as compost, the lower decomposability of biochar gives it an advantage over other organic amendments. For instance, once biochar is applied to farmlands, it could remain in soil for decades to millennia, in contrast to compost that needs to be frequently applied to maintain its effectiveness. Some argue that the pyrolysis process is costly and

energy-consuming, making the production of biochar a non-profitable approach in the real market. This argument can be misleading as biochar production also produces various other products such as bio-oil and syngas out of the same process; if the economic value of those products are all realized, the cost and required energy for biochar production are diminished. Therefore, biochar application for the remediation of Ni contaminated soil can be economically and environmentally feasible.

For biochar application to remediate Ni contaminated aquatic systems, there are also various approaches that could be used to augment the potentially high economic cost. For example, a sequential use system which utilizes the same biochar in multiple systems can be a promising approach. This approach employs the sequential application of biochar, including recovering the biochar after each use, and in ideal systems, each sequence adds value to the whole process. For instance, Wurzer et al. (2020) proposed the following sequence for a sequential biochar use system: waste to biochar (waste management) → adsorption of H_2S (biogas purification) → sulfonated catalyst (esterification) → pH buffering (composting) → soil amendment (carbon sequestration). However, this being a relatively new concept, the integration of the remediation of Ni contaminated aquatic systems into an ideal sequential biochar use system has not been documented yet.

4. Biochar for remediation of Ni contamination

4.1 Biochar for remediation of soil Ni contamination

Numerous studies show that biochar has a high potential to immobilize Ni in the soil relative to other conventional soil amendments such as limestone, organoclay, bentonite, zeolite, activated-carbon, and nano-fertilizer (Shaheen et al., 2015). In Shaheen et al. (2018), biochars produced from shell limestone and other substances (e.g., humus, perlite and clay) were applied to a silty-acidic soil at 1% and were found to decrease water-soluble and exchangeable Ni by about 59 and 34%, respectively, compared to the control treatment. In a

field study, hardwood biochar produced at 600 °C and applied to the soil at a range of 0.5-2% resulted in up to 200% greater Ni immobilization efficiency as compared to the control, three years after biochar application (Shen et al., 2016). In a pot experiment, wood biochar produced at 450 °C was applied at 1 and 2% to reduce Ni bioavailability by 22 and 33%, respectively, in a sandy clay loam soil with a neutral pH (Rehman et al., 2016). The literature clearly demonstrated that the effectiveness of biochars for Ni remediation is dependent on the feedstock type used for biochar production, while the rate of biochar application had relatively small effects; however, both the feedstock type to use and the application rate need to be tested for site-specific applications.

We extracted data from eight studies with 59 observations to evaluate the effect of feedstock type, pyrolysis temperature, and biochar pH and application rate on Ni immobilization efficiency (Figure 4). Biochars produced from wood and agricultural wastes performed well, while manure- and sewage sludge-derived biochars had a minimal effect in reducing Ni availability in the soil. Biochars produced at pyrolysis temperatures ranging from 500 to 700 °C were more effective for Ni immobilization (Figure 4), consistent with other studies (Shen et al., 2016; Uchimiya et al., 2012). Biochars with a high pH (9.5-10) were more efficient in immobilizing Ni (Figure 4), while biochar application rate had no effect on the immobilization of Ni in soil. In this case, we suggest that a low application rate (<5 t ha^{-1}) would be preferable, considering the cost for biochar production, transportation, and application when a high application rate is used.

In wetlands contaminated with Ni, the potential for biochar to immobilize Ni is affected by redox dynamics. In particular, when biochar is applied to wetlands, it would change soil pH due to its commonly alkaline nature, and alter soil redox potential owing to its redox-active surfaces (Yuan et al., 2017). These biochar induced changes in soil pH and redox potential simultaneously affect various soil properties such as dissolved aliphatic/aromatic organic carbon concentrations, and the release and transformations of Fe,

Mn, and S/SO_4^{2-} (Rinklebe et al., 2020). The release of dissolved organic carbon and the induced transformations of those elements can be accompanied by the release of associated Ni from their structures and surfaces, which in turn enhances the mobility and phytoavailability of Ni (El-Naggar et al., 2018c). Therefore, more research is required to determine whether biochar is suitable for remediation of Ni contaminated wetlands, as biochar can increase the toxicity risk of Ni under such conditions.

The nature of functional groups on biochar surfaces, and their potential to accept/donate electrons determine biochar induced changes related to those parameters and their interactions with Ni(II) in soil (El-Naggar et al., 2018c). Biochar application may also affect the Ni(II) bioavailability in soil via the redistribution of Ni(II) between colloidal and dissolved fractions. For example, biochars derived from shell limestone, perlite, and humus decreased dissolved Ni(II) concentrations in a floodplain soil by 44%, from an average of 444.5 µg L^{-1} (rang of 38.6 - 1502.9 µg L^{-1}, n=21) in the control to average of 248.9 µg L^{-1} (rang of 33.5 - 683.5 µg L^{-1}, n=19) in biochar treated soils under changing redox conditions (Rinklebe et al., 2016). However, the application of rice hull biochar (pyrolyzed at 500 °C) increased the dissolved Ni(II) concentration by 9.5%, in particular from average of 56.3 µg L^{-1} (rang of 35.2 – 76.4 µg L^{-1}, n=28) in the control to average of 61.7 µg L^{-1} (rang of 27 – 84.1 µg L^{-1}, n=32) in biochar treated soils under dynamic redox conditions (El-Naggar et al., 2018c). The variation in biochar effects on dissolved Ni(II) in those two studies was mainly related to the type of biochar and soil studied. In particular, the decreased dissolved Ni in the first study was associated with decreased dissolved organic carbon under an oxic condition, while in the second study the biochar decreased soil pH under oxic conditions that led to the acidic dissolution of Fe and Mn (hydr)oxides under aerobic-acidic conditions and the release of associated Ni(II). However, there are significant knowledge gaps in the redox-mediated transformation of Ni in biochar treated soils, the interactions between functional groups on biochar surfaces with Ni, and the factors that control its (im)moblization in the soil.

Systematic investigations are required in the future to understand the behaviour of Ni when different types of biochars are applied at varying rates to different soils.

In summary, the potential for biochars to immobilize Ni(II) is affected by many factors, including the feedstock type, pyrolysis temperature, biochar application rate , type of soil, and redox conditions in the soil. The soil- and biochar property-specific nature of the effect of biochar application on Ni(II) (im)mobilization needs to be further studied, otherwise inappropriate biochar application may pose a critical risk of soil and water contamination of Ni.

4.2 Biochar for remediation of Ni Contamination in water

Biochar has been successfully employed for the remediation of Ni contamination in aquatic systems. Biochar's high porosity and surface area, and abundance of functional groups play a major role in its potential to remove Ni from water, due to multiple mechanisms of Ni immobilization as discussed in Section 5. However, several factors influence the biochar's adsorption capacity for Ni in water. The pH of the aquatic system has a major effect, and it can stimulate/inhibit the adsorption of Ni on biochar surfaces. In particular, the potential for biochar to adsorb Ni increases gradually along with increasing solution pH from strongly acidic to neutral (e.g., from pH 2 to 7 (Yang et al., 2019)). This is attributed to pH-induced changes in biochar surface functionality. At a very low pH (pH $\leq$ 2), surface groups of biochar would be protonated by the solution H^+ ions, and thus there would be no Ni adsorption (Bogusz et al., 2017; Yang et al., 2019). When pH increases (pH = 2-5), deprotonation of the surface groups would occur, leading to less competition between Ni(II) and H^+ ions on the binding sites of biochar surfaces; hence, Ni(II) starts to be adsorbed on biochar surfaces (Higashikawa et al., 2016). At the same pH range (pH = 2-5), $Ni(OH)^+$ exhibits less electrostatic repulsion with biochar surfaces, and thus, $Ni(OH)^+$ would have a strong electrostatic attraction on electronegative surfaces of biochars at this pH level

(Bogusz et al., 2017). At higher pH (pH > 6), the hydroxides will facilitate the precipitation of Ni(II) on biochar surfaces (Shen et al., 2017). Overall, Ni can be optimally adsorbed during the wastewater treatment when the pH became neutral (pH = 7) (Yang et al., 2019). The feedstock type used for biochar production is another key factor that determines the adsorption capacity of Ni in water (Figure 5). Pristine biochars produced from rice straw, banana fruit waste, and orange peel have been reported to adsorb Ni(II) at concentrations of 54 (Deng et al., 2019), 88 (Amin et al., 2019), and 78 mg Ni g^{-1} biochar (Amin et al., 2019), suggesting that those pristine biochars have a high adsorption capacity for Ni(II). Very low adsorption capacities ranging from 0.22 to 1.09 μg g^{-1} were, however, reported for pristine sludge and olive pomace biochars (Mourgela et al., 2020), and a water hyacinth chitosan-magnetic biochar (0.48 mg g^{-1}) (Chaiyaraksa et al., 2019). The pyrolysis temperature of biochar can also affect the efficiency of biochar to adsorb Ni. Biochars prepared at low temperatures (≤ 400 °C) have low adsorption capacities, whereas those produced at high temperatures (600-800 °C) have high capacities for removing Ni(II) from aqueous solutions. For instance, biochars produced from residues of biogas production via pyrolysis at a high temperature (600 °C) achieved higher adsorption of Ni(II) than that produced at a low temperature (400 °C) (Bogusz et al., 2017). This is mainly attributed to higher pore volume and larger specific surface area of biochar pyrolyzed at a high temperature. In general, the efficiency of pristine biochars for Ni removal from aqueous solutions is highly variable as they are dependent on the feedstock type and pyrolysis temperature used for their production; in several cases, the adsorption capacity is low. As a result, the use of novel biochar composites is becoming popular due to their high capacity for removing Ni from aqueous solutions. For example, a sewage sludge biochar loaded with α-Fe_2O_3 and α-FeOOH adsorbed 35.5 mg Ni g^{-1}, while unmodified sewage sludge biochar adsorbed only 20.4 mg Ni g^{-1} in aqueous solutions at treatment dosage of 0.01 g biochar to 50 mL solution with 100 ppm initial Ni(II) concentration at neutral pH (Yang et al., 2019). A biochar produced from

Taihu blue algae impregnated with α-Fe_2O_3 and activated with KOH was highly efficient in removing 98.8% chelated Ni at pH 6.0 (Wang et al., 2020).

A pine cone-alginate hybrid biochar also showed a very high Ni adsorption capacity (156 mg g^{-1}) at pH 6.0 (Biswas et al., 2019). Acid and alkali modified date seed biochar also adsorbed 38.7 mg Ni g^{-1} at pH 6.0 (Mahdi et al., 2019). Similarly, a $KMnO_4$ and KOH modified peanut shell biochar adsorbed up to 87.2 mg Ni g^{-1} (An et al., 2019). The potential for Ni removal by modified biochars was mainly governed by inner-sphere complexation of Ni with oxygen-containing functional groups in biochar, and the combined effect of ion exchange, electrostatic attraction, and co-precipitation. An et al. (2019) described the mechanism for Ni removal as spontaneous endothermic chemisorption that was governed by amine (NH_2 – Ni) and hydroxyl functional groups in biochar. The Ni removal was better described by the Langmuir isotherm and pseudo-second-order kinetic models (Biswas et al., 2019; Yang et al., 2019), with the Sips model (Mahdi et al., 2019) and the Freundlich isotherm (Mourgela et al., 2020) also provide good fit for describing Ni removal in those studies.

In conclusion, biochar is a promising adsorbent of Ni in contaminated aqueous solutions such as industrial wastewater. Biochar surface functional groups play a major role in the removal of Ni from aqueous solutions. Therefore, the application of modified biochars via enriching them with functional groups such as acid/alkali groups, oxygen/amine containing groups, and so on, should be a better approach than the application of pristine biochar, for the remediation of Ni contaminated soil water systems.

5. Mechanisms for the physicochemical reactions between biochar and Ni

Multiple mechanisms have been suggested for the immobilization of toxic elements such as Ni following biochar application; such mechanisms include ion exchange, electrostatic attraction, surface complexation, (co)precipitation, physical adsorption, and

oxidation-reduction (Ahmad et al., 2014; He et al., 2019). We discuss below the mechanisms involved in the immobilization of Ni based on the recent literature (Figure 7).

The adsorption of Ni(II) onto surfaces of biochars produced from sugarcane bagasse, bamboo, and hickory wood chip were explained by the existence of cation-π interaction on internal and external surfaces of biochars (Lyu et al., 2018). In general, ion exchange and electrostatic attraction play a crucial role in the adsorption of Ni onto biochars, and could lead to rapid adsorption in some cases, due to ion exchange of Ni with other cations such as K^+, Mg^{2+}, and Ca^{2+} on biochar surfaces (Yang et al., 2019).

Electrostatic attraction of Ni(II) by negatively charged functional groups on biochar surfaces is also a potential mechanism for Ni(II) removal (Ahmad et al., 2014; El-Naggar et al., 2018b), partly due to increased pH after the addition of biochar with abundant active -OH functional groups on biochar surfaces (Uchimiya et al., 2010). The high electronegativity of biochar depends on the variable surface negative charges, which increase with pH (He et al., 2019). Batch experiments conducted using a sludge biochar produced at 500 °C indicated the role of electrostatic attraction in the Ni adsorption process on biochar surfaces, which gradually increased when pH increased from 2 to 7 (Yang et al., 2019). The enrichment of biochar surfaces with O-containing functional groups, such as carboxyl, lactonic, and hydroxyl, can decrease the surface zeta potential and the point of zero charge (PZC) of biochars, hence, facilitate the Ni(II) adsorption via electrostatic interaction on biochar surfaces (Lyu et al., 2018).

Surface functional groups, O-containing groups in particular, facilitate the binding of elements and could form complexes with Ni on biochars (He et al., 2019). Biochar surfaces are commonly occupied by various functional groups (eg., -COOH, -C=O, -CO- and -OH), which vary with the biochar feedstock type (Figure 7). Those functional groups can be altered as well based on the pyrolysis temperature used in producing the biochar (El-Naggar et al., 2019b). As for biochars produced at low pyrolysis temperatures, they contain more

abundant functional groups on their surfaces than those prepared at high pyrolysis temperatures. Complexation of Ni with O-containing functional groups on biochar surfaces has been reported as a mechanism for Ni immobilization in soil (El-Naggar et al., 2018b, 2018c), and Ni(II) removal from contaminated aqueous solutions (Lyu et al., 2018; Uchimiya et al., 2010). The addition of biochar produced from cottonseed hull at 350 °C to soil significantly promoted Ni immobilization due to the predominance of O-containing functional groups on biochar surfaces (Uchimiya et al., 2011). Filters made from biochars reduced the Ni(II) phytotoxicity in nutrient films; filters engineered from cottonwood biochar (pyrolyzed at 600 °C) scavenged the phytotoxicity of Ni(II) on tomato grown in hydroponic systems, with the complexation by active functional groups (C=O and —OH in particular) as a prominent mechanism (Mosa et al., 2016). Nickel(II) adsorption on surfaces of wheat straw and wood pin chip biochars produced at 500-550°C were mainly attributed to the coordination between Ni(II) and carboxyl and hydroxyl groups on biochar surfaces (Alam et al., 2018).

Inorganic components in biochar may facilitate the (co)precipitation of Ni in soil or water, forming insoluble precipitates such as phosphate, carbonates and (hydr)oxides. For example, biochar application can induce Ni precipitation with sulfate in sediments under dynamic redox conditions (El-Naggar et al., 2018c); the redox-active surfaces of biochar might affect the sulphur chemistry in the soil, and consequently, Ni associated with sulfides or sulfates is influenced via precipitation or release in the soil solution. Co-precipitation of Ni(II) with PO_4^{3-} and OH^-, derived from biochar surfaces, was also confirmed via an integrated mechanistic approach that included various chemical and spectroscopic techniques and described in the following reactions.

$$\mathrm{Ni}^{2+} + 2\mathrm{OH}^{-} \rightarrow \mathrm{Ni(OH)}_2 \downarrow$$
$$3\mathrm{Ni}^{2+} + 2(\mathrm{PO}_4)^{3+} \rightarrow \mathrm{Ni}_3(\mathrm{PO}_4)_2 \downarrow$$

Biochar application may also facilitate the precipitation of Ni with Fe, Mn, and Al, as biochar application typically increases soil pH, which would decrease the dissolution of Fe, Mn, and Al (hydr)oxides in acidic soils; thus it can control the mobilization or immobilization of Ni associated with those minerals and oxides in soil and water systems (El-Naggar et al., 2018b; Rinklebe et al., 2016).

Physical adsorption of Ni on internal surfaces of biochars in soil and water systems is one of the mechanisms for Ni removal. Biochars can reduce the bioavailability of toxic elements via sorption within their micro-pore structure (Chemerys and Baltrėnaitė, 2018). Physical adsorption was a major mechanism for the high Ni adsorption capacity (up to 15.4 mg g^{-1}) by a corncob biochar produced at 450 °C (Hu et al., 2018). The intra-particle diffusion of Ni(II) into the biochar complexed and highly variable pore system enhances the potential of biochar to adsorb Ni(II) (Lyu et al., 2018). However, the pore structure and pore volume affect the physical adsorption of Ni within the inner surfaces of biochar (Shaheen et al., 2018b). The physical adsorption by biochar plays an important role in the remediation of Ni contamination, but its contribution is less than that of chemical adsorption in remediating Ni contamination (Shen et al., 2018; Yang et al., 2019). The underlying mechanisms of physical adsorption of Ni in soil and water are still unclear and need to be further studied.

In conclusion, several mechanisms have been reported to underlie the immobilization/removal of Ni in soil/aqueous solutions. Based on the literature, Ni adsorption by biochar is governed by the synergistic effect of physical and chemical reactions that facilitate the adsorption of Ni on biochar surfaces.

6. Modifying biochars for immobilization of Ni in soil and water

The potential for biochars to remediate Ni contamination in soil and water systems depends on the chemical and physical characteristics of biochar surfaces (Shi et al., 2018), which are affected by feedstock type, pyrolysis condition, and pre- and post-pyrolysis

modification methods used in biochar production (El-Naggar et al., 2019a). Therefore, the selection of a suitable feedstock and production condition should be done before deciding on a suitable modification method. Recent modification studies have attempted to increase the surface area, porosity, and the abundance of active surface functional groups of biochars to enhance the biochars adsorption capacity for contaminants. The efficiency for immobilizing toxic elements in soil and water systems by modified biochars is greater than that of pristine biochars. Here, we highlight recent advances in the field of biochar modification in order to obtain biochar with superior properties and high Ni adsorption capacities (Figure 8).

Several physical, chemical, and biological modification methods have been reported for modifying biochars for the remediation of different toxic elements. However, only a few studies have attempted to modify biochars to increase their adsorption capacity for Ni. For instance, biochars produced from different feedstocks (sugarcane bagasse, bamboo, and hickory wood chips) and pyrolysis temperatures (300, 450, and 600 °C for a 2 h residence time) were ball milled under different milling conditions (mass ratio and milling time) (Lyu et al., 2018). The 12 h ball milling time and mass ratio of 1:100 biochar to ball was suggested as an optimal condition to maximize the capacity of modified biochars for Ni(II) adsorption (Figure 9a). The ball milling process broke down the biochar particles from millimetre to submicron scale (Figure 9b-f). The ball-milled biochars exhibited higher internal and external surface areas (up to a 33-fold increase), and greater acidic surface functional groups (Figure 9), compared to the pristine biochars. The enhancement of these acidic functional groups altered the zeta potential of the ball-milled biochar. While the zeta potential for pristine biochars ranged between 24.6 and 27.9 mV, it decreased to a range between -19.4 and -48.1 mV for the ball-milled biochar, with the pH ranging from 1.6 to 9.5 (Figure 9h). Moreover, the PZC decreased from 4 in the pristine biochar to < 0.16 in the modified biochar. The ball-milled biochars had greater Ni(II) removal, faster adsorption kinetics and greater capacity to adsorbed Ni(II) from aqueous solutions; the Ni(II)

adsorption capacity was higher than that of many commercial carbonaceous sorbents (Lyu et al., 2018).

Sewage sludge biochars have been widely employed as adsorbents for the removal of a number of toxic elements including Ni (Bogusz and Oleszczuk, 2018; Higashikawa et al., 2016); however, the adsorption capacity for Ni can be enhanced when biochar is treated with Fe (hydr)oxides (Yang et al., 2019). For instance, sludge biochar was produced at 500 °C with a 2 h residence time, and then impregnated with $FeCl_3$-$6H_2O$ and $FeSO_4$-$7H_2O$ solutions, to be charged with Fe_2O_3 and FeOOH (Yang et al., 2019). The Fe supported biochar had a surface area (35.0 m^2 g^{-1}) higher than that of pristine biochar (24.2 m^2 g^{-1}), but had lower pore volume and smaller pore size, due to Fe compounds filling pores in the biochar (Figure 10). The Fe-supported biochar had superior adsorption capacity for Ni (35.5 mg g^{-1}) compared with the pristine biochar (20.4 mg g^{-1}). The scanning electron microscopy with energy dispersive x-ray (SEM-EDX) images revealed that Fe was more intensively distributed on Fe-supported biochar surfaces, compared to the pristine biochar. The SEM-EDX images showed that Fe and Ni were closely associated, as indicated by the locations that they occupy in EDX mapping (Figure 10). This further confirms the influence of α-Fe_2O_3 and α-FeOOH employed in the modification process on enhancing the biochar's adsorption capacity for Ni.

A palm seed derived biochar-magnetic biocomposite was fabricated and tested for Ni removal (Gazi et al., 2018). In order to obtain magnetic biocomposites, Fe_3O_4 was used to modify biochar (Gazi et al., 2018). The biochar-magnetic biocomposite achieved removal efficiency of 87% (28 mg g^{-1}) at pH 3, and up to 75% (~24 mg g^{-1}) in the presence of other competing ions, due to its high micro-mesoporous structure. Biochar-based nanocomposites, such as nanometal oxide groups, have been used to immobilize toxic elements to take advantage of the benefits of both biochar and nanomaterials (El-Naggar et al., 2018b; Yang

et al., 2019). However, their suitability for immobilization/removal of Ni hasn't been proven yet, and future studies are needed to fill this knowledge gap.

Surface oxidation of biochars using ammonium persulfate and hydrogen peroxide has been used to increase the efficiency of biochars for Ni immobilization (El-Naggar et al., 2018b). Surface oxidation of peanut hull biochar using hydrogen peroxide significantly enhanced the adsorption capacity (22.8 mg g^{-1}) of Ni in contaminated water compared to activated carbon (0.8 mg g^{-1}) (Xue et al., 2012). Alkali/acid treatment of biochars could also increase biochars surface area and the abundance of specific functional groups. Post-pyrolysis activation of wood biochar surface with sodium hydroxide, for example, increased the surface area from 256 m^2 g^{-1} to 873.0 m^2 g^{-1}, cation exchange capacity from 45.7 cmol kg^{-1} to 124.5 cmol kg^{-1}, and Ni immobilization efficiency from 11 mg g^{-1} (pristine biochar) to 53 mg g^{-1} (modified biochar) (Ding et al., 2016). Alkali treatment of biochar with potassium hydroxide or sodium hydroxide increased the surface area and surface oxidation of biochar (Rajapaksha et al., 2016). Acid activation of biochar can also increase the abundance of carboxyl groups on biochar surfaces that stimulate the Ni retention in immobile forms (El-Naggar et al., 2018b). Sodium sulfide as a salt of a weak acid (H_2S) and a strong base (NaOH) was tried for surface modification of biochars. For example, Hu et al. (2018) produced biochars at pyrolysis temperatures of 300, 450 and 600 °C using different feedstocks, including crayfish shells, cotton stalks, corncob and peanut shells. They modified the biochars after pyrolysis using a Na_2S solution (2 mol L^{-1}), resulting in biochars with higher total pore volume that increased from 0.112 to 0.234 cm^3 g^{-1} and specific surface areas increased from 11.8 to 195.6 m^2 g^{-1}. Among the modified biochars, corncob biochar (pyrolyzed at 450 °C) had the maximum capacity for Ni removal (15.4 mg g^{-1}) (Hu et al., 2018). Enrichment of biochar surfaces with amino groups also plays an important role in enhancing Ni immobilization. Corn cob biochar produced via hydrothermal carbonization and modified by impregnation in a polyethylenimine solution increased the Ni adsorption

capacity (20 mg g^{-1}) in an aqueous solution by 43.7% as compared to the unmodified biochar (Shi et al., 2018). Physical modification of biochar surfaces via steam/air activation or heating was also proposed to increase the specific surface area and enhance the porous structure of biochar (Rajapaksha et al., 2016), leading to increased Ni adsorption capacities (El-Naggar et al., 2018b).

In conclusion, pristine biochars might not be efficient in the immobilization/removal of Ni in highly contaminated environments. Optimization of biochar functionality and characteristics via modification methods could maximize the potential of biochars for the remediation of soil and water highly contaminated with Ni. Alkali/acid treatment of biochars could be a promising method to increase biochar surface area and enrich surface functional groups that form complexation reactions with Ni. However, some of the modification methods are not eco-friendly and could even result in biochar being a source of contamination, such as biochar activation using mineral acids (Lonappan et al., 2019). Therefore, proper methods should be used for biochar modification. Much new research needs to be done to develop effective modification methods and to investigate the long-term impact of modified biochars on environmental quality and the health of organisms living in the ecosystem.

7. Conclusions and future research outlook

Determining the pedogenic and anthropogenic sources of Ni and understanding factors controlling their biogeochemical behaviors are of paramount importance in developing a suitable tool for remediating Ni contamination. The biogeochemical behaviors of Ni vary greatly with environmental conditions/characteristics, including the dynamics of reduction-oxidation, biological transformation, pH, presence of multi-contaminants, the chemistry of S, Fe, Mn and Al, and the content of dissolved aliphatic and aromatic carbon. The biogeochemical behavior of Ni determines the speciation and the corresponding

mobility and toxicity of Ni in the soil, bioaccumulation in plants, and its transfer to the water system. Application of biochar to Ni contaminated soil and water systems has been proven to be an effective remediation tool; however, biochar effects on the mobility and phytoavailability of Ni depend on the concentration and geochemistry of Ni, the environment condition, and biochar characteristics. The biochar to be used for a specific site for remediation of Ni contamination needs to be carefully selected, by using a suitable feedstock, production condition, and pre- and post-modification treatments. Research on optimization of biochar properties specifically for the immobilization of Ni in soil and water is gaining increasing attention. Based on our review of the literature, the following aspects need to be further studied before operational application of biochars for the remediation of Ni contaminated soil and water environments and to close the knowledge gap in this area:

1. Environmental risk assessment must be conducted before considering large-scale application of biochar to ensure that biochar application does not pose any risk to the ecosystem. Different parameters, including the concentrations of potentially toxic elements, polycyclic aromatic hydrocarbons, polychlorinated dibenzodioxins and dibenzofurans, should be determined in the soil/water to be treated with different types of pristine or modified biochars in remediation experiments.
2. Systematic investigations on the feedstock type and production condition to use for the production of biochars that are effective for remediating Ni contaminated environments are required. The optimization of biochar functionality via modification methods would maximize the potential of biochars for the remediation of soil and water contaminated with Ni. Different modification methods, however, have not been systematically tested and validated for the remediation of Ni contamination. Investigation on the long-term impact of modified biochars on

environmental quality and the health of organisms in the environment is urgently needed.

3. Soil and water contaminated with Ni often contain other co-existing organic/inorganic contaminants. Competitive adsorption among toxic elements (especially among inorganic pollutants) and Ni might occur on binding sites in biochars. Therefore, the sorption efficiency of modified biochars for Ni in multi-contaminant environments should be tested.
4. The redox-mediated transformation of Ni and its interactions among the dissolved, colloidal, and solid phases as affected by biochar application is still poorly understood. The synergistic effects of dynamic redox conditions and biochar application were found to influence the Ni forms, mobility, and phytoavailability in wetlands, and floodplains, however, systematic investigations are required in the future to improve our understanding of those transformation processes under different soil and biochar types.
5. Surface oxidation of biochars via alkali/acidic activation facilitate the binding of Ni on biochar. The O-containing functional groups on biochar surfaces are responsible for forming specific metal-ligand complexation in soil and water systems. However, the impact of this activation/modification method depends upon the feedstock type and production condition of the produced biochar, and thus, studies on elucidating the suitable biochar for these treatments will maximize the benefits of the biochar activation on Ni retention in soil and aquatic systems.
6. Meta-analysis of results in the published literature on biochar application effects on the remediation of Ni contamination is highly recommended and urgently needed. None of the published meta-analysis studies has focused specifically on the induced changes and immobilization efficiency of Ni in biochar amended soil and water systems.

7. Finally, engineered/designer biochars have higher removal efficiencies for Ni in the soil/water environment than pristine biochars. However, further research is needed to validate the efficiency and superiority of engineered/designer biochars in remediating Ni contamination in the soil/water environment. The strong binding of Ni ions onto pristine biochar matrices reduces the desorbability of the adsorbates, and hence the reusability of the spent biochar. Biochar functionalization, therefore, is needed to reduce the bond energy of sorbed Ni ions on biochar, thereby increasing the desorbability and recyclability of the spent biochar, particularly for the remediation of Ni contaminated water, where the reuse of the spent biochar would be highly desirable.

Acknowledgements

This study was supported by the National Natural Science Foundation of China (42050410315, 41877085, 41877088), the Research and Development Fund of Zhejiang A&F University (2018FR005, 2018FR006), the Open Research Fund Program of the State Key Laboratory of Subtropical Silviculture, Zhejiang A&F University (ZY20180301, ZY20180205), and the Zhejiang Postdoctoral Research Program (20120200001).

CRediT authorship contribution statement

Ali El-Naggar: Conceptualization, investigation, collecting review of literature, Writing - original draft. **Naveed Ahmed**: Visualization, Writing - review & editing. **Ahmed Mosa**: Visualization, Writing - review & editing. **Nabeel Khan Niazi**: Writing - review & editing. **Balal Yousaf**: Writing - review & editing. **Anket Sharma**: Writing - review & editing. **Binoy Sarkar**: Writing - review & editing. **Yanjiang Cai**: Visualization, Validation, Writing - review & editing. **Scott X. Chang**: Conceptualization, Funding acquisition, Supervision, Validation, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests.

References

Abbas, Q., Yousaf, B., Amina, Ali, M.U., Munir, M.A.M., El-Naggar, A., Rinklebe, J., Naushad, M., 2020. Transformation pathways and fate of engineered nanoparticles (ENPs) in distinct interactive environmental compartments: A review. Environ. Int. 138, 105646. https://doi.org/10.1016/j.envint.2020.105646

Adiloğlu, S., Turgut Sağlam, M., Adiloğlu, A., Süme, A., 2016. Phytoremediation of nickel (Ni) from agricultural soils using canola (Brassica napus L.). Desalin. Water Treat. 57, 2383–2388. https://doi.org/10.1080/19443994.2014.994110

Ahmad, M., Rajapaksha, A.U., Lim, J.E., Zhang, M., Bolan, N., Mohan, D., Vithanage, M., Lee, S.S., Ok, Y.S., 2014. Biochar as a sorbent for contaminant management in soil and water: a review. Chemosphere 99, 19–33. https://doi.org/10.1016/j.chemosphere.2013.10.071

Alam, M.S., Gorman-Lewis, D., Chen, N., Flynn, S.L., Ok, Y.S., Konhauser, K.O., Alessi, D.S., 2018. Thermodynamic analysis of nickel(II) and zinc(II) adsorption to biochar. Environ. Sci. Technol. 52, 6246–6255. https://doi.org/10.1021/acs.est.7b06261

Albanese, S., Sadeghi, M., Lima, A., Cicchella, D., Dinelli, E., Valera, P., Falconi, M., Demetriades, A., De Vivo, B., Etc., 2015. GEMAS: Cobalt, Cr, Cu and Ni distribution in agricultural and grazing land soil of Europe. J. Geochem. Explor 154, 81–93. https://doi.org/10.1016/j.gexplo.2015.01.004

Amin, M.T., Alazba, A.A., Shafiq, M., 2019. Comparative sorption of nickel from an aqueous solution using biochar derived from banana and orange peel using a batch system: Kinetic and isotherm models. Arab. J. Sci. Eng. 44, 10105–10116.

https://doi.org/10.1007/s13369-019-03907-6

An, Q., Jiang, Y.Q., Nan, H.Y., Yu, Y., Jiang, J.N., 2019. Unraveling sorption of nickel from aqueous solution by $KMnO_4$ and KOH-modified peanut shell biochar: Implicit mechanism. Chemosphere 214, 846–854. https://doi.org/10.1016/j.chemosphere.2018.10.007

Anoop Krishnan, K., Sreejalekshmi, K.G., Baiju, R.S., 2011. Nickel(II) adsorption onto biomass based activated carbon obtained from sugarcane bagasse pith. Bioresour. Technol. 102, 10239–10247. https://doi.org/10.1016/j.biortech.2011.08.069

Antoniadis, V., Levizou, E., Shaheen, S.M., Ok, Y.S., Sebastian, A., Baum, C., Prasad, M.N.V., Wenzel, W.W., Rinklebe, J., 2017. Trace elements in the soil-plant interface: Phytoavailability, translocation, and phytoremediation–A review. Earth-Science Rev. 171, 621–645. https://doi.org/10.1016/J.EARSCIREV.2017.06.005

Babaahmadifooladi, M., Jacxsens, L., De Meulenaer, B., Du Laing, G., 2020. Nickel in foods sampled on the Belgian market: identification of potential contamination sources. Food Addit. Contam. - Part A Chem. Anal. Control. Expo. Risk Assess. 37, 607–621. https://doi.org/10.1080/19440049.2020.1714751

Bandara, T., Franks, A., Xu, J., Bolan, N., Wang, H., Tang, C., 2020. Chemical and biological immobilization mechanisms of potentially toxic elements in biochar-amended soils. Crit. Rev. Environ. Sci. Technol. 50, 903–978. https://doi.org/10.1080/10643389.2019.1642832

Beesley, L., Moreno-Jiménez, E., Gomez-Eyles, J.L., Harris, E., Robinson, B., Sizmur, T., 2011. A review of biochars' potential role in the remediation, revegetation and restoration of contaminated soils. Environ. Pollut. 159, 3269–3282. https://doi.org/10.1016/j.envpol.2011.07.023

Biswas, S., Meikap, B.C., Sen, T.K., 2019. Adsorptive removal of aqueous phase copper (Cu^{2+}) and nickel (Ni^{2+}) metal ions by synthesized biochar–biopolymeric hybrid

adsorbents and process optimization by response surface methodology (RSM). Water. Air. Soil Pollut. 230, 1–23. https://doi.org/10.1007/s11270-019-4258-y

Bogusz, A., Nowak, K., Stefaniuk, M., Dobrowolski, R., Oleszczuk, P., 2017. Synthesis of biochar from residues after biogas production with respect to cadmium and nickel removal from wastewater. J. Environ. Manage. 201, 268–276. https://doi.org/10.1016/j.jenvman.2017.06.019

Bogusz, A., Oleszczuk, P., 2018. Sequential extraction of nickel and zinc in sewage sludge- or biochar/sewage sludge-amended soil. Sci. Total Environ. 636, 927–935. https://doi.org/10.1016/j.scitotenv.2018.04.072

Bolan, N., Kunhikrishnan, A., Thangarajan, R., Kumpiene, J., Park, J., Makino, T., Kirkham, M.B., Scheckel, K., 2014. Remediation of heavy metal(loid)s contaminated soils - To mobilize or to immobilize? J. Hazard. Mater. 266, 141–166. https://doi.org/10.1016/j.jhazmat.2013.12.018

Cameron, V., Vance, D., 2014. Heavy nickel isotope compositions in rivers and the oceans. Geochim. Cosmochim. Acta 128, 195–211. https://doi.org/10.1016/j.gca.2013.12.007

Chaiyaraksa, C., Boonyakiat, W., Bukkontod, W., Ngakom, W., 2019. Adsorption of copper (II) and nickel (II) by chemical modified magnetic biochar derived from eichhornia crassipes. Environment Asia 12, 14–23. https://doi.org/10.14456/ea.2019.23

Chemerys, V., Baltrėnaitė, E., 2018. A review of lignocellulosic biochar modification towards enhanced biochar selectivity and adsorption capacity of potentially toxic elements. Ukr. J. Ecol. 8, 21–32. https://doi.org/10.15421/2018_183

Cheng, S.Y., Show, P.L., Lau, B.F., Chang, J.S., Ling, T.C., 2019. New prospects for modified algae in heavy metal adsorption. Trends Biotechnol. 37, 1255–1268. https://doi.org/10.1016/j.tibtech.2019.04.007

Dähn, R., Schneidegger, A.M., Manceau, A., 2004. Uptake mechanisms of Ni(II) on montmorillonite as determined by X-ray absorption spectroscopy (abstr.). Goldschmidt.

Deng, R., Huang, D., Wan, J., Xue, W., Wen, X., Liu, X., Chen, S., Lei, L., Zhang, Q., 2020. Recent advances of biochar materials for typical potentially toxic elements management in aquatic environments: A review. J. Clean. Prod. 255, 119523. https://doi.org/10.1016/j.jclepro.2019.119523

Deng, T.H.B., van der Ent, A., Tang, Y.T., Sterckeman, T., Echevarria, G., Morel, J.L., Qiu, R.L., 2018. Nickel hyperaccumulation mechanisms: a review on the current state of knowledge. Plant Soil. 423, 1–11. https://doi.org/10.1007/s11104-017-3539-8

Deng, Y., Huang, S., Laird, D.A., Wang, X., Meng, Z., 2019. Adsorption behaviour and mechanisms of cadmium and nickel on rice straw biochars in single- and binary-metal systems. Chemosphere 218, 308–318. https://doi.org/10.1016/j.chemosphere.2018.11.081

Derakhshan Nejad, Z., Jung, M.C., Kim, K.H., 2018. Remediation of soils contaminated with heavy metals with an emphasis on immobilization technology. Environ. Geochem. Health. 40, 927–953. https://doi.org/10.1007/s10653-017-9964-z

Di Palma, L., Gueye, M.T., Petrucci, E., 2015. Hexavalent chromium reduction in contaminated soil: A comparison between ferrous sulphate and nanoscale zero-valent iron. J. Hazard. Mater. 281, 70–76. https://doi.org/10.1016/j.jhazmat.2014.07.058

Ding, Z., Hu, X., Wan, Y., Wang, S., Gao, B., 2016. Removal of lead, copper, cadmium, zinc, and nickel from aqueous solutions by alkali-modified biochar: Batch and column tests. J. Ind. Eng. Chem. 33, 239–245. https://doi.org/10.1016/j.jiec.2015.10.007

Donat, J.R., Lao, K.A., Bruland, K.W., 1994. Speciation of dissolved copper and nickel in South San Francisco Bay: a multi-method approach. Anal. Chim. Acta 284, 547–571. https://doi.org/10.1016/0003-2670(94)85061-5

Echevarria, Guillaume, 2021. Genesis and behaviour of ultramafic soils and consequences for nickel biogeochemistry, in: van der Ent, A., Baker, A.J., Echevarria, G., Simonnot, M., Morel, J.L. (Eds.), Agromining: Farming for Metals. Mineral Resource Reviews.

Springer, Cham, pp. 215–238. https://doi.org/10.1007/978-3-030-58904-2_11

Eissa, M.A., 2019. Effect of cow manure biochar on heavy metals uptake and translocation by zucchini (Cucurbita pepo L). Arab. J. Geosci. 12, 48. https://doi.org/10.1007/s12517-018-4191-1

El-Naggar, A., El-Naggar, A.H., Shaheen, S.M., Sarkar, B., Chang, S.X., Tsang, D.C.W., Rinklebe, J., Ok, Y.S., 2019a. Biochar composition-dependent impacts on soil nutrient release, carbon mineralization, and potential environmental risk: A review. J. Environ. Manage. 241, 1–10. https://doi.org/10.1016/j.jenvman.2019.02.044

El-Naggar, A., Lee, M.H., Hur, J., Lee, Y.H., Igalavithana, A.D., Shaheen, S.M., Ryu, C., Rinklebe, J., Tsang, D.C.W., Ok, Y.S., 2020. Biochar-induced metal immobilization and soil biogeochemical process: An integrated mechanistic approach. Sci. Total Environ. 698, 134112. https://doi.org/10.1016/j.scitotenv.2019.134112

El-Naggar, A., Lee, S.S., Awad, Y.M., Yang, X., Ryu, C., Rizwan, M., Rinklebe, J., Tsang, D.C.W., Ok, Y.S., 2018a. Influence of soil properties and feedstocks on biochar potential for carbon mineralization and improvement of infertile soils. Geoderma 332, 100–108. https://doi.org/10.1016/j.geoderma.2018.06.017

El-Naggar, A., Lee, S.S., Rinklebe, J., Farooq, M., Song, H., Sarmah, A.K., Zimmerman, A.R., Ahmad, M., Shaheen, S.M., Ok, Y.S., 2019b. Biochar application to low fertility soils: A review of current status, and future prospects. Geoderma 337, 536–554. https://doi.org/10.1016/j.geoderma.2018.09.034

El-Naggar, A., Rajapaksha, A.U., Shaheen, S.M., Rinklebe, J., Ok, Y.S., 2018b. Potential of biochar to immobilize nickel in contaminated soils, in: Nickel in Soils and Plants. CRC Press, pp. 293–318. https://doi.org/10.1201/9781315154664-13

El-Naggar, A., Shaheen, S.M., Chang, S.X., Hou, D., Ok, Y.S., Rinklebe, J., 2021. Biochar surface functionality plays a vital role in (im)mobilization and phytoavailability of soil vanadium. ACS Sustain. Chem. Eng. 9, 6864–6874.

https://doi.org/10.1021/acssuschemeng.1c01656

El-Naggar, A., Shaheen, S.M., Ok, Y.S., Rinklebe, J., 2018c. Biochar affects the dissolved and colloidal concentrations of Cd, Cu, Ni, and Zn and their phytoavailability and potential mobility in a mining soil under dynamic redox-conditions. Sci. Total Environ. 624, 1059–1071. https://doi.org/10.1016/j.scitotenv.2017.12.190

El-Naggar, A., Shaheen, S.M.S.M., Hseu, Z.-Y.Z.-Y., Wang, S.-L.S.-L., Ok, Y.S.Y.S., Rinklebe, J., 2019c. Release dynamics of As, Co, and Mo in a biochar treated soil under pre-definite redox conditions. Sci. Total Environ. 657, 686–695. https://doi.org/10.1016/J.SCITOTENV.2018.12.026

Elkhlifi, Z., Kamran, M., Maqbool, A., El-Naggar, A., Ifthikar, J., Parveen, A., Bashir, S., Rizwan, M., Mustafa, A., Irshad, S., Ali, S., Chen, Z., 2021. Phosphate-lanthanum coated sewage sludge biochar improved the soil properties and growth of ryegrass in an alkaline soil. Ecotoxicol. Environ. Saf. 216, 112173. https://doi.org/10.1016/j.ecoenv.2021.112173

Evans, L.J., Barabash, S.J., 2010. Molybdenum, silver, thallium and vanadium, in: Trace Elements in Soils. pp. 515–549. https://doi.org/10.1002/9781444319477.ch22

Fiyadh, S.S., AlSaadi, M.A., Jaafar, W.Z., AlOmar, M.K., Fayaed, S.S., Mohd, N.S., Hin, L.S., El-Shafie, A., 2019. Review on heavy metal adsorption processes by carbon nanotubes. J. Clean. Prod. 230, 783–793. https://doi.org/10.1016/j.jclepro.2019.05.154

Gall, L., Williams, H.M., Siebert, C., Halliday, A.N., Herrington, R.J., Hein, J.R., 2013. Nickel isotopic compositions of ferromanganese crusts and the constancy of deep ocean inputs and continental weathering effects over the Cenozoic. Earth Planet. Sci. Lett. 375, 148–155. https://doi.org/10.1016/j.epsl.2013.05.019

Gazi, M., Oladipo, A.A., Azalok, K.A., 2018. Highly efficient and magnetically separable palm seed-based biochar for the removal of nickel. Sep. Sci. Technol. 53, 1124–1131. https://doi.org/10.1080/01496395.2017.1340955

Gonnelli, C., Renella, G., 2013. Chromium and nickel, in: Alloway B. (Ed.), Heavy Metals in Soils: Trace Metals and Metalloids in Soils and Their Bioavailability. Springer, Dordrecht. https://doi.org/10.1007/978-94-007-4470-7_11

Guan, C.Y., Tseng, Y.H., Tsang, D.C.W., Hu, A., Yu, C.P., 2019. Wetland plant microbial fuel cells for remediation of hexavalent chromium contaminated soils and electricity production. J. Hazard. Mater. 365, 137–145. https://doi.org/10.1016/j.jhazmat.2018.10.086

He, L., Zhong, H., Liu, G., Dai, Z., Brookes, P.C., Xu, J., 2019. Remediation of heavy metal contaminated soils by biochar: Mechanisms, potential risks and applications in China. Environ. Pollut. 252, 846–855. https://doi.org/10.1016/j.envpol.2019.05.151

Hernández-Quiroz, M., Herre, A., Cram, S., de León, C.P., Siebe, C., 2012. Pedogenic, lithogenic - or anthropogenic origin of Cr, Ni and V in soils near a petrochemical facility in Southeast Mexico. Catena 93, 49–57. https://doi.org/10.1016/j.catena.2012.01.005

Higashikawa, F.S., Conz, R.F., Colzato, M., Cerri, C.E.P., Alleoni, L.R.F., 2016. Effects of feedstock type and slow pyrolysis temperature in the production of biochars on the removal of cadmium and nickel from water. J. Clean. Prod. 137, 965–972. https://doi.org/10.1016/j.jclepro.2016.07.205

Hooda, P.S., 2010. Trace Elements in Soils, Trace Elements in Soils. Wiley. https://doi.org/10.1002/9781444319477

Hu, X., He, M., Kong, L., 2015. Photopromoted oxidative dissolution of stibnite. Appl. Geochemistry 61, 53–61. https://doi.org/10.1016/j.apgeochem.2015.05.014

Hu, X., Xue, Y., Liu, L., Zeng, Y., Long, L., 2018. Preparation and characterization of Na_2S-modified biochar for nickel removal. Environ. Sci. Pollut. Res. 25, 9887–9895. https://doi.org/10.1007/s11356-018-1298-6

Hussain, J., Husain, I., Arif, M., Gupta, N., 2017. Studies on heavy metal contamination in

Godavari river basin. Appl. Water Sci. 7, 4539–4548.

Ibrahim, M., Li, G., Chan, F.K.S., Kay, P., Liu, X.-X., Firbank, L., Xu, Y.-Y., 2019. Biochars effects potentially toxic elements and antioxidant enzymes in *Lactuca sativa* L. grown in multi-metals contaminated soil. Environ. Technol. Innov. 15, 100427. https://doi.org/10.1016/j.eti.2019.100427

Kabata-Pendias, A., 2011. Trace Elements in Soils and Plants. CRC Press. ISBN 9781420093681

Khan, M.N., Mobin, M., Abbas, Z.K., Alamri, S.A., 2017. Fertilizers and their contaminants in soils, surface and groundwater, in: Encyclopedia of the Anthropocene. Elsevier, pp. 225–240. https://doi.org/10.1016/B978-0-12-809665-9.09888-8

Laribi, A., Shand, C., Wendler, R., Mouhouche, B., Colinet, G., 2019. Concentrations and sources of Cd, Cr, Cu, Fe, Ni, Pb and Zn in soil of the Mitidja plain, Algeria. Toxicol. Environ. Chem. 101, 59–74. https://doi.org/10.1080/02772248.2019.1619744

Li, C., Zhou, K., Qin, W., Tian, C., Qi, M., Yan, X., Han, W., 2019. A Review on heavy metals contamination in soil: Effects, sources, and remediation techniques. Soil Sediment Contam. 28, 380–394. https://doi.org/10.1080/15320383.2019.1592108

Lonappan, L., Liu, Y., Rouissi, T., Brar, S.K., Surampalli, R.Y., 2019. Development of biochar-based green functional materials using organic acids for environmental applications. J. Clean. Prod. 244, 118841. https://doi.org/10.1016/j.jclepro.2019.118841

Lyu, H., Gao, B., He, F., Zimmerman, A.R., Ding, C., Huang, H., Tang, J., 2018. Effects of ball milling on the physicochemical and sorptive properties of biochar: Experimental observations and governing mechanisms. Environ. Pollut. 233, 54–63. https://doi.org/10.1016/j.envpol.2017.10.037

Mahdi, Z., El Hanandeh, A., Yu, Q.J., 2019. Preparation, characterization and application of surface modified biochar from date seed for improved lead, copper, and nickel removal from aqueous solutions. J. Environ. Chem. Eng. 7, 103379.

https://doi.org/10.1016/j.jece.2019.103379

Man, Y., Wang, B., Wang, J., Slaný, M., Yan, H., Li, P., El-Naggar, A., Shaheen, S.M., Rinklebe, J., Feng, X., 2021. Use of biochar to reduce mercury accumulation in *Oryza sativa* L: A trial for sustainable management of historically polluted farmlands. Environ. Int. 153, 106527. https://doi.org/10.1016/j.envint.2021.106527

Manceau, A., Tamura, N., Marcus, M.A., MacDowell, A.A., Celestre, R.S., Sublett, R.E., Sposito, G., Padmore, H.A., 2002. Deciphering Ni sequestration in soil ferromanganese nodules by combining X-ray fluorescene, absorption, and diffraction at micrometer scales of resolution. Am. Mineral. 87, 1494–1499. https://doi.org/10.2138/am-2002-1028

Manceau, A., Tommaseo, C., Rihs, S., Geoffroy, N., Chateigner, D., Schlegel, M., Tisserand, D., Marcus, M.A., Tamura, N., Chen, Z.S., 2005. Natural speciation of Mn, Ni, and Zn at the micrometer scale in a clayey paddy soil using X-ray fluorescence, absorption, and diffraction. Geochim. Cosmochim. Acta 69, 4007–4034. https://doi.org/10.1016/j.gca.2005.03.018

Mandal, R., Hassan, N.M., Murimboh, J., Chakrabarti, C.L., Back, M.H., Rahayu, U., Lean, D.R.S., 2002. Chemical speciation and toxicity of nickel species in natural waters from the sudbury area (Canada). Environ. Sci. Technol. 36, 1477–1484. https://doi.org/10.1021/es015622e

Massoura, S.T., Echevarria, G., Becquer, T., Ghanbaja, J., Leclerc-Cessac, E., Morel, J.-L., 2006. Control of nickel availability by nickel bearing minerals in natural and anthropogenic soils. Geoderma 136, 28–37. https://doi.org/https://doi.org/10.1016/j.geoderma.2006.01.008

Mosa, A., El-Banna, M.F., Gao, B., 2016. Biochar filters reduced the toxic effects of nickel on tomato (*Lycopersicon esculentum* L.) grown in nutrient film technique hydroponic system. Chemosphere. 149, 254-62. https://doi.org/10.1016/j.chemosphere.2016.01.104

Mourgela, R.N., Regkouzas, P., Pellera, F.M., Diamadopoulos, E., 2020. Ni(II) adsorption on biochars produced from different types of biomass. Water. Air. Soil Pollut. 231, 1–16. https://doi.org/10.1007/s11270-020-04591-1

Munir, M.A.M., Liu, G., Yousaf, B., Mian, M.M., Ali, M.U., Ahmed, R., Cheema, A.I., Naushad, M., 2020. Contrasting effects of biochar and hydrothermally treated coal gangue on leachability, bioavailability, speciation and accumulation of heavy metals by rapeseed in copper mine tailings. Ecotoxicol. Environ. Saf. 191, 110244. https://doi.org/10.1016/j.ecoenv.2020.110244

Nawab, J., Ghani, J., Khan, S., Xiaoping, W., 2018. Minimizing the risk to human health due to the ingestion of arsenic and toxic metals in vegetables by the application of biochar, farmyard manure and peat moss. J. Environ. Manage. 214, 172–183. https://doi.org/10.1016/j.jenvman.2018.02.093

Palansooriya, K.N., Shaheen, S.M., Chen, S.S., Tsang, D.C.W., Hashimoto, Y., Hou, D., Bolan, N.S., Rinklebe, J., Ok, Y.S., 2020. Soil amendments for immobilization of potentially toxic elements in contaminated soils: A critical review. Environ. Int. 134, 105046. https://doi.org/10.1016/j.envint.2019.105046

Panagopoulos, I., Karayannis, A., Kollias, K., Xenidis, A., Papassiopi, N., 2015. Investigation of potential soil contamination with Cr and Ni in four metal finishing facilities at Asopos industrial area. J. Hazard. Mater. 281, 20–26. https://doi.org/10.1016/j.jhazmat.2014.07.040

Parades-Aguilar, J., Reyes-Martínez, V., Bustamante, G., Almendáriz-Tapia, F.J., Martínez-Meza, G., Vílchez-Vargas, R., Link, A., Certucha-Barragán, M.T., Calderón, K., 2021. Removal of nickel(II) from wastewater using a zeolite-packed anaerobic bioreactor: Bacterial diversity and community structure shifts. J. Environ. Manage. 279, 111558. https://doi.org/10.1016/j.jenvman.2020.111558

Poznanović Spahić, M.M., Sakan, S.M., Glavaš-Trbić, B.M., Tančić, P.I., Škrivanj, S.B.,

Kovačević, J.R., Manojlović, D.D., 2019. Natural and anthropogenic sources of chromium, nickel and cobalt in soils impacted by agricultural and industrial activity (Vojvodina, Serbia). J. Environ. Sci. Heal. - Part A Toxic/Hazardous Subst. Environ. Eng. 54, 219–230. https://doi.org/10.1080/10934529.2018.1544802

Pyle, G., Couture, P., 2011. Nickel, in: Wood, C.M., Farrell A.P., Brauner, C.J. (Eds.), Homeostasis and Toxicology of Essential Metals. Fish Physiology. Academic Press. 31, 253–289. https://doi.org/10.1016/S1546-5098(11)31005-9

Quantin, C., Ettler, V., Garnier, J., Šebek, O., 2008. Sources and extractibility of chromium and nickel in soil profiles developed on Czech serpentinites. Comptes Rendus - Geosci. 340, 872–882. https://doi.org/10.1016/j.crte.2008.07.013

Rajapaksha, A.U., Alam, M.S., Chen, N., Alessi, D.S., Igalavithana, A.D., Tsang, D.C.W., Ok, Y.S., 2018. Removal of hexavalent chromium in aqueous solutions using biochar: Chemical and spectroscopic investigations. Sci. Total Environ. 625, 1567–1573. https://doi.org/10.1016/j.scitotenv.2017.12.195

Rajapaksha, A.U., Chen, S.S., Tsang, D.C.W., Zhang, M., Vithanage, M., Mandal, S., Bolan, N.S., Sik Ok, Y., 2016. Engineered/designer biochar for contaminant removal/immobilization from soil and water: Potential and implication of biochar modification. Chemosphere 148, 276–291. https://doi.org/10.1016/j.chemosphere.2016.01.043

Ratié, G., Jouvin, D., Garnier, J., Rouxel, O., Miska, S., Guimarães, E., Cruz Vieira, L., Sivry, Y., Zelano, I., Montarges-Pelletier, E., Thil, F., Quantin, C., 2015a. Nickel isotope fractionation during tropical weathering of ultramafic rocks. Chem. Geol. 402, 68–76. https://doi.org/10.1016/j.chemgeo.2015.02.039

Ratié, G., Quantin, C., Jouvin, D., Calmels, D., Ettler, V., Sivry, Y., Vieira, L.C., Ponzevera, E., Garnier, J., 2015b. Nickel isotope fractionation during laterite Ni ore smelting and refining: Implications for tracing the sources of Ni in smelter-affected soils. Appl.

Geochemistry 64, 136–145. https://doi.org/10.1016/j.apgeochem.2015.09.005

Rehman, M.Z.U., Rizwan, M., Ali, S., Fatima, N., Yousaf, B., Naeem, A., Sabir, M., Ahmad, H.R., Ok, Y.S., 2016. Contrasting effects of biochar, compost and farm manure on alleviation of nickel toxicity in maize (*Zea mays* L.) in relation to plant growth, photosynthesis and metal uptake. Ecotoxicol. Environ. Saf. 133, 218–225. https://doi.org/10.1016/j.ecoenv.2016.07.023

Rinklebe, J., Antoniadis, V., Shaheen, S.M., Rosche, O., Altermann, M., 2019. Health risk assessment of potentially toxic elements in soils along the Central Elbe River, Germany. Environ. Int. 126, 76–88. https://doi.org/10.1016/J.ENVINT.2019.02.011

Rinklebe, J., Shaheen, S.M., 2017. Redox chemistry of nickel in soils and sediments: A review. Chemosphere 179, 265–278. https://doi.org/10.1016/j.chemosphere.2017.02.153

Rinklebe, J., Shaheen, S.M., El-Naggar, A., Wang, H., Du Laing, G., Alessi, D.S., Sik Ok, Y., 2020. Redox-induced mobilization of Ag, Sb, Sn, and Tl in the dissolved, colloidal and solid phase of a biochar-treated and un-treated mining soil. Environ. Int. 140, 105754. https://doi.org/10.1016/j.envint.2020.105754

Rinklebe, J., Shaheen, S.M., Frohne, T., 2016. Amendment of biochar reduces the release of toxic elements under dynamic redox conditions in a contaminated floodplain soil. Chemosphere 142, 41–47. https://doi.org/10.1016/j.chemosphere.2015.03.067

Rodríguez-Vila, A., Covelo, E.F., Forján, R., Asensio, V., 2015. Recovering a copper mine soil using organic amendments and phytomanagement with *Brassica juncea* L. J. Environ. Manage. 147, 73–80. https://doi.org/10.1016/j.jenvman.2014.09.011

Russell, R.M., Beard, J.L., Cousins, R.J., Dunn, J.T., Ferland, G., Hambidge, K.M., Lynch, S., Penland, J.G., Ross, A.C., Stoecker, B.J., 2001. Dietary reference intakes for vitamin A, vitamin K, arsenic, boron, chromium, copper, iodine, iron, manganese, molybdenum, nickel, silicon, vanadium, and zinc. A Rep. Panel Micronutr. Subcomm.

Up. Ref. Levels Nutr. Interpret. Uses Diet. Ref. Intakes, Standing Comm. Sci. Eval. Diet. Ref. Intakes Food Nutr.

Samantaray, P.K., Madras, G., Bose, S., 2019. Microbial biofilm membranes for water remediation and photobiocatalysis. ACS Symp. Ser. 1329, 321–351. https://doi.org/10.1021/bk-2019-1329.ch014

Santhosh, C., Malathi, A., Daneshvar, E., Kollu, P., Bhatnagar, A., 2018. Photocatalytic degradation of toxic aquatic pollutants by novel magnetic 3D-TiO_2@HPGA nanocomposite. Sci. Rep. 8, 1–15. https://doi.org/10.1038/s41598-018-33818-9

Schaumlöffel, D., 2012. Nickel species: Analysis and toxic effects. J. Trace Elem. Med. Biol. 26, 1–6. https://doi.org/https://doi.org/10.1016/j.jtemb.2012.01.002

Shaaban, M., Van Zwieten, L., Bashir, S., Younas, A., Núñez-Delgado, A., Chhajro, M.A., Kubar, K.A., Ali, U., Rana, M.S., Mehmood, M.A., Hu, R., 2018. A concise review of biochar application to agricultural soils to improve soil conditions and fight pollution. J. Environ. Manage. 228, 429–440. https://doi.org/10.1016/j.jenvman.2018.09.006

Shaheen, S.M., Antić-Mladenović, S., Wang, S.-L., Niazi, N.K., Tsadilas, C.D., Ok, Y.S., Rinklebe, J., 2018a. Nickel mobilization/immobilization and phytoavailability in soils as affected by organic and inorganic amendments, in: Tsadilas, C., Rinklebe, J. (Eds.), Nickel in Soils and Plants. CRC Press, Taylor & Francis Group, New York, USA, pp. 265–292. https://doi.org/10.1201/9781315154664-12

Shaheen, S.M., El-Naggar, A., Antoniadis, V., Moghanm, F.S., Zhang, Z., Tsang, D.C.W., Ok, Y.S., Rinklebe, J., 2020. Release of toxic elements in fishpond sediments under dynamic redox conditions: Assessing the potential environmental risk for a safe management of fisheries systems and degraded waterlogged sediments. J. Environ. Manage. 255, 109778. https://doi.org/10.1016/j.jenvman.2019.109778

Shaheen, S.M., El-Naggar, A., Wang, J., Hassan, N.E.E., Niazi, N.K., Wang, H., Tsang, D.C.W., Ok, Y.S., Bolan, N., Rinklebe, J., 2018b. Biochar as an (im)mobilizing agent

for the potentially toxic elements in contaminated soils, in: Biochar from Biomass and Waste. Elsevier, pp. 255–274. https://doi.org/10.1016/b978-0-12-811729-3.00014-5

Shaheen, S.M., Rinklebe, J., Selim, M.H., 2015. Impact of various amendments on immobilization and phytoavailability of nickel and zinc in a contaminated floodplain soil. Int. J. Environ. Sci. Technol. 12, 2765–2776. https://doi.org/10.1007/s13762-014-0713-x

Shen, Z., Som, A.M., Wang, F., Jin, F., McMillan, O., Al-Tabbaa, A., 2016. Long-term impact of biochar on the immobilisation of nickel (II) and zinc (II) and the revegetation of a contaminated site. Sci. Total Environ. 542, 771–776. https://doi.org/10.1016/j.scitotenv.2015.10.057

Shen, Z., Zhang, Y., McMillan, O., Jin, F., Al-Tabbaa, A., 2017. Characteristics and mechanisms of nickel adsorption on biochars produced from wheat straw pellets and rice husk. Environ. Sci. Pollut. Res. 24, 12809–12819. https://doi.org/10.1007/s11356-017-8847-2

Shen, Z., Zhang, Yunhui, Jin, F., Alessi, D.S., Zhang, Yiyun, Wang, F., McMillan, O., Al-Tabbaa, A., 2018. Comparison of nickel adsorption on biochars produced from mixed softwood and Miscanthus straw. Environ. Sci. Pollut. Res. 25, 14626–14635. https://doi.org/10.1007/s11356-018-1674-2

Shi, Y., Zhang, T., Ren, H., Kruse, A., Cui, R., 2018. Polyethylene imine modified hydrochar adsorption for chromium (VI) and nickel (II) removal from aqueous solution. Bioresour. Technol. 247, 370–379. https://doi.org/10.1016/j.biortech.2017.09.107

Shi, Z., Peltier, E., Sparks, D.L., 2012. Kinetics of Ni sorption in soils: Roles of soil organic matter and Ni precipitation. Environ. Sci. Technol. 46, 2212–2219. https://doi.org/10.1021/es202376c

Shin, W.S., 2017. Adsorption characteristics of phenol and heavy metals on biochar from Hizikia fusiformis. Environ. Earth Sci. 76, 782. https://doi.org/10.1007/s12665-017-

7125-4

Šillerová, H., Chrastný, V., Vítková, M., Francová, A., Jehlička, J., Gutsch, M.R., Kocourková, J., Aspholm, P.E., Nilsson, L.O., Berglen, T.F., Jensen, H.K.B., Komárek, M., 2017. Stable isotope tracing of Ni and Cu pollution in North-East Norway: Potentials and drawbacks. Environ. Pollut. 228, 149–157. https://doi.org/10.1016/j.envpol.2017.05.030

Singh, J., Sharma, M., Basu, S., 2018. Heavy metal ions adsorption and photodegradation of remazol black XP by iron oxide/silica monoliths: Kinetic and equilibrium modelling. Adv. Powder Technol. 29, 2268–2279. https://doi.org/10.1016/j.apt.2018.06.011

Sugawara, E., Nikaido, H., 2014. Properties of AdeABC and AdeIJK efflux systems of Acinetobacter baumannii compared with those of the AcrAB-TolC system of Escherichia coli. Antimicrob. Agents Chemother. 58, 7250–7257. https://doi.org/10.1128/AAC.03728-14

Terzano, R., Spagnuolo, M., Vekemans, B., De Nolf, W., Janssens, K., Falkenberg, G., Fiore, S., Ruggiero, P., 2007. Assessing the origin and fate of Cr, Ni, Cu, Zn, Pb, and V in industrial polluted soil by combined microspectroscopic techniques and bulk extraction methods. Environ. Sci. Technol. 41, 6762–6769. https://doi.org/10.1021/es070260h

Tessier, A., Campbell, P.G.C., Bisson, M., 1979. Sequential extraction procedure for the speciation of particulate trace metals. Anal. Chem. 51, 844–851. https://doi.org/10.1021/ac50043a017

Tongtavee, N., Shiowatana, J., McLaren, R.G., Gray, C.W., 2005. Assessment of lead availability in contaminated soil using isotope dilution techniques. Sci. Total Environ. 348, 244–256. https://doi.org/10.1016/j.scitotenv.2004.12.066

Tsadilas, C., Rinklebe, J., 2018. Nickel in Soils and Plants - 1st Edition. CRC Press, Taylor & Francis Group, New York, USA.

Uchimiya, M., Cantrell, K.B., Hunt, P.G., Novak, J.M., Chang, S., 2012. Retention of heavy

metals in a typic kandiudult amended with different manure-based biochars. J. Environ. Qual. 41, 1138–1149. https://doi.org/10.2134/jeq2011.0115

Uchimiya, M., Chang, S.C., Klasson, K.T., 2011. Screening biochars for heavy metal retention in soil: Role of oxygen functional groups. J. Hazard. Mater. 190, 432–441. https://doi.org/10.1016/j.jhazmat.2011.03.063

Uchimiya, M., Lima, I.M., Thomas Klasson, K., Chang, S., Wartelle, L.H., Rodgers, J.E., 2010. Immobilization of heavy metal ions (CuII, CdII, NiII, and PbII) by broiler litter-derived biochars in water and soil. J. Agric. Food Chem. 58, 5538–5544. https://doi.org/10.1021/jf9044217

Vasilache, T., Lazar, I., Stamate, M., Nedeff, V., Lazar, G., 2013. Possible environmental risks of photocatalysis used for water and air depollution - Case of phosgene generation. APCBEE Procedia 5, 181–185. https://doi.org/10.1016/j.apcbee.2013.05.032

Venegas, A., Rigol, A., Vidal, M., 2016. Effect of ageing on the availability of heavy metals in soils amended with compost and biochar: evaluation of changes in soil and amendment properties. Environ. Sci. Pollut. Res. 23, 20619–20627. https://doi.org/10.1007/s11356-016-7250-8

Vilvanathan, S., Shanthakumar, S., 2017. Continuous biosorption of nickel from aqueous solution using chrysanthemum indicum derived biochar in a fixed-bed column. Water Sci. Technol. 76, 1895–1906. https://doi.org/10.2166/wst.2017.289

Wang, He, Wang, Han, Zhao, H., Yan, Q., 2020. Adsorption and Fenton-like removal of chelated nickel from Zn-Ni alloy electroplating wastewater using activated biochar composite derived from Taihu blue algae. Chem. Eng. J. 379, 122372. https://doi.org/10.1016/j.cej.2019.122372

Wang, T., Liu, Y., Wang, J., Wang, X., Liu, B., Wang, Y., 2019. In-situ remediation of hexavalent chromium contaminated groundwater and saturated soil using stabilized iron sulfide nanoparticles. J. Environ. Manage. 231, 679–686.

https://doi.org/10.1016/j.jenvman.2018.10.085

WHO, 2000. Chapter 6.10 Nickel General description [WWW Document]. Air Qual. Guidel. - Second Ed. URL https://www.euro.who.int/__data/assets/pdf_file/0014/123080/AQG2ndEd_6_10Nickel.pdf (accessed 4.5.21).

WHO, 1996. Permissible limits of heavy metals in soil and plants, Geneva, Switzerland.

Wu, H., Tan, H.L., Toe, C.Y., Scott, J., Wang, L., Amal, R., Ng, Y.H., 2020. Photocatalytic and photoelectrochemical systems: Similarities and differences. Adv. Mater. 32:18, 1904717. https://doi.org/10.1002/adma.201904717

Wu, H., Zheng, Z., Tang, Y., Huang, N.M., Amal, R., Lim, H.N., Ng, Y.H., 2018. Pulsed electrodeposition of CdS on ZnO nanorods for highly sensitive photoelectrochemical sensing of copper (II) ions. Sustain. Mater. Technol. 18, e00075. https://doi.org/10.1016/j.susmat.2018.e00075

Wurzer, C., Masek, O., Sohi, S., Mašek, O., 2020. Synergies in sequential biochar systems, in: Green Carbon. ETN, pp. 147–159.

Xue, Y., Gao, B., Yao, Y., Inyang, M., Zhang, M., Zimmerman, A.R., Ro, K.S., 2012. Hydrogen peroxide modification enhances the ability of biochar (hydrochar) produced from hydrothermal carbonization of peanut hull to remove aqueous heavy metals: Batch and column tests. Chem. Eng. J. 200, 673–680. https://doi.org/10.1016/j.cej.2012.06.116

Yang, L., He, L., Xue, J., Wu, L., Ma, Y., Li, H., Peng, P., Li, M., Zhang, Z., 2019. Highly efficient nickel (II) removal by sewage sludge biochar supported α-Fe_2O_3 and α-FeOOH: Sorption characteristics and mechanisms. PLoS One 14, e0218114. https://doi.org/10.1371/journal.pone.0218114

Yang, X., Tsibart, A., Nam, H., Hur, J., El-Naggar, A., Tack, F.M.G., Wang, C.-H., Lee, Y.H., Tsang, D.C.W., Ok, Y.S., 2019. Effect of gasification biochar application on soil

quality: Trace metal behavior, microbial community, and soil dissolved organic matter. J. Hazard. Mater. 365, 684–694. https://doi.org/10.1016/j.jhazmat.2018.11.042

Yuan, Y., Bolan, N., Prévoteau, A., Vithanage, M., Biswas, J.K., Wang, H., 2017. Applications of biochar in redox-mediated reactions. Bioresour. Technol. 246, 271–281. https://doi.org/10.1016/J.BIORTECH.2017.06.154

Zama, E.F., Reid, B.J., Arp, H.P.H., Sun, G.X., Yuan, H.Y., Zhu, Y.G., 2018. Advances in research on the use of biochar in soil for remediation: a review. J. Soils Sediments 18, 2433–2450. https://doi.org/10.1007/s11368-018-2000-9

Zambelli, B., Uversky, V.N., Ciurli, S., 2016. Nickel impact on human health: An intrinsic disorder perspective. Biochim. Biophys. Acta - Proteins Proteomics. 1864, 1714–1731. https://doi.org/10.1016/j.bbapap.2016.09.008

Zelano, I.O., Sivry, Y., Quantin, C., Gelabert, A., Maury, A., Phalyvong, K., Benedetti, M.F., 2016. An isotopic exchange kinetic model to assess the speciation of metal available pool in soil: The case of nickel. Environ. Sci. Technol. 50, 12848–12856. https://doi.org/10.1021/acs.est.6b02578

Zhang, A., Li, X., Xing, J., Xu, G., 2020. Adsorption of potentially toxic elements in water by modified biochar: A review. J. Environ. Chem. Eng. 8, 104196. https://doi.org/10.1016/j.jece.2020.104196

Zhu, L., Tong, L., Zhao, N., Li, J., Lv, Y., 2019. Coupling interaction between porous biochar and nano zero valent iron/nano A-hydroxyl iron oxide improves the remediation efficiency of cadmium in aqueous solution. Chemosphere 219, 493–503. https://doi.org/10.1016/j.chemosphere.2018.12.013

Zou, Q., Xiang, H., Jiang, J., Li, D., Aihemaiti, A., Yan, F., Liu, N., 2019. Vanadium and chromium-contaminated soil remediation using VFAs derived from food waste as soil washing agents: A case study. J. Environ. Manage. 232, 895–901. https://doi.org/10.1016/j.jenvman.2018.11.129

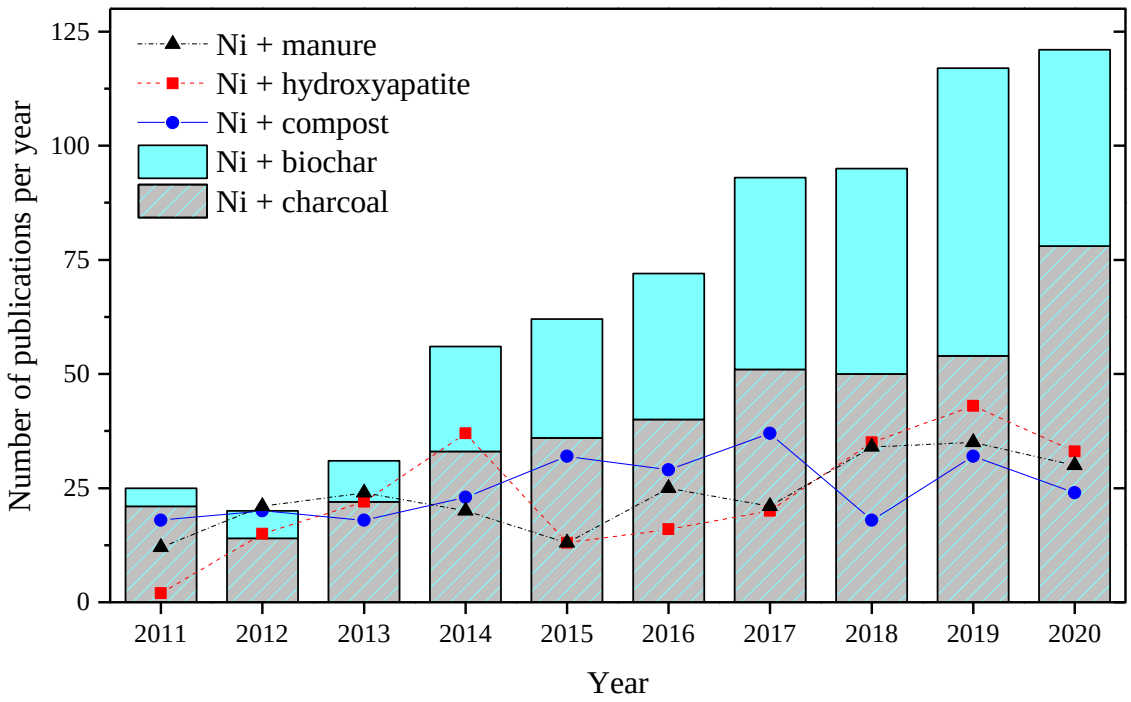


Figure 1: The number of publications in the SCOPUS database based on the following keywords: biochar and nickel, charcoal and nickel, compost and nickel, manure and nickel, and hydroxyapatite and nickel.

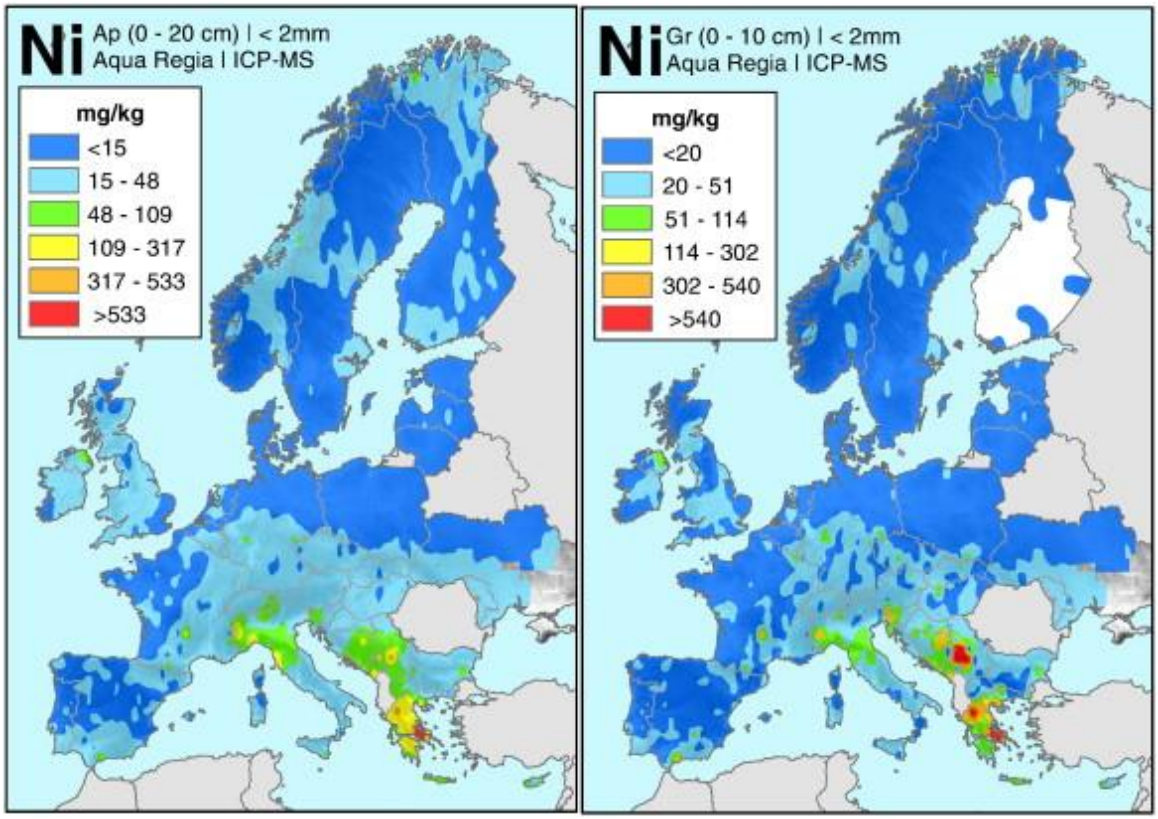


Figure 2: Maps of Ni concentrations in soils collected from agricultural (Ap) and grazing land (Gr) in Europe determined by ICP-MS following an aqua regia extraction. (reproduced from Albanese et al. (2015), with permission from the publisher)

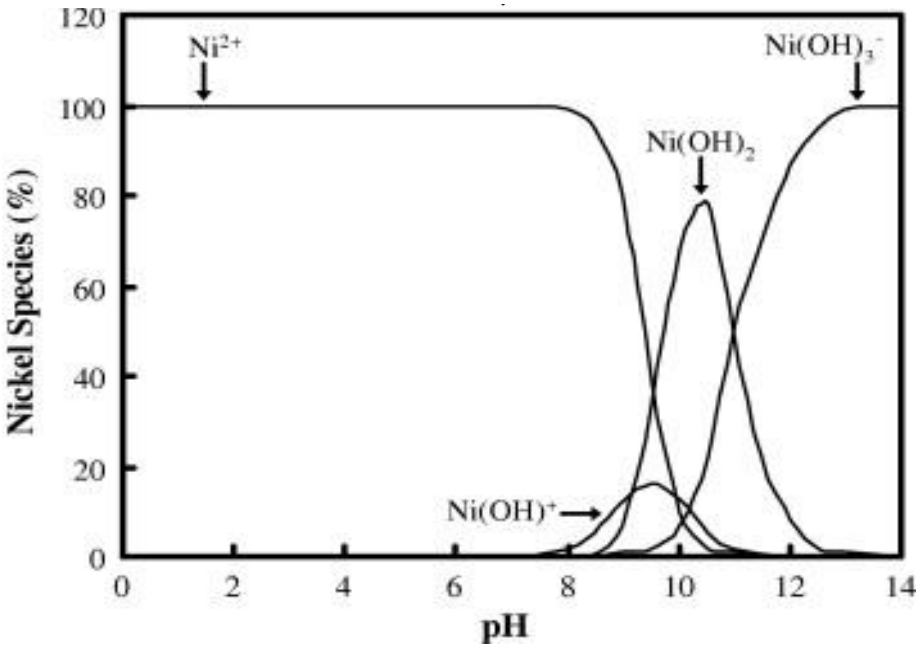


Figure 3: The speciation of Ni in aqueous solutions in the absence of oxidizing agents (Adapted from Anoop Krishnan et al. (2011), with permission from the publisher)

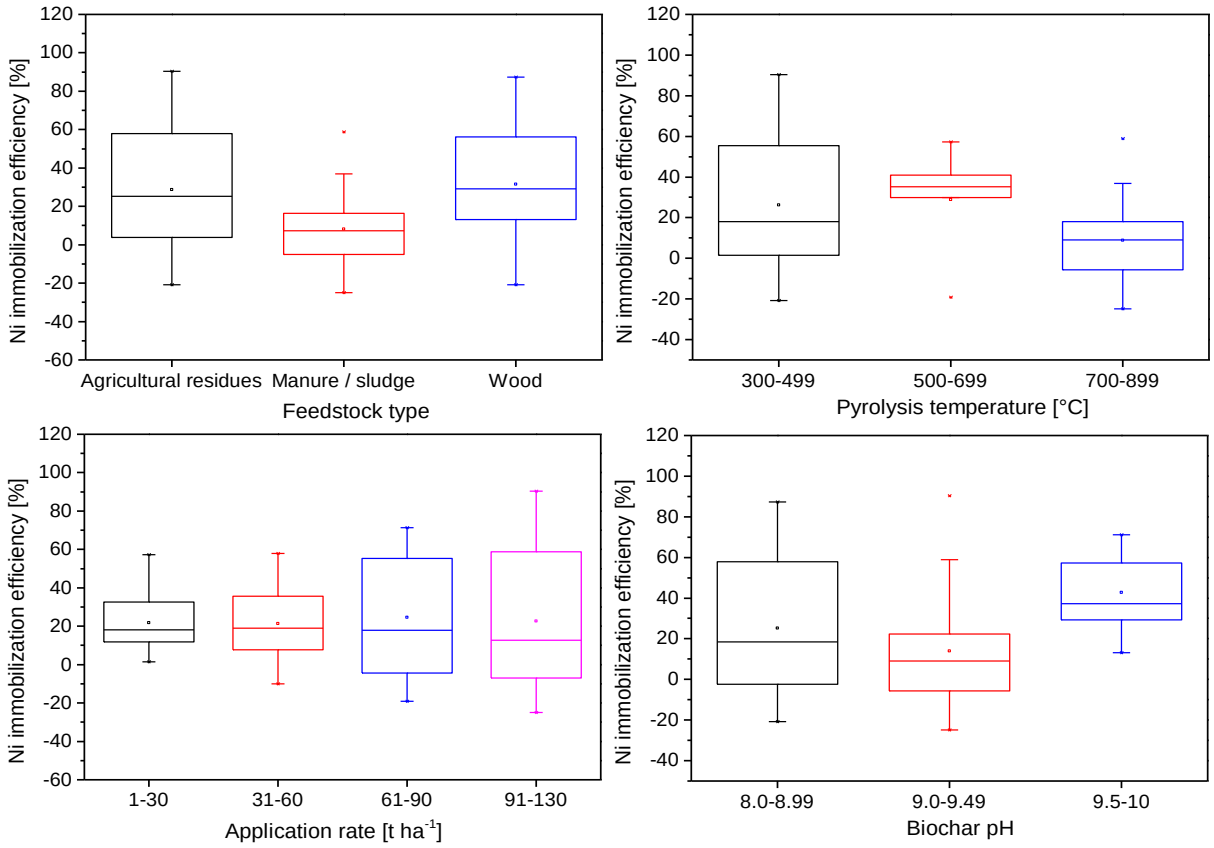


Figure 4: The nickel immobilization efficiency of biochars as affected by biochar properties. Data are extracted from eight studies (based on a total of 59 individual observations). The box chart is represented by the median (centerline), mean (dot), lower and upper quartiles (the lower and upper borders of the box, respectively), whiskers-error bars (the minimum and maximum observations) (Bogusz and Oleszczuk, 2018; Eissa, 2019; Ibrahim et al., 2019; Munir et al., 2020; Nawab et al., 2018; Rehman et al., 2016; Rodríguez-Vila et al., 2015; Venegas et al., 2016)

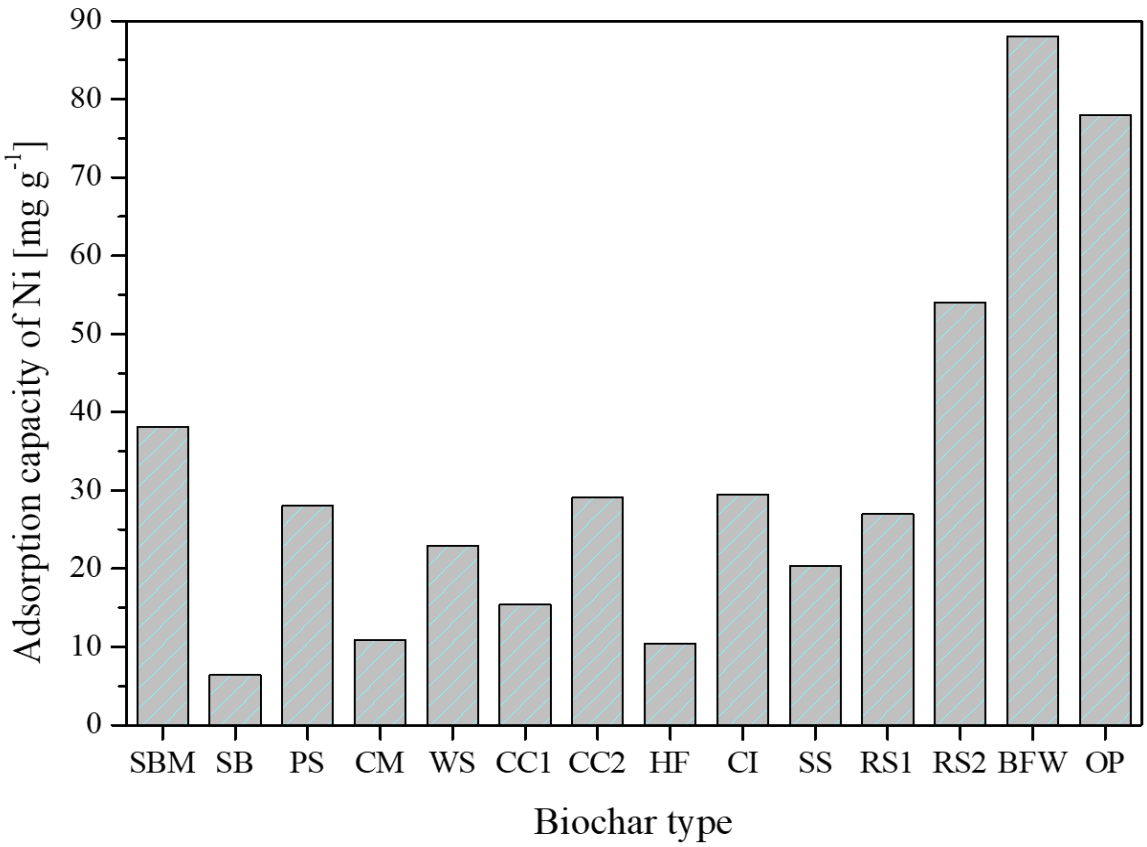


Figure 5: Adsorption capacity of Ni on biochar produced from different feedstocks. Data are obtained from different studies as fellows (abbreviation: type of biochar feedstock with references cited): SBM: Sugarcane bagasse (milled) (Lyu et al., 2018), SB: Sugarcane bagasse (un-milled) (Lyu et al., 2018), PS: Palm seed (Gazi et al., 2018), CM: chicken manure (Higashikawa et al., 2016), WS: wheat straw pellets (Shen et al., 2017), CC1: corncobs (Hu et al., 2018), CC2: corncobs (Shi et al., 2018), HF: hizikia fusiformis (Shin, 2017), CI: chrysanthemum indicum (Vilvanathan and Shanthakumar, 2017), SS: sewage sludge (Yang et al., 2019), RS1: rice straw (Deng et al., 2019), RS2: rice straw (Deng et al., 2019), BFW: banana fruit waste, and OP: orange peel (Amin et al., 2019)

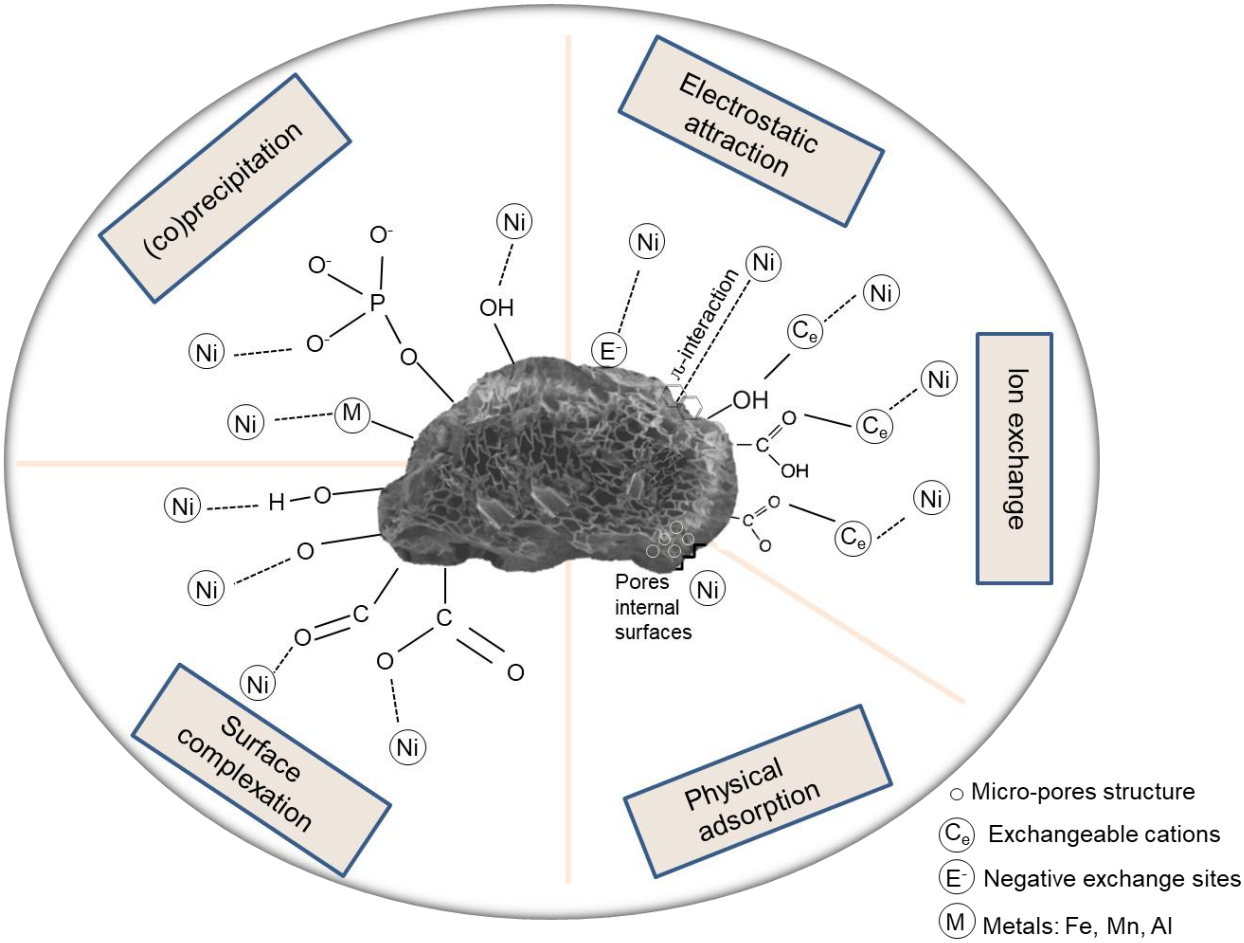


Figure 6: A schematic diagram of mechanisms of interactions between biochar and Ni

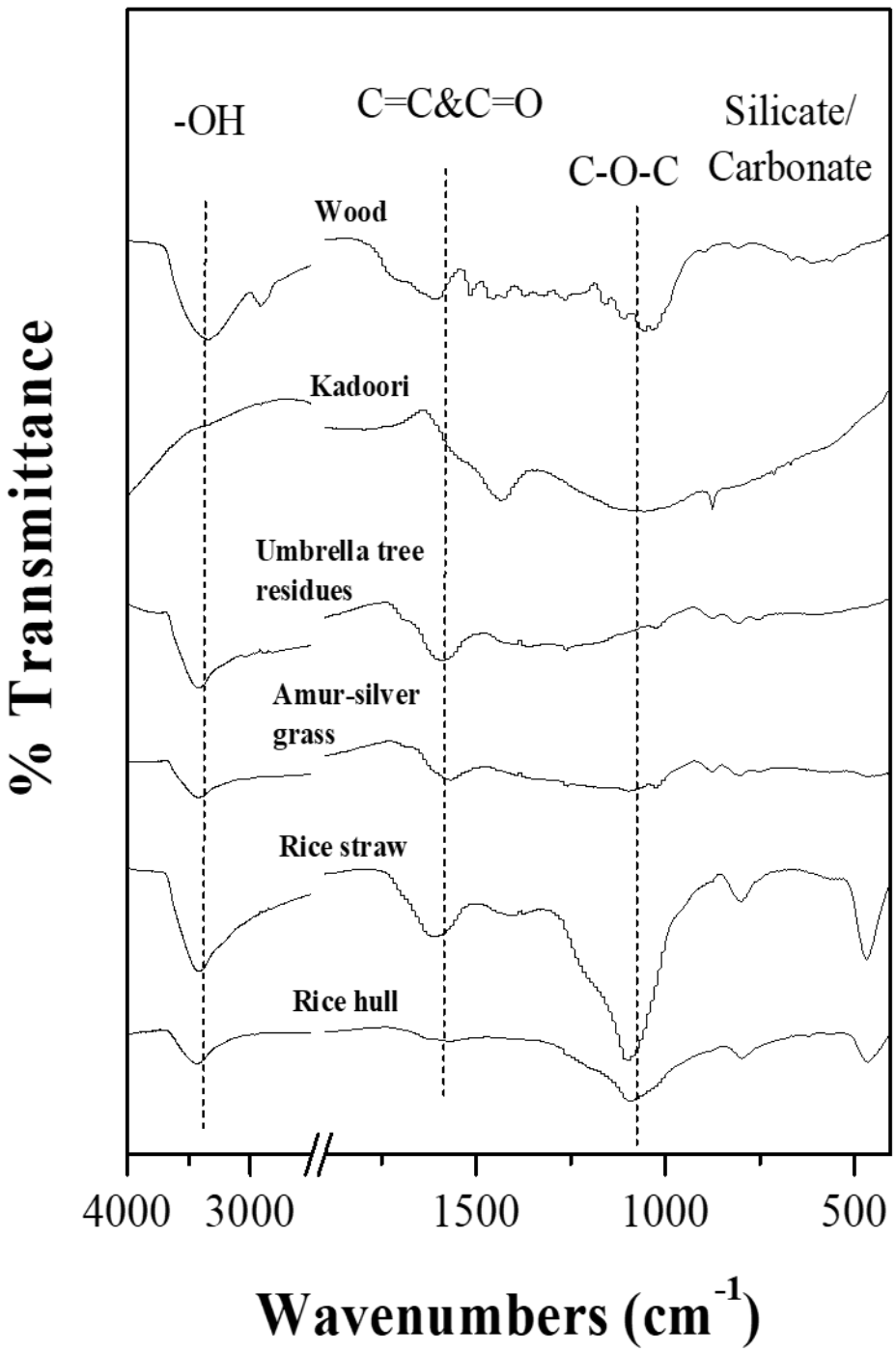


Figure 7: Fourier-transform infrared spectra of six types of biochars produced from different feedstocks. Data are obtained from previous studies (El-Naggar et al., 2018a; Rinklebe et al., 2020)

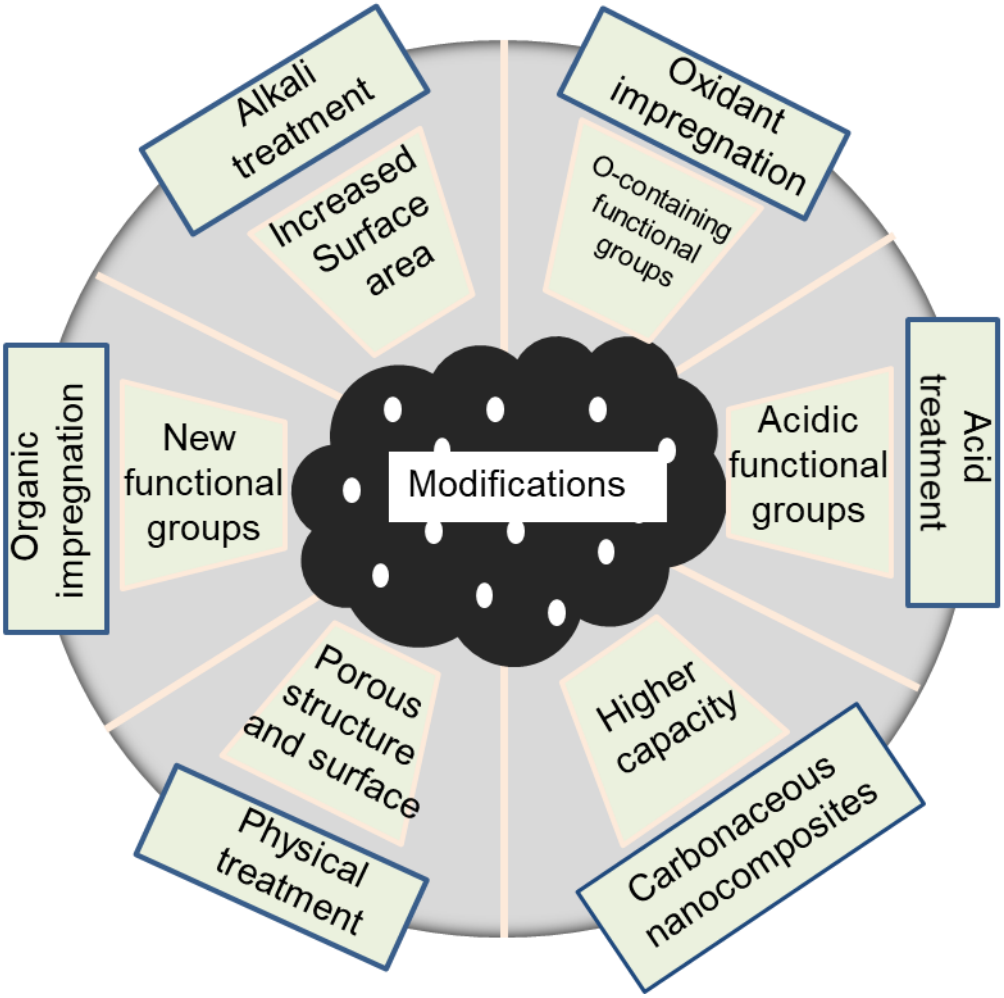


Figure 8: A schematic diagram illustrating recently proposed methods of biochar modification for the remediation of Ni contamination

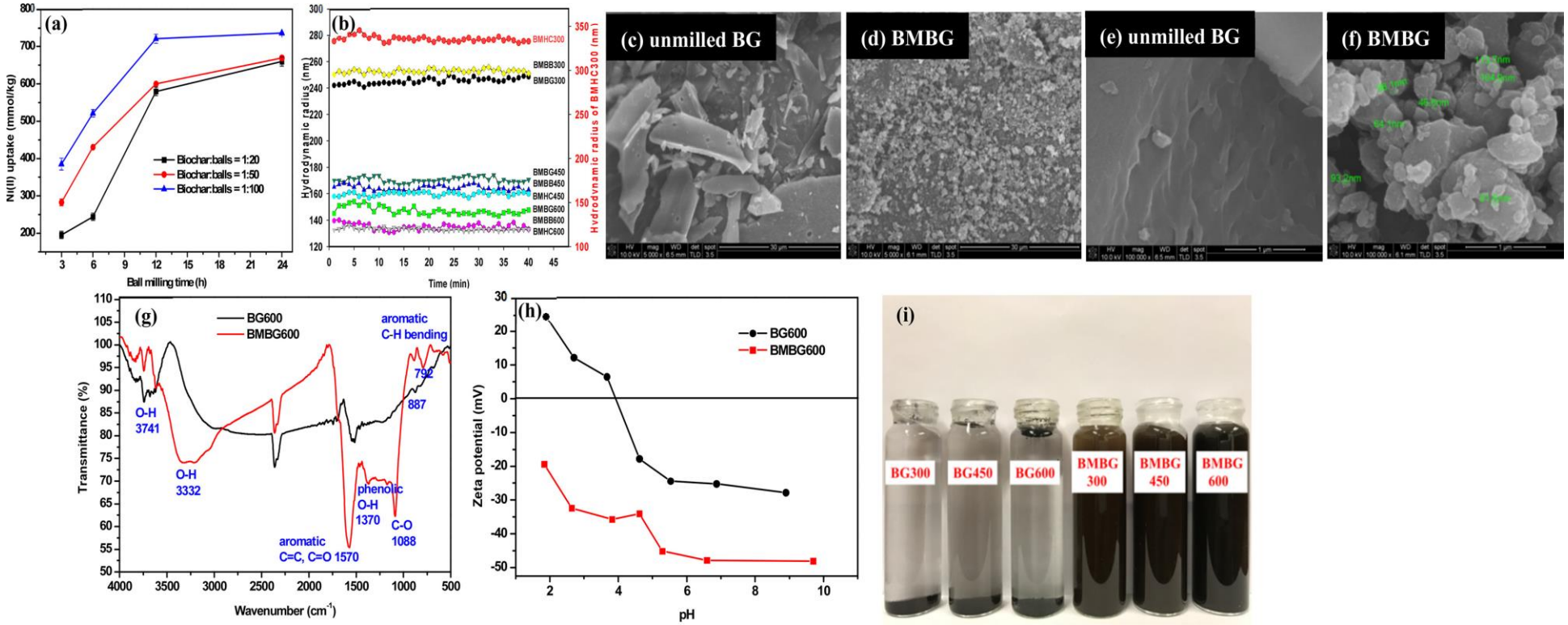


Figure 9: Biochar engineering via optimization of ball-milling conditions; (a) Effects of mass ratio and time on Ni(II) adsorption by ball-milled 600 °C bagasse biochar (BMBG600); (b) hydrodynamic radius of BMBG prepared at 0.4 g L^{-1}; (c) un-milled BG 600, (d) BMBG, and high-magnification images of (e) un-milled BG 600, (f) BMBG SEM images; (g) FTIR spectra and (h) zeta potential of un-milled BG600 and BMBG600; (i) BG and BMBG suspensions (48 h after preparation) (reproduced from Lyu et al. (2018), with permission from the publisher)

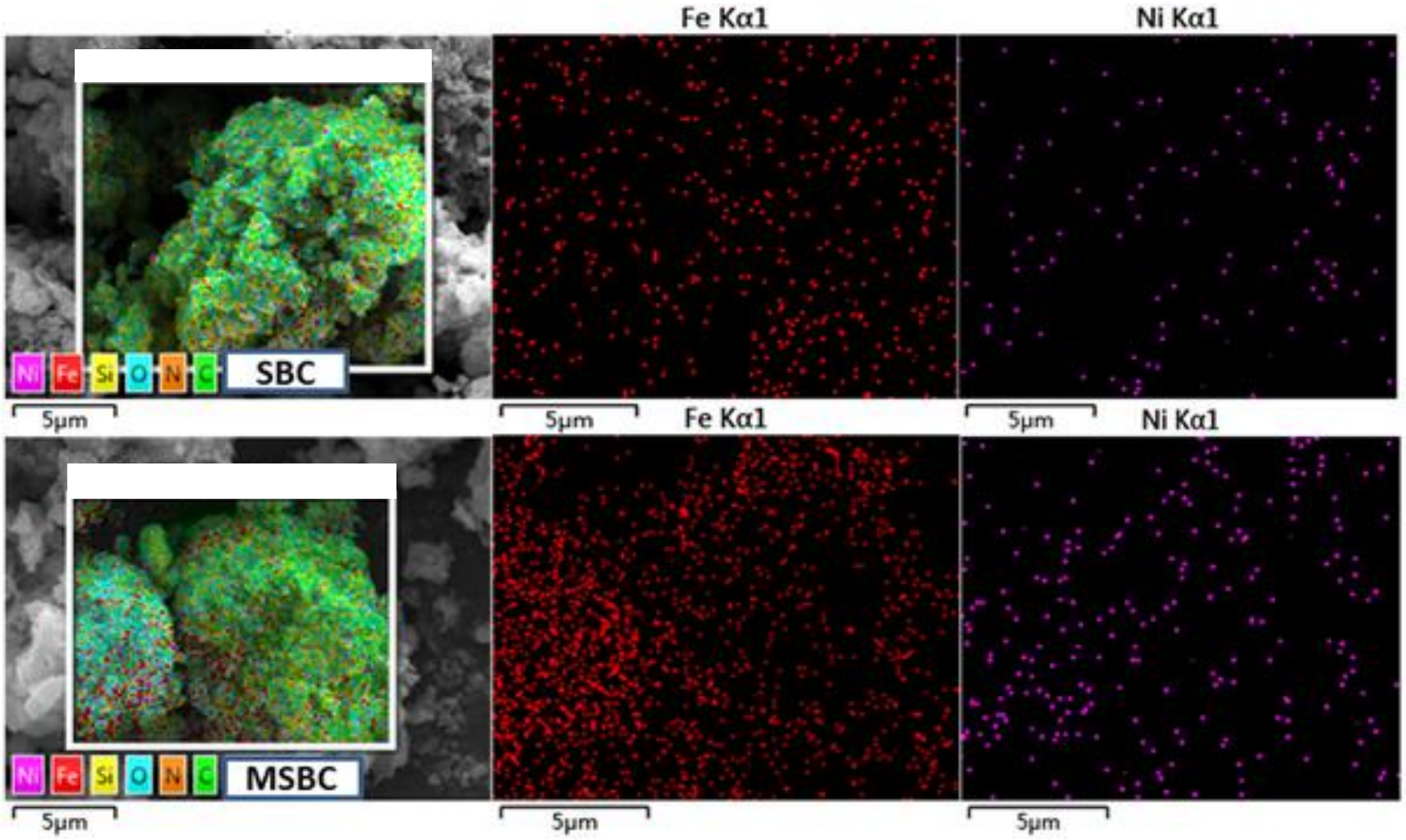


Figure 10: Scanning electron microscopy with energy dispersive x-ray analysis on the surface of sewage sludge biochar (SBC), sewage sludge biochar supported α-Fe_2O_3 and α-FeOOH (MSBC) after Ni adsorption (reproduced from Yang et al. (2019), with permission from the publisher)