A Critical Review of Challenges Faced by Converting Food Waste to Bioenergy Through Anaerobic Digestion and Hydrothermal Liquefaction

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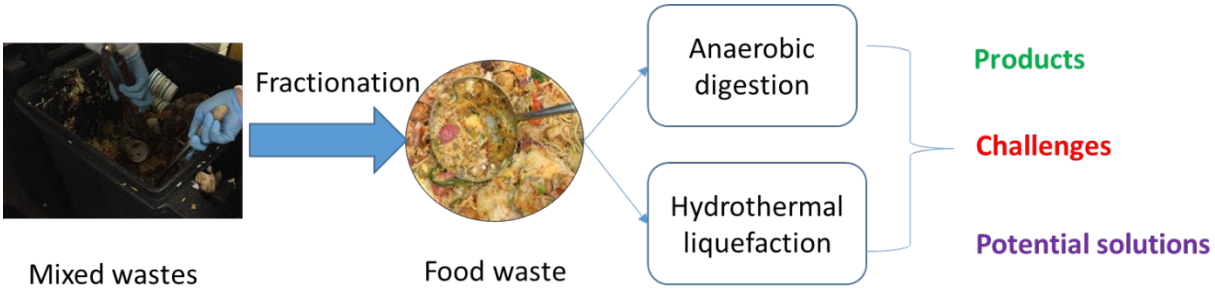
Abstract

The conventional approaches for handling food waste has been incineration, composting, landfilling, and anaerobic digestion for producing biogas. In light of organic waste bans and newly discovered presence of per- and polyfluorinated substances (PFAS) in food, food packaging materials, and compost, this review provides a critical summary of what has been investigated and reported and what needs to be considered when choosing suitable pathways for food waste. In addition to the fundamental principles inherent to anaerobic digestion and hydrothermal liquefaction, challenges for each process are identified followed by discussion of potential solutions to resolve the bottlenecks.

Keywords: food waste, anaerobic digestion, dissolved methane, hydrothermal liquefaction, per- and polyfluorinated substances (PFAS)

Graphical Abstract



Statement of novelty

Although a few review papers provided either comprehensive or specific coverage of food waste conversion, there is an urgent need to reevaluate existing technologies and explore new ones given the highly diverse nature of food waste and emerging issues related to this waste material. In particular, articles reviewing challenges faced in the field application of various technologies are significantly lacking. Thus, the novelty of this review article lies in the fact that critical challenges in valorizing real-world food waste to products were identified and potential solutions were discussed in detail. Specifically, this review manuscript aimed to draw attention to: (1) feedstock preparation in terms of fractionation and prevention of contamination; (2) potential issues related to conventional conversion pathways, such as methane in anaerobic digestor effluent and bottlenecks inherently tied to hydrothermal liquefaction; and (3) emerging contaminants, for instance per- and polyfluorinated substances (PFAS) in food packaging materials.

1. Introduction

In recent years, food waste has become a serious emerging problem due to at least three factors: the enormous and increasing quantity, issues related to conventional ways of disposal, and new concerns tied to this type of waste. The author is fully aware of reviews published on food waste recently. These reviews considered different aspects related to food waste. Topics covered include, but not limited to: food waste management from the point of understanding the food-energy-water nexus [1]; pretreatment and anaerobic digestion of food waste for production of methane, hydrogen, and ethanol [2-4]; utilization of food waste for producing large quantity commodity chemicals (biogas, biodiesel, biochar), consumer chemicals (health supplements and detergents), specialty or performance chemicals (e.g., cosmetics, coatings, adhesives, and foods), niche chemicals (e.g., chitosan, glucose, phosphate, carbohydrates and free amino nitrogen) [5]. In addition to these chemicals, other products, such as: butanol, volatile fatty acids, electricity, biofertilizer, animal feed, and biopolymers were also reviewed [6][7] [8] [6, 9]. Specific to biopolymers, such as polyhydroxyalkanoates (PHA), different types of food waste, for instance whey, waste oil, spent coffee grounds, sugar industry waste and legume waste were reviewed for producing PHA by different microorganisms, either wild or engineered strains [10].

Besides these addressing valorization of food waste from a global and comprehensive perspective, other review papers evaluated this topic for different regions, such as South and Southeast Asia [11] or targeting specific food waste, such as meat, poultry and fish processing industries [12] or fruit waste (banana peels, pineapple wastes, grape romance, orange peels, melon peels) [13, 14]; a single product from food waste: biofertilizer [15] or one conversion technology, for instance gasification followed by syngas fermentation [16].

Although these reviews provided either encyclopedia or specific coverage of food waste, there is an urgent need to reevaluate existing technologies and explore new ones given the highly diverse nature of food waste and emerging issues related to this waste material. In particular, articles reviewing challenges faced in the field application of various technologies are significantly lacking. Therefore, the objective of this review article is to fill the knowledge gaps unaddressed by researchers and practitioners. Specifically, this review focused on: (1) feedstock preparation: fractionation and prevention of contamination; (2) conversion pathways, especially problems associated with anaerobic digestion and hydrothermal liquefaction pathway; and (3) emerging contaminants, such as per- and polyfluorinated substances (PFAS) in food waste.

2. Feedstock Fractionation

2.1. Definition and current status of food waste

Different organizations, such as Food and Agriculture Organization of the United Nations, European Commission, and World Resources Institute, define food waste differently [1]. Different definition leads to different calculation of food waste generation. But globally speaking, it is estimated that around 1.3 billion tons of food waste is generated each year [5] and the total amount is predicted to be 2.5 billion tons by 2025 [2]. In the US, food waste is food not used for its intended purpose, no longer fit for human or animal consumption, and sent for disposal. These food wastes include byproducts from food and beverage processing that cannot be recycled or reused [17]. In the US, the aggregate disposal rate of food waste is estimated to be 0.615 pounds/person/day. Annually, this translates to over 35.5 million tons of food waste generation [18]. According to the US Environmental Protection Agency (EPA), a total of 35.2 million tons of food waste went to landfills in 2013. This amount is 57.5% of total food waste generated in that year [17]. In 2017, around 75.3% of food waste was landfilled [19]. These

89 wastes are generated by commercial, institutional, industrial, and residential settings and are
90 considered as the organic fraction of municipal solid waste (OFMSW). These wastes are the
91 focus of this review.

92 In view of greenhouse gas emission from organics-laden food waste at landfills, in 2015,
93 the US Department of Agriculture (USDA) and EPA adopted a domestic goal of a 50% reduction
94 of food loss and waste by 2030 [20]. In order to accomplish this goal, the EPA defined a Food
95 Recovery Hierarchy that prioritizes waste source reduction, feeding hungry people, and feeding
96 animals [21], and has provided funding for food waste reduction programs and incentives [22].
97 At state levels, several states have either developed or are in the process of developing structures
98 for organic waste bans. These states include California, Connecticut, Massachusetts, New Jersey,
99 Rhode Island, Vermont, and New York [23].

100

101 *2.2. Fractionation approaches*

102 To divert food waste from landfills, the first step is to separate these wastes from MSW (Fig. 1).
103 As reviewed by Badgett and Milbrandt [23], in 2014, there were 198 communities in the US that
104 provided curbside food waste collection. In 2019, the number increased to 326. The top five
105 states with the most communities practicing curbside collection are California, Washington,
106 Minnesota, Illinois, and Vermont. In terms of household numbers with access to curbside
107 collection, the top five states are California, Washington, New York, Texas, and Colorado.

108 New York City (NYC) has an ambitious goal of sending zero waste to landfills by 2030
109 [24]. Organics, including food scraps, food-soiled paper, and yard waste comprise 34% of total
110 MSW and the amount has been increasing over the years. In 2013, a pilot curbside organics
111 collection program served approximately 3,500 households in Staten Island. By the end of 2017,

this service reached more than 3.3 million New Yorkers. Among the collected organics, yard waste at 60% was the major component, followed by food scraps at 31%. Contamination is consisted of mainly misplaced recyclables: organics not accepted in the program, for instance diapers, textiles and construction wood; plastic bags, food wrappers; and a variety of inorganic materials. After the contaminants are mechanically sorted out, the remaining material is generally composted or sent to waste-to-energy facilities.

Processing sorted food waste for energy production was reported at a 5 ton/day pilot scale in south Korea [25]. Steps involved in the food waste to biogas were shredding the plastic wastes containing the food waste; screening through a 50-mm drum screen for rejecting coarse and light fractions, such as pieces of plastic bags, cloth and wood; and removing metals by a magnetic separator. The separated food waste was then fed to two-stage anaerobic digestion. The residence time in the first and second reactor was 5 days and 15 days, respectively. At an organic loading rate of 7.9 kg volatile solids/m^3/day, the volatile solid reduction efficiency was 70%. A total of 3.6 tons pre-sorted MSW containing 2.9 tons of food waste led to 230 m^3 biogas containing methane at 70% (v/v) and 80 kg of humus.

2.3. Bottlenecks on fractionation

Even though the anaerobic fermentation was performed successfully, problems encountered in this study [25] included: (1) unstable feed composition. Percentage change of food waste in MSW forced adjustment of the organic loading rate for the anaerobic digestion; and (2) mechanical sorting of MSW for enriching organics was difficult. On the one hand, substantial amounts of inert and organics (39.6% of the total) were rejected and sent to elsewhere for processing. On the other hand, undesired items caused serious clogging problems in the

conveying line, hoppers, and reactor bottoms. Reasonably, these problems will show up in any food waste fractionation practices. Thus, innovative approaches for separating food waste from MSW and rejecting undesired components are urgently needed. Alternatively, diverting food waste from MSW or sorting food waste at the source is an even better solution. This solution, however, requires cooperation of all food waste generators. To this end, educating the general public and raising the awareness of issues brought forth by food waste need to be initiated.

3. Conversion Pathways

As mentioned above, the majority of food waste is either disposed at landfills, incinerated, or composted. While landfilling and incineration do not enjoy good public acceptance, composting does. This is primarily due to the generation of a material that can be used in agricultural contexts either to facilitate plant growth or improve soil quality. However, detailed life cycle analysis (LCA) revealed that among the four processes evaluated: waste to energy as the baseline business as usual, composting through enclosed tunnel, composting using enclosed windrow, and anaerobic digestion (AD) of food waste followed by composting of the residual waste, the two composting schemes are not good for the environment [26]. For the studied town of Brookhaven, a suburban New York municipality, among the four processes investigated, source separation of food waste, treating it by AD, and composting the AD residuals was considered the best way to reduce the overall environmental burdens. The authors did mention that the benefits of composting may be underestimated since some aspects of compost use in the models, such as weed suppression, increased soil productivity, and water conservation were excluded. Irrespective of the conclusions made by this study, it is crucial to consider the overall environmental effect of any technologies for treating food waste. Since composting has been

158 practiced at commercial scales, the following section only covers biochemical and
159 thermochemical conversion pathways.
160
161 *3.1. Biochemical conversion: Anaerobic digestion*
162 **3.1.1. Current status and general working principle**
163 AD commercialized in the US since the late 1970s [27], is an attractive waste treatment practice
164 [28]. Currently, the U.S. has more than 2,116 active anaerobic digesters that produce biogas [29].
165 The USDA estimated that additional 11,000 biogas systems could be deployed in the US and
166 bring the total number of sites to 8,241 on dairy and swine farms, 3,681 at Wastewater Treatment
167 Plants (WWTPs), and 1,086 at landfills [27]. As stated in the Biogas Opportunity Roadmap,
168 biogas systems offer at least six benefits, such as creating additional revenues; providing a
169 renewable and sustainable source of energy; driving economic growth, especially in rural areas;
170 cutting methane emissions from uncontrolled sources; protecting the environment by preventing
171 release of greenhouse gases; and enhancing resilient communities nationwide.
172 Through numerous scientific studies and practical applications at various scales, the AD
173 process, despite its complexity of dealing with mixed feedstocks and mixed microbial
174 communities, has been understood well. In general, there are four stages involved in AD:
175 hydrolysis, acidogenesis, acetogenesis, and methanogenesis. The first two stages are performed
176 by hydrolytic and acidogenic bacteria to hydrolyze the complex substrates (carbohydrates, lipids,
177 proteins, etc.) to simple monomers (sugars, fatty acids, amino acids, etc.) and further to CO_2, H_2,
178 organic acids, and alcohols. These structurally simple compounds are then converted by
179 hydrogen producing acetogens to form acetate, H_2, and CO_2, a process termed acetogenesis.

180 Methanogenesis, conducted by methanogens, further utilize the H_2, CO_2, and acetate to produce
181 CH_4 [30].
182
183 **3.1.2. Problems associated with AD**
184 Converting food waste to biogas has been reviewed in detail by several researchers [2-4] and is
185 thus, not discussed further here. But one emerging issue of greenhouse gas (GHG) emission from
186 digesters needs to be highlighted. This emission has two outlets. First, fugitive emission of GHG.
187 Based on downwind gas measurements at a municipal WWTP, methane emissions corresponded
188 to 2.07-32.7% of methane generated in the plant. Under steady conditions, the loss of CH_4 was
189 found to be 2.1-4.4% of the gas generated. But up to 32.7% of CH4 generation could be lost
190 during periods of operational difficulties, for example release from pressure valves
191 accompanying a foaming event [31].
192 Second, methane dissolved in the effluent leaving anaerobic digesters has emerged as a
193 key concern for mainstream anaerobic processes [32]. For effluent from anaerobic membrane
194 bioreactors (AnMBR), the average dissolved methane concentration was 1.5 times oversaturation
195 relative to Henry's law. At 15^{o}C, the dissolved methane in the permeate from AnMBR was 40-
196 50% of total methane generated from the system. This large loss of methane to the effluent is due
197 to higher methane solubility at lower temperatures and oversaturation [33]. Methane
198 oversaturation has also been observed in non-membrane conventional anaerobic digesters [34-
199 36]. If not captured, the dissolved methane will eventually end up in the atmosphere.
200 **3.1.3. Approaches for recovering or removing methane from AD effluent**
201 Considering the significant mass of methane in AD effluent, various approaches have been
202 reported either for recovering methane or for removing this gas from the effluent. At the

recovering side, a hollow-fiber degassing membrane module was used for degasifying methane in the liquid outlet of an upflow anaerobic sludge blanket (UASB) reactor [37]. Under 35 °C and a hydraulic retention time (HRT) of 10 h, the average dissolved methane concentration was 15 mg chemical oxygen demand (COD)/L from an initial 63 mg COD/L. Correspondingly, total methane recovery efficiency was increased from 89% to 97%. At 15°C, the average dissolved methane concentration was 14 mg COD/L from a beginning 104 mg COD/L. The methane recovery efficiency was from 71% to 97%. Due to invasion of air, however, methane content in the collected gas was around 20%. Later on, the same group of researchers reported a collection efficiency of 41-60% using similar membrane degasification [38]. Methane in the recovered gas was around 52%, similar to 50% in the AD headspace. The dissolved methane concentration was 51 mg COD/L, which is equivalent to 13 mg CH_4/L. Although technically this membrane degasification seems promising, energy required for this membrane process needs to be calculated carefully to ensure a positive energy balance.

Besides energy intensive membrane degasification, aeration either by biogas or air could be an option. Biogas-assisted mixing was reported to lead to a methane recovery efficiency of 53.6% at 20 °C and 57.4% at 33 °C [39]. Compared to biogas, air could be used to strip methane out of and add oxygen into the effluent [40]. With air provided, the off gas from a closed vessel can be captured. It needs to be noted that this aeration is not selective. While methane is stripped, CO_2 and other gases in the effluent will partition to the gas phase, too and dilute methane in the recovered gas. This off gas could be burned together with biogas to generate heat and electricity. This simple approach could lead to low methane concentration in the effluent, the associated cost, however, needs to be computed to demonstrate its benefit.

In addition to physical separation and recovery, methane in the effluent could be utilized by biological processes. Microorganisms play key roles in the global methane cycle. It is estimated that approximately 300 teragrams (Tg) of methane are produced annually through methanogenesis. Before this huge amount of methane diffuses into oxic environments, 90% of that is consumed by methane oxidizers [41]. As a highly reduced molecule, methane can be oxidized either aerobically or anaerobically [42]. While the aerobic pathway involving methane monooxygenase enzyme has been well understood [43], anaerobic oxidation of methane (AOM) remains as one of the most controversial, scientifically intriguing, and technically challenging subjects of microbial ecology [44]. AOM was initially identified in marine sediments and water columns more than 30 years ago and was estimated to consume > 50-90% of methane released from methanogenesis in the ocean [45] [46].

There are two groups of microbes that can perform AOM. The first group is nitrite-dependent anaerobic methane oxidizing bacteria (AMOB). These bacteria, closely related to *Candidatus Methylomirabilis oxyfera*, a member of the uncultured NC phylum [47], have been found in many environments including freshwater lakes [48, 49], rivers [50], wetlands [51-56], and marine ecosystems [57, 58]. *Ca. Methylomirabilis oxyfera* is able to oxidize methane using nitric oxide as the source of molecular oxygen via the complete aerobic pathway starting with particulate methane monooxygenase (pMMO) [59, 60]. The second group is generally referred to as anaerobic methanotrophic archaea (ANME). Under the phylum of Euryarchaeota, ANME has three distinct clusters, ANME-1, ANME-2, and ANME-3. These clusters are distantly or closely related to the orders of *Methanosarcinales* and *Methanomicrobiales*.

Three mechanisms have been proposed for ANME to oxidize methane anaerobically with assistance from nearby bacteria. The first one is acetogenesis. Under this mechanism, there

248 are two hypotheses. The first one hypothesizes that methane oxidizing archaea uses two
249 molecules of methane to produce H_2 and acetic acid which are subsequently consumed by sulfate
250 reducing bacteria (SRB) [61]. The second hypothesis is reverse aceticlastic methanogenesis. In
251 this case, methane oxidizing archaea produces acetate from CO_2 and methane and SRB consume
252 the formed product [62, 63]. The second mechanism, methylogenesis, hypothesizes that methane
253 is activated as it binds to coenzyme M (CoM) and releases electrons for ATP synthesis in
254 ANME. The produced methyl sulfide from methane oxidizing and CO_2 reducing archaea are
255 transferred to SRB. The third mechanism, reverse methanogenesis, has been investigated
256 intensively and accepted broadly. This theory assumes that the initial step of methane oxidation
257 is primarily a reversal of the terminal reaction in methanogenesis which yields methane and the
258 heterodisulfide (CoM-S-S-CoB) from the reduction of methyl-coenzyme M (CoM-S-CH_3) with
259 coenzyme B (H-S-CoB) [64]. Methyl coenzyme M reductase (Mcr) is the enzyme responsible for
260 this step. Under this mechanism, based on the electron acceptors, ANME is divided into three
261 categories: sulfate dependent, nitrate dependent, or iron/manganese dependent.

262 AOM by ANME has been believed to be mediated by a consortium of ANME archaea
263 and sulfate or nitrate or iron/manganese-reducing bacteria [47, 65, 66]. Recently, however, a new
264 hypothesis was proposed that some ANME groups might mediate AOM alone. This hypothesis is
265 supported by: (1) without any bacterial partner, ANME archaea have been found alone as single
266 cells or as monospecific aggregations [67-70]; (2) ANME-1 archaea especially seem to be less
267 dependent on the activity of a closely associated bacterial partner [69, 71]; (3) ANME-2 can
268 perform AOM and reduce sulfate to disulfide which is then taken up by SRB and
269 disproportionated to sulfide and sulphate in a 7:1 ratio [72]; and (4) ANME-2d is capable of
270 independent AOM using nitrate as the terminal electron acceptor [41].

The benefit of AOM lies in the fact that methane can be removed from digester effluent while either nitrite, nitrate, or sulfate can be depleted, too in the coupled process simultaneously, thus producing treated water ready for discharge. To get a better idea on how this could be used in reality for treating digester effluent, readers are referred to these reviews and research papers [41, 47, 73-83].

3.2. Thermochemical Conversion

3.2.1. Working principles and current status

Thermochemical processes typically include gasification, combustion, pyrolysis, and hydrothermal liquefaction (HTL). While the first two approaches transform feedstock materials to various gases, the latter two strategies are used mainly to produce liquid transportation fuels. During pyrolysis and liquefaction, oil molecules that are commonly referred to as bio-oil or bio-crude are generated in addition to gaseous products and solid char. Compared to solid fuels and syngas, biocrude presents several advantages: 1) higher energy density; 2) easier to transport and store than gaseous products; 3) potential to be used as fuel oil substitute; and 4) usable in engine, turbine, and burner applications [84].

Fast or slow pyrolysis works well with biomass having moisture content of less than 40%. Higher water content requires higher heat of vaporization for this process. In contrast, HTL can handle biomass with high moisture contents in the range of 75-95% [85]. HTL, also referred to as thermochemical liquefaction or subcritical water liquefaction, benefits from the characteristics of hot compressed water, which is highly reactive near its critical point (374 ^{o}C, 22.0 MPa) [86]. During liquefaction, a series of reactions occurs in three steps: hydrolysis, depolymerization, and repolymerization/self-condensation [87].

Generally, lipids are transformed to fatty acids, proteins are turned to nitrogen heterocycles, pyrroles, and indoles while carbohydrates are converted to cyclic ketones and phenols [88]. For lignocellulosic materials, given the complicated structures of the components, the reactions are even more complex. Cellulose, as a linear polymer of glucose connected by β-1,4 –glycosidic bonds, possesses strong intra- and inter-molecular hydrogen bonds [89, 90]. As a result, crystalline cellulose is resistant to swelling in water under normal conditions. But in subcritical or supercritical water, the two types of bonds are broken. Glucose and oligomers are released as a consequence. The freed sugars are then going through a series of degradation and lead to the formation of many compounds [91], such as: acetaldehyde, acetic acid, acetone, acrylic acid, 1,2,4-benzenetriol, dihydroxyacetone, erythrose, formic acid, fructose, furfural, glycealdehyde, glycoaldehyde, glycolic acid, 5-hydroxymethylfurfural, lactic acid, levoglucosan, and pyruvaldehyde.

Different from cellulose, hemicellulose is composed of several sugar monomers, such as xylose, mannose, glucose, galactose, and others [92]. Hemicellulose does not have a crystalline structure and is much more susceptible to degradation and hydrolysis compared to cellulose due to the presence of side chains and the lack of repeating β- 1,4 –glycosidic bonds [93]. Under hydrothermal condition, this polymer is hydrolyzed to its corresponding monomers which can be reacted further to form a variety of compounds similar to those listed above for glucose. Lignin is a complex compound with a high molecular weight and an even more random structure than hemicellulose. During HTL, lignin is degraded to its three most prevalent monomers: *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol as well as other chemicals resulting from further degradation of these monomers [94].

For the purpose of producing biocrude, HTL has been investigated to liquefy several biomass materials, such as: swine manure [95], wood flour [96], sugar beet pulp [97], garbage [98], microalgae [99], and microalgal residue after lipid extraction [100]. Compared with a higher heating value (HHV) of 10 to 20 MJ/kg and an oxygen content of 30 to 50% of the processed feedstocks, the oil produced generally has a HHV of 30 to 36 MJ/kg and an oxygen content of 10 to 20% [101]. The reported oil yield, however, varies broadly from 12% to 75% depending on the feedstock tested.

For HTL, yield of biocrude can be affected by several factors. As a thermal process, temperature has been recognized as the most important one. At temperatures lower than 280 ^{o}C, incomplete decomposition of individual feedstock components suppresses the biocrude yield. At too high a temperature, above 350 ^{o}C, however, oil yield is also lowered. This is due to: first, increased gas formation as secondary decomposition and Bourdard gas reactions become active at high temperatures [102]; second, recombination of free radicals leads to formation of char. Thus, temperature for HTL of wet biomass materials has been recommended to be 300-350 ^{o}C [85].

Besides temperature, catalysts have been proven to play significant roles in increasing oil yield and improving oil quality. Various catalysts, such as organic acid (formic acid, acetic acid), alkaline (potassium hydroxide), sodium carbonate, and metals have been tested for different biomass feedstocks. The exact effect from different catalysts varies significantly for the same tested material [91, 103].

In addition to temperature and catalysts, feedstock particle size may also influence oil yield considering the fact that small particles having larger surface areas may have better accessibility to water for achieving higher degree of reaction. However, as sub/supercritical

water overcomes the heat transfer limitation in HTL, particle size is suggested to be a minor factor [104]. A particle size between 4 and 10 mm is suitable for HTL. This size requirement eliminates the need for costly grinding. Additionally, from all reported studies, biomass heating rate, residence time, pressure, and the presence of a reducing gas (hydrogen and/or carbon monoxide) are all secondary parameters. Their effects are either minor or negligible for oil yield [85].

3.2.2. HTL of food waste

Through HTL, as other wet wastes, food waste could be converted to biocrude. The resulting biocrude could be upgraded to renewable diesel and aviation kerosene. If all to kerosene, then 3.8% of kerosene-type jet fuel produced in 2016 could be from the food waste generated in the conterminous US [105]. Although the potential impact is not negligible, at the time of writing, only a few studies have evaluated HTL of food waste from different perspectives. The publications are categorized into three groups. First, characterization of the resulting products from HTL of food waste. Analysis of biocrude produced from meat, cheese, or fruits was revealed the presence of thousands of heteroatom compounds in each biocrude sample [106]. The aqueous byproducts derived from HTL of food industry waste, municipal waste, and yeast and algae cultivated on wastewater were evaluated in detail. Both organic and inorganic species in these aqueous streams were profiled and quantified using state-of-the-art technologies [107]. The solid products, the hydro-char derived from HTL of dairy waste, waste from WWTP, dining halls, fruit and alcohol manufacturing and olive oil production were characterized [108].

Second, food waste, both pure and mixed were investigated under different HTL conditions (Table 1). Model food waste molecules, such as potato starch, casein, and sunflower

oil were investigated by fast and isothermal HTL. The former at 600 ºC for 1 min led to higher biocrude energy recoveries than did the latter at 350 ºC for 60 min for all three feedstocks, especially the polysaccharides [109]. When ternary mixtures were fed to the HTL reactors, the biocrude yield ranged from 25% to 55%. The energy recovery was between 47% and 75% depending on the exact composition of the mixtures. This study also revealed that reactions did occur between molecules derived from different feedstocks in the mixture.

Posmanik et al. investigated the feasibility of coupling HTL of binary and ternary model compounds with anaerobic digestion. The AD is used to convert carbon in the aqueous phase to methane. While higher HTL temperatures favored oil production, the anaerobic degradability of the aqueous phase correlated negatively to HTL temperature. This is probably due to the presence of recalcitrant or inhibitory products formed during HTL at high temperatures [110]. The same group of researchers also evaluated HTL of carbohydrate rich food waste from university dining hall with or without an acid or base. It was concluded that oil yield is associated with different chemical pathways and dehydration reactions were enhanced by acidification through acid addition [111].

Compared to homogeneous catalysts, heterogeneous ones, such as $CeZrO_3$ was demonstrated to have better performance in terms of energy balance and economics, especially when the catalyst's lifetime is considered [112]. Compared to non-catalytic and Na_2CO_3-catalyzed HTL of synthetic institutional food waste, $CeZrO_3$ gave higher biocrude yield, energy recovery as well as an aqueous phase that contained approximately half of the organic carbon detected in those derived from Na_2CO_3 catalyzed reactions. In addition, $CeZrO_3$ nanopowder was shown to be stable and had minimal leaching to the aqueous phase. The reusability was studied using a model compound of pentanal. Even though the catalyst can be reused between runs

385 without calcination and regeneration, it is unclear how $CeZrO_3$ would be used when actual food
386 waste is processed. In this case, the solid catalyst would end up in the char. Whether this catalyst
387 embedded in char can still be reused would be highly questionable.
388 Third, two studies have attempted to model HTL of food waste. Deniel et al. used
389 monomeric and polymeric model molecules to model the behavior of wet organic residues
390 during HTL [113]. With real wet waste, - 8.0 to + 4.8 wt% of experimental yields were observed.
391 The authors pointed out that a model protein instead of a model amino acid or native extracts of
392 the biomass should be used to obtain better representativeness and accuracy of the model. In
393 another study, Aierzhati et al. developed a predictive model to accurately determine biocrude oil
394 yield from different food wastes. This regression model was developed using R software using
395 data generated from hydrothermally treating three groups: high liquid, high protein, and high
396 carbohydrate at temperatures ranging from 280 ºC to 360 ºC for 10-60 min. The response of
397 biocrude yield can be calculated from content of protein, lipid, and carbohydrate, HTL
398 temperature, and residence time. This model was also able to predict previously published data
399 with a R^2 of 94.3% [114].
400
401 *3.2.3. Bottlenecks for HTL of food waste*
402 HTL has been investigated extensively for biomass materials, such as wood, short-rotation
403 woody crops, agricultural wastes, short-rotation herbaceous crops, animal wastes [85, 115-117],
404 microalgae [118-120], and sewage sludge [121-125]. Compared to these feedstocks, research on
405 HTL of food waste is much less. Based on the few studies summarized in Table 1, it is
406 reasonable to perceive that HTL of food waste will face the same challenges as those for other
407 biomass materials. Some critical bottlenecks need to be addressed before HTL could be used at

commercial levels for food waste conversion. First, the carbon yield in biocrude needs to be improved. This issue is due to relatively low biocrude yield and less than optimal separation between solid and liquid and liquid and liquid. To solve this problem, the HTL process must be optimized followed by efficient separation steps. By doing so, organic-soluble compounds can be maximally partitioned to the water insoluble phase resulting in higher biocrude yield and less organics lost in the aqueous phase. Second, disposal of the aqueous stream. Under general HTL condition, the aqueous stream derived from HTL contains 20-50% of feed carbon [107]. This leads to a byproduct having high concentration of COD, high nitrogen, and phosphorous concentrations. Third, performing HTL under continuous operation mode suffers multiple problems, such as feed material pumpability; equipment capital cost, especially at modular or small scales; operation and maintenance cost, in particular for communities where trained professionals are hard to find to operate small food waste HTL reactors requiring high pressure.

4. PFAS in Food Waste

4.1. PFAS in diet

Concentration of PFAS in food waste, although not reported so far, should be highly dependent on the source of food and water used for preparation and/or cooking. A detailed review of literature between May 2011 and October 2016 revealed that the main route of human exposure to these compounds is dietary intake [126]. This conclusion is based on studies conducted mainly in European countries: Germany, the Netherlands, France, Spain, Sweden, Italy, Greece, Finland, and Greenland. Among all of these, four studies performed in four European countries (Belgium, Czech Republic, Italy, and Norway) are noteworthy. These four investigated 14 PFAS in 20 vegetable species [127], 21 PFAS in 50 selected pooled samples representing 15 food

commodities [128], 12 PFAS in fruits, cereals, sweets, and salt [129], and 7 PFAS from the PERFOOD EU project [130]. In south Korea, 16 PFAS in 397 food items were evaluated [131]. For another four countries: Brazil, Saudi Arabia, Spain, and Serbia, 21 PFAS were quantified in 283 foodstuff representing the diet in South America, western Asia, Mediterranean area, and southeastern Europe [132]. According to the recommendations by the European Food Safety Authority (EFSA) issued in 2012 on the maximum dietary intakes of PFOS and PFOA [133], it was concluded that for nonoccupationally exposed populations, human health risks would not be of concern. This could be true for the very limited countries for which recent data are available. However, considering the relatively high concentrations of PFAS detected in fish and other seafood, it was cautioned that certain risks could be high for individuals consuming great amounts of fish and shellfish [126].

This conclusion is in line with results disclosed by earlier studies. For example, an analysis of 252 food samples (UK-produced and imported) showed that all 11 of the targeted PFAS were detected in 75 individual food items. The highest levels found were 63 μg total PFAS/kg with 59 μg/kg for perfluorooctanesulphonic acid (PFOS) in an eel sample, and 40 μg/kg PFOS out of 62 μg total PFAS/kg in a whitebait sample [134]. Similarly, PFAS have been detected in Canadian diet, such as meat, fish, fast food, and popcorn [135]. Studies by the same Canadian research group later revealed that conventional way for cooking fish, either baking, broiling, or frying was no effective to reduce PFAS content in these food [136].

In addition to studies related to food, uptake of PFAS by vegetables has been investigated mainly from the perspective of food chain or possible human consumption. For this purpose, edible plants, such as carrots, potatoes (direct contact with soil), and cucumbers (no direct contact with soil) were grown on soil amended with sewage sludge spiked with PFOA and PFOS

[137]. It was found that PFOA had higher transfer factors than PFOS for these three plants. In particular, the vegetative compartments of cucumber (stalks and leaves) had the highest transfer factor of 0.88 for PFOA, 0.17 For PFOS. Lettuce and tomato were studied in a similar fashion by growing in industrially impacted biosolids-amended soil [138]. The soil was not spiked. But the amendment rates were generally above typical agronomic application rate. Results from this study showed that preferential uptake of perfluorinated carboxylic acids (PFCAs) over perfluorinated sulfonic acids (PFSAs) and accumulation of shorter chain PFAAs over longer chain PFAAs. Perfluorobutanoic acid (PFBA) had the highest bioaccumulation factor (BAF) of 56.8 in lettuce and perfluoroheptanoic acid (PFPeA) had the highest of 17.1 in tomato. In another study [139], the same research group grew radish, celery, tomato, and sugar snap pea in three kinds of soil: industrially impacted biosolids-amended soil, municipal biosolids-amended soil, and control soil. In edible portion of the crops grown in the first type of soil, PFOA had the highest concentration of 67 ng/g in radish root; PFBA, 232 ng/g in celery shoot; and PFBA, 150 ng/g in pea fruit.

The Minnesota Department of Health conducted a similar study by measuring PFAS in garden produce exposed to PFAS contaminated drinking water [140]. PFBA is the primary PFAS present in water followed by PFPeA. Although PFBA, PFOA, and PFOS appeared in 100% soil samples at higher concentrations than other PFASs, only PFBA was found in plants.

Apart from food, PFAS in drinking water has been a critical issue. As reported by a 2017 paper, across the US, hundreds of sites have been contaminated by PFASs and more than 6 million Americans are consuming water containing PFOS and PFOA exceeding the US EPA's provisional guidelines for these two chemicals, a maximum combined 70 ng/L [141]. Very recently, according to test results published by Environmental Working Group (EWG), out of tap

477 water samples from 44 places in 31 states and the District of Columbia, only one location had no
478 detectable PFAS, and only two other locations had PFAS below 1 ppt
479 (https://www.ewg.org/research/national-pfas-testing/).
480
481 *4.2. PFAS in food packaging or contacting materials*
482 The unique properties of PFAS being water- and grease-repelling have led to their extensive use
483 in food packaging and contacting materials (FPCM). FPCM include boxes for fries and pizzas;
484 wrappers for sandwich, burgers, dessert and bread; microwave popcorn bags; paper cups, etc.
485 [142]. In most finished FPCM, PFAS concentrations were typically found in the range of 1.0 −
486 1.5% per fiber dry weight [143]. As a result of phase-out of PFOS and its precursors by 3M
487 [144], PFAS used in FPCM nowadays are mainly acrylate polymers with fluorotelomer or
488 sulfonamido alcohol side chains or perfluoropolyether perfluoropolyether- based polymers
489 (PFPEs), a shift from PFOS precursors, such as N-ethyl perfluorooctane sulfonamido alcohol-
490 based phosphate diesters (SamPAPs) [145, 146].
491 PFAS migrating from FPCM to food has been reported. For example, results from the
492 popcorn bag experiments demonstrated that fluorotelomer in the 3 – 4 mg/kg range was found in
493 food as a result of migration of these chemicals [147]. The same researchers also reported that
494 the fluorotelomer migration from paper could be significantly enhanced by high temperatures
495 and emulsifier-containing oil [148]. In view of the potential migration of PFAS from FPCM to
496 food, several states in the United States have implemented bans on the use of these chemicals in
497 food packaging and the Danish Ministry of the Environment and Food has established a total
498 fluorine indicator value of 0.1 μg/cm^2 in food packaging [143].

PFAS in FPCM are counted as the major source of these compounds detected in finished compost samples. In one study, for composts that included food packaging, the loads of 17 perfluoroalkyl acids (PFAAs) ranged from 28.7 to 75.9 μg/kg of OFMSW. For those without food packaging, the content of these PFAAs was 2.38 to 7.60 μg/kg. Thus, apparently, food packaging was the main source of PFAS identified in these samples [149].

The PFAS concentrations detected in these compost samples, however, are lower than the screening standards set by Maine's Department of Environmental Protection (DEP) on March 22, 2019. Maine DEP's memorandum requires that sludge/biosolids program licensees and sludge/biosolids composting facilities to test their materials for PFAS. The screening standards are: 2.5 μg of PFOA/kg, 5.2 μg of PFOS/kg, or 1,900 μg of PFBS/kg [150]. Thus, it appears that the PFAS-containing compost can be safely land applied under the current regulatory atmosphere. Whether this remains true in the future, however, is uncertain given the rapidly growing knowledge of PFAS and stricter regulations toward these chemicals.

4.3. Challenges related to PFAS in food waste

Due to the highly recalcitrant nature of PFAS, only few technologies are able to remove or degrade these compounds. The top approach that has been investigated extensively and reported numerous times is sorption. A broad range of sorbents, such as activated carbon (AC), ion exchange resin, minerals, carbon nanotubes, biochar, and molecularly imprinted polymers have been tested for use in PFAS removal from surface water, drinking water, leachate, and wastewater [151-156]. Besides sorption, other reported techniques include: (1) coagulation and flocculation. Various coagulants, such as aluminum salts [157], ferric salts [157], specialty coagulants [158], and electrocoagulation [159, 160] have been investigated; (2) membrane

filtration. Within this category, reverse osmosis [161, 162], nanofiltration [163-165], microfiltration [166], and electrodialysis [162] have been studied at lab scales for ex situ PFAS treatment; (3) chemical reactions. Within this group, ozone-based systems [167, 168], sonochemical [169-172] and electrochemical [173-177] have been tested at pilot scales. Other approaches, for instance alkaline metal reduction, doped zero valence iron, plasma, photolysis [178, 179], activated persulfate, hydrogen peroxide based systems, and solvated electrons have all been studied at laboratory levels [151]; and (4) biodegradation. Both anaerobic and aerobic microbial systems have been tested for degrading PFAS. While PFAS precursors are reported to be transformed by microbial activities [180-187], PFOA is demonstrated to be microbiologically inert [188] by one research group and degradable by other researchers [189, 190]. Similarly, whether PFOS can be biodegraded or not is also highly debated [191-193]. Very recently, both PFOA and PFOS at 0.1 mg/L and 100 mg/L, are reported to be degradable by pure and enrichment cultures of *Acidimicrobium* sp. strain A6 (A6), an autotroph that oxidizes ammonium to nitrite while reducing ferric iron. This process, however, is very slow. Removal of up to 60% of PFOA and PFOS was observed during 100 day incubations [194]. Thus, for composting and AD of food waste, assuming biodegradation could take place, the percentage of degradation within the respective residence time would not be much.

Different from biodegradation, hydrothermal reactions did show promise for destruction of PFAS. While PFOS had little reactivity in pure subcritical water, amendment of iron powder (< 53 μm) at 53.6 g/L decreased PFOS concentration from 372 μM (186 mg/L) to < 2.2 μM after 6 h at 350 ºC. No PFCAs were detected. The authors concluded that the added iron provided surface for PFOS sorption. The sorbed PFOS was then decomposed with the increase of temperature with the release of fluoride ion [195]. The same researchers also reported

degradation of perfluorohexanesulfonate (PFHxS), a bioaccumulative analogue to PFOS in subcritical and supercriticial water. Compared to PFOS, PFHxS had slight reactivity in pure water at 350 ºC. At 380 ºC, the decomposition to fluoride and sulfate became more significant. Similar to PFOS, the addition of iron dramatically accelerated PFHxS decomposition at both temperatures [196].

A recent study also confirmed PFOS degradation under HTL conditions. Screening a wide ranges of amendments including acids, bases, strong oxidants, and metals revealed that NaOH, $NaBH_4$ and K_2FeO_4, each at 1 mol/L, gave the highest rate of defluorination of PFOS (50 mg/L) in water at 350 ºC for 90 min. The most reactive amendments, irrespective of their type, all shifted the solution pH to be ≥ 9, thus suggesting base-promoted mechanism [197]. PFOA was demonstrated to be more unstable than PFOS in hydrothermal water. The degradation rate was > 99% at 250 ºC for 30 min without any amendments.

Although HTL shows great promise in destructing pure PFAS in water either with or without an amendment, it is unclear whether: (1) this approach can destroy other PFAS or PFAS mixtures rather than PFOA and PFOS; (2) this method can degrade PFAS mixtures at ng/L or ng/kg concentrations in food waste; and (3) how the optimal conditions for PFAS degradation intersect with other goals of HTL of food waste, such as biocrude yield.

5. Conclusion

In light of detection of emerging contaminants, such as PFAS in food waste, more states implementing organic waste bans, stricter environmental regulations, and public pressure demanding better approach for handling food waste, it is more urgent than ever before that researchers, regulators, and practitioners should work together to address these challenges. On

the one hand, conventional approaches used at commercial levels, such as incineration, composting, and AD need to be re-evaluated to assess their suitability in dealing with food waste. On the other hand, innovative and out-of-the-box methodologies need to be proposed and tested to transform food waste into commodities that are valuable. Only when food waste is utilized in positive ways, treated as a resource rather than garbage, can this waste and the problems associated with its disposal be truly resolved.

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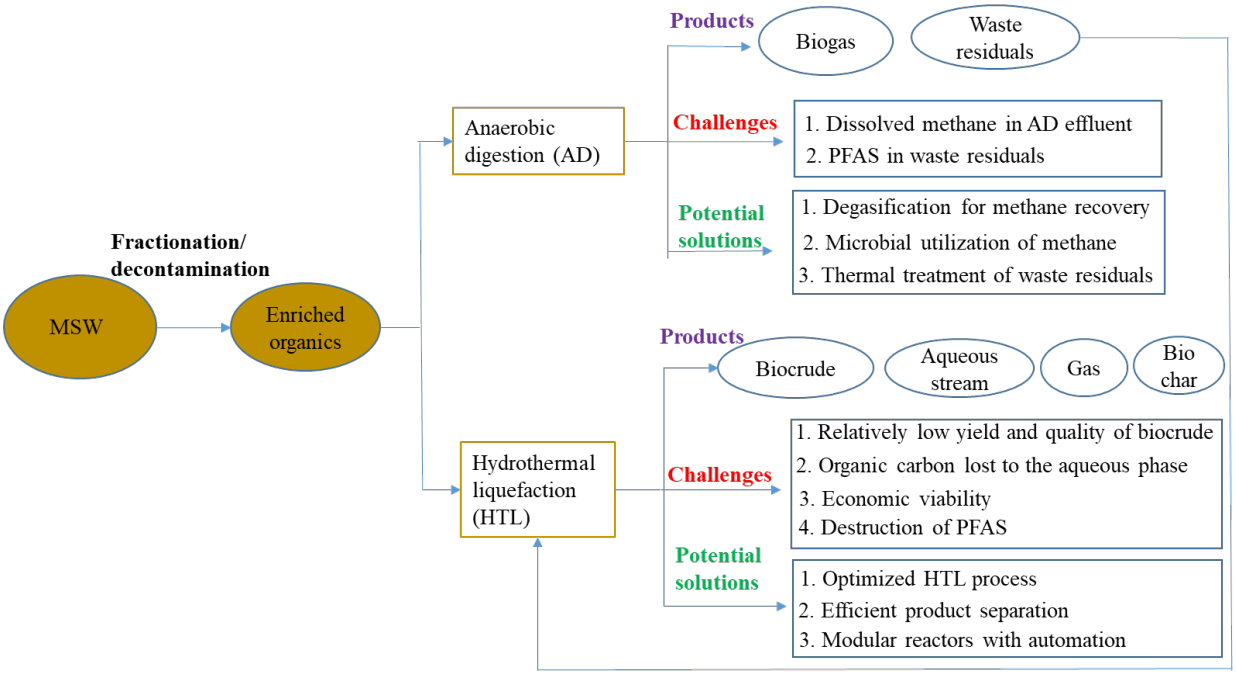

Fig. 1: The overall process diagram for converting food waste to fuels and commodities.

Table 1: Comparison of food waste conversion by HTL.

Feedstock	**Temperature** (°C)	**Residence time** (min)	**Catalyst** (%)	**Carbon yield in biocrude** (%)	**Biocrude yield** (%)	**Biocrude HHV** (MJ/kg)	**Energy recovery** (%)	**TOC of aqueous phase** (ppm)	**Reference**
Synthetic institutional food waste	300	60	$CeZrO_3$	53	1.59 x of noncatalytic	31.2	38.8	13800	109
			Na_2CO_3	39	1.12 x of noncatalytic	24.2	21.3	24200	
			No	38.8		35.6	27.6	12500	
Carbohydrate rich from dining hall	300	60	5 M H_3PO_4	43	NA	32.5	NA	NA	107
			1 M NaOH	33	NA	35.4	NA	NA	
			NO	18	NA	34.8	NA	NA	

Potato starch	600	1			18.6	37	46		
	350	30			13	36.3	38		
Casin	600	1			23	36.8	43		
	350	30	NO	NA	20	33.2	38	NA	106
Sunflower oil	600	1			91	43.5	95		
	350	30			89	42.3	86		
Ternary mixtures	600	1			25-55	NA	47-75		
Institutional food waste	360	40	NO	NA	46.9	38.15	72.2	NA	111
Blackcurrant pomace	300	60	NO	NA	27	NA	NA	NA	110