



# Production and characterization of hydrochars and their application in soil improvement and environmental remediation

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## ARTICLE INFO

### Keywords:

Hydrochar  
Biochar  
Hydrothermal carbonization  
Soil degradation  
Environment remediation

## ABSTRACT

With the rapid global population growth and industrial development, the promotion of sustainable agricultural production and environmental conservation has attracted great public and research interests. Application of carbonaceous materials (e.g., activated carbon, biochar, and hydrochar) for soil improvement and environmental remediation is highly recommended because of their economic viability and applicability. Hydrochars, carbonaceous solid materials with unique physicochemical properties and produced by hydrothermal carbonization (HTC) of biomass, have received wide attention due to their increasing applications as soil amendments, slow-release fertilizers, adsorbents, and energy sources. This review highlights the production of hydrochars from dry and wet feedstocks and summarizes the physicochemical properties including surface structure, porosity, nutrient content, and stability. Applications of hydrochars for soil improvement and environmental remediation are systematically analyzed and reviewed on the aspects of improving soil physicochemical and biological properties, affecting greenhouse gas emission, and remediating heavy metals and organic pollutants in water and soil environments. Finally, the knowledge gaps in the production, characterization, and application of hydrochars are addressed and the future research directions toward the development of hydrochar technology are proposed.

## 1. Introduction

Recently, the generation of large amounts of biowastes such as agricultural waste, municipal sludge, and food waste have raised serious management and disposal challenges globally [1,2]. For example, wastewater treatment plants (WWTPs) annually produce 12.7 million dry tons of municipal sludge in the U.S. 230 million tons in Europe, 30 million tons in China, and 3.0 million tons in Australia [3,4]. The worldwide production of municipal sludge was around 1.3 billion tons in 2017 [2]. In addition, agricultural waste, which is mostly referred as to the residues and by-products of agriculture (e.g., crop straws), is also largely produced. For instance, the annual generation of agricultural waste is 140 billion tons globally [5], while this number was about 819 megatons in 2014 in China [6]. Many of these biowastes are rich in

organic carbon (OC), nitrogen (N), phosphorus (P), and other beneficial elements (e.g., Ca, Mg, and Fe). However, the current management strategies, including burning, landfills, composting and bio-drying, are typically lack of value-added utilization technologies and easily result in serious issues, such as resource loss, environmental pollution, and greenhouse gas (GHG) emission [2,7], as well as increasing financial burdens on various levels of governments [8]. For example, it is estimated that food waste contributed to one sixth of the methane emissions from landfills in the U.S. [9]. The cost of municipal sludge management in a WWTP could reach to 57% of the total operation cost [10]. Particularly, ever-increasing biowaste such as food waste, sewage sludge, and pig manure containing high moisture ( $\geq 30\%$ ), potential source of nitrogen and phosphorus [11], makes big challenges to apply the traditional technologies like pyrolysis and incineration to recover

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energy or value-added products, because of the extra energy input for dry treatment [12]. Therefore, the development and optimization of sustainable technologies for high-value utilization of and resource recovery from biowastes (particularly for the wet biowastes) are urgently needed.

Hydrothermal carbonization (HTC) is a promising technique to achieve the target of comprehensive utilization of biowastes to produce a carbon (C) rich solid product named as hydrochar [7,13]. Converting biomass into hydrochar, a primary product of HTC, could be an environmentally-friendly measure to achieve resource utilization and produce value-added products [13–15]. Compared to other thermal technologies such as pyrolysis, gasification, and combustion, which typically require pre-drying for the wet biomass, HTC can be applied to both dry and wet biowastes [16], thus requires less energy from the feedstock [17]. Hydrochars, C-rich materials with high contents of oxygen-containing functional groups and nutrients [7] and high energy density [18], can be used as adsorbents [19], catalysts [20], soil conditioners [13], and bioenergy sources [7]. Hydrochars in certain aspects (e.g., rich C, application as adsorbents or soil amendments) are similar to biochars, a type of solid carbonaceous and recalcitrant materials derived from pyrolysis of biomass [1,21,22]. In the past decade, biochars have been highly recommended as promising solutions for sustainable agricultural production and environmental remediation [23–25]. Recently, increasing studies reported the applications of hydrochars as soil amendments [7,26]. Hydrochars have also been tested as promising adsorbents for heavy metals [27,28], organic pollutants (e.g., pharmaceuticals, dyes, pesticides) [28,29], and excessive nutrients (e.g., phosphate and nitrate) [30,31] in water and soil environments. Even though hydrochars have the same applications (e.g., adsorbent, catalyst, and soil amendment) with biochars, they are very different in feedstocks, production technologies, and characteristics [22,32].

The HTC process and its influences on physicochemical characteristics of hydrochars such as yield, morphology, surface structure, and nutrient availability, have been extensively studied [13,33–36]. Increasing studies also reported the application of modified hydrochars in improving soil fertility and remediating polluted soils and waters [15,27,28,37]. Several reviews summarized the formation of hydrochars [22,38], general application of hydrochars in energy recovery and agricultural production [7,22]. However, these reviews mainly focused on HTC conditions and physicochemical properties of hydrochars [7,17,39]. To the best of our knowledge, the comparison of hydrochars and biochars in regard to their production, characterization, and impact on soil improvement and environmental remediation is very limited.

This work aims to: 1) make a clear comparison among the technologies and feedstocks for hydrochar and biochar production; 2) summarize the physicochemical characteristics of hydrochars derived from different feedstocks; 3) illustrate the potential applications of hydrochars for soil improvement in terms of soil physicochemical characteristics, fertility, productivity, GHG emission, and microbial community; and 4) review the application of hydrochars in remediating soils and water bodies contaminated by heavy metals, nutrients, and organic pollutants. These aspects are brought together to highlight the current progresses and limitations of hydrochar research and provide future opportunities for facilitating the development of hydrochar technology; and 5) address the research gaps and provide key future directions for facilitating the sustainable development and application of hydrochar technology. Ultimately, we aim to highlight the current progresses and limitations of hydrochar research and address the question: Can hydrochars be used as sustainable alternative to biochar in agricultural production and environmental remediation?

## 2. Comparison of the technologies for hydrochar and biochar production

### 2.1. Hydrothermal carbonization (HTC) vs pyrolysis

Difference in thermal treatment conditions is one of the primary differences between hydrochars and biochars [1,22]. Thermal technologies for hydrochar and biochar production are summarized in Table 1. HTC is a thermochemical process of converting biomass with high moisture into hydrochars at the relatively low temperatures (180–375°C) in short residence time ranging from minutes to hours under autogenerated pressure (2–6 MPa) in the presence of subcritical or supercritical water environment [1,40]. It is noted that autogenous pressure of water in the inner chamber is completely sufficient in the HTC process. Pyrolysis, used for biochar production, is a thermal decomposition technology of converting biomass at the relatively higher temperatures (300–1200°C) in the absence of O<sub>2</sub> or limited O<sub>2</sub> conditions [41–43], which can be classified into slow, intermediate pyrolysis, fast, and flash pyrolysis (Table 1). Compared to pyrolysis, HTC is a promising thermal technology with attractive advantages including high conversion efficiency, elimination of pre-drying requirement, and relatively low heating temperature (HTT) [13]. HTC is generally low energy demanding due to its lower HTT (180–375°C) and possibility of direct application of wet feedstock (e.g., sewage sludge, animal manure, and kitchen wastes). However, large amount of energy input is needed for biochar production from biomass pyrolysis, particularly because of high HTT (> 400°C) for long residence time of days to weeks and necessity of the dry pre-treatments of feedstock [1,34,35]. The key parameters of HTC include feedstock, HTT, reactor, hydrous conditions, residence time, pressure, solid load, catalyst, and pH [44–46]. Among them, HTT is one of the main factors affecting physicochemical properties of hydrochars [47]. Pressure also plays an important role in the transformation of biomass during HTC [1], which can dictate reaction routes [46], and characteristics of the final products [35]. Additionally, the water in HTC is subcritical or supercritical (the critical point 374°C), lowering the activation energy level of hemicellulose and cellulose in biomass, thus facilitating the degradation and depolymerization of these components [35]. Notably, water is highly recommended as a reacting medium in HTC because it is cheap, non-toxic, and is inherently present in the wet biomass. Moreover, the solid to liquid ratio in HTC should be chosen properly and the threshold should be carefully examined. For example, Xiong et al. found that 0.1 g mL<sup>-1</sup> was an ideal solid–liquid ratio for swine manure to produce high yield of hydrochar [48]. Normally, the addition of basic additives (e.g., NaOH, Ca(OH)<sub>2</sub>) and acidic additives (e.g., HCl, H<sub>2</sub>SO<sub>4</sub>) in HTC serves different purposes such as speeding up thermal reaction, increasing bio-oil yield, and modifying the hydrochars with desired characteristics (e.g., nanopores, large surface area) [49,50]. Alkaline catalysts such as NaOH, KOH, and Ca(OH)<sub>2</sub> could facilitate water–gas shift reaction under the supercritical water conditions [51], which would result in low solid yield and production of hydrogen-rich gas by accelerating hydrolysis and decomposition of lignin [52]. Acids catalysts such as HCl and H<sub>2</sub>SO<sub>4</sub> may make a hydrochar with great surface area, high pore volume, and small pore size [52], by promoting hydrolysis, deamination, and dehydration of feedstock during hydrothermal processing [51]. However, it should be carefully considered for selecting a catalyst, which could cause the pitting of reactor or environmental pollution. Thus, more studies should be conducted to use green and environment-friendly additives during HTC. Recently, a few studies reported that hydrochars also can be obtained as byproducts from hydrothermal liquefaction (HTL) and hydrothermal gasification (HTG); these two hydrothermal techniques are used to produce bio-oil and syngas, respectively (Table 1). Thus, these two hydrothermal technologies should be paid more attention in future. Pyrolysis technology is commercially available for high-value products (e.g., biochar, bio-oil), whereas the commercial implementation of HTC technology is still in infancy. Although several studies compared the

**Table 1**  
Comparison of the thermal technologies for production of hydrochar and biochar.

	Hydrothermal treatments			Pyrolysis			
	Hydrothermal carbonization (HTC)	Hydrothermal liquefaction (HTL)	Hydrothermal gasification (HTG)	Slow pyrolysis	Intermediate pyrolysis	Fast pyrolysis	Flash pyrolysis
HTT	180–375°C	200–400°C	350–700°C	300–700°C	300–500°C	500–1000°C	400–1000°C
RT	Minutes–hours	1–120 min	30 s–30 min	Hours–weeks	< 20 s	< 1 min	< 30 min
Pressure	autogenous pressure (2–6 MPa)	10–25 MPa	20–50 MPa	0.1 MPa	0.1 MPa	< 5 MPa	< 0.5 MPa
Main products	Hydrochar	Bio-oil	Syngas	Biochar	Bio-oil	Bio-oil	Syngas
Feedstock	Dry Wet	Agricultural wastes, woody wastes, crop residue Fresh vegetable wastes, sewage sludge, animal wastes, algae		Agricultural wastes, woody wastes, crop residue Fresh vegetable wastes, sewage sludge, animal wastes, algae (Note: wet feedstock needs to be dried before pyrolysis)			
Reaction mechanism	Hydrolysis, dehydration, decarboxylation, condensation, polymerization, and aromatization			Dehydration, aromatization, decarboxylation, polymerization, intramolecular condensation, and rearrangement reactions			
Reference	[1,17,22,54] [1,55] [54,56]			[22,57] [43,58] [41,42,59] [42,60]			

HTT: heating temperature; RT: residence time of HTC process. Pressure: the pressure for fast and flash pyrolysis is generally higher than 1 atmospheric pressure (0.1 MPa), but lower than 5 MPa and 0.5 MPa, respectively.

differences of these technologies [22,53], the understanding of their economic feasibility and energy cost at different scales for commercial hydrochar production is still limited. Therefore, more future studies should be focused on the commercial and large-scale hydrochar production with commercial implementation of HTC technology. Notably, reducing energy input and economic cost in HTC should be fully considered, and the application of solar energy, continuous reactors, and deep learning techniques can be expected.

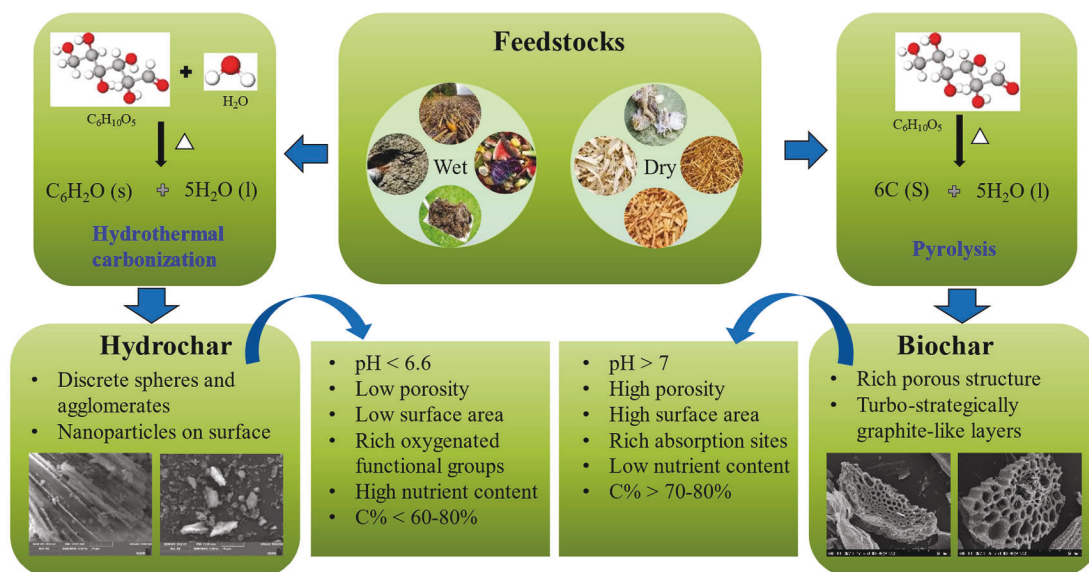
Formation of biochars from biomass pyrolysis mainly consists of three reaction stages, dehydration and decarboxylation at the first stage, depolymerization at the second stage, aromatization and intermolecular rearrangement at the third stage, which has been extensively reviewed in previous studies [21,57]. A wide range of reactions, including hydrolysis, dehydration, decarboxylation, aromatization, and condensation polymerization, may occur during HTC [40,44,61,62]. In the hydrolysis stage, hemicellulose starts to hydrolyze at 180°C, whereas cellulose starts at around 230°C and lignin starts at above 260°C, leading to the formation of oligomers like cellobiose, celotriose, cellotetraose, cellopentaose, and cellohexaose [40,63]. Water at this stage in the form of hydronium ions has high values of ionic product of  $H^+$  and  $OH^-$ , facilitating the hydrolysis process [40]. Then dehydration and decarboxylation occur immediately, involving removal of water and carbon dioxide from the biomass matrix [64], accompanied by the production of organic acids (e.g., acetic, lactic, propionic, levulinic and formic acids) and aldehydes [65,66]. Condensation and polymerization are the next stage of HTC, which are influenced by intermolecular dehydration or aldol condensation. Soluble polymers are formed when the monomers such as glucose and fructose undergo these reactions. Finally, aromatization takes place to form solid hydrochars because of the decomposition of the oligo and monosaccharides, [39,54]. However, these reactions for different feedstocks (e.g., sewage sludge, animal manure, plant residues) are much complicated and still unclear. Further studies should be conducted to understand the underlying reactions of hydrochar formation from different feedstocks using different hydrothermal technologies.

HTC inevitably produces a large quantities of process water, mainly containing phenolics, acetic acid, formic acid, glycolic acid, levulinic acid, 2,5-hydroxyl-methyl-furfural (HMF), furans, heavy metals (e.g., Cu, Zn, As, Ni, Cd, and Pb), and nutrients (e.g., P, N, Ca, Mg, and K) during decomposition of biomass polymers [1,67]. Products like levulinic acid and 2,5-HMF, as the high-quality intermediate compounds, could act as potential precursors for producing value-added liquid fuel and chemicals [68–70]. Moreover, nutrient characteristics of process water imply their potentials as liquid fertilizers in agricultural application [71–73], expanding the utilization pathway of the byproducts of hydrochars. However, occurrence of the potential toxic compounds such

as heavy metals and organic compounds could pose great environmental risks if they were not effectively treated before discharge [74]. At present, most of these studies on process wastewater focused its recirculation [75–77] and environmental risk assessment [78,79]. However, the treatment of process water from hydrochar production received little attention. In the future, an industrial HTC plant will face big challenges for its operation due to the continuous production of process water. Therefore, it is critically urgent to develop effective technologies to treat or recycle the process water in order to avoid potential environmental risks and decrease cost for industrial scale production and application of hydrochars. Notably, recovering value-added products like levulinic acid, 2,5-HMF, two of the top 12 value added chemicals from biomass proposed by US Department of Energy, and P from the process water could be a feasible strategy to reduce the economic cost of HTC, which should be further explored.

## 2.2. Wet biomass vs dry biomass

Besides the thermal techniques, type of biomass feedstock is another important difference for hydrochar and biochar production (Fig. 1). HTC can be applied to a broad range of conventional biomass feedstocks (e.g., crops straws, forest wastes) and unconventional biomass feedstocks (e.g., food wastes, sewage sludge, and algae) without any pre-drying [54]. Animal wastes, sewage sludge, kitchen wastes, microalgae/macroalgae, and fresh crop residues, are considered as wet feedstocks, which generally contain high content of moisture (> 30%) [1]. Dry biomass such as air-dried agricultural residues and woody wastes generally containing < 30% moisture content is suitable for biochar production using pyrolytic technologies (Table 1). Notably, wet and dry biomass can be used in HTC for hydrochar production with less energy since drying pre-treatments of the wet biomass is not required (Table S1). In contrary, pre-treatments are required for biochar production via pyrolysis, ultimately increasing the demand of energy, labor, and the cost of biochars [22,39]. Additionally, increasing number of studies showed that the hydrochars prepared from single feedstock could have some uncertainties in their properties and applications in soil improvement and remediation [80,81], and could also limit their sustainable industrial production. Thus, the blended feedstock containing two or more types of biomass wastes are proposed to improve the yield and physicochemical properties of hydrochars, because of the potential of synergistic and antagonistic effects between the different feedstocks [82–84]. However, more studies need to be conducted to understand the critical variables determining the hydrochar properties, as well as the potential mechanisms responsible for upgrading the hydrochar properties. Overall, these aforementioned differences among the thermal techniques and feedstock would result in significant differences regarding their



**Fig. 1.** Conceptual diagram illustrating the differences between biochar and hydrochar, including their typical feedstock, production processes, and characteristics. For pyrolysis, dry feedstocks are typically treated at high temperatures (300–1000°C). The final product is a C-rich biochar with high surface area but low nutrient content [41,43]. For HTC, wet and dry biomass is treated at lower temperature (180–350°C). The final product is a C-rich hydrochar with more O-containing functional groups and high nutrient content [34,61].

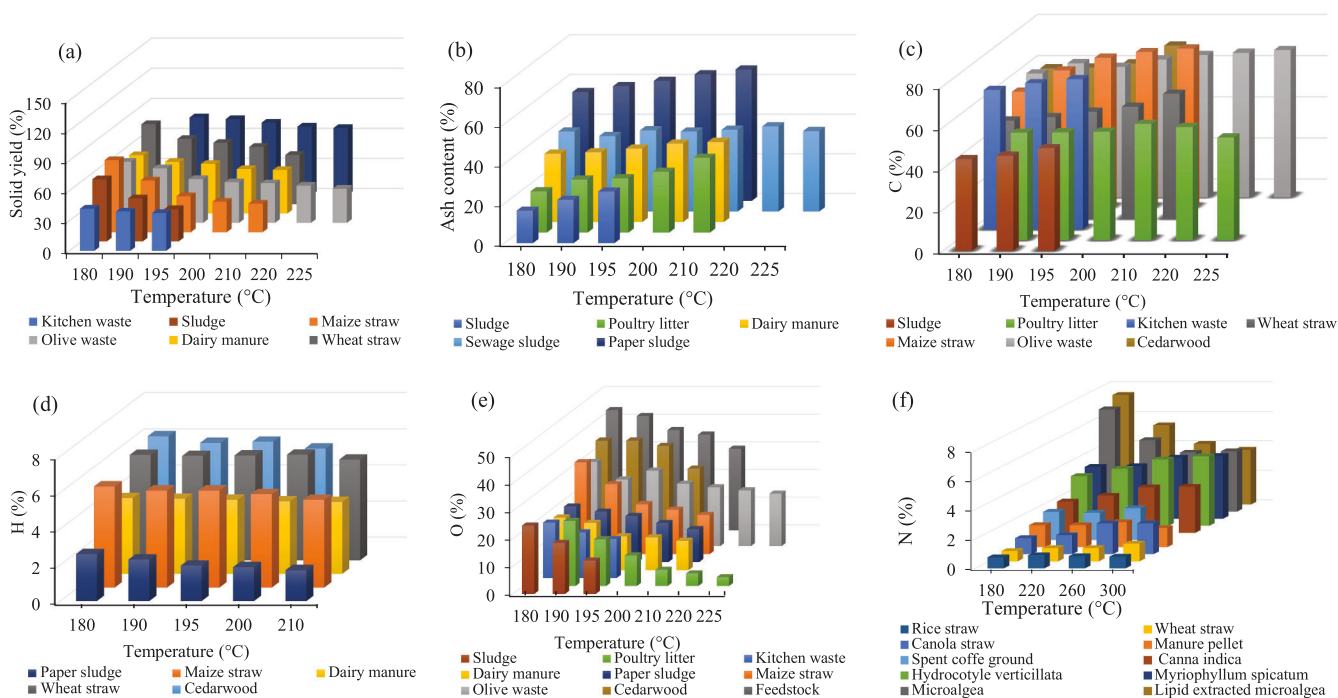
characteristics, applications, and implications between hydrochars and biochars [39].

### 3. Characteristics of hydrochars

#### 3.1. Yield

The solid mass yields of hydrochars production from HTC greatly vary, ranging from 28.6 to 79.9% (Fig. 2a), whereas those of biochars produced from pyrolysis ranges at 25–45% [85]. HTT plays a key role in

hydrochar yields [1]. Regardless of the types of feedstocks, the mass yields of hydrochars generally decrease with increasing HTT (Fig. 2a). Ash contents of hydrochars vary at 14.5–66.2% (Fig. 2b). As HTT increases from 150 to 200°C during HTC, generation of organic acids (e.g., acetic, formic, lactic, and levulinic acid) via dehydration and decarboxylation facilitates the dissolution of mineral components in the feedstock [1], consequently decreasing the mass yields (Fig. 2a) and ash contents (Fig. 2b). Notably, the ash contents of hydrochars derived from the feedstocks containing high contents of non-soluble minerals such as animal manure, paper sludge, and sewage



**Fig. 2.** Effect of heating temperature on (a) hydrochar yield, (b) ash content, (c) C content, (d) H content, (e) O content, and (f) N content. Generally, the changes in hydrochar characteristics depend on the type of feedstock and HTC conditions. Data obtained from the reported studies [14,33,47,85,88-101]



sludge, increase with increasing HTT (Fig. 2b). In addition, generation of organic acids (e.g., acetic, formic, lactic, and levulinic acid) via dehydration and decarboxylation results in the acidity of hydrochars at pH 4.6–7.4 (Fig. S1). The feedstock is another important factor determining hydrochar yield [39,86]. Lignocellulosic biomass (e.g., woody wastes, crop straws) generally results in higher yields of hydrochars than those of non-lignocellulosic biomass (e.g., sewage sludge and kitchen wastes) under the similar hydrothermal conditions [39]. For example, Tag et al. found that the yields of hydrochars derived from sunflower stalk were 40.5–68.1%, higher than those (32.8–66.2%) from poultry litter and algal biomass at the same HTC conditions [87]. Thus, HTT needs to be determined based on the types of feedstocks to optimize hydrochar yields. Similarly, biochar yields are also mainly controlled by feedstock and HTT during pyrolysis, which has been well reviewed [21]. Among the pyrolysis technologies, slow pyrolysis is widely performed for producing biochars that widely used as adsorbents in remediation of water and soil pollution and as soil amendments in improving soil quality and productivity (Table 1).

### 3.2. Elemental composition

Like biochars, hydrochars are mainly composed of C, H, O, N, and other mineral elements such as K, Ca, Mg, Fe, and Al, originating from biomass feedstock (Table S1). Total C, H, and O contents in hydrochars largely vary, ranging 44.6–77.4%, 1.7–6.1%, and 3.2–44.6%,

respectively (Fig. 2c-e). Along with elemental C, N, O, and H, other elements like K, Na, Mg, Ca, Al, Si, S, and Fe are also present in hydrochars [102], but the contents of these elements are generally much lower than those in biochars [103,104]. HTT largely affects hydrochar elemental compositions [39]. As the HTT increases, C contents of hydrochars generally increase due to the enrichment via carbonization (Fig. 2c). In contrast, H and O contents decrease due to decarboxylation and dehydration (Fig. 2d, e). C contents of hydrochars also depend on the type of feedstocks. Hydrochars derived from lignocellulosic biomass (e.g., woody wastes, crop straws) contain more C than those derived from non-lignocellulosic biomass such as manure and sewage sludge [33,39]. Compared to hydrochars, biochars have higher contents of C ranging 30–90% because of the higher degree of carbonization resulting from the relatively higher HTT (Table 1). This is further evidenced by the results of the van-Krevelen diagram (Fig. 3). The atomic ratios of O/C and H/C of biochars are distinctly lower than those of hydrochars, confirming lower degree of carbonization of hydrochars relative to biochars [53]. These results also implied that hydrochars would be less stable than biochars in soils when used for C sequestration. However, more efforts are still needed to explore the hydrochar potentials for long term C sequestration in combating with global climate change.

The N contents of hydrochars, ranging 0.7–7.5% (Fig. 2f), are determined by the feedstocks and HTC conditions. Hydrochar feedstocks generally contain inorganic N (e.g.,  $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N, and  $\text{NO}_2^-$ -N) and organic N (e.g., proteins, amino sugars, and nucleic acids) [97,98,100],

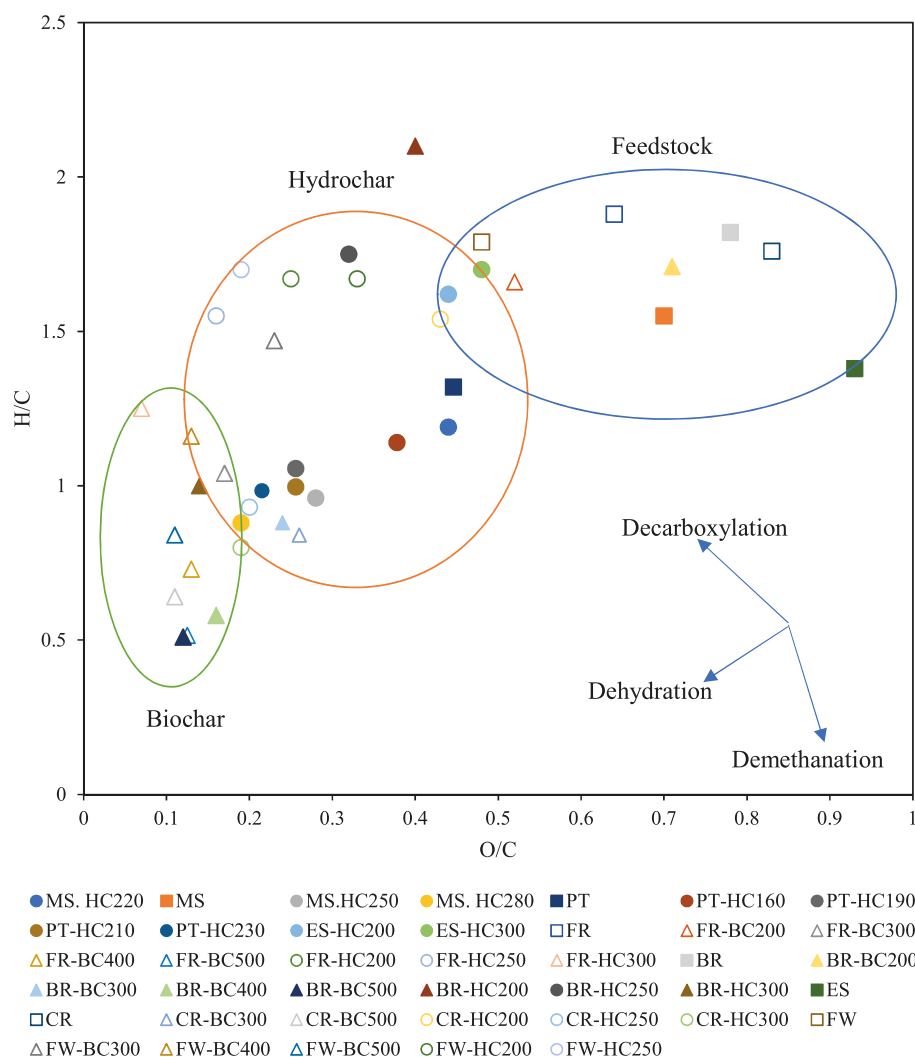


Fig. 3. van-Krevelen diagram for biochars (triangles) and hydrochars (circles) produced from different feedstocks (squares) reported in the literature. MS: maize straw, PT: peat, ES: egg shell, FR: fish residue, BR: breadcrumbs, CR: cooked rice, FW: food waste. BC stands for biochar and HC stands for hydrochar. For example: BR-BC300 represents the biochar derived from breadcrumbs at 300°C, whereas BR-HC250 stands for the hydrochar derived from bread crumbs at 250°C. Data were obtained from the reported studies [33,105,106].

which would be subjected to complex transformation during HTC (Fig. 4). Inorganic N with low thermal stability will be converted to liquid and oil phase by hydration and condensation of biomass [107]. For organic N, several reactions would occur during HTC (Fig. 4). For example, the deamination of proteins results in the formation of amino acids, which are solved in water and then hydrolyzed to  $\text{NH}_4^+\text{-N}$  [108]. HTC can also hydrolyze proteins to amino acids by breaking the C-N bonds via ring condensation and sequential cyclization, transforming into heterocyclic-N species like quaternary-N, pyrrole-N, and pyridine-N in hydrochar [108]. Similar heterocyclic-N and inorganic N species were also found in biochars [21,109], which have been well reviewed [110,111]. Total P in hydrochars range 5–95.4  $\text{mg g}^{-1}$  (Fig. 5a). The reported P species in hydrochars include organic P (e.g., nucleic acids and phospholipids) and inorganic P (e.g., octacalcium phosphate, apatite, and hydroxyapatite) [14,112,113]. The most abundant P species in hydrochars is octacalcium phosphate (20–80%), and its content increases with increasing HTT due to stabilization of P with other elements such as Ca and Mg in the feedstock like sludge (Fig. 5b).  $\text{NaHCO}_3$  and NaOH extractable P contents, representing the moderately labile fraction of P and Fe/Al associated P respectively, increase as HTT increases due to the stabilization of  $\text{H}_2\text{PO}_4^-$  and  $\text{HPO}_4^{2-}$  (Fig. 5c). Also, the content of apatite (AIP), one of the most stable forms of P, increases with increasing HTC temperature (Fig. 5d). Additionally, it would be a promising strategy to sustainably achieve the goals of P recovery and recycling with the help of hydrochars prepared from the biowaste like sludge and food waste [114,115]. However, high levels of heavy metals in the feedstocks could concentrate in the P-rich hydrochars (Table S2), complicating the recovery processes and application in soils. More investigations in future should be merited for exploring the hydrochar-

based technology for P recovery and recycling without heavy metal pollution.

Heavy metals such as Cu, Zn, As, Ni, Cd, and Pb have been widely reported in hydrochars (Table S2). They are mainly derived from feedstocks contaminated by heavy metals, such as sewage sludge, swine manure, and poultry litter [86,116]. For instance, the concentration of Cu, Zn, Cd, Pb, Ni and As in the hydrochars derived from sewage sludge ranges 377–438, 1581–2841, 4.98–6.39, 81–90.9, 41.7–52.3, 7.10–11.5  $\text{mg kg}^{-1}$ , respectively. For the hydrochars derived from the other biomass (e.g., cellulose, lignin, xylan, sunflower residue, and municipal solid waste), the concentration of Cu, Zn, Pb and Ni was 2.51–168, 21.2–47.7, 11.4–28.7 and 1.59–24.4  $\text{mg kg}^{-1}$ , respectively [117–119]. The occurrence of heavy metals in hydrochars can pose serious environmental risks during their application [120], which is ignored in the past. Hence, future studies on minimizing heavy metal contents in hydrochars are warranted to avoid their environmental risks.

### 3.3. Morphological and surface characteristics

Morphological characteristics of hydrochars, including shape, particle size, pore structure, and surface area, can be considered for their targeted applications such as adsorbents [53], catalysts [53], and soil amendments [122]. The microstructures of hydrochars derived from different feedstocks are summarized in Table 2. The surface morphology and structure of hydrochars are largely controlled by the type of feedstocks [32]. Hydrochars with spherical morphology generally show granular surfaces with floccular, lamellar, or honeycomb structures, resulting from the decomposition of carbohydrates in lignocellulosic feedstock [123]. In contrast, biochars derived from lignocellulosic materials

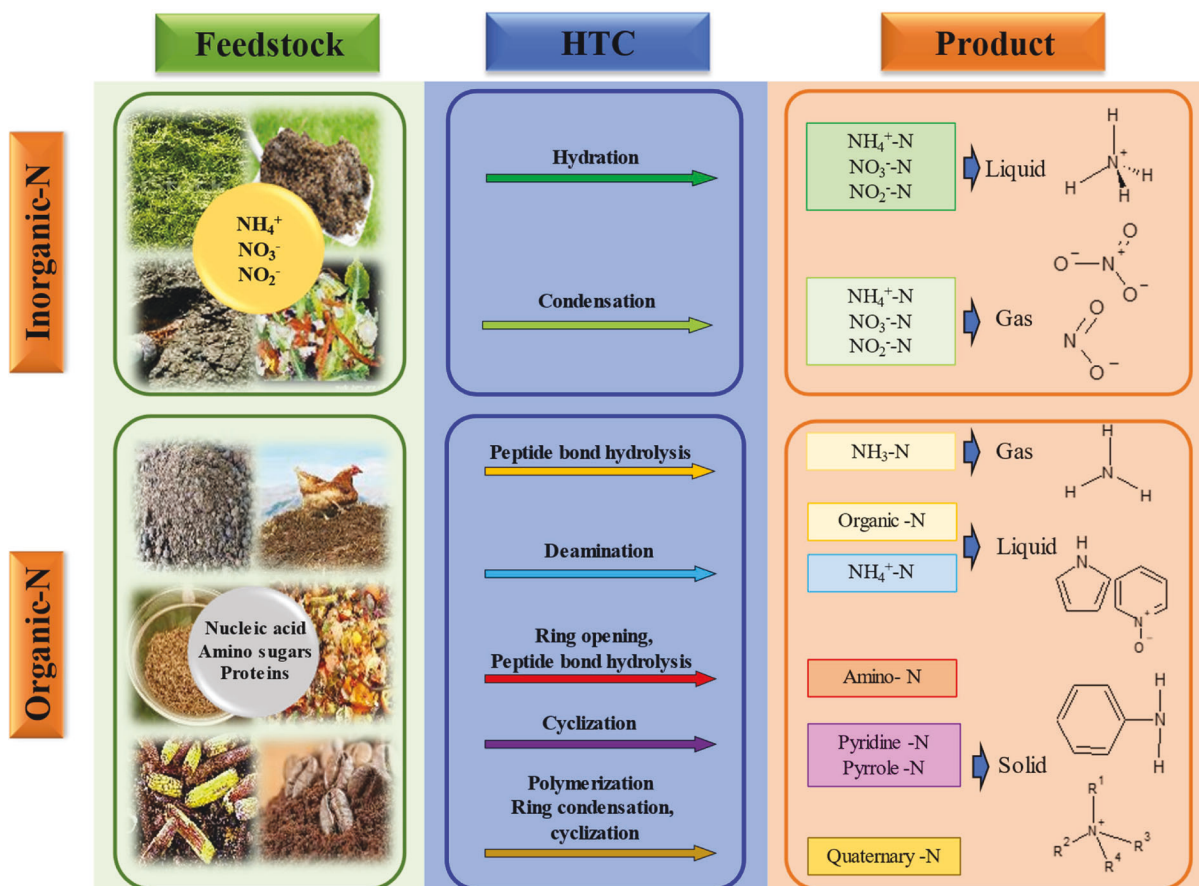
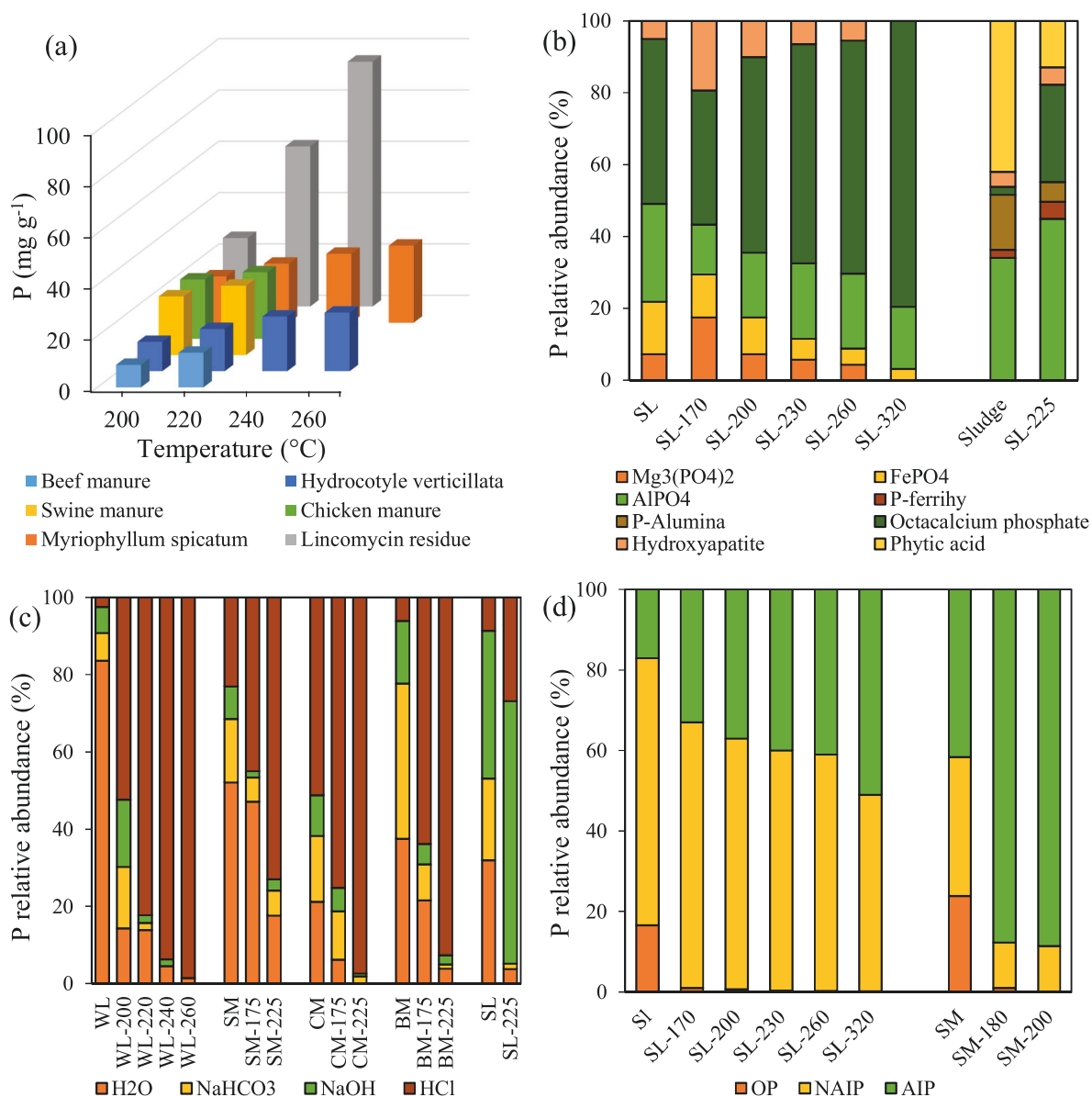


Fig. 4. Reactions of N in hydrochar feedstock during HTC. Inorganic N could be converted to liquid and gas phases at the initial stage of HTC. Some parts of the organic N may go through chemical reactions of hydrolysis and deamination, and other parts would be hydrolyzed to small fractions via ring opening, ring condensation, polymerization, and cyclization, thus resulting in the formation of heterocyclic N in hydrochars [97,98,100,109,110].



**Fig. 5.** P species and contents in different feedstocks and their derived hydrochars produced at different heating temperatures (HTTs). (a) total P content in hydrochars derived from different feedstocks. (b) Different organic and inorganic P species in hydrochar analyzed by P K-edge XANES. P in hydrochar generally bonds with various metals, such as Mg, Al, Ca, and Fe to form different inorganic fractions ( $\text{Mg}_3\text{PO}_4$ : Mg-associated, P;  $\text{FePO}_4$ : Fe-associated, P;  $\text{AlPO}_4$ : Al-associated P; P-ferrihy: phosphate sorbed on ferrihydrite; P-Alumina: phosphate sorbed on  $\gamma$ -alumina). SL: sludge. (c) Chemical forms of P extracted by  $\text{H}_2\text{O}$ ,  $\text{NaHCO}_3$ ,  $\text{NaOH}$ , and  $\text{HCl}$  solutions following the Hedley sequential extraction method [121], representing readily soluble P, exchangeable P, Fe/Al mineral adsorbed P, and insoluble P, respectively. SL: sludge; WL: wetland plants; SM: swine manure; CM: chicken manure; BM: beef manure. (d) Relative abundance of organic and inorganic P (OP: organic P; NAIP: non apatite inorganic P; AIP: apatite inorganic P). SL: sludge; SM: swine manure. The data after the names of the feedstock represent the HTC temperature. For example, SL-170 stands for the sludge hydrochar produced at 170 °C. The data were obtained from the reported studies [14,96,101,112,113,116].

generally processes turbo-strategically arranged graphite-like layers [1,32,54]. Hydrochars display as small particle sizes with discrete spheres or agglomerates [36], while biochars exhibits flattened particles, particularly those produced at higher temperatures [21]. HTC at low temperatures of 150–200 °C triggers the degradation of carbohydrate, protein, and lipids in feedstocks (e.g., sewage sludge, animal manure, and plant residues), resulting in rough surfaces, high contents of pores and cavities, and filamentous structure [86]. HTC at relatively high temperatures of 200–250 °C produces rougher surfaces and more cavities and micropores due to dehydration, deformation, fusion, and volatile matter release [36]. Above 250 °C, the pore structure of hydrochar starts to collapse and shrink due to the reformation of biopolymers, thus decreasing its porosity and surface area [124]. In addition to HTT,

the pH of process water in HTC induces significant changes in hydrochar morphology [125]. Acidic water during HTC causes spherical and granular porous structures [125], while alkaline water produces cambium lamellar structures of hydrochar [125]. Surface areas of hydrochars range 1.1–30.6 m<sup>2</sup> g<sup>-1</sup> (Fig. S2). Compared to biochars, hydrochars generally have low surface areas due to the relatively low HTT and short residence time, resulting in incomplete pore development [126,127]. Type of feedstock plays an important role in hydrochar surface areas (Fig. S2). Hydrochars derived from lignocellulosic materials (e.g., canola straw, wheat straw, hickory, peanut hull, and rice straw) exhibit higher surface areas than those of non-lignocellulosic materials (e.g., sewage sludge, animal manure). In addition to feedstock, HTT also significantly regulates hydrochar surface area [99]. As

**Table 2**  
Summary of the morphological structure of hydrochars derived from different feedstocks.

Feedstock	HTT (°C)	RT (h)	SLR	Morphology of hydrochar	Reference	
Types	Structure and morphology					
Wood sawdust	Fibrous, non-porous	220	1.5	1:4	Disordered fibers, slightly porous	[123]
Walnut shell	Non-porous	220	1.5	1:4	Circular pores	
Tea stalk	Fibrous, non-porous	220	1.5	1:4	Honeycomb shaped, thick-wall pores	
Olive pomace	Fibrous	220	1.5	1:4	Presence of channels and thick walled pores	
Apricot seed	Layered, non-porous	220	1.5	1:4	Presence of microspheres	
Hazelnut husk	Nonporous	220	1.5	1:4	Thick walled and circular pores	
Spent coffee grounds	Rough and irregular surface morphology	180, 200, 220	1, 3, 5	1:10	Tunneling, microstructural fragments, enlarged pores	[36]
Corncoobs	–	230	0.5	1:6	Slightly opened channels, fine pores, microspheres	[130]
Corncoobs	–	260	0.5	1:6	Fine pores, broken and rough surface with cracks and channels	
Food waste	Aggregated matrix, irregular particles, few pores, and pathways	180, 260	1	1:5	Peanut like microparticles, microspheres, porous	[34]
Pine wood	Amorphous	180	20	1:8.5	Irregular structure, porous, layered, nanopores, short-range ring structure, irregular structure, micrometer particles and pores	[131]
Corn stover	Micro fibrous, cellulose, semi cellulose and lignin chains	180, 260	4	1:8	Carbon spheres, nano and micro spheres	[132]
Swine manure	clustered aggregates and few pore structures on its surface	280, 200, 220	10	1:4	Small fragments, different sized pores	[116]
Maize straw	Smooth, flat, and highly organized fiber structure with few dense pores	220, 340	0.25, 0.33	1:3	Microsphere structures, highly porous	[33]
Sewage sludge	–	270	2	1:9	Granular, floc and lamellar structure, honeycomb structure, porous structure	[125]

HTT: heating temperature; RT: residence time of HTC process; SLR: solid of feedstock to liquid ratio (w/v) during HTC.

the HTT increases to 250°C, the enhanced carbonization of biomass would result in development of abundant pores in hydrochars [128], while higher temperature (> 250°C) decreases the surface area due to the blockage of pores by condensed volatile matters and sedimented minerals [129]. Moreover, low pH of the process water will facilitate the hydrolysis of carbohydrate and enhance microsphere formation, thus increasing hydrochar surface area at the early stage of HTC [126]. However, relative to biochars, the morphological characteristics of hydrochars have not been fully understood yet. Future research should address the customized production of hydrochars with desired morphological characteristics for targeted applications.

### 3.4. Surface functional groups

Surface functional groups, one of the most important characteristics of hydrochars, contribute to their high activity and reactivity in environmental remediation and soil conditioning [21,53]. O- and N-containing functional groups are the two most important groups in hydrochars and biochars [17,53,55]. O-containing functional groups, including hydroxyls (–OH), carboxyls (–COOH), ketones (–C = O), and ethers (C–O), are mainly derived from hydrolysis, dehydration, condensation, and polymerization of organic components such as carbohydrates and lignins in biomass during HTC [7,17,53,86]. However, high temperature (e.g., 500–700°C) of pyrolysis produces lower O-containing functional groups in biochars. For instance, Zhang et al. reported that the contents of O-containing functional groups in a hydrochar derived from coffee ground waste at 160°C were 13.1–104% higher than those of the biochar derived from the same feedstock at 400°C and 500°C [104]. Thus, low contents of these groups trigger higher stability of biochars toward microbial and chemical degradation relative to hydrochars [47,86]. Compared to O-containing functional groups, N-containing groups such as pyridinic-N, pyrrolic-N, quaternary-N, and pyridinic-N-oxide in hydrochars have received less attention [133]. Thus, more efforts are necessary to understand the formation and function of N-containing functional groups in hydrochars to expand the potential benefits of hydrochars in different applications.

## 4. Application of hydrochars for soil improvement

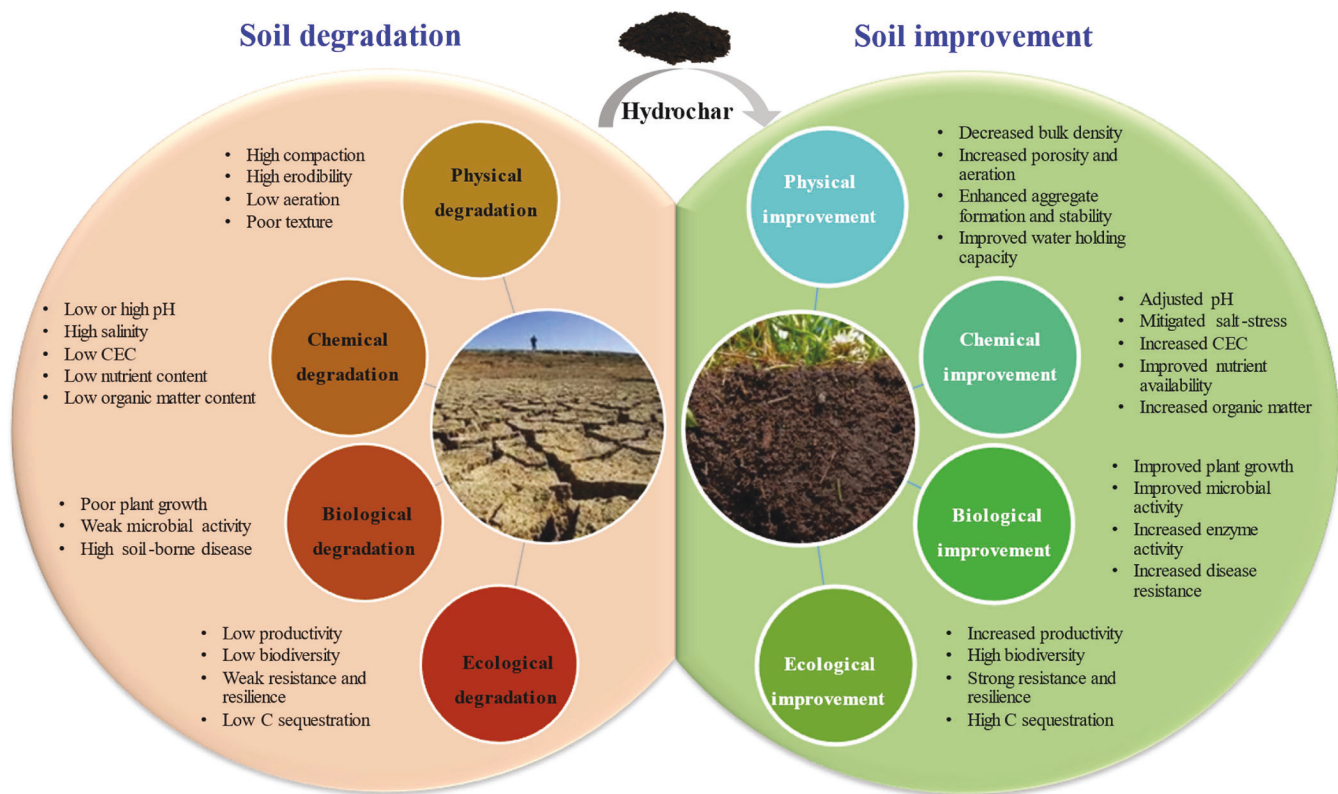
Increasing soil degradation has posed serious threats to agricultural

production, ecosystem sustainability, and global climate [134]. Supplement with soil organic carbon (SOC) is one of the most important and feasible strategies to improve soil quality, increase crop production [135], enhance C sequestration, and mitigate GHG emissions [136]. Biochars are promising soil amendments and have been extensively studied and reviewed [21,137]. Recently, increasing studies evidenced that hydrochars can also be promising multifunctional soil amendments (Fig. 6). Application of hydrochars can improve soil physical, chemical, and biological properties [122,138], enhance C sequestration, [134], decrease bioavailability and toxicity of contaminants [53], and restore ecosystem structure and function [13,134]. Feedstock and HTC conditions play key roles in the performance of hydrochar application in soil improvement [7,122,138]. However, the large variety of feedstocks for hydrochar production induces highly varied performance of different hydrochars.

### 4.1. Effects of hydrochars on soil physical properties

Degraded soils generally show poor physical characteristics in texture, structure, porosity, bulk density, and water holding capacity (WHC). Increasing studies reported that hydrochar amendment might effectively improve these soil physical properties [122,139]. Heavy textured soils (e.g., clayey soils) with significantly low porosity and high bulk density (~1.6 g cm<sup>-3</sup>) are at high risks of compaction, water-logging, and erosion [138]. Recent studies demonstrated that hydrochar application increased soil porosity by 6.3–11.5% [138], decreased bulk density by 8.2–18.9% [140], and promoted the formation and stability of soil aggregates [122,137,140]. These positive changes have been observed in soils of different textures, such as clay soils [139,140], sandy soils [138], and loamy soils [141]. High temperature (≥ 200°C) hydrochars processing rich porous structure and low bulk density (0.1–0.2 g cm<sup>-3</sup>) are more suitable for improving these soils [32,140]. Hydrochar application may also improve soil WHC and thus increase plant available water capacity (AWC) due to water retention by hydrochar pores [13,130], as well as the enhancement of soil aggregation [122]. In addition, hydrochar application may increase soil macropores, thus improve soil drainage [137,139] and water uptake by plants [13]. These positive effects are mainly controlled by internal porosity, specific surface area, and the hydrophilic surface of hydrochars [13]. Char particle size is an important parameter in controlling soil





**Fig. 6.** Summary of the issues of the degraded soils and effects of hydrochar application in these soils. Hydrochar amendment can benefit physical, chemical, biological, and ecological characteristics of the degraded soils. These improvements can result in high soil fertility, biological activity and diversity, thus increase crop productivity and improve ecosystem structure and function.

water retention capacity and permeability [139]. Hydrochars produced at high temperatures ( $\geq 200^\circ\text{C}$ ) usually have smaller particle sizes [138,142] and can block soil micropores and simultaneously decrease water entrance and retention [13,142]. Moreover, the blockage of soil micropores by small-sized hydrochars can result in lower porosity and aeration, increasing soil compaction [139,143]. Hydrochar application in sandy soils can increase WHC and AWC more effectively than clay and loamy soil [13]. Several studies also evidenced that hydrochars enhanced the aggregate formation and stability of loamy and clay soils [122,134]. On the one hand, surface functional groups of hydrochars such as hydroxyl ( $-\text{OH}$ ) and carboxyl ( $-\text{COOH}$ ) triggers the interaction of cationic bridges mainly responsible for the formation of micro-aggregates/macroaggregates in soils [134]. On the other hand, the improvement of soil aggregation may have resulted from a variety of organic substances such as organic acids and fats produced by soil bacteria, fungi, and plant roots [122], which could be enhanced by the addition of hydrochars [122]. Moreover, hydrochars can improve soil aggregate stability better than biochars due to their richer functional groups and mineral contents [134].

So far, although positive effects of hydrochars on soil physical properties have been demonstrated, huge knowledge gaps regarding the responses of soil physical properties to the application of different types of hydrochars should be further considered. The effects of interactions among soil components such as SOC, minerals, and microorganisms with hydrochar particles on soil physical properties are still unclear. Given the diversity and complexity of soil environments, the mechanisms underlying hydrochar behavior in soils under different conditions such as temperature and moisture require further investigations to establish the relationships between the hydrochar characteristics and soil physical properties.

#### 4.2. Effects of hydrochars on soil chemical properties

Extensive studies have been conducted to investigate the effects of hydrochar application on improving the chemical properties of degraded soils, such as pH, cation exchange capacity (CEC), electrical conductivity (EC), and SOC [143,144]. Depending on feedstock types and HTC conditions, hydrochars have been demonstrated to effectively improve highly weathered soils with poor chemical properties such as high EC and low CEC and SOC [15,143,145,146]. Compared to hydrochars, biochars have less effects on soil CEC, due to their inherent lower CEC resulted from higher decomposition rate of organic matter during pyrolysis [83]. Application of biochars in acidic soils has been extensively studied and highly recommended due to the inherent alkalinity of biochars resulted from the concentrated minerals during biomass pyrolysis [137,147]. Hydrochars, generally having an acidic nature ( $\text{pH} < 7.38$ ) due to the presence of organic acids [1], may effectively decrease soil pH (such as alkaline and calcareous soils) [141,145,148], thus alleviate salt stress and increase nutrient availability [149]. However, Rilling et al. observed an increase in soil pH from 7.2 to 7.6 following the application of a hydrochar with pH 4.39 [149]. They attributed the increased pH to the proton consuming reduction activities of soil microorganisms, which decreased the release of acidic metabolites [149]. Studies on the application of hydrochars in improving soil EC, an indicator of soil salinity, are very limited [148]. Because of the lower mineral contents in hydrochars than biochars, it is reasonable to hypothesize that hydrochars would lower EC enhancement than biochars [140,148].

Recent studies showed that soil CEC, an indicator of soil capacity for retaining and providing nutrients to crops, may increase upon hydrochar application [15,54,142]. The ability of hydrochars for increasing soil CEC, attributed to the high surface areas and surface O-containing functional groups [145], strongly depends on feedstock types, HTC conditions, and soil characteristics and interactions [54,141]. Generally,

hydrochars derived from lignocellulosic feedstock (e.g., crop straw and woody chips) exhibit higher CEC than sewage sludge and municipal waste [145,150]. Lower CEC is expected for hydrochars produced at higher temperatures ( $\geq 200^\circ\text{C}$ ) because of decreased reactive functional groups [151]. Thus, the plant-derived hydrochars at low temperature ( $< 200^\circ\text{C}$ ) is more likely to improve soil CEC than the sewage sludge-derived hydrochars produced at high temperature ( $\geq 200^\circ\text{C}$ ) [151]. Due to the high diversity of degraded soils and technical limitations for functionalizing hydrochar functionality, the application of an individual hydrochar might not always achieve the expected positive effects in improving soil qualities [152]. The combined application of hydrochars with other soil amendments (e.g., compost and lime) and/or chemical fertilizers could be an alternative strategy, which warrants future exploration.

#### 4.3. Effects of hydrochars as slow-release fertilizers on soil nutrient availability

Hydrochars generally contain nutrients such as N, P, K, Ca, and Mg [102,146] and can be directly used as slow-release fertilizers for plants, especially those grown in infertile soils [146,152]. The fertilization potential of hydrochars is highly controlled by the feedstock types and HTC conditions [141]. For instance, the hydrochars derived from manure are richer in nutrients, including N, P, K, Ca, and Mg, than those from plant biomass [32]. Hydrochars could directly provide N to crops because of their inherent inorganic N (e.g.,  $\text{NH}_4^+$  and  $\text{NO}_3^-$ ) and organic N (e.g., amino acids, phospholipids, and amino sugars) originated from the feedstock such as sewage sludge, animal waste, and plant residues (Fig. 2f, 4). Meanwhile, hydrochars can enhance  $\text{NH}_4^+$  and  $\text{NO}_3^-$  retention in soils by sorption via electrostatic attraction and pore-filling [26,30]. This results in slow-release of N in soils for plant uptake [152], and decreases N leaching from soils [153]. Compared to biochars, hydrochars could have higher adsorption capacities for  $\text{NH}_4^+$  due to their abundant O-containing functional groups such as carboxyl and ketone groups [154]. Modified hydrochars showed better performance for retaining N in soils than the un-modified hydrochar [154], which should be further studied in the future.

The advantage of hydrochar application as P fertilizer outweighs its application as an N fertilizer [155]. P in hydrochar, mostly presented in Al-associated P and Ca-associated P forms, can provide plant-available P over time (Fig. 5a) [112]. For example, Fei et al. reported that a sewage sludge-derived hydrochar increased available soil P by 130%. They also showed that 86.8% of the available P accounted for 2% of total P in the hydrochar released into the soil [151]. While hydrochars are considered and applied as slow-release P fertilizers, low-temperature hydrochars ( $< 200^\circ\text{C}$ ) could release P faster than high-temperature hydrochars ( $> 200^\circ\text{C}$ ) [156] due to the stabilization of water-soluble P in hydrochar as the HTT increases during HTC (Fig. 5b). Notably, little information was available for the effects of hydrochar on soil P cycling [151], which should be further explored in future.

More studies regarding the inherent nutrient potential of hydrochars and associated effects on soil fertility are still needed. A systematic comparison of hydrochars from different feedstocks and HTC conditions should be conducted to assess their nutrient availability as slow-release fertilizers and predict the potential impacts on soil nutrient availability in different soil environments. Additionally, hydrochars as slow-release fertilizers would interact with other soil substances such as chemical fertilizers and pesticides, affecting their biogeochemical cycling and efficiency. Thus, further research is needed to investigate the fertilizing potential of hydrochars in the presence of common soil substances.

#### 4.4. Effects of hydrochars on crop productivity

Improving soil quality, nutrient availability, and crop productivity are the main targets of sustainable agriculture following hydrochar application [142,154]. Effects of hydrochar application on plant growth

are summarized in Table 3. Increased crop productivity following hydrochar application is often observed in infertile or degraded soils than fertile soils [142]. Effects of hydrochars have been studied on various crops such as barley [157], leek [146], beans [143], mastic [144], myrtle [144], lettuce [158], rice [159], and alfalfa [122]. The positive response of hydrochars in crop productivity, accounting for 62% of the selected studies ( $n = 14$ , Table 3), is mainly attributed to the direct supply of essential nutrients in hydrochars for crops [146,154], as well as the improvement of soil physical and chemical properties [141,142]. If hydrochars stay in soils for long term (e.g., more than three months), the plant growth improvement is even better due to the slow release of nutrients in hydrochars and their aging effects on native soil nutrient availability [157].

Hydrochars can also inhibit crop growth and decrease their production [146,163]. These negative effects, accounting for 38% of the selected studies ( $n = 14$ , Table 3), were observed for oat [160], alfalfa [122], dandelion [149], sugar beet [163], mastic [144] and leek [146]. These studies highlight the potential environmental risks of hydrochar as soil amendments. The negative effects of hydrochars on plant growth could be attributed to the following reasons. On the one hand, the decreased plant growth may be attributed to the adverse effect of hydrochars on soil properties such as increasing soil C/N ratios and decreasing soil pH, thus leading to the enhanced microbial N immobilization and decreased N uptake by plants [146,163]. On the other hand, the negative effects could be ascribed to the inherent contaminants of hydrochars, such as heavy metals [144], PAHs, phenols, and furfural [146,163]. Hydrochars derived from sludge, and poultry litter generally contain high contents of heavy metals, showing detrimental effects on grass seed growth following hydrochar application in soils [102,164]. Heavy metals in hydrochars can also be leached to the deep soil and pollute groundwater [166]. In these cases, besides selecting the suitable feedstock without heavy metal contamination, modification of hydrochars could reduce the potential environmental and health risks [37,167], which merits further study. For example, Lang et al. found that a swine manure hydrochar modified by CaO addition decreased the leached amounts of Cu, Zn, and Mn by 93.6%, 89.6%, and 79.8%, respectively. They attributed this reduction to the increased surface negative charges, surface areas and O-containing functional groups [168].

Overall, the application of hydrochars for crop production shows inconsistent results for various plants, mainly due to the feedstock types, HTC conditions, crop species, soil, environmental conditions, and complicated interactions. Research efforts are needed to reveal the relationships between the characteristics of hydrochars and the responses of different crops, and the functions and mechanisms of hydrochars in enhancing plant growth and productivity. It is worth mentioning that neither biochars nor hydrochars can meet all the application needs and demands in agricultural production. Hence, a suitable hydrochar should be used at optimum dosage for a given crop in a specific soil.

#### 4.5. Effects of hydrochars on soil greenhouse gas emission

The annual GHG emission from agricultural activities is estimated to be 619 million metric tons  $\text{CO}_2$ -equivalent [169], largely contributing to global warming. Hence, reduction of GHG emissions from agricultural soils is necessary. Promisingly, biochars can sequester C in soils for hundreds to thousands of years due to their high stability [170]. Extensive studies have evidenced the good performance of biochars on GHG emissions from various soils [136,171]. Recently, the potential of hydrochars in reducing soil GHG emissions has received increasing attention [154,163,172]. Emissions of  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$  from the soils amended with hydrochars were highly variable in paddy soils [172,173], eroded agricultural soils [174], and grasslands [136]. Generally, hydrochar application increases  $\text{CO}_2$  [136,175] and  $\text{CH}_4$  emission [172-174] from different soils such as forest soils, agricultural soils, and grasslands. Higher gas emissions are mainly attributed to the

**Table 3**  
Summarized effects of hydrochar application on plant growth.

Hydrochar feedstock	HTT (°C)	RT (h)	Application rate	Plant type	Response of plant	Reference
Poplar wood dust	200	2	1%, 2.5%, 5% (w/w)	Oat ( <i>Avena sativa</i> L.)	Decreased shoot dry matter by 14–50%	[160]
Maize silage	230	1.25	30t ha <sup>-1</sup>	Poplar ( <i>Populus alba</i> L.)	Increased shoot dry matter by 37% and shoot length by 20%	[161]
Maize	200	4	0.7% (w/w)	Soybean ( <i>Glycine max</i> )	Increased total dry biomass by 13%.	[162]
Wood	200	4	0.7% (w/w)	Soybean ( <i>Glycine max</i> )	No significant effect on dry biomass.	
Spent coffee grains	220	12	5%, 10% (w/w)	Alfalfa ( <i>Medicago sativa</i> )	Decreased shoot dry biomass by 20–30% Increased leaf tip necrosis by 400–600%.	[122]
Beet root chips	180–200	11	2%, 4%, 10%, 20%, 30%, 80% (v/v)	Dandelion ( <i>Taraxacum</i> )	Decreased total biomass by 0.5–82%	[149]
Beet root chips	180–200	11	10% (v/v)	Clover ( <i>Trifolium</i> )	Decreased shoot dry weight by 36%, number of leaves by 9% and root dry weight by 44%.	
Beet root chip	190	4	2%, 4%, 10% (v/v)	Summer barley ( <i>Hordeum vulgare</i> )	Increased shoot dry matter yield by 4.0%, 6.0% and 0.2% respectively.	[146]
Beet root chip	190	12	2%, 4%, 10% (v/v)	Summer barley ( <i>Hordeum vulgare</i> )	Increased shoot dry matter yield by 32%, 46% respectively in 2 and 4% hydrochar application respectively. Decreased dry biomass production by 3.0 % in 10% hydrochar application.	
Beet root chip	190	4	2%, 4%, 10% (v/v)	Phaseolus beans ( <i>Phaseolus</i> )	Increased shoot dry matter yield by 88%, 147% and 37% respectively.	
Beet root chip	190	12	2%, 4%, 10% (v/v)	Phaseolus beans ( <i>Phaseolus</i> )	Increased dry matter yield by 53%, 107% and 61% respectively.	
Beet root chip	190	4	2%, 4%, 10% (v/v)	Leek ( <i>Allium ampeloprasum</i> )	Decreased shoot dry matter yield 3%, 10% and 77% respectively.	
Beet root chip	190	12	2%, 4%, 10% (v/v)	Leek ( <i>Allium ampeloprasum</i> )	Increased shoot dry matter yield by 61%, 25% in 2% and 4% hydrochar treatment respectively. Decreased dry biomass production by 1% in 10% hydrochar treatments.	
Spent brewer grain	190	4	2%, 4%, 10% (v/v)	Summer barley ( <i>Hordeum vulgare</i> )	Increased shoot dry matter yield, by 31%, 26% and 29% respectively.	
Spent brewer grain	190	12	2%, 4%, 10% (v/v)	Summer barley ( <i>Hordeum vulgare</i> )	Increased shoot dry matter yield by 32%, 17% and 1% respectively.	
Spent brewer grain	190	4	2%, 4%, 10% (v/v)	Phaseolus beans ( <i>Phaseolus Vulgaris</i> )	Increased shoot dry matter yield by 14%, 60% and 103%, respectively.	
Spent brewer grain	190	12	2%, 4%, 10% (v/v)	Phaseolus beans ( <i>Phaseolus Vulgaris</i> )	Increased shoot dry matter yield by 59%, 58% and 52% respectively.	
Spent brewer grain	190	4	2%, 4%, 10% (v/v)	Leek ( <i>Allium ampeloprasum</i> )	Decreased the shoot dry matter yield by 34%, 22% and 65%-respectively.	
Spent brewer grain	190	12	2%, 4%, 10% (v/v)	Leek ( <i>Allium ampeloprasum</i> )	Increased shoot dry matter yield by 4% in 2% hydrochar treatments. Decreased dry biomass production by 15% and 64% in 4% and 10% hydrochar treatments.	
Sugar beet pulp	190	12	1.32% (w/w)	Sugar beet ( <i>Beta vulgaris</i> L.)	Decreased yield by 97% and plant total N content by 25%. Increased total plant P content by 10%.	[163]
Beer draff	190	12	1.32% (w/w)	Sugar beet ( <i>Beta vulgaris</i> L.)	Decrease dry matter yield by 40% and plant total N content by 8%. Increased total plant P content by 2%.	
Forest wastes	–	–	50% (v/v)	Myrtle ( <i>Myrtus communis</i> L.)	Decreased seed germination by 23%, seedling survival by 22%, and stem dry weight by 75%.	[144]
Forest wastes	–	–	25% (v/v)	Myrtle ( <i>Myrtus communis</i> L.)	Decreased seed germination by 6%, seedling survival by 5%, and stem dry weight by 56%, respectively.	
Forest wastes	–	–	10% (v/v)	Myrtle ( <i>Myrtus communis</i> L.)	Increased seed germination by 13%. Decreased seedling survival by 22% and stem dry weight by 61%.	
Forest wastes	–	–	50% (v/v)	Mastic ( <i>Pistacia lentiscus</i> L.),	Decreased seed germination by 34%, seedling survival by 37%, and stem dry weight by 48%.	
Forest wastes	–	–	25% (v/v)	Mastic ( <i>Pistacia lentiscus</i> L.)	Decreased seed germination by 21%, seedling survival by 13%, and stem dry weight 28%.	
Forest wastes	–	–	10% (v/v)	Mastic ( <i>Pistacia lentiscus</i> L.)	Increased seed germination by 18%. Decreased seedling survival by 12%, and stem dry weight by 11%.	
Sewage sludge	200	0.5	0.8%, 4% (w/w)	Grass seeds ( <i>Lolium perenne</i> )	Increased dry biomass by 40–95%.	[164]
Sewage sludge	200	3	0.8%, 4% (w/w)	Grass seeds ( <i>Lolium perenne</i> )	Increased dry biomass by 42–85%.	
Sewage sludge	260	0.5	0.8%, 4% (w/w)	Grass seeds ( <i>Lolium perenne</i> )	Increased dry biomass by 42% for 4% hydrochar application. No significant effect on dry biomass for 0.8% hydrochar application.	
Sewage sludge	260	3	0.8%, 4% (w/w)	Grass seeds ( <i>Lolium perenne</i> )	Decreased dry biomass by 1% and 5% respectively.	
Biosolid from WWTP	190	4	0.8%, 1.6% (w/w)	Phaseolus beans ( <i>Phaseolus Vulgaris</i> )	Increased total dry biomass by 96–112%.	[142]
<i>Miscanthus and giganteus</i>	200	2	14.5t ha <sup>-1</sup>	Perennial ryegrass ( <i>Lolium perenne</i> )	Increased dry biomass by 32%.	[136]
Beet root chip	180–200	11	1%, 10% (v/v)	Plantain ( <i>Plantago lanceolata</i> )	Increased dry biomass by 60% for 10% hydrochar application. No significant effect on shoot and root dry biomass by 1% hydrochar application.	[165]
Poultry litter	180	1	0.5%, 1% (w/w)		Increased shoot dry matter by 145–146%.	[158]

(continued on next page)



Table 3 (continued)

Hydrochar feedstock	HTT (°C)	RT (h)	Application rate	Plant type	Response of plant	Reference
Sawdust	220	1	5%, 15% (w/w)	Lettuce ( <i>Lactuca sativa</i> )	Increased grain yield by 16.6–19.3%.	[159]
	250					
	260					

HTT: heating temperature; RT: residence time of HTC process.

high contents of labile C in hydrochars, providing extra substrates for soil microorganisms such as actinomycetes, fungi, N-fixing bacteria, and methanogens [172–174]. Hydrochars produced at high temperature ( $\geq 200^\circ\text{C}$ ) containing less labile C and more aromatic C, may release less  $\text{CO}_2$  compared to the hydrochars produced at low temperature ( $< 200^\circ\text{C}$ ) [47,86,175]. Washing hydrochars to remove the labile C fraction before their applications could decrease  $\text{CO}_2$  emissions due to the inherent labile C [172]. Moreover, hydrochar modification can significantly decrease  $\text{CO}_2$  emission from soils. Vieillard et al. observed that a hydrochar modified by grafting aminosilane increased the  $\text{CO}_2$  adsorption via intraparticle diffusion [176], which showed good potential in effectively decreasing  $\text{CO}_2$  emission from soils.

$\text{CH}_4$  emission originated from human activities (e.g., coal mining, biomass burning, and garbage disposal) accounts for 20% of the global anthropogenic warming effect [170]. To date, only a few studies reported the effects of hydrochars on  $\text{CH}_4$  release from soils [159,177], and most of these studies focused on paddy soils [172,173,178]. They found that hydrochars showed inconsistent effects on  $\text{CH}_4$  emission, including promotion [172,173], inhibition [177,178], and no effect [159]. For example, Ji et al. reported that the application of hydrochars derived from rice straws at 200, 250 and  $300^\circ\text{C}$  into a paddy soil increased the cumulative  $\text{CH}_4$  emission by 150–430% [37]. They explained the enhanced emission by the released labile organic carbons of the hydrochars and shifted microbial communities to  $\text{CH}_4$ -producing communities (e.g., *Euryarchaeota*, *Janibacter*, *Anaeromyxobacter*, *Anaerolinea*, and *Sporacetigenium*). Consistently, they further observed that the corresponding water-washed hydrochars had little effect on  $\text{CH}_4$  emission from the same paddy soil. Therefore, it would be necessary to pretreat (e.g., washing) hydrochars before their applications, which could be an efficient method to avoid the enhanced  $\text{CH}_4$  emission from paddy soils [172,178]. Another study reported that the higher rate (3%) of hydrochar application results in a larger amount of  $\text{CH}_4$  emission relative to a lower rate (0.5%) application due to the high content of labile C available for methane producing microorganisms [173]. On the contrary, Chen et al. observed that a poplar sawdust derived hydrochar applied at 0.5% into a paddy soil fertilized with urea inhibited the cumulative  $\text{CH}_4$  emissions by 14.8%, mainly due to the reduced expression of the methanogenic *mcrA* gene [173]. However, little information is available on the effect of hydrochars on  $\text{CH}_4$  emission from natural wetlands such as coastal wetlands and estuarine areas, important parts of blue C ecosystems, which should receive more efforts in future studies.

Application of biochars as soil carbon sequestration materials has attracted a great deal of worldwide attention in past decades as a strategy for  $\text{CO}_2$  mitigation [179–182], because of their high recalcitrance against microbial decomposition and negative priming effects on native SOC [171,182–184]. Thus, the high temperature biochars are preferentially recommended from a C sequestration perspective [185]. However, the application of hydrochars for sequestering  $\text{CO}_2$  in soils is still at infancy, and the limited studies showed the controversial effects [136,174,186]. For example, Sun, et al. found that hydrochar addition at 0.5% and 1.5% decreased the labile SOC fraction by 15.6–33.6% and increased the stable SOC fraction by 10.3–27.0% in a paddy soil [145]. Furthermore, they found that SOC in the hydrochar-amended soils contained more aromatic compounds but fewer carbohydrates and lower polarity. Accordingly, they demonstrated that hydrochars could

have low carbon sequestration potentials from a long-term perspective, because of their high decomposability and positive priming effects on the mineralization of native SOC. Moreover, Malghani et al. reported that  $33 \pm 8\%$  of the added corn silage hydrochar C was lost from two coarse and fine textured soils after one year incubation, but the hydrochar-amended soils preserved  $15 \pm 4\%$  more native SOC relative to the controls, showing negative priming effects [187]. This study highlighted soil C sequestration potential of hydrochar at least on decadal timescales. However, these studies only considered limited soil type under controlled laboratory incubation conditions in the relatively short-term scale. Therefore, the benefits of hydrochars in C sequestration should be carefully examined in future, and long-term laboratory and field-scale investigations with more types of soils and hydrochars should be explored.

Effects of biochar applications on  $\text{N}_2\text{O}$  emission from soils were well documented [136,188]. A meta-analysis ( $n = 88$ ) reported that the overall  $\text{N}_2\text{O}$  emissions reduction in soils following biochar applications was 38%, and biochar strongly reduced  $\text{N}_2\text{O}$  emission in paddy and sandy soils [188]. To date, limited studies investigated the effects of hydrochar application on soil  $\text{N}_2\text{O}$  emissions, and these results were inconsistent [26,175,189]. Several reports indicated that hydrochars lowered the soil  $\text{N}_2\text{O}$  emissions due to the increased sorption of  $\text{NH}_4^+$  and  $\text{NH}_3$  by pore-filling and electrostatic attraction of hydrochars [136,175]. Additionally, the enhanced N immobilization [26,190] and decreased nitrifying and denitrifying enzyme activities have been reported [190]. For instance, a study reported that a hydrochar produced from beet chip at  $200^\circ\text{C}$  significantly decreased the activity of denitrification enzymes, which was ascribed to the decreased active sites of enzymes resulted from the surface adsorption on the hydrochar [161,174,191,192]. Similarly, a poplar hydrochar produced at  $180^\circ\text{C}$  was reported to inhibit the activity of nitrification enzymes, probably due to the leaching of toxic substance (e.g., polycyclic aromatic hydrocarbon) from the hydrochar [161,174,191,192]. The high content of labile C in hydrochars provides more energy to the heterotrophic denitrifiers, resulting in full reduction of  $\text{N}_2\text{O}$  to  $\text{N}_2$ , thus mitigating  $\text{N}_2\text{O}$  emission [136,175]. On the contrary, a few studies also reported that hydrochars increased  $\text{N}_2\text{O}$  emission from paddy fields [175,189], which were dominated by denitrification. Hydrochars can increase the activity of denitrifying bacteria and consequently enhance denitrification [189]. However,  $\text{N}_2\text{O}$  emission from the hydrochar amended soils is still not well known, and the key characteristics of hydrochars in determining soil  $\text{N}_2\text{O}$  emission are not clear. Moreover, all these studies examining the effects of hydrochars on GHG emission are limited to laboratory or greenhouse scale, which cannot reflect practical environmental conditions. Thus, future studies should be conducted on GHG emission from soils amended with hydrochars at field scale under different climatic conditions (e.g., drought, flood).

#### 4.6. Effects of hydrochars on soil microbial communities

Soil microbial community has been largely studied to assess soil quality since they can play significant roles in soil health, fertility, and productivity [149]. Extensive studies have been conducted regarding the effects of biochars on soil microbial communities [23,193]. However, studies on hydrochar interactions with soil microbes are very limited. Results from several studies indicated that application of



hydrochars to the soil increased the abundance of archaea and bacteria (e.g., *Bacillus*). Other changes in microbial community included a motivated spore germination of arbuscular mycorrhizal fungi [148,149], and increased bacterial and archaeal diversity and activity [175]. For example, Sun et al. found that the application of both hydrochars from poplar wood dust and wheat straw increased fungi diversity but decreased bacterial diversity in paddy soil [145]. They suggested that the acidic nature (pH 3–5) of hydrochars is more favorable for fungi growth and activity, while bacterial species mostly prefer neutral conditions. These effects were attributed to the high contents of nutrients, labile C fractions of hydrochars [23]. Owing to the characteristics of abundant pores and high specific surface areas, hydrochars could also provide good habitats for soil microbial colonization and prevent bacteria leaching from soil or consumption by predators [148]. Notably, hydrochars may adversely affect soil microbial growth due to the release of toxic substances. For example, a study revealed that a hydrochar produced from sewage sludge at 180°C containing high content of heavy metals decreased the microbial activities and population abundance in a soil [194]. Also, these toxic compounds in hydrochars could also pose toxic effects on soil animals, such as altering the ecological behavior of earthworms [195], decreasing the abundance of collembola *Protaphorura fimata* [196], and reducing feeding and growth of soil terrestrial isopod (*Porcellio scaber*) [197]. Therefore, more targeted studies are still needed to avoid the potential of ecological risk prior to the practical applications of hydrochars into soil ecosystems. Additionally, the direct effects of hydrochars on soil properties, such as pH, CEC, WHC, and bulk density, may also indirectly influence the soil microbial community [198]. For example, a sandy loam soil amended with a pine sawdust hydrochar produced at 200°C could hold more water in the pores under dry conditions and thus prevent microbial dormancy and death [54]. Moreover, various soils and types of hydrochars need to be evaluated to compare the microbial response in the rhizosphere and bulk soils amended with hydrochar, which has been ignored in the past.

## 5. Application of hydrochars in environmental remediation

Hydrochars have been extensively studied as low-cost sorbents for contaminant removal from soil [199] and water [15,17,19,200]. The high sorption capacities of hydrochars can be utilized for the remediation of heavy metals and organic pollutants in terrestrial [199] and aquatic environments [29]. Surface area, porosity, functional groups, aromaticity, polarity, and mineral components are the critical characteristics influencing the adsorption capacities of hydrochars to various pollutants [7,13,22].

### 5.1. Inorganic contaminants

#### 5.1.1. Heavy metals

Hydrochar application in soils and waters can decrease the availability and toxicity of heavy metals to plants [102] and microbes [15,21,102,200]. Recent studies have successfully applied different hydrochars derived from various plant materials and municipal wastes to remediate the pollution of heavy metals such as Cu, Pb, Cd, and Zn in soils and waters (Table S3). The decreased bioavailability and toxicity of heavy metals were mainly due to specific and non-specific adsorption mechanisms, including pore filling [17,200], cation- $\pi$  bonding [201], precipitation/co-precipitation [166], complexation [37,166], ion exchange [19], and electrostatic attraction [200]. Porous structure, highly reactive O-containing functional groups (e.g., hydroxyl, carboxyl), and aromatic surfaces of hydrochars can benefit heavy metal adsorption in soils and waters [27,201–203]. Surface functional groups trigger ion exchange of heavy metals with cations such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  on hydrochars [19], thus, hydrochars generally have higher adsorption affinities for heavy metals relative to biochars [166,167]. Alcohols, aldehydes, ketones, carboxylic, phenolic, and ether groups on hydrochar

surfaces can form complexes with heavy metals by donating electron pairs [19,166]. Precipitation or co-precipitation of heavy metals with minerals (e.g., phosphates and carbonates) in hydrochars is another important mechanism responsible for heavy metal remediation [29]. For example, the P-rich hydrochars derived from animal manure resulted in the precipitation of Pb as  $\text{Pb}_{10}(\text{PO}_4)_6(\text{OH})_2$  and  $\text{Ca}_2\text{Pb}_8(\text{PO}_4)_6(\text{OH})_2$  in the contaminated soils [204]. Also, modification of hydrochars using catalysts such as acids (e.g.,  $\text{HNO}_3$ ,  $\text{H}_3\text{PO}_4$ ) and bases (e.g.,  $\text{KOH}$ ) is a feasible strategy to enhance heavy metal sorption or immobilization in soils and waters [27,167].

Besides direct adsorption of heavy metals, hydrochars can also indirectly enhance adsorption or immobilization of heavy metals in soils by affecting soil properties [205]. Hydrochar application can increase soil CEC, naturally results in more cation exchange sites in the soil for heavy metal adsorption via cation exchange [15,31]. Accordingly, increasing soil CEC by hydrochar application is recommended to decrease the availability and toxicity of heavy metals [102]. Moreover, hydrochar application increases SOC content, which decreases the mobility and bioavailability of heavy metals due to their complexation with SOC [53]. For example, Xia et al. found that an amino-functionalized hydrochar derived from pinewood sawdust at 200 °C significantly increased soil CEC by 8% and SOM by 59.6%, and decreased heavy metal contents in plants by 45.9–52.5% [15]. Therefore, modification of hydrochars to increase their adsorption capacities has been proposed as an effective strategy to enhance their efficiencies in heavy metal remediation. Chemical modification using acidic and alkaline reagents (e.g.,  $\text{HNO}_3$ ,  $\text{KOH}$ ) and oxidizing agents (e.g.,  $\text{H}_2\text{O}_2$ ) was generally used to increase the species and abundances of surface O-containing functional groups and surface area [7,37,53]. Notably, modification of hydrochars should be conducted based on the target of their specific applications, which should be further explored.

#### 5.1.2. Nutrients

Nutrient pollution such as  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{PO}_4^{3-}$  in waters has caused great concerns because of the eutrophication and toxicity [30,31]. Biochars as low-cost adsorbents can effectively remove these contaminants from waters [21], enhance their sorption and decrease their leaching from soils due to the adsorption via pore-filling [170], electrostatic interaction [170], ion exchange [206], and precipitation [206]. These studies have been well-reviewed [21,170]. However, only a few studies examined the applications of hydrochars in the remediation of water polluted by  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{PO}_4^{3-}$  (Table S4S3). The sorption mechanisms of hydrochars for  $\text{NO}_3^-$  and  $\text{NH}_4^+$  include ion exchange, electrostatic attraction, hydrogen bonding, and surface complexation [30,31]. A review summarized  $\text{PO}_4^{3-}$  adsorption capacities of a series of hydrochars derived from feedstocks under different HTC conditions, ranging 14–386  $\text{mg g}^{-1}$ , whereas the adsorption capacities for biochars ranged 3–887  $\text{mg g}^{-1}$  [115]. The sorption mechanisms of hydrochars for  $\text{PO}_4^{3-}$  include precipitation, electrostatic attraction, and ion exchange [30]. Compared to biochars, hydrochars have less abilities to remediate excessive nutrients in waters due to the lower adsorption capacities and higher inherent nutrient contents [22,30]. This would weaken the remediation efficiency of hydrochars in aquatic environments [22,32]. Factors affecting nutrient sorption by hydrochars include hydrochar properties (e.g., surface area, surface functional groups, and CEC) and environmental conditions (e.g., pH, organic matter) [30,207]. HTT is a critical factor affecting hydrochar properties [22] and adsorption capacities for nutrients [32]. For example, Fei et al. reported that the hydrochar produced from sludge at 250 °C had a higher adsorption capacity (21.8  $\text{mg g}^{-1}$ ) for  $\text{PO}_4^{3-}$  than the hydrochar produced at 150 °C (15.8  $\text{mg g}^{-1}$ ) [151]. Hydrochars prepared from the nutrient-rich feedstocks such as sewage sludge and animal manure at low temperature (< 250 °C) usually have less adsorption capacities to  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{PO}_4^{3-}$  because of their high contents of these nutrients [30]. Thus, these hydrochars would be less suitable for nutrient remediation in waters. In this case, the feedstocks (e.g., saw

dust, woody chips) with low nutrient contents are warranted for preparing hydrochars with efficient adsorption capacities towards these nutrients [115]. By contrast, the hydrochars prepared at high temperature ( $> 250\text{ }^{\circ}\text{C}$ ) may play better roles in the adsorption of  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{PO}_4^{3-}$  due to the higher surface areas and less nutrient contents [208]. Still, hydrochars generally possess relatively lower adsorption capacities compared with biochars containing developed pore structures and great surface areas. Furthermore, several modification methods used to enhance the hydrochar adsorption capacities have been reported, including chemical impregnation or doping, calcination, electrochemical modification or a combination of one or more technologies [62,209]. For example, the hydrochars modified with sulfuric acid increased their microporosities, specific surface areas, and negative surface charges, thus providing more adsorption sites for  $\text{NH}_4^+$  [154]. A modified sewage sludge derived hydrochar by Mg-citrate and  $\text{H}_2\text{SO}_4$  showed better performance for retaining soil N than the un-modified one due to the enhanced  $\text{NH}_4^+$  adsorption resulted from the increased surface areas and carboxyl groups [101]. Although the pristine hydrochars generally have low P adsorption capacities due to the electrostatic repulsive interaction between them [210], increasing evidences showed that the chemical modification could enhance P adsorption performance of hydrochars. For instance, a hydrochar derived from waste corncob modified by  $\text{MgCl}_2$  showed a higher adsorption capacity to  $\text{PO}_3^{4-}$  compared with the un-modified control [211]. These enhanced adsorption capacities for  $\text{PO}_4^{3-}$  were mainly ascribed to electrostatic interaction, ion exchange, pore filling, complexation, and precipitation [31,211,212]. However, compared to biochars, studies regarding nutrient remediation are still very limited for hydrochars. Future studies are warranted to focus on the interactions of hydrochars from different feedstocks and HTC conditions with more nutrient pollutants in the water environment in practical application (e.g., constructed wetlands, biofilter, and green roof).

## 5.2. Organic pollutants

### 5.2.1. Removal of organic pollutants in water

Water pollution by organic chemicals such as pesticides, pharmaceuticals, dyes, personal care products, endocrine disruptors, flame retardants, and volatile organic compounds (VOCs) has caused great concerns globally [27,213]. Because of high porosity and rich O-containing functional groups, hydrochars have been proposed as promising adsorbents for many organic pollutants, including antibiotics, pesticides, dyes, fumigants, and polycyclic aromatic hydrocarbons (PAHs) (Table S5). Sorption of organic pollutants by hydrochars mainly attributes to pore filling [214], surface complexation [19,28], electrostatic interaction [214],  $\pi$ - $\pi$  interaction [27-29], hydrophobic interactions [104], H-bonding [27,28], and ion exchange [215]. Surface areas, pore-volumes, and O-containing functional groups are critical factors controlling organic pollutant adsorption capacities by hydrochars [216]. Hydrochars with rich O-containing functional groups show high affinities for dyes [27], pharmaceuticals [29], and pesticides [217] because of H-bonding and surface complexation between O-functional groups and these chemicals [27,28,53]. These potential mechanisms are similar to biochars, which have been well-reviewed previously [21,170]. Water conditions, including pH, ions type and strength, and dissolved organic matter (DOM), largely affect the adsorption of organic pollutants by hydrochars in the water environment [29].

Hydrochars may also degrade organic pollutants (e.g., antibiotics, dyes) in waters [20]. Application of hydrochars in the presence of daylight can increase the generation of reactive oxygen species (ROS) in water via transferring electrons to dissolved O, which can further react with  $\text{H}^+$  and produce  $\text{H}_2\text{O}_2$ , thus enhancing oxidative degradation of organic pollutants [20,218]. For example, Chen et al. found that a hydrochar derived from *Platanus acerifolia* leaf and woodchips released a large amount of  $\text{H}_2\text{O}_2$  and  $\bullet\text{OH}$  from photoactive surface O-containing functional group under daylight irradiation. The change in  $\text{H}_2\text{O}_2$  and

$\bullet\text{OH}$  was six times higher compared with dark condition [20]. Degradation of organic pollutants is also attributed to the formation of persistent free radicals (PFRs) formed on the surface of hydrochars during HTC [131] and biochars during pyrolysis [219]. PFRs, acting as electron donors, lead to ROS generation and subsequent degradation of organic contaminants [131]. However, studies in photodegradation of organic pollutants by hydrochar application in water remediation are still limited. More research needs to be conducted to fully understand the roles of ROS and PFRs of hydrochars derived from different feedstocks in the degradation of different organic pollutants in practical water remediation.

### 5.2.2. Remediation of organic pollutants in soil

Soil contamination by organic compounds such as pesticides, bio-cides, pharmaceuticals, flame retardants, surfactants, and dyes, greatly threaten soil health and food safety [220,221]. Soils, as one of the most complex environments, is more difficult to remediate than water due to various elemental and organic components, different living organism's habitat, and environmental conditions [221]. Many studies have investigated the remediation potential of biochars for different soils contaminated by various pollutants such as pesticides, antibiotics, PCBs, PAHs [206,222], which have been well reviewed [21,220]. Recently, several studies reported the remediation of polluted soils using hydrochars (Table S3). Hydrochars can increase the sorption capacity of soils to organic contaminants and consequently decrease the bioavailability and toxicity of these compounds to plants and microbes [53]. Pesticides, one of the most important organic pollutants in agricultural soils due to their excessive usage and low efficiency [217], have raised great attention in soil remediation using hydrochars [199]. Hydrochars can decrease the mobility of pesticides such as aldrin, chlordane, dichlorodiphenyl trichloroethane (DDT), dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, and toxaphene in soils [199,217,223]. The potential remediation mechanisms of organic contaminants in soils following hydrochar application are jointly contributed by the adsorption of organic contaminants onto the hydrochars [104,217] and the enhanced biodegradation of the organic pollutants by microbes [223]. Hydrochars may increase soil microbial abundances and activities [175], and enhance plant growth and excretion of root exudates [142], thus resulting in the enhanced degradation of organic pollutants in soils [223]. However, little information is available on the chemical degradation of organic contaminants in soils via PFRs of hydrochars, which have been well-reported for biochars [21,219]. Hence, the degradation of organic contaminants in soils aided by PFRs of hydrochars should be considered in future studies. The rhizosphere plays a key role in plant growth and nutrients uptake. Thus, more studies are needed to focus on the fate of organic pollutants in the rhizosphere following hydrochar application. Overall, amending soils with hydrochars can increase the adsorption capacities of organic pollutants, thus decreasing their availabilities and toxicities. However, the fate of hydrochars in soil and water environments needs to be further studied to gain a full insight into the long-term consequences. Notably, hydrochars may also carry toxic compounds such as heavy metals, phenols, hydroxymethylfurfural, and furans [157]. Thus, understanding the environmental risks of hydrochars is also crucial to its successful application in soil and water remediation.

## 6. Current gaps and future perspectives

With the increasing studies on the production, characterization, and application of hydrochars in soil improvement and environmental remediation, there are still several gaps needed to be filled in the future. Several suggestions on the future development of hydrochar technology are proposed.

(1) *Production and characterization*: Although a wide variety of biomass (particular wet biowaste) with different chemical compositions are applicable in hydrochar production, there is inadequate information

with respect to the transformation of these biomass for hydrochar formation, elemental composition, surface structure and reactivity during HTC. Moreover, the blended feedstock for hydrochar production and characterization is still limited. Hence, more detailed comparative studies on hydrochars produced from different single or blended feedstocks should be conducted to establish the relationships between the physicochemical properties of hydrochar and their feedstocks and HTC conditions. Also, the formation mechanisms of hydrochars from different feedstocks under different HTC conditions using different additives still need to be illustrated.

(2) *Application in soil improvement:* Although hydrochars show promising potential in improving soil quality and productivity, different types of soils need to be used and studied. Besides the fertilization potential of hydrochars, more research is needed to obtain insights into the effects of hydrochar on more categories of soil properties, including soil structure, salinity, microbial community, and soil animals, particularly in the rhizosphere soils. Moreover, the benefits of hydrochars in mitigating GHG emissions and enhancing C sequestration should be fully assessed in future with long-term laboratory and field-scale investigations.

(3) *Application in water and soil remediation:* Although hydrochar showed great potentials in remediating organic and inorganic pollutants in soil and water environments, there is still a lack of knowledge regarding the effects of hydrochars on bioavailability and stabilization of contaminants in the environment over a long-term. Longer-term studies are necessary to examine the stability of contaminants adsorbed or immobilized by hydrochar in water and soil environments. More contaminants, particularly for emerging contaminants such as flame retardant, plasticizer, and pathogenic microorganisms, need to be considered in future studies. Moreover, to avoid or minimize the possible risks of hydrochar during water and soil remediation, hydrochars with minimal toxic components should be carefully selected. Notably, the potential environmental and ecological risks of hydrochar regarding contamination and adverse interaction with water and soil biota need to be carefully assessed before any large-scale application.

(4) *Industrialization and marketization:* Despite the rapid development and application, the use of hydrochar is still an emerging field. Most studies have been limited to the laboratory scale. There is still no industrial production or utilization unit now. Hence, more efforts are needed to examine the feasibility of hydrochar production and application at industrial scale and to develop commercial and large-scale HTC technology. Future research on reactor design, catalysts and process water recycling is recommended to overcome technological and economic constraints. The application of solar energy, continuous reactors, and deep learning techniques can be expected.

(5) *Environmental and ecological risks:* The potential environmental and ecological risks of hydrochars should be further assessed from a perspective of the whole life cycle of hydrochars, including feedstock collection and transport, production using HTC, post-treatment and transport, and application in soils and waters. For example, fine particles like nano hydrochars can be produced and easily released into surrounding environments during the production and application, but the potential risks of these fine particles are still not clear. Studies also need to develop effective technologies to treat or recycle the process water during HTC in order to decrease the potential environmental risks and costs of hydrochars. Moreover, more studies are merited to examine the environmental and ecological risks of hydrochars containing potential toxic components prior to the practical applications into soil and water ecosystems.

## 7. Conclusions

The development of sustainable thermal technologies for high-value utilization of biomass waste, particularly for the wet biowastes, are necessary to mitigate environmental challenges and sustain management of solid wastes in a circular economy approach. Hydrochars from

biomass using HTC are promising solutions for these issues. In this review, the current research progress of hydrochars were presented, and the feedstock, HTC technology, characteristics, application of hydrochars in soil improvement and environmental remediation were discussed in comparison with biochars, a type of char materials produced from pyrolysis of dry biomass. Hydrochar production from HTC is a promising way to manage dry and wet biowastes sustainably. Hydrochars can offer tremendous advantages to the agricultural and environmental fields, including soil improvement, crop productivity enhancement, and environmental remediation. Hydrochars can be considered as a tool for improving soil health by directly providing essential nutrients and indirectly improving soil physical and chemical properties and microbial community. Hydrochars also offer tremendous benefits for remediating polluted water and soil via adsorption and degradation. Based on these, hydrochar technology has showed the promising prospects in application in soil and environmental sectors, and more studies are warranted in future to fill the gaps in the production and application of hydrochars.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgments

This work was supported by National Science Foundation (CBET 1739884), U.S. Department of Agriculture Hatch Program (MAS 00549), and Shandong Province Natural Science Foundation (ZR2019MD017, ZR202102280041).

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2021.133142>.

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