# Anaerobic digestion of food waste for bio-energy production in China and Southeast Asia: a review

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# Abstract

Rapid economic growth in Asia and especially in China, will lead to a huge increase of food waste (FW) production that is expected to increase by 278 - 416 million tonnes. Among various waste management practices, anaerobic digestion (AD) is a useful method to transform food waste, producing renewable energy/biofuel and bio-fertilizers. This review aims to investigate some of the key factors in proposing FW for anaerobic digestion, with particular reference to China and South East Asian countries.

Food waste showed variable chemical composition and high content of biodegradable material (carbohydrates, protein and lipid) led to consistent biogas production (as potential) that was reported as average for Chinese FW, of  $480 \pm 88$  LCH<sub>4</sub> kg<sup>-1</sup> VS (n = 42) being this data higher than those for energy crops ( $246\pm36$  LCH<sub>4</sub> kg<sup>-1</sup> VS), makes FW a good candidate to substitute energy crops, avoiding food-energy conflict. FW co-digestion with different substrates improved total biomethane production (on average), i.e. from  $268 \pm 199$  mL g<sup>-1</sup> VS to  $406 \pm 137$  mL g<sup>-1</sup> VS. Food waste pretreatment, also, seems to be very useful in increasing total biogas production and physical and thermal treatments were the best increasing biogas of +40 % and + 30 %, respectively. Techno economic evaluation seems to indicate the feasibility in substituting EC with FW for producing biogas and reducing total biomass cost. To achieve this, separate collection sources need to be implemented, assuring high FW quality to promote a Circular Economy approach in FW management.

# **Keywords:**

Anaerobic digestion. Biomethane. Biomethane potential. Energy balance. Food Waste. Food waste co-digestion. Food waste pre-treatment.

# Abbreviations

AcoD: anaerobic co-digestion

AD: anaerobic digestion

BMP: biochemical methane potential
CFW: canteen food waste
CM: cattle manure
COD: chemical oxygen demand
CSTR: continuous stirred tank reactor
EC: energy crop
FOG: fat, oil and grease
GHG: greenhouse gas
HRT: hydraulic retention time
KFW: kitchen food waste
FW: food waste
MA: microalgae
MSW: municipal solid waste
OFMSW: organic fraction of municipal solid waste
OLR: organic loading rate
RFW: restaurant food waste
SFW: synthetic food waste
S/I: substrate/inoculum
SS: sewage sludge
TS: total solids
VFA: volatile fatty acid
VS: volatile solids
WAS: waste activated sludge

# Word count

## 1. Introduction

The utilization of fossil fuels creates environmentally negative effects on the planet because of greenhouse gas emissions (GHG) [1]. GHG increased a lot in the last century, exceeded 400 ppm in 2006 and reached 417 ppm in May 2020, the highest values ever registered [2,3]. GHG increases lead to climate change, the consequences of which have been widely scientifically discussed [4,5]. Reducing GHG emissions has become a priority for the world: to date 175 parties have ratified the Paris Agreement (Climate Summit held in Paris in 2016 - COP 21) [6]. Renewable energy and combustible production represent an option in reducing GHG emissions [7,8]. Several types of environmentally friendly and sustainable renewable energy production options such as photovoltaic, wind, biomass, geothermal, have recently received attention [7]. Renewable energy and combustibles are those produced from renewable resources. Biomasses represent a well-known and widely diffused renewable source. Between different available biomasses, food waste (FW) represents a resource that needs to be safely disposed of, otherwise, if landfilled, it can contribute extensively to GHG emissions [10]. The contribution of landfilled wastes to GHGs was reported to be 5% of the total GHGs emitted [11] and 6-18% of the total methane from anthropogenic sources [12]. Food waste (FW), is the main component of municipal solid waste and it has been described as "the decrease in quantity or quality of food"; it is part of food loss and refers to discarded or non-food use of food [13].

Due to rapid economic growth, FW generation in Asia is expected to increase by 278 - 416 million tonnes [14] contributing to 8-10% of the global anthropogenic emissions [15]. Therefore, particularly in China, i.e. the largest economy and country in Asia, the FW problem is expected to increase severely in the coming years [16,17]. A correct approach to FW management, i.e. source separate collections to guarantee high quality material, represents a priority for China (and other countries in

Asia) to avoid the problems connected with FW disposal and to allow the recovery of this precious resource in terms of both energy content and material (nutrient and organic matter contents).

The chemical characteristics of FW, i.e. macromolecular composition (e.g. sugar, proteins etc.), total solid and total organic matter contents, and the presence of inorganic and/or organic pollutants, makes it more or less possible to convert the organic matter into biogas through anaerobic digestion (AD). Anaerobic digestion is a biological process conducted in the absence of oxygen, leading to organic matter degradation and stabilization. The overall anaerobic conversion process is described as a four-step process: hydrolysis, acidogenesis, acetogenesis and methanogenesis [18], which may occur simultaneously in a single stage process or separately in a multi-stage process. A fine balance between the microbial communities operating these steps, as well as the nutrients provided, is necessary for a successful digestion process which will give the highest conversion rate of organic material to methane [19]. However, the challenges to obtaining high efficiency and stability in performing the anaerobic digestion of FW are currently being debated. In fact, many causes can negatively affect the biological process, e.g. high labile organic matter, salt, oil and protein content, low C/N ratio and micronutrients deficiency.

Theoretically, one tonne of FW could be converted into 847 kWh of electricity and 89.78 GJ of heating potential [20]. To assess such matters in China, demonstration projects for FW disposal were initiated in 2010 and since then 100 cities have been chosen as pilot cities and more than 90% of those have adopted AD technology for FW disposal and energy production [21]. Among several biological valorisation methods, AD technology was demonstrated to be one of the most advantageous technologies to maximise the energy recovery from FW [22].

Anaerobic digestion of food waste has been extensively reviewed in the past and many reviews have appeared in this journal (e.g. by Zhang in 2014 and by Xu in 2018) [18,23]. Very good work presented AD of food waste taking into consideration above all biological processes and process parameters, giving less importance to other topics such as pre-treatment (e.g. Zhang in 2014, and Jain in 2015) [18,24]. Other reviews tried to consider AD of food waste from different points of view, doing very good work in terms of the logical approach used but failing on data collection with reference to food waste, i.e. data were mainly referring to co-digestion with other substrates (e.g. Tyagi and colleagues, in 2018) [25]. Again, some authors reviews focused on the separation of AD phases to enhance performance [26]. Other authors focused the attention more on political, environmental and social aspects [27]. Many other reviews represented tentatives in considering more extensively the AD of food waste but with limited data reported (e.g. Paritosh in 2017, and Xu in 2018) [23,28].

The aim of this review is to fill the gaps by proposing to cover the state of the art of AD of food waste in China and Asiatic countries in a complete manner in order to give a complete and logical picture of the potential for recovering food waste to produce renewable energy. To do so, this review starts by reviewing why food waste can be a useful substrate in producing biogas in substitution for energy crops, i.e. chemical composition and biogas potential productivity, moving then to describe the real biogas productivity, taking into consideration, process parameters and the AD approach (one stage vs. two stage). How to enhance biogas production with co-digestion and/or by applying biomass pretreatments ends the review, proposing, also, a raw energetic approach. This review offers a full picture of the potential of biogas production from recovered food waste that can be useful not only for academics but, also, for decision makers to guide them in planning future waste management policies and the energy transition to renewable energy in a sustainable manner.

International literature has been critically studied to provide a comprehensive understanding of the AD of food waste, with the awareness that this biotechnology could represent a great potential in safely recovering FW, reducing its environmental impact and contributing to renewable energy production (Figure 1).

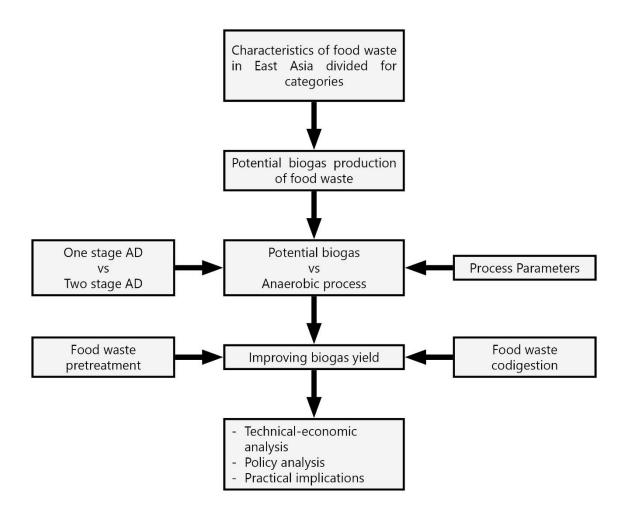


Figure 1. Flow chart summarizing the structure of the work

#### 2. Food waste characteristics and composition

Globally, around 2 billion tonnes of municipal solid waste (MSW) are produced annually throughout the world, with 32-56% [29,30] of MSW being organic biodegradable waste. Food wastes are mainly collected from households and restaurants and the composition may vary depending upon the country [31]. Furthermore, the composition of food waste also varies depending on the time of year, cultural habits, region etc. as well as climate and economic level of the region/country in question [32]. Generally, taking into consideration that moisture plus total solids (TS) contents represents 100 % of wet weight (wet basis) (wb), FW moisture content is of 70-80% wb and TS are around 20-30% wb, 90% of which are volatile solids (VS) [18]. FW is composed mostly of easily degradable carbohydrates (50-60% TS), proteins (15-25% TS) and lipids (13-30% TS) and it has a low C/N ratio [33]. It is also rich in macro-elements but lacks trace elements [34–36]. Moreover, FW is generally an acid or sub-acid substrate, which is suitable for biodegradation but sub-optimal for methanogens that operate at slightly higher pH (6.5-7.2) [37]. Table 1 displays the results of physical and chemical analysis from the literature and for different food waste sources. Moreover, characteristics of FW in East Asia were compared to the characteristics of FW in western countries. Statistical data underlined that the composition was comparable between countries, although the lack of data for carbohydrates, proteins and lipids did not allow a deeper analysis of the data sets. Within different FW categories, statistical analyses underlined similar average data for TS and VS, probably because of high variability in moisture content of waste leading to high standard deviations (Table 1). On the other hand, when VS were referred to TS content (%TS), statistical differences appeared indicating that organic wastes from canteens and restaurants showed higher VS content than those coming from domestic kitchens, but in line with those of synthetic organic wastes and with data reported for western countries. These results could indicate the presence in the KFW of inert substances or impurities lowering VS. Also interesting was the higher O content for SFW with respect other categories, which seems to indicate that from a chemical point of view SFW did not well represent real organic wastes, although the limited number of data available do not allow us to speculate on these figures.

Several previous studies have already attempted to characterize the composition of FW [38–40]. However, the methods used for such classifications are very different and the data are difficult to compare [41]. Thus, the lack of international standardization for FW classification is currently a serious barrier for waste management research. In the following chapters, FW have been classified, taking into consideration their origin and the same classification was used throughout the review. To do so, canteen food waste (CW), restaurant food waste (RFW), kitchen food waste (KFW) and synthetic food waste (SFW) categories have been considered because composition can be different due to different sources.

Chinese household FW is mostly made up of vegetables (54% wb), rice (13% wb), pork, legumes and fruit (13% wb), and other items such as wheat, beverages, fish and dairy products (the remaining 20%).

Restaurant Food Waste in China is estimated to be of 0.15 kg day<sup>-1</sup> per capita in urban areas [42], and a survey in 2011 showed that 28.3% of canteen food ended in rubbish bins in Chinese campuses [43]. Since FW generation depends a lot on consumption, it is possible to affirm that FW depends on the diet, which may vary between rural and urban environments [44]. Furthermore, very few studies used KFW in East Asian countries (only 5 papers out of 34 in this study) (Table 1), whereas KFW was more commonly included in western countries (10 papers out of 15 in this study) (Table 1). However, RFW or CFW was much more common in East Asia (21 papers out of 34 in this study). In this paper, canteen food waste is considered as being part of the group of RFW.

Studies considered showed that RFW ranges from 10% to 17% of the amount ordered by consumers (on a wet basis), and that lipids contained in FW from East Asia averaged  $17.9 \pm 5.8$  % of TS. Even though the BMP (Biochemical Methane Potential) of lipids is twice as high as that of starch, cellulose and hemicellulose [45], it is possible that lipid-rich wastes will produce low methane yields, due to an inhibitory effect on methanogens [46,47]. Furthermore, RFW presents a higher proportion of

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cooked and starch-rich material than other FWs, resulting in a higher hydrogen yield (+6% of carbohydrate removal) [48]. Lastly, a study of 2015 conducted by Fisgativa and Tremier, demonstrated that there were no significant statistical differences between restaurant FW and household FW [37]. The study further stated that the variability of FW cannot be fully explained by its origin. Thus, further research is needed to investigate the factors affecting FW biodegradability. On the other hand, it should be noted that the preparation of synthetic food waste (SFW) can be beneficial in order to reduce experimental bias caused by FW composition [49]. Many studies prepared SFW by following the FW composition (e.g. carbohydrates, protein, fat etc.) indicated in previous research independently of the countries concerned [50–54], while others based SFW composition on FW composition analysis for the particular country considered, either using the National Food guide or standard formulations. In studies from Asian countries, SFW was composed to be as representative of the traditional cuisine as possible. For example some based SFW on locally produced FW (Thailand), while others used typical ingredients from a Chinese market or from a typical Korean dish [9,55–57].

As concluded by this literature review, FW and its characterization are highly variable, which makes comparisons between studies difficult. Such obstacles could be partially overcome by using a formula for Synthetic Food Waste which could be standardized for its macromolecular composition so as to facilitate the comparisons between different studies.

Туре	TS	VS	VS	TI	С	Н	0	Ν	C/N	Carbohydrates	Proteins	Lipids	Country	Reference
of FW	(% wb)	(% wb)	(%TS)	рН		(%	TS)			(*	%TS)			
CFW	29.32	26	88.8 <sup>a</sup>	N/A	50.5	N/A	N/A	2.8	17.8	N/A	N/A	N/A	China	[58]
CFW	24.13	22.6	93.6	N/A	N/A	N/A	N/A	N/A	N/A	N/A	21	17.5	China	[59]
CFW	25.95	24.5	94.3	N/A	N/A	N/A	N/A	N/A	N/A	N/A	18.7	19.1	China	[59]
CFW	23.70	23.1	97.5	6.6	44.1	N/A	N/A	2.5	17.2	44.9	14	14.1	China	[60]
CFW	28.41	26.5	93.1 ª	6.4	N/A	N/A	N/A	N/A	14.7	53.4	19.9	19.9	China	[61]
CFW	25.94	24.6	94.8 <sup>a</sup>	N/A	51.1	7.4	37	3.4	17.5	48 <sup>a</sup>	15.1	10.6	China	[62]
CFW	25.7	24.1	93.8	5.3	45.8	N/A	N/A	2.4	18.8	46.5	13.2	17.2	China	[63]
RFW	7.63	6.1	79.7	N/A	45.5	6.9	25.24	2.1	22.1	N/A	N/A	N/A	China	[64]
CFW	32.98	31.7	96	5.3	N/A	N/A	N/A	N/A	21.5	N/A	N/A	N/A	Singapore	[65]
RFW	22.1	20.4	92.2	5.9	50.8	7.2	32	1.8	28.2	41.6	15.5	31.8	China	[65]
CFW	18.4	17.5 <sup>a</sup>	95.2	N/A	N/A	N/A	N/A	N/A	16.7	N/A	N/A	N/A	China	[66]
CFW	22.4	21.9 <sup>a</sup>	98.1	N/A	N/A	N/A	N/A	N/A	16.7	N/A	N/A	N/A	China	[66]
CFW	7.62	7.2	94.6 <sup>a</sup>	3.6	N/A	N/A	N/A	N/A	N/A	33.4 <sup>a</sup>	14.9 <sup>a</sup>	13.5 <sup>a</sup>	Japan	[67]
CFW	20.00	19.5	97.5 <sup>a</sup>	N/A	49.8	N/A	N/A	3.6	N/A	42.6	22.1	17.1	China	[68]
CFW	21.2	19.6 <sup>a</sup>	92.8	4.7	N/A	N/A	N/A	13.4	N/A	N/A	N/A	N/A	China	[69]
CFW	17.20	16.7	95.6	4.1	50.0	21.5	N/A	2.8	17.8	N/A	N/A	N/A	China	[70]
CFW	18.5	17	91.9	5.2	46.5	N/A	N/A	2.2	21.1	N/A	N/A	N/A	China	[71]
CFW	23.1	21	90.9 <sup>a</sup>	4.2	56.3	N/A	N/A	2.3	24.5	N/A	N/A	N/A	China	[72]
CFW	18.1	17.1	94	N/A	N/A	N/A	N/A	N/A	13.2	111.7	32.9	23.3	Korea	[73]
CFW	18.1	17.1	94	6.5	46.7	6.4	36.4	3.5	13.2	61.7 <sup>a</sup>	18.2 <sup>a</sup>	12.9 ª	Korea	[36]
CFW	20.5	19.5	95	N/A	51.4	6.1	38.9	3.5	14.7	N/A	N/A	N/A	Korea	[74]
<b>RFW</b> and														
CFW	21.5±6.	20.1±5.	93.5±3.	5.3±1	49±3.4a	9.2±6	33.9±5.	3.6±3°	18.5±4a	53.7±23	18.7±5.6	17.9±	_	_
Mean ± Std	<b>1</b> a	9a	8b	3 <b>.</b> 3±1	ч)±3.ча	а	5a	5.0±5	10.J± <b>-</b> a	55,1±45	10.7±3.0	5.8	_	_
(n=21)				/ -										
KFW	25.7	N/A	N/A	N/A	47.2	7	34.8	2.3	20.5	N/A	4.5	8.3	Japan	[75]
KFW	12.5	9.6	79.4 <sup>a</sup>	5.86	41.1	N/A	N/A	2.05	20	N/A	N/A	N/A	China	[76]
KFW	26	22.7	86.3	N/A	52.9	7.9	22.5	2.6	20.3	31.6	16	35.5	China	[77]
KFW	22.6	17.9	79.1	N/A	30.2	N/A	N/A	2.6	11.5	41.9	14.7	28.8	China	[78]
KFW	12.4	11	89	N/A	47.8	6.1	40.9	5.2	9.2	N/A	N/A	N/A	Korea	[79]
KFW Mean ±Std (n=5)	19.8±6. 9a	15.3±6. 1a	83.4±5a	5.8	43.8±8. 7a	7±0.9 a	25±17.1 °	3.2±1. 3°	16.3±5. 5a	36.7±7.2	11.7±6.3	24.2 ±14.2	-	-
SFW	18.3	17	92.9	3.67	46.3	4.21	38.09	3.62	12.8	65.3	16.4	11.2	China	[80]
	17.1	16.9	98.6	5.87	44.9	6.17	44.06	3.01	14.9	76.6	18.2	5.2	China	[58]
SFW	1/.1	10.7	20.0	5.07										

SFW	14.9 <sup>a</sup>	N/A	92.8 <sup>a</sup>	N/A	N/A	N/A	N/A	N/A	13.9	N/A	N/A	N/A	China	[56]
SFW	40	39.2 <sup>a</sup>	98	N/A	45.9	N/A	N/A	N/A	N/A	74	18	10	China	[82]
SFW	14.9	14.4	96.5	5	45.2	12.3	N/A	3.8	12.1	N/A	N/A	N/A	China	[70]
SFW	10.3	9.2	92.3	3.77	45	N/A	N/A	3.2	N/A	N/A	N/A	N/A	Japan	[83]
SFW	14.3	13.1	91.6	N/A	47.4	6.6	43.7	1.9	24.9	N/A	N/A	N/A	Japan	[84]
SFW Mean ± Std (n=8)	17.6±9. 5a	17.1±10 .2a	94.6±2. 7b	4.5±0. 9	45.8±0. 9a	7.3±3. 5a	42±3.3b	3.1±0. 7°	15.7±5. 2a	73.7±6	20.7±36.5	9.7±9. 2	-	-
(11-0)														
$\frac{(n-6)}{\text{All data}}$ $\frac{\text{Mean} \pm \text{Std}}{(n=34)}$	20.3±7 a	18.9±6. 9a	92.5±4. 9b	5±1	47±4.9a	8.1±4. 2a	33±11.2 °	3.4±2. 3°	17.5±4. 5a	56.8±20.7	18±6	17.1± 8	East Asia	(all the above)

2 <sup>a</sup>Calculated according to the data available in the paper.

**3** <sup>b</sup>Data from:[29,73,93,85–92].

4 <sup>C</sup>only two data points were available.

5 <sup>a</sup>Values followed by the same letter are not statistically different for p<0.05:

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7 **3.** Biochemical Methane Potential

Biochemical Methane Potential is used for assessing about the potential producible biogas of a
biomass performed under standardized conditions. It represents the maximum producible biogas
independently of the conditions adopted in full-scale applications.

Different approaches have been proposed in the past to measure BMP. Data for food waste categories 11 reported in Table 2 for this work have been carried out by using an Automatic Methane Potential Test 12 System II (AMPTS II) (Bioprocess Control AB, Sweden) [56,80,94], a lab test, "Anaerobic Lab Work 13 14 1992 and one of the most adopted methodologies [56,95–98]. Other data have been obtained applying 15 the above-mentioned principles with some modifications [87]. Independently of the method adopted, 16 all the tests are quite similar, so that the results acquired can be compared. Tests are generally performed in batch form under mesophilic conditions (35-37 °C) using an inoculum in order to avoid 17 inhibition due to VFA accumulation and pH dropping. Hydraulic Retention Time (HRT) adopted 18 varied ranging from 12 to 65 d. BMP data showed some variability, generally because of different 19 food waste composition. Different potentials are obtained from a cluster of substrates (SFW). For 20 21 instance, differences reported for BMP (Table 2) are due to the differences in terms of crude fat content in wastes [99]. Also, the composition in terms of carbohydrates, protein, and lipids and their 22 relative ratios within the investigated FW was directly correlated to the ultimate BMP. Congruently 23 24 to different ratios, the potential of the investigated SFW ranged between a minimum, i.e. 440 L<sub>CH4</sub> kg<sup>-1</sup> VS and maximum 628 L<sub>CH4</sub> kg<sup>-1</sup> VS [21]. Meanwhile BMP data ranging between 435 L<sub>CH4</sub> kg<sup>-1</sup> 25 VS and 684 L<sub>CH4</sub> kg<sup>-1</sup> VS depended on the different origins (canteens) of the substrate [80], while the 26 range between 116  $L_{CH4}$  kg<sup>-1</sup> VS and 435  $L_{CH4}$  kg<sup>-1</sup> VS reported by Kawai and colleagues in 2014 [100] 27 was due to the effect of the labile organic fraction in food waste and to the different 28 29 substrate/inoculum (S/I) applied.

The potential producible biomethane of FW produced in China and South-East Asia is characterised
by a wide range between the minimum and maximum values (116-684 L<sub>CH4</sub> kg<sup>-1</sup> VS), while BMP

from European FW has a narrower range (425-529 L<sub>CH4</sub> kg<sup>-1</sup> VS). These differences can be linked to
 the compositional differences reported in Table 1.

Different food waste types, i.e. SFW, KFW and CFW did not show statistically different average 34 35 values (Table 2), because of the very high standard deviations characterizing each waste class. Total bio-methane production ranged from 461 L<sub>CH4</sub> kg<sup>-1</sup> VS to 517 L<sub>CH4</sub> kg<sup>-1</sup> VS and so was very similar 36 for each of the classes, with a Grand Mean of  $480 \pm 88$  (*n*=42) L<sub>CH4</sub> kg<sup>-1</sup> VS (Table 2). These data are 37 in line with those reported for FW coming from western countries, i.e. 479±43 L<sub>CH4</sub> kg<sup>-1</sup> VS (Table 38 39 2). Interestingly, this value was much higher than those calculated for energy crops, i.e. 237±58 L<sub>CH4</sub> kg<sup>-1</sup> VS. This indication is very important in view of proposals to use food waste as a substitute for 40 41 energy crops in producing biogas without reducing biogas plant productivity [101]. Energy crop (EC) substitution, in fact, is important in saving not only soils producing food but also in lowering the total 42 costs in producing renewable energy because of crop production costs. 43

44	Table 2. Biochemical Methane Potential	BMP	) data from literature for food wastes from east Asia
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Substrate	Reactor	Vol (L)	Temp (°C)	HRT (d)	Organic Loading Rate; Substrate to inoculum ratio (S/I)	VS Removal (%)	BMP (L <sub>CH4</sub> kg <sup>-1</sup> VS)	Country	Reference
SFW	Batch	0.5	37	30	S/I = 0.5	67 <sup>a</sup>	440	China	[102]
SFW	Batch	0.5	37	30	S/I = 0.5	67 <sup>a</sup>	531	China	[102]
SFW	Batch	0.5	37	30	S/I = 0.5	67 <sup>a</sup>	536	China	[102]
SFW	Batch	0.5	37	30	S/I = 0.5	67 <sup>a</sup>	628	China	[102]
SFW	Batch	0.5	36	12	15 g VS L <sup>-1</sup> d <sup>-1</sup>	89 <sup>a</sup>	407	China	[58]
SFW	Batch	0.5	36	12	5 g VS L <sup>-1</sup> d <sup>-1</sup>	89 <sup>a</sup>	443	China	[58]
SFW	Batch	0.25	37	65	10 gVS L <sup>-1</sup> ; S/I= 0.5	90	499	China	[99]
SFW	Batch	0.25	37	65	10 gVS L <sup>-1</sup> ; S/I= 0.5	63	508	China	[99]
SFW	Batch	0.25	37	65	8 gVS $L^{-1}$ ; S/I= 0.5	68	524	China	[99]
SFW	Batch	0.25	37	65	8 gVS $L^{-1}$ ; S/I= 0.5	91	531	China	[99]
SFW	Batch	0.25	37	65	6 gVS $L^{-1}$ ; S/I= 0.5	89	570	China	[99]
SFW	Batch	0.25	37	65	6 gVS $L^{-1}$ ; S/I= 0.5	91	580	China	[99]
SFW	Batch	0.25	37	65	4 gVS $L^{-1}$ ; S/I= 0.5	89	606	China	[99]
SFW	Batch	0.25	37	65	4 gVS $L^{-1}$ ; S/I= 0.5	92	630	China	[99]
SFW	Batch	0.5	35	40	S/I = 0.2	N/A	489	Korea	[103]
SFW	Batch	N/A	37	25	2 gVS L <sup>-1</sup>	N/A	472	Korea	[97]
SFW	Batch	2	37	45	Seeded sludge; 9.8 g VS FW	N/A	116	Japan	[100]
SFW	Batch	2	37	45	Seeded sludge; 9.8 g VS FW	N/A	214	Japan	[100]
SFW	Batch	2	37	45	Seeded sludge; 9.8 g VS FW	N/A	257	Japan	[100]
SFW	Batch	2	37	45	Seeded sludge; 9.8 g VS FW	N/A	269	Japan	[100]
SFW	Batch	2	37	45	Seeded sludge; 9.8 g VS FW	N/A	435	Japan	[100]
Mean ± SD							461.2±139.7a <sup>a</sup> (n=21)		
KFW	Batch	1	37	30	3 g VS L <sup>-1</sup> ; S/I =0.5	87	541	China	[77]
KFW	Batch	0.25	35	N/A	150 mL inocula; S/I=1:5	88	568	China	[104]
KFW	Batch	0.4	41	45	9.2 g of substrate; S/I =1:1.4		479 (TS)	China	[105]
KFW	Batch	1	35		10 g VS L <sup>-1</sup>	68	313	China	[106]
Mean ± SD							474±140.1a (n=4)		
CFW	Batch	N/A	37	30	S/I= 0.5	N/A	684	China	[58]
CFW	Batch	N/A	37	30	S/I = 0.5	N/A	605	China	[58]

ODW	D (1	NT / A	27	20		<b>NT / A</b>	506	<b>C1</b> :	[20]
CFW CFW	Batch	N/A	37 35	30 27.5	S/I= 0.5 8 g VS L <sup>-1</sup>	N/A N/A	586 581	China China	[58]
	Batch	1							[71]
CFW	Batch	0.5	35	20	10 g VS L <sup>-1</sup>	N/A	560	China	[107]
CFW	Batch	0.25	37	16	1 g VS	N/A	531	China	[108]
CFW	Batch	N/A	37	30	S/I= 0.5	N/A	515	China	[58]
CFW	Batch	0.25	35	N/A	S/I=1:4	N/A	507	China	[109]
FW	Batch	0.15	35	50	20 g VS L <sup>-1</sup> , food-to-microorganism ratio = 1	N/A	497	China	[110]
CFW	Batch	N/A	37	30	S/I= 0.5	N/A	435	China	[58]
FW	Batch	0.15	35	50	20 g VS L <sup>-1</sup> , food-to-microorganism ratio = 1	N/A	435	China	[110]
RFW	Batch	0.5	37	30	10 g VS L <sup>-1</sup>		668	China	[111]
CFW	Batch	1	35	8	600 ml sludge and 12 g VS food waste	N/A	260	China	[112]
RFW	Batch	1	35	N/A	500 mL of inoculum and 15 g of FW	54	268	China	[99]
CFW	Batch	0.16	35	35	35 mL of anaerobic sludge, 30 mL culture medium and 200 mg solid FW	N/A	610	Singapore	[113]
CFW	Batch	0.12	35	40	I/S = 2.0 (VS-basis)	N/A	576	Japan	[114]
RFW	N/A	N/A	N/A	N/A	N/A	N/A	479	Singapore	[36]
FW	Batch	0.5	35	30	17.7 g L <sup>-1</sup>	N/A	410*	Singapore	[115]
Mean ± SD (FW as a subgroup)							517 ± 119a (n=18)		
Gran Mean ± SD (SFW, KFW, RFW, CFW, and generic FW)							480 ± 88C (n=42)		
					FW FROM OTHER COUNTRIES				
FW	Batch	0.4	37	N/A	S/I = 0.5	N/A	529	Ireland	[86]
FW	Batch	0.4	37	35	S/I=1	N/A	501	UK	[91]
FW	Batch	0.4	37	30	S/I = 0.5	N/A	493	Ireland	[94]
RFW	Batch	1	50	28	10.5 gVS L <sup>-1</sup>	N/A	445	USA	[93]
RFW	Batch	1	50	28	6.8 gVS L <sup>-1</sup>	N/A	425	USA	[93]
Mean ± SD							479 ±43C (n=5)		

#### **BMP FROM OTHER SUBSTRATES**

					ENERGY CROPS and GREEN RESID	UES			
Switchgrass	Batch	1	37	30	3 g VS L-1 S/I =0.5	87	246	China	[77]
Straw	Batch	1	35	N/A	600 ml sludge + 12 gVS straw	N/A	281	China	[112]
Corn stover	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	241	China	[77]
Rice straw	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	281	China	[77]
Wheat straw	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	245	China	[77]
Yard waste	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	183	China	[77]
Mean ± SD							246±36A (n=6)		
					FOOD INDUSTRY WASTE				
Cheese whey	Batch	0.25	35	38	S/I = 1	N/A	424	USA	[116]
Ice Cream	Batch	0.25	35	30	S/I = 1	N/A	502	USA	[116]
Used vegetable oil	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	811	China	[77]
Used vegetable oil	Batch	0.25	35	40	S/I = 1	N/A	648	USA	[116]
Fresh Dog food	Batch	0.25	35	30	S/I = 1	N/A	427	USA	[116]
Dog food	Batch	1	35	13	Substrate+ inoculum and tap water to working volume 750 mL	87	652	China	[117]
Mean ± SD							577 ± 152D (n=6)		
Fruit and vegetable waste	Batch	0.5	35	20	10 g VS L <sup>-1</sup>	N/A	300	China	[107]
Fruit and vegetable waste	Batch	1	37	30	3 g VS L <sup>-1</sup> ; S/I =0.5	87	342	China	[77]
Fruit and vegetable waste	Batch	0.25	37	16	1 g VS	N/A	443	China	[108]
Mean ± SD							362 ±73b (n=3)		
					MANURE				
Dairy manure	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	87	51	China	[77]
Caw manure	Batch	0.25	37	16	1 g VS	N/A	182	China	[108]
Chicken manure	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	N/A	295	China	[77]
Pig manure	Batch	0.25	37	16	1 g VS	N/A	385	China	[108]

Swine manure	Batch	1	37	30	3 g VS L <sup>-1</sup> S/I =0.5	N/A	322	China	[77]
Brown Water	Batch	0.5	35	30	17.7 g L <sup>-1</sup>	N/A	280	Singapore	[115]
Mean ± SD							253 ±119A		
Mean ± 5D							( <b>n=6</b> )		
<sup>a</sup> Values followed by	the same la	ttor are no	t statistica	11 different	for n<0.05; lowercase latters ind	icoto difforancas	hotwoon SEW	KEW and CEW: 11	processo lattors indicato

45 <sup>a</sup>Values followed by the same letter are not statistically different for p<0.05: lowercase letters indicate differences between SFW, KFW and CFW; uppercase letters indicate

46 differences between different biomass typologies.

# 47 **4.** Biomethane production from food waste

### 48 *4.1 Single-Stage Anaerobic Digestion*

49 Single-stage AD systems have relatively simple designs; to date this process is thought to be the 50 easiest to build and operate. About 90% of full-scale plants rely on single-stage systems for the 51 anaerobic digestion of organic waste [118].

In the single stage process, the maximum achievable Organic Loading Rate (OLR) is highly 52 dependent on reactor configuration [119]; the OLR of 5 g VS L<sup>-1</sup>d<sup>-1</sup> was a typical value of plants 53 relying on wet systems [120], for which the OLR seldom exceeded 10 g VS L<sup>-1</sup> d<sup>-1</sup> [17,121,122]. An 54 upper limit for OLR seems to exist around 15 g VS L<sup>-1</sup> d<sup>-1</sup>, but the achievable OLR can be greatly 55 affected by the overall digestibility of the waste [119]. Operational stability and efficiency, when 56 OLR is lower than the upper limit mentioned, are widely reported in the literature. For instance, a 57 high methane yield of 455 L kg<sup>-1</sup> VS, in a single wet CSTR operating at 37°C for 187 days with 92.2% 58 of VS reduction was obtained at an OLR of 9.2 g VS (15.0 kg COD) L<sup>-1</sup> day<sup>-1</sup> by Nagao and 59 colleagues in 2012 [83]. Congruently 7 g VS L<sup>-1</sup> d<sup>-1</sup> was the optimal OLR to apply in a semi-60 61 continuous CSTR fed with FW from student canteens run in a dual solid-liquid system [71].

The initial loading of the process can be provided as either OLR or substrate and inoculum concentration, the resulting mean value calculated is the average of those experiments providing the specific OLR. It was found that, when FW, as a category, is anaerobically digested by a mesophilic single stage process, the OLR of  $7.7 \pm 6.4$  g VS L<sup>-1</sup> d<sup>-1</sup> (*n*=10) corresponded to the mean methane yield of 414 ± 31 LCH<sub>4</sub> kg<sup>-1</sup> VS (*n*=10) (Table 3). Meanwhile, when the substrate is SFW and the OLR of  $7.7 \pm 3.5$  g VS L<sup>-1</sup> d<sup>-1</sup> (*n*=5), the methane yield corresponded to  $434 \pm 15.8$  L<sub>CH4</sub> kg<sup>-1</sup> VS (*n*=5) (Table 3), these data being very similar.

HRT varies accordingly to the substrate's composition: an HRT of 10-15 up to 30 days is considered
a typical range of values for a main single wet anaerobic process [119] (Table 3). As shown in Table
3, the mesophilic single stage process applied to the category FW, provided an average methane yield

of  $375\pm107 L_{CH4} kg^{-1}VS$ , with the corresponding average HRT of 30 d when the treated substrate is 72 KFW, not so far from the bio-methane data calculated, as average, for the other categories of food 73 wastes (Table 3), i.e. 390±76 L<sub>CH4</sub> kg<sup>-1</sup> VS. Synthetic food wastes (SFW) showed higher data, i.e. 74 442±21 L<sub>CH4</sub> kg<sup>-1</sup>VS, than those measured for "real food wastes" although there was no statistical 75 difference between the different categories (Table 3). It must be noted that the ultimate methane yield 76 77 fluctuates in ranges with different widths depending on the substrate used. The widest range between 78 the minimum and the maximum methane yield pertains to KFW, which fluctuates between 232 L<sub>CH4</sub> kg<sup>-1</sup>VS and 591 L<sub>CH4</sub> kg<sup>-1</sup>VS, while FW's is between 250 L<sub>CH4</sub> kg<sup>-1</sup>VS and 551 L<sub>CH4</sub> kg<sup>-1</sup> VS. SFW 79 80 apparently seems to have a reduced variability, as its methane yields fluctuate in the narrowest of the 81 ranges from 417 L<sub>CH4</sub> kg<sup>-1</sup>VS to 477 L<sub>CH4</sub> kg<sup>-1</sup>VS due to the origin of "ad hoc made" waste. With reference to the KFW data, it is interesting to underline that thermophilic conditions allowed higher 82 performances than mesophilic ones, i.e.  $473 \pm 74 L_{CH4} \text{ kg}^{-1} \text{VS}$  (n = 5) vs.  $320 \pm 81 L_{CH4} \text{ kg}^{-1} \text{VS}$  (n = 5) 83 9), respectively (Table 3) (p < 0.05). The fact that mesophilic conditions produce lower methane yield 84 is confirmed by the average data calculated from Table 3 for RFW, CFW and CF groups under 85 mesophilic condition, i.e.  $374 \pm 62 L_{CH4} \text{ kg}^{-1} \text{VS}$  (*n* = 18), that did not differ statistically from the data 86 87 above reported from mesophilic AD of KFW.

Biogas from FW is composed mainly by  $CH_4$  (60% v/v) and  $CO_2$  (40% v/v) [123,124], and it can be directly burned producing electrical energy (EE) or upgraded with no additional cost (upgrading unit cost is equal to engine cost to produce EE) to bio-methane for direct injection in the gas grid or to be used as biofuel [124].

Substrate	Reactor	Vol (L)	T (°C)	HRT (d)	Initial loading and/or ORL	VS Removal (%)	CH4 Yield (L <sub>CH4</sub> kg <sup>-</sup> <sup>1</sup> <sub>VSadded</sub> )	Energy (MJ kg <sup>-</sup> <sup>1</sup> <sub>VSadded</sub> )	Country	Reference
SFW	Single stage Semi -Continuous feeding mode	4	55	30	1.5 L inocula + 1.5 L substrate	83	477	17.10	China	[81]
SFW	Single stage Semi -Continuous feeding mode	4	37	30	1.5 L inocula + 1.5 L substrate	82	461	16.52	China	[81]
SFW	Single stage Semi -Continuous feeding mode	3	37*	16	9.2 gVS L <sup>-1</sup> d <sup>-1</sup>	91.8	455	16.31	Japan	[83]
SFW	Single stage Semi -Continuous feeding mode	3	37*	16	7.4 gVS L <sup>-1</sup> d <sup>-1</sup>	90	444	15.91	Japan	[83]
SFW	Single stage Semi -Continuous feeding mode	3	37*	16	12.9 gVS L <sup>-1</sup> d <sup>-1</sup>	92.5	432	15.48	Japan	[83]
SFW	Single stage CSTR	12	55	30	N/A	N/A	430	15.41	Japan	[52]
SFW	Single stage Semi -Continuous feeding mode	3	37*	16	5.5 gVS L <sup>-1</sup> d <sup>-1</sup>	89	421	15.09	Japan	[83]
SFW	Single stage Semi -Continuous feeding mode	3	37*	16	3.7 gVS L <sup>-1</sup> d <sup>-1</sup>	84.4	417	14.95	Japan	[83]
Mean ± SD (n=8)							442±21a	15.85±0.75		
KFW (dinner)	CSTR in Batch	0.4	55	29	20.12 g VS L <sup>-1</sup>	N/A	591	21.18	China	[125]
KFW (lunch)	CSTR in Batch	0.4	55	29	20.12 g VS L <sup>-1</sup>	N/A	501	17.96	China	[125]
KFW (dinner)	CSTR in Batch	0.4	35	29	20.12 g VS L <sup>-1</sup>	N/A	493	17.67	China	[125]
KFW (lunch)	CSTR in Batch	0.4	55	29	9.54 g VS L <sup>-1</sup>	N/A	420	15.05	China	[125]
KFW (breakfast)	CSTR in Batch	0.4	55	29	20.12 g VS L <sup>-1</sup>	N/A	419	15.02	China	[125]

# **92 Table 3.** Methane production from anaerobic digestion performed in single stage.

KFW (dinner)	CSTR in Batch	0.4	55	29	9.54 g VS L <sup>-1</sup>	N/A	434	15.55	China	[125]
KFW (lunch)	CSTR in Batch	0.4	35	29	20.12 g VS L <sup>-1</sup>	N/A	371	13.30	China	[125]
KFW (breakfast)	CSTR in Batch	0.4	35	29	20.12 g VS L <sup>-1</sup>	N/A	370	13.26	China	[125]
KFW	Laboratory-scale anaerobic batch test	1	35	40	10 g VS L <sup>-1</sup> d <sup>-1</sup>	67.7	313	11.22	China	[106]
KFW (dinner)	CSTR in Batch	0.4	35	29	9.54 g VS L <sup>-1</sup>	N/A	306	10.97	China	[125]
KFW (lunch)	CSTR in Batch	0.4	35	29	9.54 g VS L <sup>-1</sup>	N/A	276	9.89	China	[125]
KFW (Breakfast)	CSTR in Batch	0.4	35	29	9.54 g VS L <sup>-1</sup>	N/A	264	9.46	China	[125]
KFW	Single stage CSTR	N/A	35	25	1.11 g VS L <sup>-1</sup> d <sup>-1</sup>	N/A	257	9.21	China	[99]
KFW (breakfast)	CSTR in Batch	0.4	55	29	9.54 g VS L <sup>-1</sup>	N/A	232	8.32	China	[125]

Mean ± SD (n=14)							375±107a	13.43±3.83		
CFW	Lab scale bottle fed every 24 hours	0.5	55	35	1.5 g VS L <sup>-1</sup> d <sup>-1</sup>	N/A	551	19.75	China	[126]
CFW	Lab scale bottle fed every 24 hours	0.5	55	35	$1 \text{ g VS } L^{-1} d^{-1}$	N/A	513	18.39	China	[126]
FW	Single stage Semi -Continuous feeding mode Helix type mixer	6	35	30	6.4 g VS L <sup>-1</sup> d <sup>-1</sup>	86	465	16.67	China	[69]
RFW	Single stage CSTR	6	35	30	2.4 g VS L <sup>-1</sup> d <sup>-1</sup>	73	448	16.06	Japan	[67]
FW	Single stage Semi -Continuous feeding mode	6	35	20	8.4 g VS L <sup>-1</sup> d <sup>-1</sup>	82	439	15.73	China	[69]
RFW	Single stage Semi -Continuous feeding mode	0.5	37	20	4-4.1 g VS L <sup>-1</sup> d <sup>-1</sup>	80	425	15.23	China	[127]
FW	Single stage Semi -Continuous feeding mode	6	35	16	10.8 g VS L <sup>-1</sup> d <sup>-1</sup>	79	416	14.91	China	[69]

CFW	Single stage Semi -Continuous feeding mode	1	35	30	8 g VS L <sup>-1</sup>	N/A	410	14.69	China	[72]
CFW	Single stage Semi -Continuous feeding mode	1	35	21-28	$7 \text{ g VS } L^{-1} d^{-1}$	N/A	405	14.52	China	[72]
FW	Single stage Semi -Continuous feeding mode	6	35	12	13.6 g VS L <sup>-1</sup> d <sup>-1</sup>	77	405	14.52	China	[69]
RFW	Single stage Semi -Continuous feeding mode	0.5	37	20-40	190 mL seed sludge and 10 mL substrate	75.6	396	14.19	Korea	[36]
CFW	Lab scale bottle fed every 24 hours	0.5	37	35	1.5 g VS L <sup>-1</sup> d <sup>-1</sup>	N/A	387	13.87	China	[126]
FW	Single stage Semi -Continuous feeding mode	6	35	8	21.8 g VS L <sup>-1</sup> d <sup>-1</sup>	74	377	13.51	China	[69]
CFW	Lab scale bottle fed every 24 hours	0.5	37	35	1 g VS L <sup>-1</sup> d <sup>-1</sup>	N/A	370	13.26	China	[126]
CFW	Single stage Semi -Continuous feeding mode	1	35	30	8 g VS L <sup>-1</sup>	N/A	347	12.44	China	[72]
CFW	Batch AD	1	35	50	35 g VS L <sup>-1</sup>	N/A	331	11.86	China	[109]
FW	Single stage Semi -dry batch test	N/A	30	60	N/A	N/A	314	11.25	China	[128]
CFW	Single stage - continuously shaken bottle	1	35	N/A	5 g VS L-1	N/A	281	10.07	China	[112]
CFW	Batch test	1	35	30	10 g VS L <sup>-1</sup>	N/A	277	9.93	China	[72]
CFW	Batch AD	1	35	50	45 g VS L <sup>-1</sup>	N/A	250	8.96	China	[109]
Mean ± SD (n=20)							390±76a	13.99±2.74		

95 The two-stage AD approach has been proposed, reporting benefits in terms of: i. multiple products (H<sub>2</sub> and CH<sub>4</sub>) production; ii. better stabilization of the AD; iii. better regulation of the methanogenic 96 process vs. overload; iv. AD time reduction; v. increase of the total energy produced [32]. All this 97 because the separation of the hydrolysis/fermentation phase from methanogenesis allows better 98 process conditions, and should increase process performance in terms of total energy produced from 99 100 biomass [129]. In particular, it seems that performing the first stage before anaerobic digestion 101 under controlled acidogenic conditions producing H<sub>2</sub>, allowed benefits in enhancing the subsequent 102 methanogenic process.

From the literature, it is not very clear whether, effectively, the two-stage approach allows an increase 103 in total energy produced. Many works demonstrated energy increases (from 20% to 60%) using two-104 105 stage approaches [130–132]. Schievano and colleagues in 2014 reported that two-stage AD recovered 106 8%–43% more energy than one-stage and never significantly less, testing this approach on different biomass types [129]. On the other hand, previous work of ours [133] performed at lab-scale did not 107 108 find any differences between the one- and two-stage approaches. In this review, data from China and 109 some East Asian countries have been collected (Table 4) and discussed for energy efficiency. To do so, the total energy produced in the processes (MJ kg<sup>-1</sup>v<sub>Sadded</sub>) was calculated from H<sub>2</sub> and CH<sub>4</sub> 110 111 produced by the two-stage approach, assuming the lower heating value of hydrogen to be equal to 50 MJ kg<sup>-1</sup> and the lower heating value of bio-methane to be equal to 119.9 MJ kg<sup>-1</sup> [134] (Table 4). The 112 113 results obtained, as averages of different literature data, were then compared to energetic data reported 114 for single stage AD (Table 3). The results obtained indicated that restaurant/canteen wastes produced more energy than other FW groups (p>0.05). This is can be due to different composition of RFW 115 116 with respect the other groups, i.e. higher lipids content (see chapter 2).

117 More interesting was the fact that, on average, there was no significant difference (p>0.05) in total 118 energy produced by two-stage (Table 4) vs. one-stage (Table 3). Therefore, it can be concluded that 119 taking into consideration data from the literature (Table 3 and 4) and making a complete energy

- balance, there were no differences in total energy recovered due to performing anaerobic digestion in
- 121 one vs. two stages, with the average data being as follows:  $14.1 \pm 4.2$  MJ kg<sup>-1</sup> VS<sub>added</sub> (n = 23) and
- 122 15.97  $\pm$  0.7 MJ kg<sup>-1</sup> VS<sub>added</sub> (*n* = 42), respectively.

<b>123 Table 4.</b> Bio-hydrogen and Bio-methane yield from food wa	te.
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		Bio-H <sub>2</sub>	1		Bio-CH	4	Total energy (H <sub>2</sub> + CH <sub>4</sub> )	Experiment	Country	Reference
Type of FW	Yield (mL g <sup>-</sup> <sup>1</sup> VSin)	HRT (d)	Temperature (°C)	Yield (mL g <sup>-</sup> <sup>1</sup> VSin)	HRT (d)	Temperature (°C)	(MJ kg <sup>-1</sup> <sub>VSadded</sub> )			
KFW	not measured	5	35	380 <sup>a</sup>	20	35	13.6	two-stage CSTR	Korea	[135]
KFW	not measured	5	35	440 <sup>a</sup>	20	55	15.8	two-stage CSTR	Korea	[135]
KFW	not measured	5	55	370 <sup>a</sup>	20	37	13.3	two-stage CSTR	Korea	[135]
$KFW^2$	66	2	55	364	10	55	13.7	two-stage CSTR	Japan	[136]
KFW <sup>3</sup>	20	2	55	329	10	55	12	two-stage CSTR	Japan	[136]
$KFW^4$	85	2	55	338	10	55	13	two-stage CSTR	Japan	[136]
KFW							-		*	
Mean ± Std (n=6)	-	-	-	-	-	-	13.5 ± 1.2ab <sup>b</sup>	-	-	-
CFW	125	70	55	526	70	35	20.2	two-stage CSTR	China	[137]
CFW	104.5	5	55	512	30	35	19.5	two-stage CSTR	China	[138]
RFW	34.7	N/A	35	387.9	N/A	35	14.3	two-stage batch	China	[64]
RFW <sup>5</sup>	Undetected	6	55	690	24	35	24.7	two-stage CSTR	Japan	[67]
RFW	121.1	N/A	37	321	N/A	37	12.8	batch	China	[139]
CFW	147.3	2.87	55	383.0	14.4	35	15.3	two-stage CSTR	Japan	[140]
RFW	114	1.9	55	450.6	7.7	55	17.3	batch, then semi- continuous	Japan	[141]
CFW	205	1.3	55	464	5	35	18.8	two-stage CSTR	Japan	[142]
RFW and CFW Mean ± Std (n=8)	-	-	-	-	-	-	$17.8 \pm 3.5b$	-	-	-
SFW	not measured	-	-	477	30	55	17	two-stage CSTR	China	[81]
SFW	69	N/A	N/A	269.7	N/A	N/A	10.4	batch	China	[82]
SFW	292.7	0.5	N/A	391.6	1	N/A	17.2	CSTR	Thailand	[9]
SFW	not measured	-	-	290	16	35	10.4	batch	China	[143]
SFW	not measured	-	-	270	17	35	9.7	batch	China	[144]
SFW	55.1	N/A	37	94.8	N/A	37	4	batch	Thailand	[55]
SFW	not measured	-	-	180	17	35	6.5	batch	Hong Kong	[145]
SFW Mean ± Std (n=7)	-	-	-	-	-	-	10.7 ± 5a	-	-	-

- <sup>124</sup> <sup>a</sup>such values were calculated according to the data available in the paper; 1, Organic Fraction of Municipal Solid Waste was considered as KFW; 2, garbage was considered kitchen FW; 3, okara
- 125 was considered kitchen FW; 4, potato was considered kitchen FW; 5, oily food waste was considered restaurant FW. Numbers followed by the same letter do not show statistically significant
- 126 differences per  $\alpha$ =0.05.
- <sup>b</sup>values followed by the same letter are not statistically different for p<0.05.

# 128 *4.3 Anaerobic Co-digestion of food waste with different substrates.*

The performance of AD is significantly affected by the C/N ratio: an appropriate balance between 129 130 carbon and nitrogen is required for effective digestion [146]. Therefore, an adjustment of the C/N 131 ratio is needed for stable AD in long-term operations. Such an adjustment could be considered as a 132 pre-treatment itself [147]. Tanimu and colleagues found that the activity of methanogens was 133 enhanced during the anaerobic digestion of FW for C/N=30, followed by C/N=26 and lastly C/N=17, with the average biogas yields obtained equal to 1,002, 620 and 479 L kg<sup>-1</sup> VS, respectively [148]. 134 135 Hence, it is possible to improve biogas production from FW by co-digesting it with additional 136 substrates [149], which can optimize the C/N ratio, usually by raising the relative C content. For 137 example, Zhang in a work published in 2013 [71] found the optimal C/N ratio to be 15.8 when codigesting FW with cattle manure (CM). Li and colleagues in 2009 noticed an improvement of 138 methane yield by around 44% when co-digesting FW with CM compared to mono-digestion of FW 139 [150]. In addition, Banks found in 2011 [151] a decrease of greenhouse gas emissions when co-140 141 digesting FW with cattle slurry.

Furthermore, the occasional addition of a third component to co-digestion (FW+CM+oil or FW+CM+fat) may enhance the methane yield [152]. Similar findings were reported by Amha and colleagues in 2017 [153], in which the addition of fat, oil and grease (FOG) positively impacted methane production.

146 Several other substrates have been found to be favourable to the anaerobic co-digestion (AcoD) of 147 FW. For example, Kim and Oh in 2011 [154] mixed FW and livestock waste to reduce VS and improve the yield. Yong and colleagues in 2015 co-digested FW with straw and found the optimal 148 ratio to be FW:straw of 5:1 [112]. The findings of this study were also validated in 2014 by Zhan-149 150 jiang, who found the highest methane yield in the mass ratio of FW: rice straw of 3:1 [155]. Volatile solids reduction was also obtained by adding green waste, which influences the C/N ratio [156]. 151 Furthermore, mixing yard waste (green waste) with FW relieves VFA accumulation, thus improving 152 153 methane yield [86]. It was also found that adding the sludge of wastewater treatment plants to FW

could reduce the negative effects of Na<sup>+</sup> [69]. Optimal ratios of food waste and tall fescue were
observed to be 1.52/1 and 1/1 by Chen and colleagues in 2016 [157]. Biogas production enhancement
was also observed by adding distiller's grain and piggery wastewater into FW [36,158].

157 A recent study used food waste in co-digestion with microalgae (MA), trying several substrate ratios [114]. It was found that increasing the amount of FW can enhance the AD performance, with 158 159 maximum methane yield obtained with the ratio MA:FW of 0.2:0.8, achieving the value of 640 L kg<sup>-</sup> <sup>1</sup>VS<sub>added</sub>, almost 5 times more than microalgae alone. Jang and colleagues in 2015 [159] analysed the 160 161 co-digestion of food wastewater and waste activated sludge (WAS) and found that co-digestion 162 provided higher gas production than WAS alone. In 2016 Abudi and colleagues [160] set up a batch 163 experiment using OFMSW, thickened WAS and rice straw. The three substrates were compared at 164 three different ratios: 1:1.5:1.5, 1:0.5:0.5 and 3:0.5:0.5, respectively. The ratio 3:0.5:0.5, which was 165 characterized by a high fraction of OFMSW, achieved the greatest biogas production, of 558 L kg<sup>-</sup> <sup>1</sup>VS. An overview of substrates used in co-digestion with food waste is shown in Table 5 and Table 166 6. The tables underlined the percentages of increase/decrease in methane yield from FW alone to FW 167 168 co-digested. The values of increases in yield were found to be extremely variable amongst different studies. Furthermore, not all values were positive numbers, indicating that in some cases the co-169 170 digestion of FW with other substrates resulted in a decrease of yield (e.g. a study of 2014, where the 171 decrease was of 16%) [161]. However, not all cases presenting such characteristics resulted in an absolute decrease of yield. For example, Zhang and colleagues in 2013 [71] found that the methane 172 yield of mono-digested FW (mL CH<sub>4</sub>  $g^{-1}VS$ ) was higher than that of FW + cattle manure, but on the 173 contrary, the total methane (mL) was higher in co-digestion rather than in mono-digestion. 174

However, the anaerobic co-digestion (AcoD) of FW with sewage sludge (SS) (Table 6) gives the opportunity of fixing such obstacles by adjusting C/N ratio, mediating hydrolysis and diluting harmful substances [100,162]. It is possible to improve the performance of FW, which is characterized by high solids concentration and high C/N ratio varying from 13.2 to 24.5 [163], with the addition of sludge (SS), which is known to be characterized by a lower C/N ratio (from 6 to 9). Anaerobic co-

180 digestion of FW with SS appeared to be a very promising technology, as it can produce higher methane yields. Indeed, in a work of 2016, Prabhu and Muturi found methane increases in biogas of 181 60% and 73% with the addition of SS to FW; while in 2015 Koch and colleagues found accelerated 182 183 methane production rates [164,165]. Moreover, AcoD of FW with SS is beneficial to process stability, particularly in thermophilic conditions [138]. Anaerobic co-digestion of FW with SS appeared to be 184 185 an advantageous way of managing sewage sludge, especially in China [166] and in South Korea [30] 186 where such matters raise a lot of concern. Given the many benefits of the co-digestion of FW with 187 SS, it is still a matter for investigation to define what the ratio FW:SS should be. A work of 2003 by 188 Kim and colleagues for example [167] found optimal ratios in thermophilic (39.3% FW) and 189 mesophilic (50.1% FW) conditions respectively. Furthermore, in 2004 Kim and colleagues 190 demonstrated that adding SS (13-19%) to FW can enhance potential hydrogen production [74]. 191 Again, in 2014 Kuo-Dahab and colleagues found that by adding 50% of SS to FW total solids, biogas production and stability increased, while VS decreased [168]. Other research also recommended 192 setting the blend ratio of FW:SS to 1:1 for high-solids anaerobic co-digestion, since it showed good 193 194 synergistic effects and increased biogas production [66]. Silvestre and colleagues in 2015 [169] 195 accounted for an increase of 200% methane production rate and 59% methane yield with the addition of 54% FW to the FW+SS mixture. Prabhu and Mutnuri in 2016 indicated that a 1:2 mixture of 196 197 FW:SS was optimal to produce biogas with a methane content of 60% (v/v) [164]. However, as shown in Table 6, this does not necessarily translate into a higher amount of methane: there was in fact a 198 199 21% reduction in methane yield from FW to FW + SS (1:2 mixing ratio). In a work of 2016, Zahan 200 and colleagues also suggested that the presence in the mix of at least 47-48% of FW improved methane yield of SS [170]. However, another work in 2013 found a biogas and VS linear decrease 201 202 with the increase in the presence of FW in the mix during co-digestion [69]. In 2003, Heo and 203 colleagues [57] also reported decreasing methane yield by 85% to 50% when increasing FW from 10% to 50%. Such different conclusions on the matter of the mixture ratio can be attributed to the 204 205 different C/N ratios of FW and SS used in the research cited [66]. Table 6 reports research on the

206	matter of co-digestion of FW with SS in different experimental conditions and relative biogas yields,
207	with particular focus on mixture ratios. Statistical analysis showed that there were no differences for
208	bio-methane production by adding SS in co-digestion with FW but that the very high standard
209	deviations in the work reported did not permit any conclusions to be reached at this stage.

**Table 5.** Performance of anaerobic co-digestion of FW with different co-substrates as available in recent literature

<b>Co-substrates</b>	Mixing ratio	CH4 yield before co-digestion (mL g <sup>-1</sup> VSin)	CH4 yield after co-digestion (mL g <sup>-1</sup> VSin)	Yield increase (%)	Temperature (°C)	Experiment	Country	Reference
FW+cow manure	N/A	54	247	357	55°C	Batch	Malaysia	[171]
FW+microalgae	0.8:0.2	575.7	639.8	11.1	35°C	Batch	Japan	[114]
FW+waste activated sludge	70:30	N/A	254	N/A	35°C	Batch	China	[172]
FW+raw straw	5:1	281	392	39.5	35°C	Batch	China	[112]
FW+green waste	40:60	326.4	272.1	-16.6	mesophilic	24.5 days HRT	China	[161]
FW+pulp and paper sludge	1:1	229.7	432.3	88.2	thermophilic	Batch	China	[173]
FW+cattle manure	2:1	410	388	-5.4	35°C	Batch	China	[71]
FW+rice straw	3.88:1	N/A	559	N/A	35°C	CSTR	China	[157]
FW+landfill leachate	N/A	1.1	466	42,264	35°C	Batch	China	[174]
Mean ± Std	-	$268 \pm 199a^{a}$ (n=8)	406 ± 137b (n=9)	-	-	-		-

<sup>a</sup>values followed by the same letter are not statistically different for p<0.05.

FW: SS	Methane yield of FW before co-digestion mL g <sup>-1</sup> VSin	Methane yield after co-digestion mL g <sup>-1</sup> VSin	Increase of yield (%)	Temperature	HRT/days	Country	Reference
3:1	N/A	336	N/A	30°C	90	India	[175]
50:50	N/A	107	N/A	35°C	N/A	Pakistan	[176]
3:1	N/A	407	N/A	Thermophilic	30-3	China	[102]
1:7	N/A	710	N/A	35°C	20	Hong-Kong	[177]
3:1	271	264	-2.5	35°C	7	China	[178]
1:2	625	490	-21.6	Mesophilic	20	India	[164]
1:1	70.7	297	319.6	37°C	37	China	[162]
3:2	70.7	316	347.2	37°C	37	China	[162]
1:0.4	N/A	352	N/A	35°C	30-8	China	[69]
85:15	322	353	9.8	37°C	N/A	China	[139]
50:50	N/A	321	N/A	35°C	13	Korea	[103]
Mean ± Std	$272 \pm 228a^{a} (n=5)$	359 ± 149a (n=11)	-	-	-		-

214 <sup>a</sup>values followed by the same letter are not statistically different for p<0.05

215

# 216 5. Pre-treatments of food waste to increase methane yield.

Pre-treatment is fundamental to improve biodegradability of the substrates. For example, Song and 217 Zhang, in a work of 2015, demonstrated that pre-treating co-substrates using wet-state H<sub>2</sub>O<sub>2</sub> can 218 219 improve biodegradability and methane yield [179]. The pre-treatment with 3% H<sub>2</sub>O<sub>2</sub> (w/w) allowed 220 an increase in methane yield by 50.3%. With this method, VFAs concentration may also decrease. 221 An alkaline solution of H<sub>2</sub>O<sub>2</sub> can also be used to improve biomass delignification [180]. Lipids 222 removal from food waste can be beneficial to the AD process by giving it more stability, as found by 223 Algapani and colleagues in 2017 [138]. Biological co-treatment (biological solubility pre-treatment) 224 could improve the hydrolysis performance of FW and SS, since alkalis generated by sludge can buffer 225 VFAs and maintain optimum pH for hydrolysis [181]. According to Zhang and colleagues, the 226 increase in biological co-treatment time can improve hydrolysis and acidogenesis, with optimum pre-227 treatment time of 24h [65]. With such treatment, it is possible that the digestibility of the substrates 228 is improved. With this co-pre-treatment, the methane yield of the AD process increased by 24.6% and the VS reduction increased by 10.1%. Microwave pre-treatment at maximum power and 229 230 temperature of 1000 W and 100°C was also found to be favourable to methane production by codigestion of FW with sludge, with the optimized ratio of 3:2= FW: SS [162]. In this case, methane 231 232 production increased by 6.9% compared with the untreated substrates. Other pre-treatment methods 233 that solve the problem of sludge solubility include ultrasound [182], heat and free nitrous acid combined [183] as well as other mechanical, thermal, chemical and biological pre-treatments [184]. 234 235 Such a variety of methods can also be applied to FW [49].

Table 7 reports data referring to the effects of pre-treatment on bio-methane production by using different approaches. A representation of different pre-treatments with a brief explanation for each one was also reported by Parthiba Karthikeyan and colleagues in 2018 [32]. In brief, pre-treatment can be classified as i. physical treatment, able to modify the surface area available for biological reaction; ii. thermal treatments that allow solubilizing macromolecules, the disintegration of cell membranes, the hydrolysis of macromolecules enhancing mass homogenization; iii. chemical treatments able to hydrolyse macromolecules destroying 3D structure of organic matter so that
subsequent hydrolysis becomes easier; iv. biological treatments that acted like chemical treatments
but worked more slowly [32].

245 Pre-treatments showed a general positive effect, enhancing biogas/biomethane production (Table 7). 246 The positive effects varied a lot and depended upon the method and conditions adopted. It is 247 impossible to give any unique indication about what is the best pre-treatment. Based on the data 248 collected in this review, it seems that physical, thermal, chemical and physical-chemical treatments 249 and pre-treatments plus co-digestion gave good results in terms of biomethane produced in 250 comparison with untreated biomass, i.e. on an average: physical + 40±9 % (n = 3), thermal: + 27±22% 251 (n = 3), chemical + chemical/physical: + 42±31% (n = 5), and combined pre-treatment: +34 ±21% (n = 5)= 18). On the other hand, biochemical treatments gave lower effect: + 12.5  $\pm$ 12 % (n = 6) and 252 "Process": + 15  $\pm$ 13% (*n* =5) (Table 7). 253

Pre-treatment allows the production of more bio-methane, increasing the total energy produced, but it requires energy to be performed, which should be lower than the energy gained. Previous reviews and work on the topic did not report specifically on the energetic balance, or reported only a qualitative approach regarding the effect of the pre-treatment (bio-methane production increase). Therefore, in this review a first attempt has been made to perform an energy balance, although the lack of information on pre-treatment energy requirements with particular reference to the organic wastes made it very difficult.

From the literature, it was possible to extract data regarding energy consumption for performing pretreatments, referring to the total solid unit (MJ ton<sup>-1</sup> TS). Therefore, taking into consideration the biomethane added because of the pre-treatment (% of total bio-methane produced without pre-treatment) (Table 7), the average bio-methane production for the organic wastes ( $395 \pm 84 \text{ L kgVS}^{-1}$ ; *n*=42) (Table 3), the average total solid and volatile solid contents of organic wastes ( $20.3 \pm 7$  % wet weight and  $92.5 \pm 4.9$  % TS; *n* = 34) (Table 1), and lower calorific power of methane (119.9 MJ kg<sup>-1</sup>), the total energy gained (MJ ton<sup>-1</sup> TS) has been calculated for some of the pre-treatments listed in the Table 7. The results obtained (on average for each pre-treatment category) were then discussed taking
 into consideration the energy (MJ ton<sup>-1</sup> TS) necessary for the pre-treatment, obtaining the net energy
 produced (on average).

271 Physical pre-treatments increased, on average, the amount of CH<sub>4</sub> produced by  $40 \pm 10\%$  (*n*=3) (Table

272 7) that considering the energy necessary for the pre-treatment i.e. 40 - 162 MJ ton<sup>-1</sup> TS (n=11) [185–

187], gave, on average, a net energy gain of 5,000 MJ ton<sup>-1</sup> TS. Again, heat pre-treatments require

274 144-709 MJ ton<sup>-1</sup> TS (n=6) [187,188], that taking into consideration an increase of CH<sub>4</sub> produced by

275  $30 \pm 16\%$  (*n*=4) gave a net energy gain of 3,500 MJ ton<sup>-1</sup> TS. Chemical pre-treatments instead gave

a bio-methane gain of  $22 \pm 14$  % (*n*=2) requiring an amount of energy of 2,500-4,260 MJ ton<sup>-1</sup> TS (*n* 

- 277 = 2 [189], which means a net energy gain of -500 MJ ton<sup>-1</sup> TS, i.e. in this case pre-treatments were
- 278 not energy-efficient [189].

# **Table 7.** Pretreatments to enhance methane production during anaerobic digestion of food wastes.

Pre-treatment Country		Treatment description and substrate specifications	Reactor Design	Temperature (°C)	Results on process factors	Biogas / H <sub>2</sub> / CH <sub>4</sub> production		Increment (%)	Reference
						Before Process	After Process		
				PHYSICAL					
Shredding	Japan	FW standard. Size reduction by beads mill to mean particle size of 0.718 mm (control 0.888 mm)	Batch	Mesophilic	40% higher COD solubilisation	375 mL g <sup>-1</sup> total COD <sup>-1</sup> cumulative biogas	503 mL g <sup>1</sup> total COD <sup>-1</sup> cumulative biogas	+34%	[190]
High voltage pulse discharge	China	Canteen FW. Optimized discharge conditions: the pulse voltage of 40 kV, the electrode distance of 5 mm, the pulse frequency of 400 Hz, and the pre- treatment time of 30 min.	Batch	35	Higher concentration of SCOD, soluble protein, and soluble sugars than the control, i.e. 107.3%, 171% and 24.8% respectively,	240 mLCH <sub>4</sub> g <sup>-1</sup> COD <sub>removed</sub>	315 mL CH <sub>4</sub> g <sup>-1</sup> COD <sub>removed</sub>	+35%	[191]
Ultrasonic	Korea	Ultrasonic homogenizer used for 30 min at 360 kJL <sup>-1</sup> energy intensity.	Batch	35	Additional reduction: +11.1%COD; + 6.5 VSS; +3.7 %TSS	108 mL cumulative CH4	163mL cumulative CH4	+51%	[192]
				THERMAL/HEA	хт				
Thermal	China	KW; fluid circulating process at constant temperature of 120 °C for 50 min	Two stage	35	Solubility rate of 26.63% on TS basis and of 49.21% on VS basis; 74.92% (v/v) CH <sub>4</sub> concentration in biogas	911 mL g <sup>-</sup> <sup>1</sup> VS total biogas (KFW without pre-	1,200 mL g <sup>-</sup> <sup>1</sup> VS total biogas (KFW with pre-	+31.7%	[193]
Thermal	Korea	FW; heated in an autoclave at 120 °C for 60 min.	Batch	35	+9.9 % COD removal; +7.2 VSS reduction; +5.2% TSS reduction	treatment) 108 mL cumulative CH4	treatment) 160 mL cumulative CH4	+48%	[192]

Hydrothermal	China	Air-dried rice chopped, immersed in the distilled water for 12 h, heated in reactor to 180 °C, with heating rate of 10 °C min <sup>-1</sup> for 15 min.	Batch	35	Iintensified hydrolysis of water- insoluble fractions, cumulative methane yield, + 9.5%, higher at 180°C	160 mL g <sup>-1</sup> TS cumulative CH4	176 mL g <sup>-1</sup> TS cumulative CH4	+10%	[194]
Hydrothermal	China	FW; Stirring speed of 500 rpm at 140°C up to 30 min	II dark fermentation + AD	35	The COD solubilization yield increased from 54.32% to 70.38%	388 mL g <sup>-</sup> <sup>1</sup> VS CH4 yield	512 mL $g^{-1}$ VS CH <sub>4</sub> yield	+32%	[64]
				CHEMICAL					
Alkali treatment	Korea	FW and the WAS; Addition of 0.4 mole $L^{-1}$ NaOH to batch reactor: pH 12.7 after treatment.	Batch	35	+3 %TSS reduction; +0.8 % VSS reduction, +3.4% COD reduction	108 mL cumulative CH4	121 mL cumulative CH4.	+12%	[192]
Alkali treatment	China	Addition of NaOH.	Batch	35	Enhanced buffering capacity of KFW	313.4 mL g <sup>-1</sup> VS cumulative CH <sub>4</sub>	458.4 mL g <sup>-1</sup> VS cumulative CH4	+32%	[150]
			P	HYSICAL+CHEMI	ICAL				
Alkali-thermal	N/A	+ NaOH 0.2 mole $L^{-1}$ ; heat up 170 °C for 60 min (autoclave).	N/A	N/A	COD removal for FW of 49.8% and 51.9% for FW + Waste Activated Sludge	82 mL g <sup>-1</sup> VS	164 mL g <sup>-1</sup> VS.	+100%	[195]
Alkali Thermal treatment	Korea	NaOH added to the reactor. The feeds were heated up in an autoclave at $120^{\circ}$ C for 30 min.	Batch	35	+10.7% COD removal +3% VSS reduction; TSS reduction +5.6 %TSS removal	108 ml cumulative CH4	159.9 mL cumulative CH4	+48%	[192]
Micro-aeration	China	Between -17 and -67 mV throughout the 4-day micro-aeration pretreatment where the majority of the waste came from Chinese, Indian, Indonesian and Malay food stalls.	Batch	35	10% more VS degradation	385 L <sub>biogas</sub> kg <sup>-1</sup> VS cumulative biogas	456 L <sub>biogas</sub> kg <sup>-1</sup> VS cumulative biogas	+18.4%	[196]

**BIO-CHEMICAL ON FOOD WASTE COMPOSITION** 

Lipid extraction	China	Lipids from food waste extracted using methanol and chloroform.	Batch	55	Decreased in lipid content in FW from 37 g $L^{-1}$ to 26 g $L^{-1}$	426 mL g <sup>-1</sup> VS <sub>added</sub> CH <sub>4</sub> yield	531 mL g <sup>-1</sup> VS <sub>added</sub> CH <sub>4</sub> yield	+25%	[138]
Lipid extraction	China	Lipids from food waste extracted using methanol and chloroform.	Batch	35	Decreased in lipid content in FW from 37 g $L^{-1}$ to 26 g $L^{-1}$	400 mL g <sup>-1</sup> VS <sub>added</sub> CH4 yield	418 mL g <sup>-1</sup> VS <sub>added.</sub> CH4 yield	+5%	[138]
Lypase addition	China	FW; lypase addition	Batch	37	TS and VS reductions 36.6– 48.6% and 24.6–29.1% higher	498 mL g⁻ ¹VS CH₄ yield	500 mL g <sup>-1</sup> VS CH4 yield	+0.4%	[110]
Biological co- pretreatment	China	FW; 500 g FW + 500 g Waste Activated Sludge added into the biological co-pre- treatment reactor, purged with pure nitrogen gas for 30 min. After seeding with 300 ml of inoculum, the reactor was operated at 35°C, for 24 hours.	Batch	35	Acceleration of the solubilization of particulate organic matter and improvement of the hydrolysis rate of FW and Waste Activated Sludge prior to AD	230 mL g <sup>-1</sup> VS	294 mL g <sup>-1</sup> VS.	+28%	[65]
Saccharification	Japan	FW + 50 mL of distilled water; pH adjusted to 5.0 with 5 M HCl; saccharification (Amyloglucosidase) at 60 °C for 6 h; separation of the saccharified liquid from residue.	Single stage Continuous feeding mode	Mesophilic	Prevention of acidification due to the labile organic fraction of the FW and reduction of the volume and operating cost of the AD reactors	252.6 mL g <sup>-1</sup> VS cumulative CH4	248.4 mL g <sup>-</sup> <sup>1</sup> VS cumulative CH4	-1.7%	[67]
Thermophilic digestion	China	Co-digestions mixing ratio of food, wastewater and waste activated Sludge at 75:25.	Single stage Continuous feeding mode	Mesophilic	High methane content (68.24% $v/v$ ) in thermophilic digestion and (65.21% $v/v$ ) in mesophilic digestion	1,233 L CH4 L <sup>-1</sup> d <sup>-1</sup>	1,423 L CH4 L <sup>-1</sup> d <sup>-1</sup>	+15%	[197]

### PROCESS

Recirculation	China	FW; recirculation rate at 0.3.	Temperature Phased AD with digestate Recirculation	55-35	Stable hydrogen and methane production 70% VS reduction	125 $H_2$ yield	135 H <sub>2</sub> yield	+8%	[198]
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Recirculation	Japan	Raw FW + cardinal elements; effluent from stage II of the TPAD-R was recycled to stage I with the same flow rate as the influent of the whole system.	Temperature Phased AD with digestate Recirculation	55-35	Same methane production as single stage; in addition to methane, hydrogen was also produced in stage I of the TPAD-R	$\begin{array}{l} 0 \mbox{ mL } H_2  g^{-1} \\ VS_{added} \end{array}$	$\begin{array}{l} 50 \ mLH_2 \ g^{-1} \\ VS_{added} \end{array}$	Total	[67]
Recirculation	Japan	Raw FW + cardinal elements; effluent from stage II of the TPAD-R was recycled to stage I with the same flow rate as the influent of the whole system.	Temperature Phased AD with digestate Recirculation	55-35	Relief of toxic effect of Long Chain Fatty Acids, higher biogas yield Temperature Phased AD with digestate recirculation system compared to the same process without recirculation	0.69 L g <sup>-1</sup> VS <sub>added</sub> Biogas yield	0.74 L g <sup>-1</sup> VS <sub>added</sub> . Biogas yield	+7%	[67]
Recirculation + heat bacterial inactivation	Japan	Raw FW; methanogenic sludge from a methane reactor inactivated in oven at 100°C for an hour; recirculation ratio approximately 2.9.	Two stage	55-55	Smallest butyrate conversion compared to other phases were at the same level	115 L g <sup>-1</sup> VS <sup>added</sup> Hydrogen yield	147 L g <sup>-1</sup> VS <sup>added</sup> Hydrogen yield	+28%	[140]
Solid /Liquid separation	China	FW; solid and liquid fractions firstly separated by a sieve with 2: 2 mm lattice.	Single stage in Semi- Continuous feeding mode	35	Solid fraction provided higher methane yield compared to the entire original food waste	405 mL g <sup>-1</sup> VS	540 mL g <sup>-1</sup> VS	+33%	[72]
Solid /Liquid separation	China	FW; solid and liquid fractions firstly separated by a sieve with 2: 2 mm lattice.	Single stage Semi - Continuous feeding mode	35	Liquid fraction provided low methane yield compared to the entire original food waste	405 mL g <sup>-1</sup> VS	390 mL g <sup>-1</sup> VS	-4%	[72]
			СОМ	BINED PRETREA	TMENT				
Three stage AD + co- digestion	China	FW and horse manure (wet mass ratio 1:1); first hydrolysis in chamber level 1; acidogenesis in chamber 2; methanogens of hydrolized and acidified mix in chamber 3.	III stage	35	Increased VFAs production; enhanced methanogenic activity subsequent to separated hydrolysis and acidogenesis; enhanced solubilization of the mix	300L CH <sub>4</sub> g <sup>-1</sup> VS (One stage)	370 L CH <sub>4</sub> g <sup>-1</sup> VS (Three stages)	+23%	[65]
Three stage AD + co- digestion	China	FW and horse manure (wet mass ratio 1:1); first hydrolysis in chamber level 1; acidogenesis in chamber 2;	III stage	35	Increased VFAs production; enhanced methanogenic activity subsequent to separated	340 L CH4 g <sup>-</sup> <sup>1</sup> VS (Two stage)	370L CH4 g <sup>-1</sup> VS (Three	+9%	[65]

		methanogenesis of hydrolized and acidified mix in chamber 3.			hydrolysis and acidogenesis; enhanced solubilization of the mix		stages)		
TPAD + Recirculation + co-digestion	Japan	FW+ de-oiled grease trap waste.	Temperature Phased AD with digestate	55-35	Biogas of 940 L kg <sup>-1</sup> VS 64.5% v/v (CH4), 1.7% v/v (H2) and 33.4% v/v (CO <sub>2</sub> )	620 L kg <sup>-</sup> <sup>1</sup> VS biogas yield (single stage 35°C)	600 L kg <sup>-</sup> <sup>1</sup> VS biogas yield	-3%	[66]
Autoclave thermal + co- digestion	India	FW from hostel mess, co-digestion with 30% of poultry manure. Autoclave with fixed temperature of 120°C and pressure at 10 bar for 30 min.	Recirculation Batch	50	TS removal of 59.27 % (non- treated of 57.94%); VS removal of 62%, (non-treated 59%)	8,921 mL maximum cumulative biogas	9462 mL maximum cumulative biogas	+6%	[199]
Microwave irradiation + co- digestion	India	FW from hostel mess, co-digestion with 30% of poultry manure; household microwave oven: 1,460 W, 2,450 MHz, with wavelength 12.24 cm.	Batch	50	TS removal of 58.83% (non- treated of 57.94%) VS removal by 61% (non-treated of 59%)	8,921 mL maximum cumulative biogas	9,287 mL maximum cumulative biogas	+ 4%	[199]
Ultrasonic + co- digestion	India	FW from hostel mess, co-digestion with 30% of poultry manure; ultrasonication at 20 kHz and power output of 130W.	Batch	50	TS removal of 61.83%, (non-treated of 57.94%); VS removal of 65%, (non-treated of 59%)	8921 mL maximum as cumulative biogas	9.926 mL maximum as cumulative biogas	+11%	[199]
Biological pre- treatments + co- digestion	Singapore	FW + activated sludge; after seeding with 300 ml of inoculum, the reactor was operated at 35 °C, for 24 hours.	Single stage Semi - Continuous feeding mode	35	Higher methane yield at higher OLR; accelerated solubilization of particulate organic matter and improved hydrolysis prior to AD	230 mL g <sup>-1</sup> VS	294 mL g <sup>-1</sup> VS	+28%	[65]
Fungal mash rich in hydrolytic enzymes	Singapore	Fungal mash rich in hydrolytic enzymes produced in-situ with cake waste autoclaved, inoculated with Aspergillus awamori hen directly used to hydrolyze the mixture of sludge, and food waste.	Batch	35	Eliminate the drawbacks of high energy consumption, and the use of hash chemicals of other pre-treatments	412.5 mLCH <sub>4</sub> g <sup>-1</sup> VS (No pre- treated mixed waste)	600.5 mL CH <sub>4</sub> g <sup>-1</sup> VS (Pretreated mixed waste)	+45.6%	[113]
Fungal mash rich in hydrolytic	Singapore	Fungal mash rich in hydrolytic enzymes	Batch	35	Eliminate the drawbacks of	610.3 mL CH <sub>4</sub> g <sup>-1</sup> VS.	817 mLCH <sub>4</sub> g <sup>-1</sup> VS.	+33.9%	[113]

enzymes	produced in-situ with cake waste autoclaved, inoculated with Aspergillus awamori hen directly used to hydrolyze the mixture of sludge, and food waste.			high energy consumption, and the use of hash chemicals of other pre-treatments	No pre- treated FW	Pretreated mixed waste		
Fungal mash rich Singa in hydrolytic enzymes	Fungal mash rich in hydrolytic enzymes produced in-situ with cake waste autoclaved, inoculated with Aspergillus awamori hen directly used to hydrolyze the mixture of sludge, and food waste.	Batch	35	Eliminate the drawbacks of high energy consumption, and the use of hash chemicals of other pre-treatments	817 mL CH <sub>4</sub> g <sup>-1</sup> VS. (Pre- treated FW)	600.5 mL CH <sub>4</sub> g <sup>-1</sup> VS. (Pretreated mixed waste)	-26.5 %	[113]
Thermophilic + China co-digestion	FW + waste activated sludge; lab-scale CSTRs working at controlled temperatures (35 and 55 °C).	Single stage Semi - Continuous feeding mode	55	Enhanced methane yield	260 L CH4 g <sup>-1</sup> VS <sub>added</sub> .	$\begin{array}{l} 400\\ L\ CH_4\ g^{-1}\\ VS_{added.} \end{array}$	+54%	[200]
Ultrasonic + co- Korea digestion	FW + waste activated sludge; ultrasonic homogenizer used for 30 min at 360 kJ L <sup>-</sup> <sup>1</sup> energy intensity.	Batch	35	Additional reduction: +12%COD; +4.7 VSS; +3.9 %TSS	108 mL as cumulative CH4	197 mL cumulative CH4	+82%	[192]
Thermal + co- Korea digestion	co-digestion of FW and waste activated sludge; autoclave at 120 °C for 60 min.	Batch	35	Additional reduction: +12% COD; +7 VSS; +1.7 %TSS	108 mL as cumulative CH4	193mL as cumulative CH4	+78%	[192]
Microaeration + China co-digestion	FW + brown water; introduction of small amounts of O <sub>2</sub> ; aeration intensity 0.0375 $L O_2 L^{-1}$ <sub>reactor</sub> d <sup>-1</sup> .	Batch	35	Increased solubilization and acidification efficiencies, enhanced breakdown short- chain fatty acids to acetic acid	431 L kg <sup>-1</sup> VS <sub>fed.</sub> cumulative biogas	530 L kg <sup>-1</sup> VS <sub>fed</sub> cumulative biogas	+23%	[196]
Alkali-thermal+ Korea co-digestion	FW + waste and waste activated sludge + NaOH added to the reactor; autoclave at 120° C for 30 min.	Batch	35	Additional reduction: +13% COD; +4.9 VSS; +4.3% TSS +5%	108 mL cumulative CH4	177 mL cumulative CH4	+63%	[192]
Fungal mash rich Singa in hydrolytic enzymes (Crude enzymes cocktail)	ore FW; pre-treatment with fungal mash	Batch	35	Significantly improved solid hydrolysis	610.3 mL CH <sub>4</sub> g <sup>-1</sup> VS. Experimenta l yield	817 mL CH <sub>4</sub> g <sup>-1</sup> VS. Experimental yield	+34%	[113]

Fungal mash rich in hydrolytic enzymes (Crude enzymes cocktail)	Singapore	FW; fungal mash rich in hydrolytic enzymes + untreated activated sludge at 1:1 ratio.	Batch	35	Food waste C/N ratios from 24.9 to and 16.5 after 24 h hydrolysis; co-digestion with activated sludge make up the C/N ratio to the optimum range	412 mL CH4 g <sup>-1</sup> VS. Experimenta l yield	600.5 mL CH4 g <sup>-1</sup> VS. Experimental yield	+46%	[113]
Lipid extraction+ co-digestion	China	FW from canteen + Sewage sludge; using methanol and chloroform.	Batch	55	Significantly faster reactions than those in mesophilic conditions	426 mLCH4 g <sup>-</sup> <sup>1</sup> VS <sub>added.</sub>	483 mLCH4 g <sup>-</sup> <sup>1</sup> VS <sub>added</sub>	+13%	[138]
Lipid extraction+ co-digestion	China	FW from canteen + Sewage sludge; using methanol and chloroform.	Batch	35	Possible reduction of VFA accumulation, preventing media acidification	400 mL CH4 g <sup>-</sup> <sup>1</sup> VS <sub>added.</sub>	467 mL CH4 g <sup>-</sup> <sup>1</sup> VS <sub>added</sub>	+16%	[138]

#### 6. Trends and Perspectives. 281

282

#### 6.1 Techno-economic analysis 283

FWs need to be economically and safely disposed of. High moisture content discourages FW 284 285 incineration, landfilling has high cost and leads to environmental issues such as the production of GHG and leachate [201–203]. Another option is represented by composting, which has high energy 286 287 consumption contributing to GHG emission and does not produce renewable energy. It has been 288 reported that 260.79, 82.21, and -86.21 thousand tons of GHG emissions are emitted from food waste landfill, composting, and AD, respectively [204]. 289

Data reported in previous chapters highlighted that FW has a chemical composition that allows a 290 291 valuable biogas production, contributing to renewable energy production and to GHG reduction, fighting climate change. Previous chapters also indicated that FW can be proposed to substitute EC 292 293 that were economically and environmentally unsustainable in producing biogas [205]. Therefore, FW 294 could become a useful feedstock in producing renewable energy by AD, allowing, also, its safe 295 disposal and lowering total costs for biomass [94].

296 Food wastes have been reported successfully substituting energy crops at full scale AD plants. A feed 297 mix with an energy crop has been substituted introducing 58% (w/w) of kitchen food waste from source-separated collection, with no modification in total biogas production [101], confirming the 298 299 technical feasibility.

300 Economic evaluation indicated for the EU context (e.g. Italy), that energy crops largely used in EU producing biogas, contributed by 47.6-63.7% to total energy production cost, depending on the crop 301 302 used, since the value was null for food waste [129]. In addition, feed-in tariffs, credits for carbon 303 reduction, and tax exemptions have been adopted by different countries to sustain waste management, 304 reducing AD costs [27]. For example, in the Italian context an in-feed tariff of 70 € Mg<sup>-1</sup> FW further 305 reduces costs for FW-AD [206]. Comparing three full scale AD plants using different feeding mixes, i.e. i. FW + recirculated digestate (60 % + 40 % w/w), ii. swine manure + energy crops + agro-306

industrial wastes (63 % + 15 % + 22 % w/w) and iii. swine + manure + maize silage + agro-industrial by-products (48 % + 10 % + 10 % + 32% w/w), the net profit (electricity benefit of 280  $\in$ MWhe<sup>-1</sup>) was of 0.397  $\in$  kW h<sup>-1</sup> EE for plant i, 0.160  $\in$  kWe h<sup>-1</sup> for plant ii and 0.111  $\in$  kWe h<sup>-1</sup> for plant iii, indicating the net improvement of economic balance in using food waste as feedstock. In a Chinese context, a recent cost-benefit analysis (build–operate–transfer financed modality) reported an attractive net profit margin and an internal rate of return (IRR) of 31% and 12%, respectively, for an AD plant, underlying, again, the attractiveness of FW-AD [27].

314

### 315 6.1 Policy analysis

316 Food waste can be used to produce bioenergy via biogas production. This approach represents, also, a solution for FW disposal, if it is designed in a sustainable way, i.e. lowering or nulling 317 environmental impacts and reducing GHG emission [207]. On the other hand, the residue of biogas 318 319 production, i.e. digestate, need to be safely disposed of or used as it represents by itself a problem. Digestate contains organic matter (OM) and nutrients (e.g. N, P, K etc.) that could be recovered in 320 321 producing biofertilizers and promoting a Circular Economy approach [202]. There are numerous examples of using digestate and digestate-derived fertilizers in agriculture, closing OM and nutrient 322 323 loops [208,209]. To do so, high quality FW needs to be collected, avoiding the presence of inorganic 324 and organic pollutants. Municipal solid waste management (MSW) in China is characterized above all by MSW landfilling (60.16 %), followed by incineration (29.84 %), untreated discharge (8.21 %) 325 and other treatments (1.79 %) [203], with these figures recently confirmed [27,202]. Anaerobic 326 digestion is used only for a tiny part of total waste treated in China, although the trend is positive, i.e. 327 from <3 % of total waste in 2005 to 3% of total waste in 2020. In addition, global perspectives are 328 329 very good as the size of the global market for biogas was reported to be likely to increase by 43 % in 2026 in comparison with 2018, i.e. from 20,853 US\$ Million to 29,954 US\$ Million [210]. 330

331 Chinese MSW contains about 62% of organic matter, which is much higher than the data reported

for OECD countries, i.e. 27% [203] but unfortunately it is mixed with other waste so that only a

333 very low-quality FW not suitable for AD production in a Circular Economy frame, is available.

FW quality improvement can be successfully achieved by implementing "source separate" FW
collection, above all if door-to-door systems are used, allowing the effective reduction of the presence
of plastics and heavy metals, and resulting in FW becoming more suitable for AD purposes [211].
Introducing compostable material allows the further increasing of FW quality [212].

Today's MSW collection system in China is only a mixed collection system that does not allow high quality FW collection, although some cities have started pilot projects in separate collection. However, in the 13th Five-Year Plan (2016–2020) of the Chinese government it is reported that all municipalities, cities and provincial capitals have to plan for safe solid waste disposal at a rate of 100% in solid waste in urban areas, and 80% in rural areas in 2016 [27]. In particular the Chinese government reported indications to safely dispose of urban solid waste by developing the use of waste to provide energy for the anaerobic digestion industry [27].

345 The encouragement of source separated collection is strongly recommended, because it will allow

the obtaining of a high-quality FW, that can be exploited to produce biogas and recover nutrients.

347 This in turn promotes the Circular Economy and environmental sustainability in terms of both GHG

348 reduction (renewable energy production) and FW safe disposal (biofertilizers). In the Chinese

349 context it has been reported that about 23 % GHG emissions can be decreased by source-separated350 collection compared with the base scenario [204].

Separate collection of food waste has been proved possible to be done in highly densely inhabited areas, that are typical of China. For example, Milan (northern Italy), that is characterized by a density of 2,063 inhabitants per km<sup>-2</sup> similar to that of Beijing municipality (China), i.e. 1,458.59 km<sup>-2</sup>, started FW separate collection in 2012, reaching 100% of FW collection on 2019, and pushing the total source separate MSW collection at the regional level to 70 % of total MSW. Door to door collection permitted high FW quality with less than 5 % of impurities, of which 3% was constituted by plastic.

- 357 The ban on non-biodegradable shoppers in Italy in 2011, is further reducing this amount in favor of
- the presence of compostable material, making FW suitable to be used for AD purposes and recycling
- 359 OM as a nutrient, adopting Circular Economy approaches (Figure 2).

361



**Figure 2.** Circular economy model proposed in this paper. The model uses food waste instead of

364 energy crops as a substrate for anaerobic digestion.

# 6.3 Practical implications of this study

366

This work highlights the great potential of food waste for its conversion into methane by anaerobic 367 digestion, as suggested by data collected for China and South East Asia. In particular, results reported 368 indicated the high potential of food waste in producing bio-methane, so that it can be considered a 369 370 good candidate to substitute for energy crops that are less sustainable and conflict with food 371 production. Decoupling AD process in a two-stage approach did not seem to lead to any advantage in terms of total energy produced so that a simpler one stage approach would seem preferable. On the 372 373 other hand, a two stage process could be considered in producing different products, i.e. H<sub>2</sub> and CH<sub>4</sub>. 374 Furthermore, biogas yield can be enhanced by co-digestion of FW with other organic wastes or by 375 adopting physical, thermal, chemical and physical-chemical FW pre-treatments that showed positive 376 energy balance. Data collected indicated the AD of FW to be economically sustainable in a Chinese 377 context, taking into consideration feed-in tariffs, credits for carbon reduction and/or tax exemption 378 tools.

To promote food waste-AD in a Circular Economy frame, avoiding waste production, FW management should be redesigned in order to get high quality food waste (low impurity presence), that can be used to produce both renewable energy but also biofertilizers. High quality source separate collection (e.g. door to door) seems to be a viable road, also, for highly densely populated area. The introduction of using biodegradable shoppers and in the future the implementation in using singleuse biodegradable plastics is, also, strongly recommended.

385

# 386 6. Final remarks and Conclusions

In general, some useful conclusions about the use of food waste in anaerobic digestion can besummarized as follows:

- 1. Potential bio-methane production is high, i.e.  $480\pm88 L_{CH4} kg^{-1} VS$  (n=42), when compared to that of energy crops,  $246\pm36 L_{CH4} kg^{-1} VS$  (n = 6). So that FW uses should be encouraged through high quality separate collection to assure high quality biomass to substitute EC.
- 392 2. Anaerobic digestion allowed the production of about 81% ( $395 \pm 84 \text{ L kgVS}^{-1}$ ) of the potential 393 producible bio-methane, so that AD is considered to be a well-consolidated bioprocess. Both co-
- digestion and FW pretreatment improve total biogas yield.
- **395 3**. Data collected did not seem to indicate differences in total energy production using two-stage vs.
- 396 one stage AD process. However, two-stage AD allowed the production of  $H_2$  and  $CH_4$  instead of
- **397** only CH<sub>4</sub>.
- 398 4. Energy balance indicated that pre-treatments gave a valuable net energy increase with respect to
- 399 untreated biomass, except for chemical pre-treatments that resulted in a negative balance.
- 400 5. Source separate collection of FW should be implemented assuring high quality feedstock for AD
- 401 purpose in a Circular Economy frame.
- 402
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