



Recycling food waste to agriculture through hydrothermal carbonization sustains food-energy-water nexus

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ABSTRACT

Recycling underutilized resources from food waste (FW) to agriculture through hydrothermal carbonization (HTC) has been proposed to promote a circular economy (CE) in food-energy-water (FEW) nexus. However, most HTC studies on FW were conducted at laboratory scale, and little is known on the efficacy and feasibility of field application of HTC products from FW, i.e. the aqueous phase (AP) and solid hydrochar (HC), to support agriculture production. An integrated pilot-scale HTC system was established to investigate practical HTC reaction conditions treating FW. A peak temperature of 180 °C at a residence time of 60 min with 3 times AP recirculation were recommended as optimal HTC conditions to achieve efficient recovery of nutrients, and desirable AP and HC properties for agriculture application. Dilution of the raw AP and composting of the fresh HC are necessary as post-treatments to eliminate potential phytotoxicity. Applying properly diluted AP and the composted HC significantly improved plant growth and nutrient availability in both greenhouse and field trials, which were comparable to commercial chemical fertilizer and soil amendment. The HTC of FW followed with agricultural application of the products yielded net negative carbon emission of $-0.28 \text{ t CO}_2\text{e t}^{-1}$, which was much lower than the other alternatives of FW treatments. Economic profit could be potentially achieved by valorization of the AP as liquid fertilizer and HC as soil amendment. Our study provides solid evidences demonstrating the technical and economic feasibility of recycling FW to agriculture through HTC as a promising CE strategy to sustain the FEW nexus.

1. Introduction

Food-energy-water (FEW) nexus are interconnected and interdependent due to their fundamental roles in supporting human life, economic activities, and environmental sustainability [1]. The nexus aims to relieve conflicts and improve synergies among the three sectors, and can enhance the use efficiency of resources and energy [2]. Facing the unprecedented limitation of resource availability, the FEW has been widely recognized as the key to solve the crisis caused by population

growth, global climate change and environmental pollution [3].

A huge amount of food waste (FW) is generated in the food supply chain, with an estimated one third of food production, or 1.3 billion tons, globally [4]. The FW causes approximately \$100,000 billion in social, economic, and environmental consequences globally each year, with environmental costs of \$70,000 billion and social costs of \$90,000 billion [4]. This massive waste stream contains significant resources that could be recycled to support industrial and agricultural production. For instance, it is estimated that 3.58 million tons of nitrogen (N) and 0.54

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million tons of phosphorus (P) could be recovered from China's FW in 2022, which equates to approximately 10.43 % and 5.12 % of China's annual chemical fertilizer production for N and P, respectively [5].

The FW has high moisture and complicated composition, therefore, they are hard to be effectively treated and valorized with the methods commonly adapted for other municipal solid wastes [6]. Improper disposal of FW will cause the contamination to environment and the spread of infectious diseases. Landfilling of FW is limited by land use, long process time and emission of greenhouse gas (e.g. CO₂, CH₄). Incineration of the FW produces energy, but energy-intensive pre-drying is required due to its high moisture. In both cases, nutrient recovery is not realized [7]. Biological treatments, such as composting and anaerobic digestion, require low energy input and can generate value-added products like organic fertilizers [8,9]. However, the relatively long processing time (several weeks) limits their efficiency and capacity to handle the large amounts of FW generated daily [10]. Overall, facing the challenges during processing and potential pollutant emissions, the variety of products produced through the utilization of FW is limited [11].

In recent years, much attention has been paid to hydrothermal carbonization (HTC) treatment of FW due to its advantages in recycling the energy and resources contained in high moisture solid waste [12,13]. During the HTC process, wet feedstocks are heated to temperatures between 180 °C and 250 °C, while several thermochemical reactions occur, including hydrolysis, dehydration, decarboxylation, polymerization, condensation, and aromatization [14,15]. As a result, three products of value are generated: a solid phase called hydrochar (HC), an aqueous phase (AP), and a gaseous phase that is primarily composed of CO₂ [12]. The HC is a complicated mixture of various organic compounds with a "core-shell" chemical structure, where large part of recalcitrant aromatic compounds forms the core and a small part of labile oxygen-containing functional compounds forms the shell [16]. Besides, the "microsphere" physical structure is commonly developed on the HC surface. Compared with the biochar (BC) produced by pyrolysis, the HC has much lower specific surface area and porosity [17]. The AP is actually the processing water which contains considerable dissolved organic compounds and inorganic solutes. Because the HTC process is operated as a batch, closed loop system, the nutrients embedded within the feedstock is distributed primarily in the AP and HC products.

Recycling the HTC products from FW to improve soil fertility and support crop production has been proposed as a way to sustain the FEW nexus [18–21]. Application of woody and grassy biomass HC could facilitate the formation of more stable native soil organic carbon [22]. We have recently shown that the HC of FW could saturate hydraulic conductivity and water holding capacity of soil [23], thus HC can be applied as a soil amendment in improving soil aggregation [24]. The effect of applying HTC products on plant growth, however, has been controversial. Some studies have demonstrated that HC stimulates plant growth and increases crop yield [19,25], while phytotoxicity of HTC products to seed germination and plant growth has also been reported [26]. In a meta-analysis of the HC application effects on plant growth, Luutu et al. [27] found that on the average, HC negatively impacted seed germination and shoot biomass. These inconsistencies are likely attributed to the differences in feedstock composition, HTC reaction conditions, and pre- or post-treatments, as well as the application method and amount of products, soil types, and plant species.

The yield of AP converted by HTC is generally 7 ~ 8 times more than the solid HC in volume. Some attempts have been done to recover the dissolved nutrients in the AP, such as P through chemical precipitation [28], while the recovery processes require extra chemical materials, energy and equipment [29]. The valorization of the AP, especially in agriculture application, is still one of the main challenges limiting the wide extension of HTC treating FW [30]. Furthermore, it should be realized that the water contained in the AP or FW (~80 %) is also an important recyclable resource to be used in agriculture.

The circular economy (CE) concerns a closed-looped reutilization system for natural resources and focus on the strategy of reduce, reuse

and recycle concept [2]. The CE has received much attention in many waste treatments (e.g. pyrolysis, composting) because of the great potential in pollution reduction and resources recycling to support economic growth [31]. Therefore, this concept has potential for evolving CE management of HTC treating FW. However, the main focus of HTC valorization has revolved in the application of products as source of energy [32]. The lack of the studies from large scale HTC treatment with raw FW materials makes it hard to identify the practical HTC conditions for FW with the purpose to reuse the products in agriculture, and therefore the reliable evaluation of its economic feasibility is unexpected. Besides, the studies investigating the effects of HTC products on plant growth was limited within pot experiments [27], which make it difficult to extrapolate their results to crop production in the field.

The current study aims (1) to establish a pilot-scale integrated HTC module for HTC of FW; (2) to identify the practical HTC reaction parameters and physio-chemical properties of the HC and AP with the purpose to valorize these products in agriculture; (3) to verify the efficacy of applying the HC as a soil amendment and AP as a liquid fertilizer to support crop production with field experiments; (4) to evaluate the environmental impacts and economic feasibility of large scale HTC treatment of FW with agriculture application of the products in field for a sustainable CE in the FEW nexus.

2. Materials and methods

2.1. Raw food waste and the HTC feedstock

The raw FW was obtained from a campus canteen in Nanjing Agricultural University capable of serving 30,000 people daily (Fig. 1a). To ensure samples are representative of the temporal variation in FW supply, a total of twelve monthly samples of raw FW were collected from the canteen over four seasons in 2021 (winter, spring, summer, and fall). Bones and plastic bags were removed from the raw FW, after which the FW was crushed using a commercial crusher (YL9022, Yifa, China). Waste cooking oil was present in the raw FW (~10 vol%) and was extracted for oil recycling using a separator (HYX-400, Yinxu, China) without extra heating (Fig. 1b). The crushed raw FW after oil removal was used as the FW feedstock for HTC process (Fig. 1c). For composition analysis, samples of the FW feedstock were filtered to separate solid and liquid FW phases. Then, the liquid FW feedstock was centrifuged to remove suspended solids. The collected solid FW feedstock was dried in an oven at 105 °C for 24 h. Composition analysis of the dry solid FW feedstock was performed according to the Chinese National Food Safety Standards (GB 5009–2010). Elemental analysis of both solid and liquid FW feedstocks was conducted using an elemental analyzer after digestion by H₂SO₄-H₂O₂ (ICP-OES, PerkinElmer, USA).

2.2. HTC processing of the FW

The pilot-scale HTC system was a 50-L stainless steel batch reactor with a 9 kW electrical heater manufactured by Chaoyang Chemical Engineering Company (Shandong, China) (Fig. 1d). For each run, 15 kg of the FW feedstock (~80 % water content) was homogenized immediately after collection, using a mechanical grinder (2800 rpm, with a 5 mm grind size), and transferred into the reactor using a sludge pump (JTP-7000, Sunsun China, 50 W, head 4.4 m, flow rate 7000 L h⁻¹). Afterwards, an additional 15 kg of process water (tap water or AP) was added to the reactor to achieve a 1:1 ratio between wet feedstock to water to ensure sufficient HTC reactions [33]. The reactor was sealed and heated by an electric heater installed inside to reach peak temperature within 6 h controlled with a power regulator. The heating was stopped after the system temperature being kept at peak temperature for a residence time. Then, reactor was allowed to naturally cool down to 70 °C, which took approximately 6 h. Then, the reactor contents were extracted and separated into solid and aqueous phases using a gauze filter press (Fig. 1e). For each run, the electricity consumption was

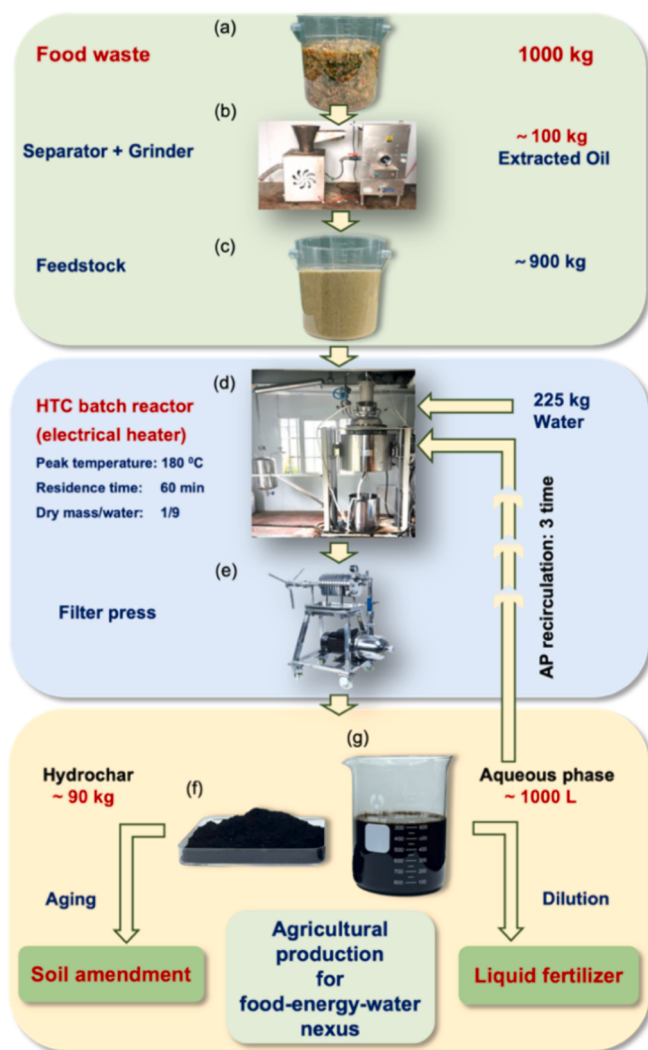


Fig. 1. Schematic flow of hydrothermal carbonization (HTC) processing 1 metric ton food waste (FW) in a pilot-scale production. (a) raw FW material; (b) pre-treatment system; (c) FW feedstock; (d) HTC reactor; (e) dewater system; (f) hydrochar; (g) aqueous phase (AP). The FW was collected from Nanjing Agricultural University across an entire year. The pre-treatment system was used to remove the bones and waste cooking oils. The peak temperature of HTC reaction was 180 °C and resident time was 60 min. The 15 L AP was recirculated as process water for 3 times. The calculation of water consumption is based on the following practices: for every 4 HTC runs, only 15 kg water was added to the system in the first run, thus, for 3 times of AP recirculation, the water consumption was $900/15/4 \times 15 = 225$ kg.

recorded by an electric meter.

Five combinations of peak temperature and residence time were set for the HTC reaction: (1) 180 °C, 30 min; (2) 180 °C, 60 min; (3) 180 °C, 90 min; (4) 200 °C, 30 min and (5) 220 °C, 30 min. According to [34], the HTC reaction severity, R , can be calculated as:

$$R = \log(t \times \exp((T - 100)/14.75)) \quad (1)$$

where T and t represents the peak temperature (°C) and residence time (min), respectively. These five HTC reaction conditions are referred as $R_{3.8}(180-30)$, $R_{4.1}(180-60)$, $R_{4.3}(180-90)$, $R_{4.4}(200-30)$ and $R_{5.0}(220-30)$ with the first number in the subscript indicating reaction severity and the two numbers in parenthesis indicating the peak temperature and residence time. The FW feedstock was carbonized at each reaction condition in triplicate.

According to previous studies [35–38], recirculating AP has been

utilized as a method to reduce water and energy requirements. Thus, AP recirculation was conducted during HTC processing, where 15 L of the AP after first HTC run was used in subsequent runs. This procedure was repeated for a total of 5 runs. After each run with AP recirculation, chemical properties, including electrical conductivity (EC), pH and concentration of nutrients, were measured for the AP and HC.

2.3. Measurements of the yield and properties of the HTC products

After solid–liquid separation, the weight of the AP, W_{AP} (kg), was measured, and the yield (wt. %) of the HC, $Yield_{HC}$, was calculated as:

$$Yield_{HC} = \frac{W_{HC,dry}}{W_{feedstock,dry}} \times 100\% \quad (2)$$

where $W_{HC,dry}$ and $W_{feedstock,dry}$ is the weight of the HC and FW feedstock after oven drying at 105 °C for 24 h.

The AP samples were filtered through a 0.45- μ m membrane for further analysis. The pH and EC were measured using portable probes (PB-10, Sartorius, Germany; DDS-11A, Yixin, China). Total organic carbon (TOC) was measured using a TOC analyzer (Multi N/C 3000, Jena, Germany). Concentrations of NH_4^+ and NO_3^- were measured using a continuous flow analyzer (San++, Skalar, Netherlands). Free amino acids (FAAs) were measured using an automatic amino acid analyzer (L-8900, Hitachi, Japan). Organic components in the AP were analyzed using gas chromatography–mass spectrometry (GC–MS) with an HP-5MS column (30 m $\times$ 250 μ m $\times$ 0.25 μ m) (7890A-5975C, Agilent, USA). The column temperature was initially held at 40 °C for 5 min, then programmed to 280 °C at a rate of 10 °C min^{−1} with a hold time of 5 min. Helium was used as the carrier gas with a flow of 1 ml min^{−1}. Electron impact ionization energy was 70 eV, and mass spectra were scanned from 20 to 450 u. The injection sample volume was 1.0 μ L.

Dissolved organic matter (DOM) in the AP were determined using fluorescence spectrometer (Fluoromax-4, Jobin-Yvon, French) with excitation wavelengths of 220–600 nm in 5 nm intervals and emission wavelengths of 200–600 nm in 5 nm intervals. Blank full term excitation emission matrix (EEM) spectra of deionized water were tested before sample analysis, and the scans were repeated 3 times. Rayleigh and Raman scattering were removed using Matlab (MATLAB R2020b, Toolbox). The region integration method was then used to calculate the biological index (BIX) to the qualify the biodegradability of the DOM [39].

The fatty acids were analysed using GC–MS (Trace 1310-ISQ 7000, Thermo, USA). In addition, several plant biostimulants in the AP, such as Zeatin, Brassinolide, and Methyl Jasmonate, were detected using a liquid chromatography–mass spectrometry (LC–MS) method [40,41].

The solid HC samples were ground using an agate mortar to pass through a 2 mm sieve for laboratory analysis. Total carbon content of the HC was determined using an elemental analyzer (multi EA 5000, Elementar, Germany). The pH and EC were measured using 1:5 (w/w) suspension of the HC in deionized water. Microstructure of the HC particles was visualized using a scanning electron microscope (SEM-JSM IT100, JEOL, Japan). Volatile compounds of the HC were identified using pyrolysis and gas chromatography coupled with mass spectrometry (Py-GC/MS) following Olszewski et al. [42].

The HC and AP samples were digested with H_2SO_4 – H_2O_2 for the measurements of nitrogen (N), phosphorus (P), potassium (K), calcium (Ca), magnesium (Mg), and sodium (Na) using an inductively coupled plasma–optical emission spectrometer (ICP–OES, PerkinElmer, USA). The available N (AN) of HC was extracted by alkaline hydrolysis diffusion method and quantified by 0.005 mol L^{−1} H_2SO_4 [43]. The available P (AP) of HC was extracted by 0.5 mol L^{−1} $NaHCO_3$ at a ratio of 1:20 (w/v), followed determination by molybdenum antimony anti-color methods [44]. The available K (AK) of HC was extracted by ammonium acetate at a ratio of 1:10 (w/v) followed determination by plasma emission spectrometry (ICP–OES, PerkinElmer, USA).

2.4. Phytotoxicity test and pot experiment of the AP and HC

Phytotoxicity of the AP was tested with seed germination of barley and rapeseed according to Kong et al. [45]. Preliminary tests showed that the full-strength AP was extremely detrimental to seed germination with 0 % germination rate and thus the AP must be diluted for agriculture use. For each test, 20 seeds were uniformly distributed on a 9 cm filter paper in a petri dish. The seeds were moistened with 10 ml of deionized water as a control (CK) and AP diluents with AP to water ratio (w/w) of 1: 50 (AP₅₀), 1: 150 (AP₁₅₀), and 1: 250 (AP₂₅₀), respectively. All treatments were kept in a dark room controlled at 27 °C in triplicates. The seed germination rate were recorded after 3 days and root length of 10 randomly selected seedlings were measured after 5 days. Relative germination rate (RGR, %), relative root length (RRL, %) and relative germination index (RGI, %) were calculated as:

$$RGR = \frac{NGS_{AP}}{NGS_{CK}} \times 100\% \quad (3)$$

$$RRL = \frac{MRL_{AP}}{MRL_{CK}} \times 100\% \quad (4)$$

$$RGI = \frac{RGR \times RRL}{100} \quad (5)$$

where, NGS_{AP} and NGS_{CK} are the number of germinated seeds treated with AP diluents and water, respectively; MRL_{AP} and MRL_{CK} are the mean root length in the treatments of AP diluents and CK, respectively. The hydrolase activities (protease and amylase) of barley seeds were further measured by testing kit from Cominbio Technology Co. LTD, Suzhou, China (<https://www.cominbio.com>). Phytotoxicity of the AP was further tested with pot experiment using cucumber seedlings irrigated with AP diluents. Cucumber seedlings with similar sizes were transplanted into pots (18 cm in diameter and 15 cm in height) filled with commercial coconut husk as growing substrate. The seedlings were irrigated twice a week with 250 ml each time of deionized water (CK) or AP₅₀, AP₁₅₀, and AP₂₅₀, respectively. Each treatment had 3 replicates. After 4 weeks, the fresh biomass of the shoots and roots were measured [46].

The plant growth experiments of the pot filled with soil were conducted using topsoil (0–20 cm) collected from Baguazhou, an island located in the middle-low region of Yangtze River in Nanjing, China. The soil was sandy loam in texture [23]. Soil sample was air-dried, and ground to pass a 1 mm sieve for uniform packing. Each pot (18 cm in diameter and 15 cm in height) was packed with 3 kg soil to reach a bulk density (BD) of 1.3 g cm⁻³. Lettuce seedlings (*Lactuca sativa* L. var. *ramosa* Hort.) with similar sizes were transplanted into the pots. To evaluate the nutritional and phytotoxic effects of the AP, three different liquids were used: tap water as a control (CK), 1/2 Hoagland solution (1/2H), and 150 times diluent of the raw AP (AP₁₅₀). The formulation of Hoagland solution is generally recognized as ideal solution to meet the nutrient demand of common plants and half of its concentration is adequate to supply growth at seedling stage [47]. Each liquid was applied twice a week with 250 ml each time for 4 weeks.

The effects of HC as a soil amendment on soil nutrients and plant growth were investigated in the greenhouse with pot experiments using the same sandy loam soil. To eliminate the potential phytotoxicity of HC, the fresh HC (F_{HC}) was composted with rice straw (10:1, w%) for 30 days to obtain aged HC (A_{HC}). The aging processes were performed in a 12 L fermenter. The hydrochar was built into piles and flipped every three days. When the temperature of the pile stopped the change for five consecutive days, the HC was recovered after air drying.

Each pot (18 cm in diameter and 15 cm in height) was packed with the 3 kg soil (CK) and 1 w% of the F_{HC} and A_{HC}, respectively, to reach a BD of 1.3 g cm⁻³. Besides, equivalent base fertilizer (N-P₂O₅-K₂O, 12-12-20 mg) was homogeneously mixed with the soil for each pot. Tap water was applied twice a week with 250 ml each time for 4 weeks.

The lettuce plants were harvested after 30 days of growth with AP and HC applications. The fresh biomass weight of root and shoot were measured, respectively. Soil samples were taken from the pots, air dried and ground to pass 1 mm sieve for lab measurements. Soil total N (TN) were determined by H₂SO₄ digestion-Kjeldahl method. Soil available N (SAN) was extracted by alkaline hydrolysis diffusion method and quantified by 0.005 mol L⁻¹ H₂SO₄ [43]. Soil available P (SAP) was extracted by 0.5 mol L⁻¹ NaHCO₃ at a ratio of 1:20 (w/v), followed determination by molybdenum antimony anti-color methods [44]. Soil available K (SAK) was extracted by ammonium acetate at a ratio of 1:10 (w/v) followed determination by plasma emission spectrometry (ICP-OES, PerkinElmer, USA).

2.5. Field application of the AP and HC

Field experiment was conducted at Huaian Experimental Station (33°22'N and 118°56'E), Jiangsu Province from June 2023 to October 2023. The mean annual temperature and rainfall are 14.5 °C and 900 mm, respectively. The soil in the 0–20 cm soil layer is silt loam with organic carbon of 8.7 g kg⁻¹, total N 0.51 g kg⁻¹, total P 0.64 g kg⁻¹ and total potassium 8.1 g kg⁻¹.

The field experiment was divided into two parts, including applying AP as liquid fertilizer and HC as soil amendment, respectively. The seeds of maize (cv. Jiangyu 877) were sowed with a 40 cm row spacing and a 40 cm plant spacing on 25th June. For the AP application experiment, drip lines were installed with a space of 40 cm in row accompanying with the plant rows. Then, the drip irrigation was divided into three 240 m² plots (6 m × 40 m) with 1500 plants in each plot. Before sowing, the compound fertilizer was applied at the rate of 140 kg N ha⁻¹; 84 P₂O₅ kg ha⁻¹ and 73.5 K₂O kg ha⁻¹ as basal fertilization for all plots, which was the common practices of local area for maize production. Topdressing fertilizer was applied at the rate of 60 kg N ha⁻¹; 36 kg P₂O₅ ha⁻¹ and 31.5 kg K₂O ha⁻¹ to two of the blocks using the aqueous phase (AP) and soluble chemical fertilizer (CF, N-P-K, 16-16-14), respectively. The topdressing was applied 4 times with 2500 L liquid fertilizer each time at jointing, flared, heading and milk stage, respectively. The other block irrigated with same amount of water was marked as control-1 (CK-1). Details of the topdressing timing and amount of liquid fertilizer applied at these four growing stages are provided in Table S1.

The HC application experiment included three treatments: HC, the aged HC applied at 0.5 wt% mixed in 0–20 cm soil layer before sowing; BC, applied at 0.5 wt% mixed in 0–20 cm soil layer before sowing; and the control-2 (CK-2) without any soil amendment. The BC was derived from wheat straw and its properties can be found in Ji et al. [48]. Each treatment was performed in a 48 m² plot (6 m × 8 m) with 300 plants. Basal fertilizer was applied in the same way as the AP application experiment.

After harvest on 15th October, undisturbed and disturbed soil samples were randomly taken from the 0–20 cm topsoil at eight sites across each plot. Soil bulk density (BD, g cm⁻³) was determined after oven-drying at 105 °C for 24 h. The distribution of soil water-stable aggregates (%), i.e. >0.5 mm, 0.5–0.1 mm, and <0.1 mm, was measured with wet-sieving procedures [49]. Leaf area index at different growing stages was calculated by measuring each leaf area and number of leaves and the planting density [50]. The maize grain yields in each plot of the treatment were measured after harvesting, threshing, and air-drying of the grains. Substance composition of the grain was measured by near-infrared spectroscopy (DA7250, Perten, USA). Soil organic carbon (SOC) was measured using the Walkley-Black method [51]. Available N, P and K (mg kg⁻¹) was measured for the soil and total content of N, P and K (mg g⁻¹) were measured for the maize plant.

2.6. Environmental and economic analysis

During the HTC processing of FW, the gaseous phases were collected to measure CO₂ emission by static chamber-gas chromatography

method under $R_{4.1(180-60)}$ [12]. During field application of the AP and HC, the greenhouse gas (GHG) emissions flux ($\text{mg m}^{-2} \text{hr}^{-1}$) of CO_2 , CH_4 and N_2O were measured at five growing stages of sowing, jointing, flared, heading and milk, respectively. Then, the cumulative GHG emission ($\text{kg CO}_2\text{e ha}^{-1}$) and soil carbon sequestration ($\text{kg CO}_2\text{e ha}^{-1}$) were calculated following Zhang et al. [52]. Net carbon emission ($\text{t CO}_2\text{e t}^{-1}$ wet feedstock) was calculated for the HTC process and other FW treatments, i.e. landfilling, incineration, aerobic composting and anaerobic digestion.

An economic analysis was conducted to assess the feasibility of HTC treatment of FW followed with the agriculture application of the products. The analysis accounted for the costs of construction and operation, and incomes of potential economic benefits of the extracted cooking oil, government subsidies and the sale of HC as soil amendment and AP as liquid fertilizer. The calculation was conducted under the circumstance of a 10-ton daily disposal capacity and a 10-year depreciation period of the HTC facility. The total investment to run the project was 1.82 million yuan, assuming 30 % of which was from self-raised fund and the other 70 % was loaned from bank with a loaning rate of 4.9 % (Bank of China, <https://www.Boc.cn>). The commodity price (yuan t^{-1}) of the extracted cooking oil from the raw FW was obtained from market survey during 2020–2022. The commodity prices of the AP (yuan L^{-1}) and HC (yuan t^{-1}) were referred to the average prices of the liquid fertilizers and organic soil amendments available from the Alibaba online market, respectively.

2.7. Statistical methods

Statistical analyses were performed using SPSS 16.0. One-way analysis of variance (ANOVA) was conducted to determine the significant differences ($p < 0.05$) among treatments with Duncan's multiple range test.

3. Results and discussion

3.1. Food waste properties and plant essential nutrients

The average yield of the raw FW from the canteen was 150 tons per month with little temporal variation during the year, except for winter (Jan to Feb) and summer (July to Aug) vacations (Fig.S1). The raw FW contained a variety of wasted cooked food (e.g. rice, vegetables, meat, and noodles) and condiments (e.g. soy and chili sauce) (Fig. 1a). As shown in Fig. 2a, main substance components of the FW feedstock were carbohydrates ($\sim 48.5\%$, i.e. starch, cellulose, hemi-cellulose, lignin and others), lipids ($\sim 26.9\%$), and proteins ($\sim 18.5\%$). Interestingly, the FW collected from various university canteens and restaurants across China contains the similar amount of carbohydrates ($52.8 \sim 69.5\%$) and proteins ($14.4 \sim 20\%$) [20], indicating that the FW used in this study represents a common diet food wastes in China and other countries with the similar diet habitats.

Notably, the content of lipids in the FW feedstock was much higher than those reported in other HTC studies using sewage sludge and plant residuals as feedstocks. The high content of lipids in the FW feedstock may affect HTC reaction and consequently make the products' properties different from those derived from other types of feedstocks [53]. Additionally, the temporal variation of each component over seasons was constrained within a narrow range (Fig. 2a), which indicates a relatively stable chemical composition of the FW feedstock and consequently consistent properties of the HTC products under certain reaction conditions.

The FW contains large amount of plant essential nutrients: 23.8 mg N g^{-1} , 3.7 mg P g^{-1} , 6.0 mg K g^{-1} , 5.3 mg Ca g^{-1} , 0.5 mg Mg g^{-1} in the solid FW feedstock (Fig. 2b), and 4480 mg L^{-1} (N), 1468 mg L^{-1} (P), 1306 mg L^{-1} (K), 3283 mg L^{-1} (Ca) and 198 mg L^{-1} (Mg) in the liquid FW feedstock (Fig. 2c). In addition, Na content in the FW feedstock was 19.4 mg g^{-1} in solid and 2940 mg L^{-1} in liquid, which were higher than

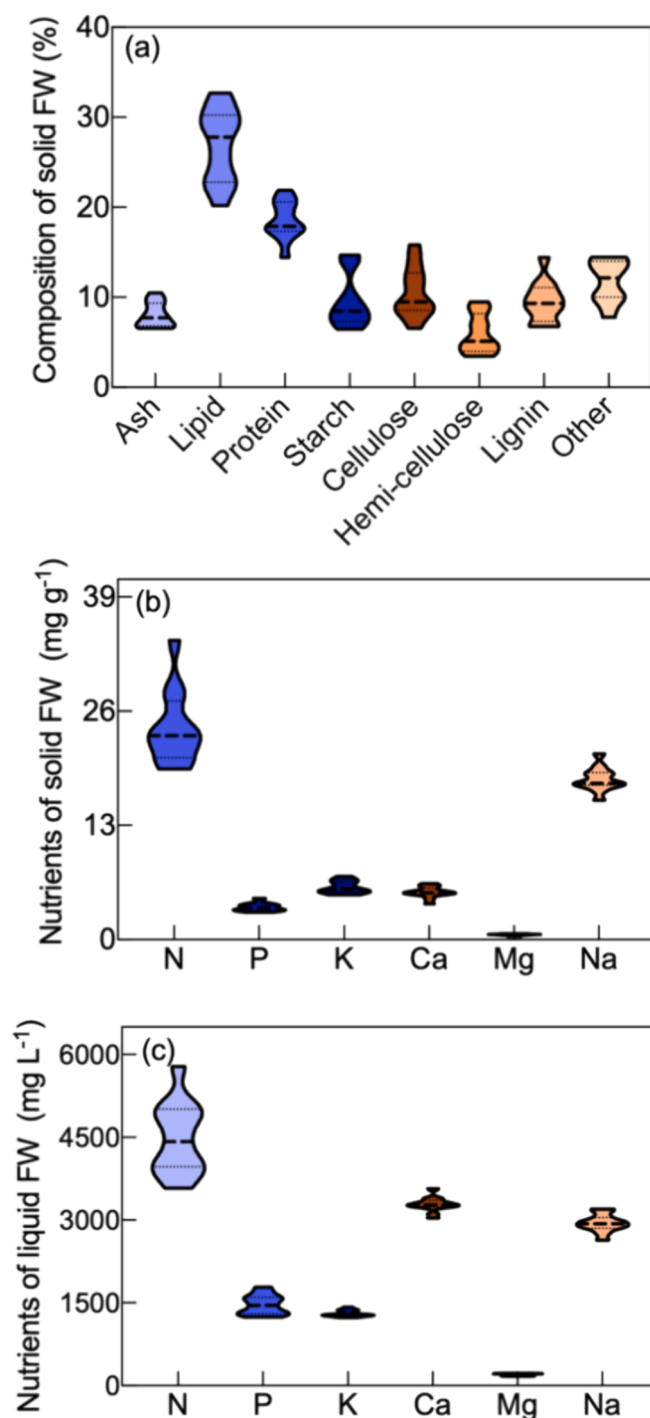


Fig. 2. Proximate composition and nutrients in the food waste (FW) feedstock for hydrothermal carbonization. (a) proximate composition of solid FW feedstock; (b) nutrients in solid FW feedstock; (c) nutrients in liquid FW feedstock. The FW samples were taken every quarter during 2021 from a campus canteen in Nanjing Agricultural University with a daily supply capacity for 30,000 students. Values denote mean $\pm$ standard error ($n = 12$).

other types of feedstocks reported for HTC treatment [54]. It should be attributed to the salt added during cooking. The heavy metals in the FW feedstock were $0.26 \text{ mg Cr kg}^{-1}$, $0.002 \text{ mg As kg}^{-1}$, $0.02 \text{ mg Cd kg}^{-1}$, $0.15 \text{ mg Hg kg}^{-1}$ and $0.05 \text{ mg Pb kg}^{-1}$, respectively (Table S2), which were much lower than the other high moisture feedstocks for HTC, such as sludge and livestock manure. Furthermore, the heavy metals in the FW feedstock were also much lower than the maximum threshold values

required for the organic fertilizers (NY/T 525-2021) set by the Ministry of Agriculture and Rural Affairs of the People's Republic of China. The results demonstrate that there would not be any risk of heavy metal contamination by recycling HTC products derived from FW in

agriculture.

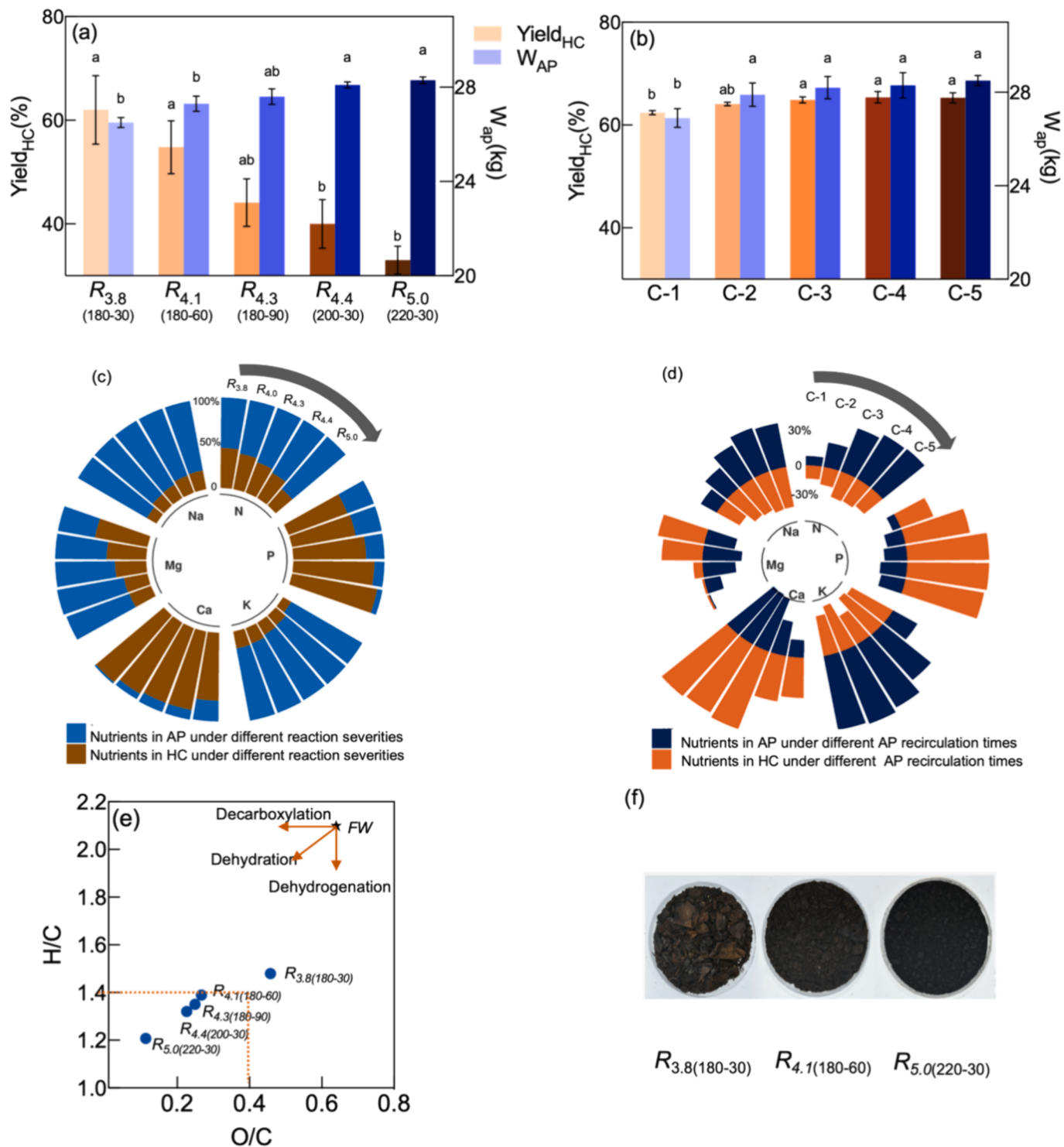


Fig. 3. Effects of hydrothermal carbonization (HTC) reaction severity (R) and aqueous phase (AP) recirculation times on the yield and nutrients fate of the aqueous phase and hydrochar (HC) derived from food waste (FW). (a) the yields of AP and HC under different HTC reaction severities. The peak temperature and residence time for the severity of $R_{3.8}$, $R_{4.1}$, $R_{4.3}$, $R_{4.4}$ and $R_{5.0}$ are indicated in X-axis; (b) the yields of AP and HC under different AP recirculation time. C-1, C-2, C-3, C-4 and C-5 represent 1 to 5 times of the aqueous phase recirculation; (c) the nutrients fate of AP and HC under different HTC reaction severity; (d) the variation of nutrients of AP and HC under different AP recirculation times; (e) the Van-Krevelen diagram of FW and HC under different HTC reaction severities; (f) phenotypes of HC under reaction severity of $R_{3.8}$, $R_{4.1}$ and $R_{5.0}$, respectively. The error bars represent standard error and different letters indicate statistically significant ($p < 0.05$) differences among treatments.

3.2. Effects of reaction severity and AP recirculation on the HTC products from FW

3.2.1. Effects on the yields of AP and HC

The yields of the AP and HC from the HTC of FW are largely affected by reaction severity (Fig. 3a). Increasing the reaction severity from $R_{3.8}$ to $R_{5.0}$ decreased the HC yield from 62 % to 33 % with the simultaneous increase of AP volume (mass). Reductions in HC yields at higher temperatures may be attributed to the enhanced hydrolysis of the feedstock, increased dissolution of soluble intermediates and compound volatilization. Similar results were reported in studies on the HTC products of other feedstocks in laboratory-scale experiments [55–58], suggesting carbonization with this pilot-scale system occurs as expected. The $R_{3.8}$ (180-30), $R_{4.1}$ (180-60) and $R_{4.3}$ (180-90) have statistically similar AP mass and HC yields, indicating that the increase in residence time has no significantly discernible impact at a 180 °C and suggesting the temperature ramp-up and cool-down periods may dominate the HTC reactions at these conditions. The $R_{4.3}$ (180-90), $R_{4.4}$ (200-30) and $R_{5.0}$ (220-30) show statistically similar AP masses and HC yields, but with $R_{4.4}$ (200-30) and $R_{5.0}$ (220-30) statistically different from $R_{3.8}$ (180-30) and $R_{4.1}$ (180-60). Increasing the reaction temperature from 180 °C to 200 °C with a 30 min residence time decreases HC yield and increases the AP mass, but differences between 200 °C to 220 °C are not statistically significant. These results suggest that operating at shorter reaction times and/or lower temperatures, both with long (6 hr) cool-down and ramp-up periods as operated in this study (Fig.S2), may be a viable approach to achieve desired HTC products while reducing overall system energy needs. Additionally, depending on the properties of the desired end-products, lower reaction temperatures should be used to generate a larger amount of HC and less AP. It was also found that under AP recirculation, the mass of AP remained stable, while the yield of HC increased from C-1 to C-3 (Fig. 3b), due to the strong adsorption capacity of HC and dissolved organic matter from AP underwent HTC to form additional solid products [59].

3.2.2. Effects on the pH and electrical conductivity of AP and HC

The HTC reaction severity affected several fundamental chemical properties of the AP and HC (Fig. S3). The APs and HCs from the HTC of FW were both acidic with pH values ranging from 3.8 to 5.2 and from 5.8 to 6.2, respectively. Similar results were reported by Wang et al. [60] who also used FW as the feedstock for HTC treatment. In contrast, the HCs derived from other types of feedstock, such as sewage sludge, was found weakly alkaline [39]. The acidity of the products from HTC of the FW should be attributed to the amino acids and fatty acids from the hydrolysis of proteins [60]. Reaction severity significantly affected the pH of the AP, with higher pH values under higher intensities of $R_{4.4}$ and $R_{5.0}$, while the acidity of the solid HC was hardly affected by reaction intensity. The EC value of both APs and HCs was not significantly affected by the reaction intensity. Apparently, the EC ranging from 4.1 to 4.8 ms cm⁻¹ of the raw APs was too high to be directly applied for irrigation or fertigation in agriculture [61]. Increasing HTC reaction severity could increase total organic carbon (TOC) of the raw APs, but not increased total carbon content of the HC. The results indicate that chemical composition of the HC is relatively stable under altered HTC conditions. The formation of more soluble organic matters under higher reaction intensity could explain the decrease in the yield of HC and increase in the TOC of the AP [62]. The pH of the AP remained within a narrow range from 4.02 to 4.18 and was not significantly impacted by AP recirculation (Table S3). This change may be attributed to the consistent content of acidic substances in the dynamic states under a given reaction severity [63]. The EC of the AP increased from 4.13 ms cm⁻¹ at C-1 to 4.62 ms cm⁻¹ at C-3 and then kept stable afterwards. The increased EC could be due to the accumulation of inorganic ions and incomplete ionization of organic acids.

3.2.3. Effects on nutrient fates of AP and HC

The HTC reaction severity affected nutrient fates in the AP and HC in different manners (Fig. 3c). Proportion of total N in the AP increased from 55 % to 67 % with elevating reaction severity. It was probably due to the enhanced cyclization and deamination of the proteins under higher HTC reaction severity, which leads to N dissolvment and formation of amino acids, ammonia and nitrogenous compounds [64]. In contrast, elevating the reaction severity reduced the proportions of total P, Ca and Mg in the AP, meanwhile increased their proportions in the HC, which likely due to their precipitation and absorption with other metal ions [29,65]. Because of high mobility and solubility, the fates of K and Na in both AP and HC were relative less affected by the HTC reaction intensity [18]. As expected, most of the Na was distributed to the AP (78–83 %), with a concentration ranging from 2.6 to 3.8 g L⁻¹ (113–165 mM). Since Na is a beneficial nutrient and supply of several mM Na in nutrient solution can promote plant growth [66], properly dilution of the AP above 50 times (2.2–3.2 mM) should not result in harmful effect of Na to plants. In addition, Na concentration in the HC ranged from 5.9 to 10.6 mg g⁻¹, which is much lower than the threshold values of the organic fertilizer standard allowed for salts in China (NT/Y 4077–2022). The data indicate that field application of the HC at a reasonable amount would not cause salinity damage to the soil.

The proportion of total N in the AP increased by 31 % and kept stable after 3 times of AP recirculation. Meanwhile, the proportions of nutrient elements, especially P and Ca, in the AP and HC was remarkably affected by AP recirculation times (Fig. 3d). Mau et al. [67] utilized poultry litter as the feedstock for HTC and also found that 3 times of AP recirculation could reach the highest content of nutrients in the AP. Thus, with the purpose to achieve efficient recovery of nutrients in the AP and HC, 3 times of AP recirculation was recommended as an applicable and effective produce in the HTC of FW.

3.2.4. Determination of practical HTC reaction conditions for FW

The Van-Krevelen diagram in Fig. 3e shows the coalification degree of the FW feedstock under different HTC reaction severity. The HC under higher reaction severities had lower H/C and O/C atomic ratios. It should be attributed to the enhanced dehydration and decarboxylation of the feedstock with the increased reaction severity during HTC. As shown in Fig. 3f, the FW feedstock was not fully carbonized under $R_{3.8}$ (180-30) as compared with those under $R_{4.1}$ (180-60) and $R_{5.0}$ (220-30). The characteristic H/C atomic ratio (to 0.4) and O/C atomic ratio (to 1.4) of lignite (Fig. 3e) are commonly used as a reference to identify HC [54]. Therefore, the solid product under $R_{3.8}$ (180-30) can not be classified as HC (Fig. 3f), while the solid products yielded under $R_{4.1}$ (180-60) with the same temperature of 180 °C but longer residence time (60 min) could meet the standard for HTC of FW.

Ultimately, selection of reaction conditions for the HTC of FW depends on system energy balances and desired product properties. It is expected that lower temperatures would be desired to decrease operational costs of the process. As described earlier, operating at shorter reaction times with the longer cool-down and ramp-up periods may be advantageous from an energy perspective.

Based on these results above, the reaction severity, $R_{4.1}$, with a peak temperature of 180 °C and a residence time of 60 min, and 3 times AP recirculation was recommended as the practical reaction conditions for large-scale HTC treatment of FW. Whereby, the HC and AP produced under this practical HTC conditions were used in the subsequent experiments to evaluate their effects in agriculture application.

3.3. Characteristics of the AP and HC from FW for agriculture application

3.3.1. Characteristics of the AP

The AP solution contains large quantity of N in the ratio of inorganic form (~20 %) and organic form (~80 %) (Fig. 4a). For inorganic N, the $\text{NH}_4^+\text{-N}$ was the dominate component (~95 %). The sum of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ ranged from 989 to 1213 mg L⁻¹ and accounted for ~30 % to the

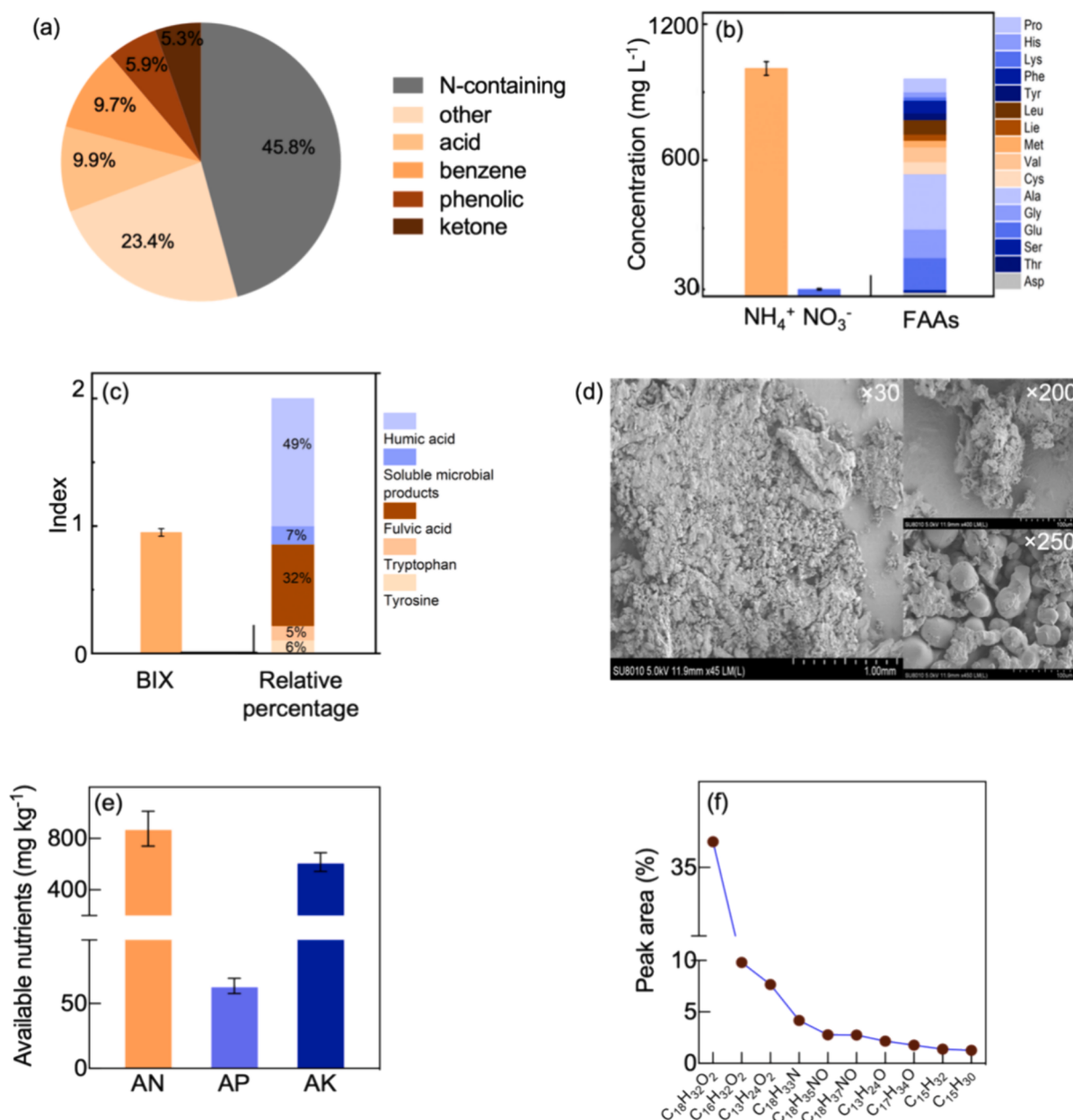


Fig. 4. The physio-chemical characteristics of the aqueous phase (AP) and hydrochar (HC) derived from food waste by hydrothermal carbonization. (a) proportions of organic components in the AP; (b) concentrations of soluble NH₄⁺-N and NO₃⁻-N and free amino acids (FAAs) in the AP; (c) biogenic index (BIX) and relative percentage of each region of dissolved organic matter components in the AP; (d) scanning electron microscopy (SEM) images of the HC at ×30, ×200 and ×250 magnification, respectively; (e) available nutrients of the HC. AN, AP, AK: available N, available P, available K, respectively; (f) the top 10 substances from Py-GC/MS analysis of HC.

total N in the AP (Fig. 4b). In addition, other essential major nutrients are also highly abundant in the solution of AP, including 288 mg P L⁻¹, 1351 mg K L⁻¹, 430 mg Ca L⁻¹, 150 mg Mg L⁻¹.

A total of 16 different free amino acids were detected in the AP, among which alanine (Ala), glycine (Gly), and glutamic acid (Glu) were the three leading components (Fig. 4b). These three amino acids are major components involved in plant root acquisition of organic N, N allocation and assimilation [68]. Plenty of studies have proved that amino acids can be directly acquired by plants through root zone application or leaf spray and function in improving plant growth [69,70], therefore, the promoting effects of AP solution to plant growth may be partially resulted from its FAAs.

The AP contained multiple soluble organic compounds (Fig. 4a). The N-containing compounds, including pyridine, pyrrole, and pyrazine,

accounted for 45.8 % of the total organic matter, while organic acids, benzenes, phenols, and ketones accounted for 30 % of the total organic matter in the AP. In the studies using other feedstocks, the phenols, especially the guaiacol and catechol, were identified as phytotoxic substances contained in the HTC products leading to inhibition of seed germination and negative impacts on plant growth [71,72]. However, these phytotoxic substances are water soluble and prone to chemical and biological decomposition [73]. Thus, it is reasonable to expect that suitable post-treatments of the HTC products, such as washing/composting of the HC and dilution of the AP, could efficiently eliminate the potential detrimental effects caused by phytotoxicity in agriculture application [19].

Various fatty acids were found in the AP, where 10-Transpentadecenoate (8.85 mg L⁻¹), Myristoleate (5.05 mg L⁻¹) and Pentadecano (4.19

mg L⁻¹) had the highest concentration. (Table S4). The dominant DOM in the AP were humic acid-like substances (~49 %) and fulvic acid-like substances (~32 %) (Fig. 4c). The BIX was close to 1.0, indicating high biodegradability of the DOM in the AP. In addition, three types of phytohormones, i.e. zeatin-riboside (ZR), methyl jasmonate (MeJA) and aminocyclopropane carboxylic acid (ACC), were detected in the AP with concentrations ranging from 0.25 to 0.30 ng mL⁻¹ (Table S5). These substances may positively affect plant growth by promoting a series of physiological and biochemical processes [74–76].

3.3.2. Characteristics of the HC

Cracks and microsphere structures were observed on the HC surface (Fig. 4d). The specific surface area (SSA) of the HC was 1.983 m² g⁻¹, and the pore volume was 0.033 cm³ g⁻¹ with pore size of 6.62 nm, indicating the pores were mainly mesopores. Compared with the biochar produced by pyrolysis, we found that the HC derived from the FW had a much lower SSA due to the lower reaction severity and weaker pore development during HTC [23]. However, the HC derived from FW generally has rich functional groups that can enhance soil fertility by increasing soil cation exchange capacity [77]. Considerable plant

essential nutrients were reserved in the HC, including 822 mg N kg⁻¹, 66 mg P kg⁻¹ and 616 mg K kg⁻¹, respectively (Fig. 4e).

The HC contained unsaturated fatty acids, including linoleic acid (~36.6 %), palmitic acid (~9.8 %) and tridecanedioic acid (~7.6 %) (Fig. 4f). These compounds could be steamed from oxidative cleavage of short aldehyde like hexanal or octanal and lipids [42], and potentially cause phytotoxicity of HC [53].

3.4. Evaluation of the HTC products from FW in agriculture application

3.4.1. Pre-field evaluation the AP and HC on plant growth

Compared with the treatment of tap water as blank control (CK), the AP₅₀ diluent significantly decreased the seed germination rate and root length of the rapeseed and barely (Table S6). Besides, the AP₅₀ inhibited the hydrolase activities of protease and amylase of the seeds (Fig. 5b), which could be attributed to the high concentrations of salts (EC 0.2 ms cm⁻¹), organic acids (pH 6.1) and NH₄⁺-N (20 mg L⁻¹) in the AP₅₀. On the contrary, the AP₁₅₀ significantly increased the seed germination rate and root length increased by 21 %~28 % and 7 %~18 %, respectively (Table S6). Similar promoting effects of the AP on seedling and plant

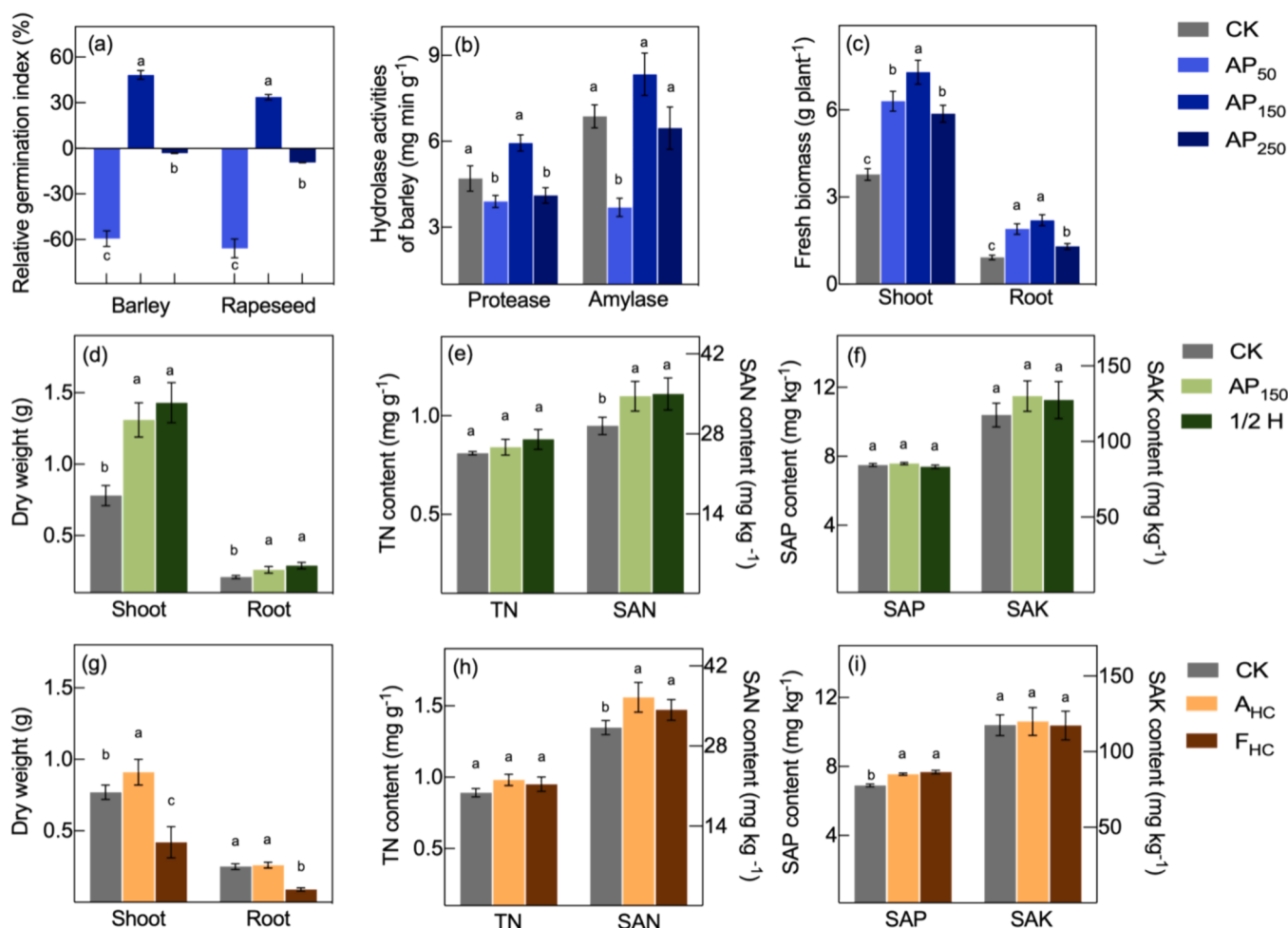


Fig. 5. Effects of diluted aqueous phase (AP) and post-treated hydrochar (HC) derived from hydrothermal carbonization of food waste on seed germination, plant growth and soil nutrients. (a) germination rate of barley seeds and rapeseed; (b) hydrolase activities of barley; (c) fresh biomass of shoot and root of cucumber; (d) dry biomass of shoot and root of lettuce under different AP treatments; (e) soil total nitrogen (TN) and soil available nitrogen (SAN) under different AP treatments; (f) soil available phosphorus (SAP) and soil available potassium (SAK) under different AP treatments; (g) dry biomass of shoot and root of lettuce under different HC treatments; (h) soil total nitrogen (TN) and soil available nitrogen (SAN) under different HC treatments; (i) soil available phosphorus (SAP) and soil available potassium (SAK) under different HC treatments. CK was tap water as blank control; AP₅₀, AP₁₅₀ and AP₂₅₀ denotes the AP diluted by deionized water with 50, 150 and 250 times, respectively; 1/2H was half intensity of standard Hoagland solution containing sufficient and balance essential nutrients; F_{HC} was the fresh produced HC without any post-treatment; A_{HC} was the aged HC by composting with rice straw (10:1, w%) for 30 days. The error bars represent standard error and different letters indicate statistically significant ($p < 0.05$) differences among treatments.

growth were reported by Gao et al. [78]. No significant difference was found for seed germination rate and root length between the CK and AP₂₅₀ (Table S6). The results indicate that the proper dilution of the AP from FW at about 150 times could not only eliminate its potential phytotoxicity but also significantly promote plant growth.

Applying the AP diluents, even at the lowest dilution rate of 50 times, did not have negative effect on the root and shoot growth of cucumber in the pot experiment with growing media (Fig. 5c). Instead, shoot and root growth of cucumber were significantly stimulated at 4 weeks after the application of AP diluents. It implies that the seedlings are more tolerant to the potential phytotoxicity of the AP than the seeds. Thus, the application of the AP diluents as topdressing to crops in field would not have the risk of phytotoxicity. In addition, the buffering effect of the porous growth media may contribute to phytotoxicity elimination of the AP. Moreover, the aerobic condition in the growth media could promote the chemical and biological degradation of these liable substances responsible for phytotoxicity. These mechanisms eliminating phytotoxicity would be also valid in soils during field application of the AP. Notably, significantly higher fresh shoot biomass was found in all the AP diluent treatments compared with the CK. Fresh biomass of the shoot and root of cucumber were both significantly higher in the AP₁₅₀ than those in the AP₅₀ and AP₂₅₀. These results demonstrate that the nutrients in the AP could support plant growth when applied as liquid fertilizer.

Furthermore, in the pot experiment with soil, the lettuce plants that were fertigated with the AP₁₅₀ for 4 weeks showed similar growth with those supplied with 1/2H, and significantly higher shoot and root biomass than the CK (Fig. 5d). Similar positive effects on plant growth have also been reported for the AP from HTC of other feedstocks in pot experiments [67,73]. In addition to the nutritional functions, the positive promoting effects of the AP on plant growth may be also related to the potential functions of its bio-stimulants and amino acids (Fig. 4). With the purpose to reuse the AP as a liquid fertilizer in agriculture, a ~150 times dilution was recommended as a proper post-treatment for the raw AP produced from HTC of FW. The changes of soil nutrients influenced by the application of AP diluent are shown in Fig. 5e-f. There was no significant differences in soil total N, available P and K among the AP₁₅₀, 1/2H and the CK. This could probably due to the limited application amount of the liquid fertilizers and short growing period. In contrast, soil available N content was significantly increased by the application of AP₁₅₀ compared with the CK.

Biomass of the lettuce grown in sandy soil with different post-treated HCs at an application rate of 1 w% are shown in Fig. 5g. The soil applied with F_{HC} yielded significantly lower dry weight of the plant shoot and root than the CK. It indicates that direct application of fresh HC derived from FW would inhibit plant growth even at a relatively low application rate. The detrimental effects of fresh HCs produced from various feedstocks on plant growth have been commonly observed [16]. The potential phytotoxic substances in the HC derived from FW should be one important factor causing the inhibited plant growth in our study. Similarly, phytotoxicity has been widely reported for the fresh HCs derived from other feedstocks, including maize silage, sewage sludge, wheat straw, orange peel and beetroot chips [16]. Some intermediate products during HTC reaction, such as 5-hydroxymethylfurfural (5-HMF), guaicol, levulinic, glycolic acid, acetic acid, glycoaldehyde dimer and catechol, have been identified as the substances responsible for the phytotoxicity of the HC [79]. However, due to the high complexity and diversity of the intermediates in the HTC, it is still hard to clarify the substances and underlying mechanisms causing phytotoxicity of the HTC products. Additionally, application rate of the HC as soil amendment has been considered as a key factor affecting the HC's phytotoxicity [79]. Besides, the significantly decreased biomass (Fig. 5g) could be also related to the considerable amount of liable organic matter with high C/N ratio (~25) in the F_{HC} derived from FW (Table S7), which would enhance microbial N immobilization in the soil and thus decrease N availability to the plants [80].

In contrast to the F_{HC}, the application of A_{HC} significantly increased

the shoot biomass, while had no detrimental effect on the root biomass (Fig. 5g). Similar promoting effect on plant growth has been reported in other studies using HCs from different feedstocks [81]. Since there was no significant difference of the nutrient status between the soils applied with fresh HC and post-treated HCs (Fig. 5h-i), the promoting effects in the post-treated HCs could not be contributed by the increased available nutrients after HC application. Thus, on one hand, the promoting effects could be related to the phytohormones absorbed on the HC surface or released from decomposition of the liable organic components in the HC [22]. On the other hand, HC application could probably affect soil physical properties by improving water holding capacity, aeration conditions and mechanical characteristics to facilitate plant growth [23,82].

Our results from pre-field experiments (Fig. 5) demonstrate that the toxicity would not be a hindrance for agriculture application of the AP and HC from FW. A proper dilution of the AP and composting of the fresh HC can be considered as effective post-treatments prior to their field application.

3.4.2. Field application of the AP and HC from FW

To testify if the HTC generated AP and HC products of FW can be effectively and safely used as liquid fertilizer and soil amendment to promote crop production in farmland, we produced large amount of AP and HC by our pilot-scale facilitator of HTC for being applied during the entire growth season of maize in field. The maize growth at harvest period in the field fertigated with the AP from FW and soluble CF is shown in Fig. 6. Although all the testing plots were supplied with high amount of compound fertilizer as basal fertilization and several times of heavy rains resulted in partial leaching of applied soluble AP and chemical fertilizers (CF) through topdressing fertigation, the growth of maize was promoted by AP and CF in comparison to the treatment of control (CK-1: irrigated with same amount of water) (Fig. 6a,b). The yields in the AP and CF treatments were significantly higher than that in the CK-1, while no significant difference was found between the AP and CF treatments (Fig. 6c). Similarly, contents of available nutrients in the soil, total nutrients in the shoot and the leaf area index were significantly higher in the AP and CF treatments than those in the CK-1, while no significant difference was found between the AP and CF treatments (Fig. 6d,e,f). These results indicate that the effects of AP on plant growth of maize was equivalent to the pure nutritional functions of the CF. Besides, the protein content in grain was significantly higher in the AP and CF treatments than that in the CK-1. Notably, the lipid content in the grains was the highest in the AP treatment (Fig. 6g), which could be attributed to the contribution of slow-released N from DOM in the AP (Fig. 4c) to promote the synthesis of fatty acids and amino acids during grain-filling stage [83].

In comparison to the control treatment without any soil amendment (CK-2), application of composted HC and BC also improved the maize growth and grain yield (Fig. 7a,b,c). The applications of HC and BC increased the yield of maize by 12.5 % and 13.4 %, respectively, compared with the CK-2 (Fig. 7c). Notably, both the soil available N (SAN) and K (SAK) were significantly higher in the HC and BC treatments than those the CK-2, while the soil available P (SAP) was significantly higher in the HC treatment than those in the BC treatment and CK-2 (Fig. 7d). The higher soil nutrient contents could be on one hand attributed to the nutrients contained in the HC and BC, while on the other hand, to the reduced leaching due to the high absorption ability of the HC and BC particles [16]. Additionally, the total N and P contents in the shoot were significantly higher in the HC and BC treatments than those in the CK-2, while no significant difference was found between the HC and BC treatments (Fig. 7e).

Since the density of both HC and BC particles is much lower than that of soil particles, application of the HC and BC decreased soil bulk density (Fig. 7f) and promoted soil aggregation to form more >0.5 mm macroaggregates (Fig. 7g). The improvement of soil structure by the HC application would further lead to enhanced soil ability of water

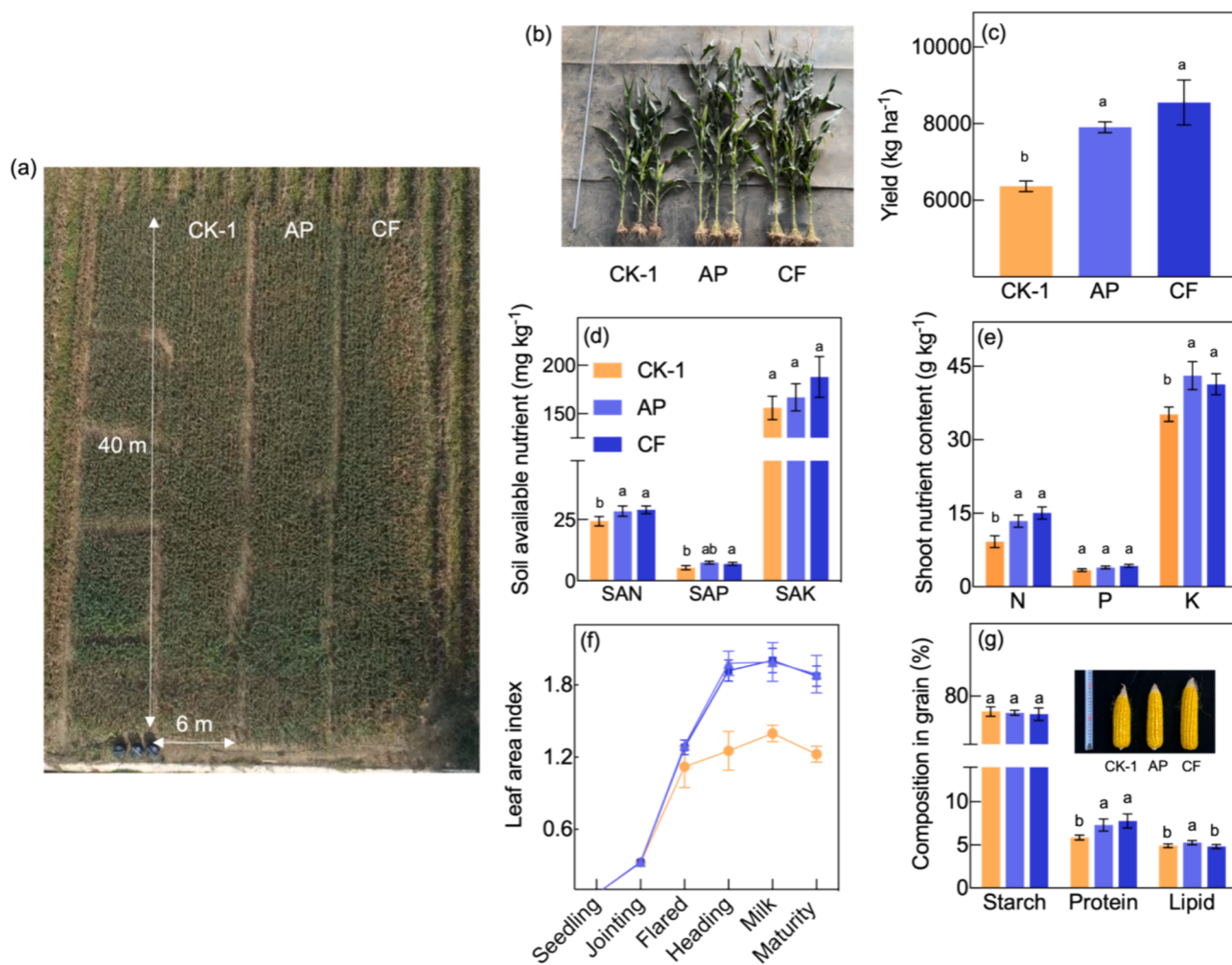


Fig. 6. Effects of applying aqueous phase (AP) derived from hydrothermal carbonization of food waste on maize growth and soil nutrients in the field application. (a) aerial photograph of the field experiment growing with maize; (b) phenotype of the maize; (c) the yield of maize; (d) soil available nutrients; SAN, soil available nitrogen; SAP, soil available phosphorus; SAK, soil available potassium; (e) the N, P and K in the shoot; (f) the leaf area index of maize; (g) the proximate composition of maize grain, under different liquid fertilizer treatments in the field. CK-1 was the control without topdressing liquid fertilizer; AP and CF denote the aqueous phase from hydrothermal carbonization (HTC) of food waste and compound chemical fertilizer supplied as topdressing liquid fertilizer, respectively. The error bars represent standard error and different letters indicate statistically significant ($p < 0.05$) differences among treatments.

retention, aeration and nutrient supply, and decreased resistance against root penetration [82,84].

The results from our field experiment demonstrate the feasibility of recycling the HTC products from FW to support agriculture production, and potential valorization of the AP as liquid fertilizer and the HC as a soil amendment.

3.5. Environmental impact and economic feasibility of recycling FW to agriculture through HTC

3.5.1. Environmental implication in carbon neutrality

The equivalent carbon emissions of different sectors along the HTC processing of FW and agriculture application of its products are shown in Fig. 8a. The actual carbon emission from the HTC reaction was 0.01 t CO₂e t⁻¹ based on the measured CO₂ concentration in our pilot-scale study. The equivalent carbon emission of the electricity consumption during HTC processing was 0.34 t CO₂e t⁻¹. The other three sectors, i.e. fertilizer replacement, GHG emission reduction and soil carbon sequestration were related to the agriculture application of the HTC products, and defined as carbon sinks with equivalent carbon emission of -0.06 CO₂e t⁻¹, -0.28 CO₂e t⁻¹ and -0.54 CO₂e t⁻¹, respectively. The detailed measurement and calculation of GHG emission during the

growth season of maize can be found in [Supplementary Materials](#). Fig. 8b outlines the net carbon emission of different scenarios of FW treatments. The HTC and anaerobic digestion yielded negative net carbon emission of -0.28 t CO₂e t⁻¹ and -0.49 t CO₂e t⁻¹, respectively, which were much lower than those yielded by aerobic composting (-0.02 t CO₂e t⁻¹), incineration (0.58 t CO₂e t⁻¹) and landfilling (0.78 t CO₂e t⁻¹) [85–87]. These data implicate that large-scale HTC treatment of FW followed with agricultural application of its products will sever as an environmentally friendly route to achieve carbon neutrality [88].

3.5.2. Economical feasibility

Based on the practices of our pilot scale operation and estimation of the revenue and expenditure currently in China, assuming to run a 10 t day⁻¹ HTC project treating FW, an initial capital investment of 2.13 million yuan will be required for the equipment and construction ([Supplementary Material](#)), and another 2.6 million yuan should be cost for annual operation, including feedstock collection (0.32 million yuan), energy consumption (0.98 million yuan), labor (0.45 million yuan) and others (0.75 million yuan) (Fig. 8c). The energy consumption is the largest cost and is always the major concern for the HTC treatment. However, it should be noted that the energy cost for treating the unit weight of FW feedstock would be reduced in large-scale HTC scenario

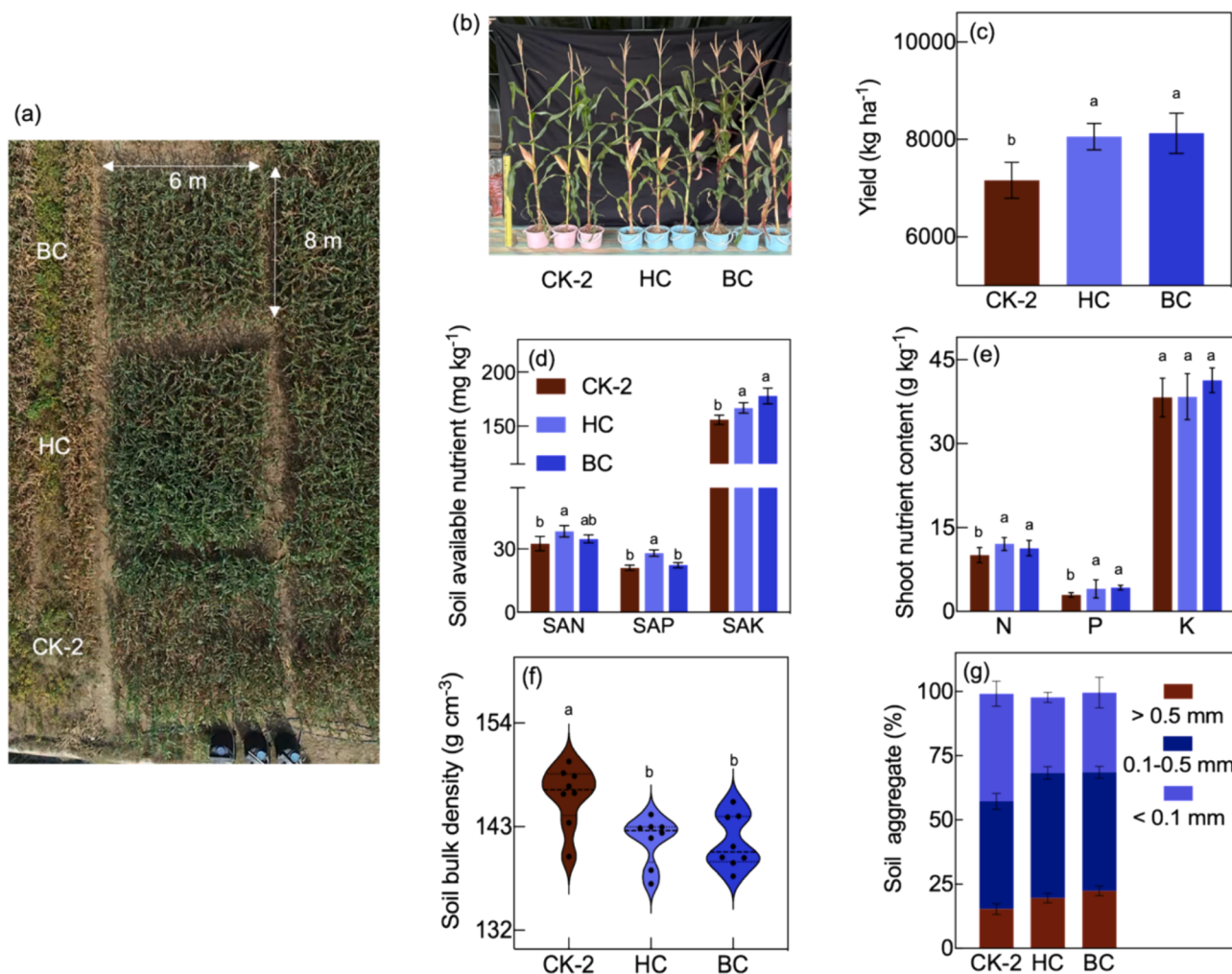


Fig. 7. Effects of the hydrochar (HC) derived from hydrothermal carbonization of food waste on maize growth and soil physio-chemical properties in the field application. (a) aerial photograph of the maize; (b) phenotype of the maize; (c) the yield of maize; (d) soil available nutrients; SAN, soil available nitrogen; SAP, soil available phosphorus; SAK, soil available potassium; (e) the N, P and K in the shoot; (f) soil bulk density; (g) soil aggregates distribution, under different soil amendment treatments in the field application. CK-2 represents the treatment without soil amendment as blank control; HC and BC denote the treatments of aged food waste derived HC and biochar mixed in the topsoil (0–20 cm) at 0.5 wt% before planting, respectively. The error bars represent standard error and different letters indicate statistically significant ($p < 0.05$) differences among treatments.

due to the less system heat loss [89]. Furthermore, the vigorous development of clean energy projects, such as solar power and wind energy [49], shall further reduce the cost for the heating, enhancing the feasibility of large-scale HTC treatment of FW and reaching the carbon neutrality in the future. The revenue of recycling FW to agriculture through HTC mainly come from commercialization of the AP as liquid fertilizer and the HC as soil amendment (Fig. 8c). The price of the AP liquid fertilizer is expected to be 0.7 yuan per liter referred to the price of commercial liquid fertilizer with similar major nutrient contents. The price of the HC is 1000 yuan per ton referred to the price of commercial BC in China (Table S8). Besides, the sale of waste cooking oil can achieve an income of 0.14 million yuan. There will be a government subsidy of 0.98 million yuan for treating FW in China. Overall, the annual revenues would sum up to 4.15 million yuan after tax deduction. Based on these data, the return period of the project is estimated to be 3.5 years, and will make a profit of 6.2 million yuan within ten years (Fig. 8d).

3.5.3. Circular economy for HTC of FW with agriculture application

Following the CE principle, the application of HTC of FW in agriculture presents several potential avenues. Firstly, HTC of FW provides a rapid treatment method without waste generation and emission. So, it will mitigate both the environmental burden of FW accumulation and

the release of greenhouse gases (GHGs) in society. Secondly, conversion of FW by HTC into AP and HC based fertilizer rich in N, P, K, and applying the HTC products to soils could replace the chemical fertilizer input and improve soil fertility [90]. Farmer will benefit because of pollution reduction and adverse effects of linear production processes, i. e. soil acidification and global warming. Furthermore, the design of economically efficient and eco-friendly processes requires joint efforts to expand the value in CE. By combining HTC with other waste treatment methods of FW such as anaerobic digestion, incineration, and wet oxidation, it can potentially offer benefits in terms of energy efficiency and sustainability [91].

4. Conclusion

In this study, an integrated pilot-scale HTC system was successfully operated to valorize FW to products for agriculture application. A peak temperature of 180°C at a residence time of 60 min, with 3 times of AP recirculation, were identified as the practical conditions to achieve desirable AP and HC properties. Post-treatment measures, such as dilution of the raw AP and composting of the fresh HC, effectively eliminated potential phytotoxicity, the detrimental effects on seed germination and plant growth. Properly diluted AP used as a liquid

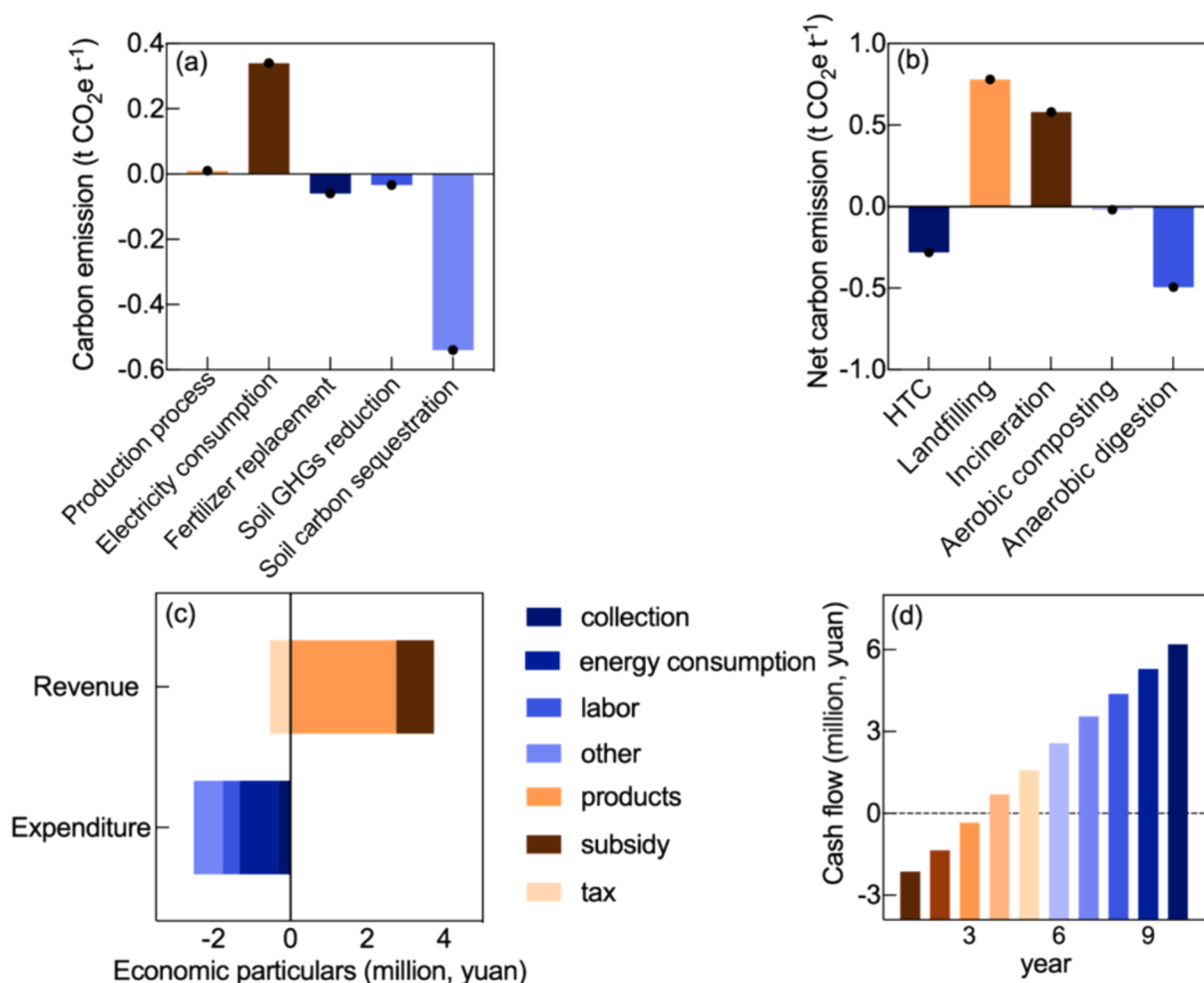


Fig. 8. Environmental assessment and economic evaluation on the hydrothermal carbonization of food waste with the application of its products in agriculture. (a) the equivalent carbon emission during the hydrothermal carbonization (HTC) of food waste (FW) with agriculture application of the products; (b) the net carbon emission under different scenarios of FW treatments (the dates were average values collected from Nordahl et al., 2020; Munir et al., 2023; Schott et al., 2016; Wang et al., 2023; Havukainen et al., 2017); (c) the revenue and expenditure, and (d) cash flow analysis of the HTC of FW with the valorization of the products in agriculture under a 10 t/day treatment capacity over ten years.

fertilizer were comparable to commercial fertilizers in promoting plant growth in both greenhouse and field. Applying the post-treated HC increased plant growth and soil nutrient availability, making it an effective soil amendment. Economic feasibility and positive environmental implication in carbon neutrality were proved for HTC treatment of FW followed with agriculture application of its products. Our integrated study provides solid evidences to support that HTC is a promising method in treating FW with great potential benefits in environment, agriculture, and economy.

The efficacy and underlying mechanisms of the AP and HC from FW in increasing crop yield should be further investigated under different climate conditions, as well as for various soil types and crop species. A more efficient and energy-saving system is expected to be adapted for large-scale HTC plant to treat FW. Additionally, the HTC of FW should be also developed by coupling clean energy sources such as wind and solar energy to reduce the dependence on non-renewable fuels. In addition, legislation and policy restrict the development of HTC of FW, the openness attitude is expected to fill the gap in industrialization and marketization in the future.

CRedit authorship contribution statement

Hao Xu: Writing – original draft. **Tong Chen:** Investigation. **Yide Shan:** Investigation. **Kang Chen:** Investigation. **Ning Ling:** Writing –

review & editing. **Lixuan Ren:** Writing – review & editing. **Hongye Qu:** Investigation. **Nicole D. Berge:** Writing – review & editing. **Joseph R.V. Flora:** Writing – review & editing. **Ramesh Goel:** Writing – review & editing. **Lubo Liu:** Writing – review & editing. **Zhipeng Liu:** Writing – review & editing. **Guohua Xu:** Writing – review & editing.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2024.153710>.

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