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**Effect of rice husk and palm tree-based biochar addition on the anaerobic digestion of food waste/sludge**

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## **Abstract**

This study explored the impact of different types of biochar on the anaerobic digestion (AD) of food waste and sludge and the biochar-enhanced mesophilic AD of food waste for methane production. A particular focus was placed on the process and underlying mechanisms. Rice husk biochar (RHB) and palm tree biochar (PTB) were loaded at levels of 5, 10, and 15 g/L. The results showed that 5 g/L of RHB led to an increase in methane yields of  $455.83 \pm 17.9$  mL/gVS<sub>add</sub> compared to the control yield of  $333.32 \pm 14.9$  mL/gVS<sub>add</sub>. The addition of an adequate amount of biochar improved the cumulative methane yield, whereas excessive amounts inhibited the AD process. RHB550 biochar improved the process stability, which was favourable for syntrophic volatile fatty acid (VFA) oxidation, and consequently increased the cumulative methane yield by up to  $36.75 \pm 2.9\%$ . Earlier stagnation was also shown to be caused by the elimination of VFAs and ammoniacal-nitrogen and the degradation of chemical oxygen demand (COD). Increasing the concentrations of RHB550 biochar enabled significant improvement in methane yield by increasing the pH and COD removal, which could be a useful strategy in AD to further increase the organic loading rate.

**Keywords:** Food waste; biochar; anaerobic digestion; methane yield; trace metals.

## **1. Introduction**

Food waste (FW), such as the raw food materials and food residues of homes, restaurants, and food processors, is one of the main components of municipal solid waste (MSW). As economic development has increased, the population rises, living standards in cities grow, and the amount of FW increases. This is due to the enormous annual global volume of FW, which is estimated at 1.33 billion tons [1] and represents 33% of total FW produced for human consumption purposes

[2]. In South Korea, the average FW generation has reached 13,697.4 ton/day, accounting for 27.4% of the MSW over an 11-year period from 2003 to 2014 [3]. Ocean dumping and direct landfilling of organic waste have been banned since 2014 in accordance with policy formulation. Landfilling is still the primary disposal process, although it is not a sustainable solution when considering nutrient cycling and greenhouse gas emissions. Anaerobic digestion (AD) is an economic and ecological technology that has been suggested for the simultaneous treatment of FW and production of renewable energy [4]. There are two key issues in the processing of FW: (i) increased volatile fatty acid (VFA) accumulation system stability and (ii) high organic loading rate reactor efficiency (OLRs) [5]. AD with sewage sludge, which has high alkalinity and improved buffering capacity, is considered more efficient than the mono-digestion of sludge and FW [6]. Furthermore, the low organic load of sewage sludge and the resulting unused capacity (30%) of sludge digesters in wastewater treatment plants are additional drivers of FW/sludge co-digestion for reduced waste discharge and optimal methane production. Recent studies on FW AD show that high concentrations of intermediate metabolites, that is, VFAs, exist in co-digestion systems at the end of the digestion period [7,8]. This accumulation inhibits methane production in AD. Therefore, the acceleration of VFA degradation during AD is crucial for improving the system performance [8].

In recent years, the addition of biochar to AD has attracted more attention because of its potential benefits in the AD process, such as stabilising the digestion process, alleviating the inhibitory effect, enhancing the buffering capacity, and improving the methane yield. Biochar in anaerobic digesters reduces the time delay to start methane production, and the total methane production increases [9,10]. Methane production may be enhanced by adding biochar through the following mechanisms: (a) reduction of acid stress, which increases the methanogenic activity in AD reactors; (b) facilitation of electron transfer between bacteria and methanogens attached to the

biochar surface, (c) enhanced resistance to ammonia stress due to the attachment of microorganisms to biochar, and (d) the enabling of microorganisms to utilise long-chain fatty acids during AD [8]. Inspired by the buffering principle, some recent investigations have focused on the addition of biochar to FW digestion, in which the over-acidified state was efficiently adjusted by the buffering capacity of biochar, thereby achieving better digestion performance [11]. The effect of biochar addition on AD has been studied with different types of organic waste residues such as FW [11], sewage sludge [12], poultry waste [13], and other AD. Furthermore, the use of biochar in anaerobic digesters has been investigated, in addition to its traditional use as a soil amendment. Most of these studies used a single biochar type and pyrolysis temperature to investigate its effect on AD performance, with a few exceptions that compared the effects of different biochar extents [12,13]. The properties of biochar differ depending on the raw material and pyrolysis temperature used for its production. Consequently, this affects its function when added to an anaerobic digester [10]. Therefore, it is necessary to understand how biochar produced from different feedstocks has different effects on AD. In addition to its conventional use as a soil amendment, the addition of biochar to AD systems can enable pyrolysis kilns and gasifiers to become more economically viable.

Biochar dosage is an important parameter for biochar-assisted AD digestion because it has an important influence on the adsorption efficiency of biochar. Therefore, to achieve a satisfactory and stable methane yield, it is necessary to optimise the biochar dosage concentration during the AD of FW. Different types of biochar were added during the start-up with FW, the optimal amount (10 g/L) of biochar was found for start-up, and both methanogenic pathways and potential microbial species were elucidated which could contribute to direct interspecies electron transfer (DIET) [8]. This would enable optimal biochar selection and dosage optimisation to improve the

AD. By adding an AD process, the production of biogas is improved in terms of quality and quantity, whereas by adding 5–18 g/L of dry mass biochar, methane production increases by 20–40% [11,14]. Additionally, biochar in AD can adsorb inhibitors, such as ammoniacal nitrogen ( $\text{NH}_4^+\text{-N}$ ) and VFAs, by increasing the buffer potential and stabilising the AD system [14,15], thereby improving the quality of methane production. Biochar additions of 2–20 g/L have been used in highly solid anaerobic digesters to test their performance. A significant improvement in biomethane production rate was observed with 0.7 g/g of  $\text{RS}_{\text{sludge}}$  biochar added to the cardboard AD batch. Excessive biochar (3.86 g/g  $\text{TS}_{\text{sludge}}$ ) resulted in a decrease in biomethane production and prolongation of the lag phase. Using biochar at a ratio of <0.5% or >20% (w/w) has been reported to decrease methane yield [16]. Many studies have reported improving AD processes by adding biochar at different pyrolysis temperatures; however, few studies have examined the effects of different doses or concentrations of biochar on the stability of AD processes. Hence, choosing an optimal biochar dosage concentration for VFA degradation during AD is crucial for cost-effective applications. As biochar dosages increase in quality, the cost also increases. In addition, there is a shortage of studies on biochar-amended FW/SS co-digestion, while there has been an increase in studies on FW mono-digestion with biochar. While the promotion of the AD method by utilising different biochar pyrolysis temperatures has been long noted, few analyses have evaluated the effects of biochar from optimal conditions on the stability of the AD phase at various doses or concentrations. AD in FW batch experiments indicated that biochar is capable of catalysing AD by reducing VFA and supporting archaeal growth. Therefore, using an optimal biochar dosage concentration for VFA and organic matter degradation during AD digestion is crucial for its cost-effective application.

However, to the best of the authors' knowledge, the impact of biochar addition on the various biochars and the consequences of varying extents under the same conditions were not documented in FW AD. The objective of this study was to determine the effects of biochar made from two different feedstocks on the AD of FW and sewage sludge under mesophilic conditions and the effects of different types of biochar and biochar dosage additions on the methane production and stability of an anaerobic digester to improve FW organic matter decomposition. Biochar is the first step toward developing an efficient organic loading system for the AD of FW.

## **2. Materials and Methods**

### ***2.1. Substrate and inoculum***

Liquid food waste (including rice, noodles, pork, vegetables, and eggshells) was collected from the school cafeteria at Kyonggi University, South Korea. In AD studies, anaerobically digested seed sludge was used as the inoculum. Waste activated sludge (WAS) was obtained from a wastewater treatment plant (Incheon landfill area) in South Korea. The substrate and inoculum were kept at 4 °C in a 10 L tank before utilisation. Both FW and sludge substrates were characterised separately for different physicochemical parameters before being included in the digestion, as described in previous protocols [17]. **Table 1** lists the characteristics of the sludge and FW.

### ***2.2. Biochar characterisation***

To represent economic agricultural waste disposal, rice husk and palm trees were selected for biochar production. The biochars were collected from the Yougiind company, South Korea, and consisted of two types of biochar, rice husk biochar (RHB) and palm tree biochar (PTB), which were first air-dried and cut into 5–10 mm pieces and then prepared using a scalable 20-minute method, ideal for research, consisting of an indirectly and electrically heated revolving kiln. The

PTB comprised 25 cm fronds that were completely burned to ashes; however, aluminium-wrapped fronds were converted to biochar after 4 hr [18]. The biochar was pyrolyzed at 550 °C and 400 °C in consideration of the optimum nutrient retention to increase rice yield [6]. The resulting pH, EC, bulk density, porosity, and water holding capacity of the rice husk and palm tree biochars were 9.7, 0.59 mS/cm, 0.23 g/cm<sup>3</sup>, 70%, and 2.68 g water/g dry sample and 7.96, 0.359 mS/cm, 0.34 g/cm<sup>3</sup>, 81%, and 2.48 g water/g dry sample, respectively. **Table 2** provides the various physicochemical properties of the biochar. The biochar was then ground and sieved to obtain a fraction of 1–2 mm.

### ***2.3. Experimental setup***

Because the influence of biochar varies with loading volumes on AD of FW, a total of six groups were defined as T1, T2, T3, T4, T5, and T6 with RHB and PTB rates of 5, 10, and 15 g/L (w/w), respectively [8]. As a control reactor, no biochar was loaded onto C<sub>0</sub>. Similar setups have been used in 0.5 L digesters with biochar in laboratory-scale anaerobic batch reactors with a working volume of 0.3 L. The inoculum was added to the reactor to maintain a 1.0% inoculum/substrate (I/S) ratio, based on volatile solids (VS). The digester was placed in a digital water bath and magnetically stirred at 120 rpm at 35 °C for a digestion period of 35 days. All reactors were purged with N<sub>2</sub> gas for 3 min to create an anaerobic environment in the bottle and sealed with a rubber septum. The pH of the FW digestate was evaluated every 48 h. NaHCO<sub>3</sub> was mixed to mitigate the rapid acidification of the FW digestion mixture every 24 h if required to ensure a neutral pH (6.5–7.0) before the methanogenesis phase. During the AD of FW and sludge, the biogas volume was collected daily using a gas bag and was measured using a gas flow meter.

### ***2.4. Biodegradability of food waste/sludge***



The methane output was determined by dividing the total methane volume generated by the initial VS mass of the substrates. The calculations were corrected to account for the digestate excluded during sampling. The free ammonium nitrogen (FAN) concentration in the reactors was measured using Equation 1 according to the VS removal efficiency.

$$\text{Total solids (TS) (or VS) removal (\%)} = \frac{(\text{TS (or VS)}_{\text{initial}} - \text{TS (or VS)}_{\text{final}}) \times 100}{\text{TS (or VS)}_{\text{initial}}} \dots (1)$$

### 2.5. Analytical methods

Samples were taken from each reactor to measure the VS and TS on the first and last day of co-digestion. Samples were also drawn on alternate days to measure VFAs (acetic acid, butyric acid, propionic acid, iso-butyric acid, valeric acid, iso-valeric acid, and caproic acid) and total ammoniacal-nitrogen (NH<sub>4</sub><sup>+</sup>-N) and to evaluate the kinetics of the chemical oxygen demand (COD) removal process following standard protocols [21] and using methods as mentioned by [22]. To measure protein and carbohydrates, the Folin-Lowry and phenol-sulfuric methods were used, respectively [20]. Gas chromatography was used to analyse the composition of the biogas (GC-TCD, Agilent 7890A, Agilent Technologies, Inc., USA), using an HP-PLOT/Q column, with a thermal conductivity detector and helium as the carrier gas [6].

### 2.6. Gompertz model of methane production

The Gompertz model was used to describe the experimentally observed accumulated methane performance mathematically as proposed by [9] Cai et al. (2016). The model has the equation form..... (1)

$$M(t) = P \times \exp \left\{ -\exp \left[ \frac{R_{\max} \times e \times (1-t)}{P} + 1 \right] \right\} \dots \dots \dots (1)$$

where M(t) signifies the cumulative production of methane (mL CH<sub>4</sub> g/L VS<sub>added</sub>) at time t,

$P$  is the ultimate methane yield (mL CH<sub>4</sub> g/L VS<sub>added</sub>) at the end of incubation,  $R_{max}$  is the maximum methane production rate (mL CH<sub>4</sub> g/L VS<sub>add</sub>),  $K$  is the lag phase (d),  $t$  is the time (d), and  $e$  is a constant (2.71828).  $Y$ ,  $R_{max}$ , and  $P$  were calculated using Origin Pro 9.0 through curve fitting.

## ***2.7. Statistical analysis***

All tests were performed in triplicate, and the findings were evaluated using a variance analysis in SPSS software, with  $p < 0.05$  considered to be statistically significant.

## **3. Results and discussion**

### ***3.1. Comparison of biochar properties and their implications in co-digestion***

Biochar, a combustible carbon-rich solid material generated through the gasification or pyrolysis of biomass waste, is a promising additive with several desirable features for the enhanced AD method. Biomass feedstock and pyrolysis conditions influence the physical properties and chemical composition of the biochar produced. In this work, the biochars were sourced from RHB and PTB residues, and these were produced at pyrolysis temperatures of 550 and 400 °C. Biochar from solid agricultural residues has received great attention from scientific communities [15,8] because it leads to useful products and simultaneously contributes to solving pollution problems arising from biomass accumulation. AD is affected by the physicochemical properties and structures of biochar, such as pH, porous structure, chemical composition, and surface area [22] (Wang et al., 2018). RHB550, the alkaline nature of biochar, contributes significantly to the buffering action upon its inclusion in FW AD [8]. The presence of elevated nutrient concentrations, such as N and K contents of 1.53% wt. and 1.35% wt., respectively, contributes to the additional alkalinity. Another important characteristic is the O/C ratio of 0.78, which implies high hydrophobicity of biochar and is a desirable property of absorption capacity which should be

useful in AD systems [11]. The conductivity of biochar is another useful property. **Table 2** shows that the rice-husk-based biochar had a much higher conductivity than the PTB, which may be a key factor for facilitating VFA oxidation and assimilation into methane via DIET in AD [22]. In addition, the physical properties of biochar differ. The surface area of biochar can be used as a physical indicator to assess its structure for microbial attachment. Compared to PBT400 biochar, RHB500 has a 1.5-fold higher surface area ( $19.8 \text{ m}^2/\text{g}$ ) (**Table 2**). Furthermore, secondary organic matter in biochar, particularly if it contains selected properties associated with AD, such as high surface area, alkalinity, and hydrophobicity, may promote syntrophic VFA degradation and improve methane production efficiency.

### ***3.2. Influence of biochar addition on the degradation of organics in the anaerobic digestion system***

Generally, the presence of FW along with sewage sludge in AD promotes higher hydrolysis and acidogenesis, thereby leading to the generation of soluble products. Soluble and total COD were determined in this study and efficiently indicated the decomposition of macromolecular organic matter as well as the utilisation of dissolved organic matter that may eventually affect methane production at different dosages of biochar-modified digestive conditions [8]. The sCOD levels of the biochar at different concentrations showed a significant difference ( $p > 0.05$ ) in the first 10 days. The sCOD accumulated and reached a high concentration on day 10 for all the biochar addition reactors, after which the concentrations decreased due to methane assimilation during the subsequent period of digestion. The sCOD concentrations were comparatively higher in the RHB500 biochar and control reactor during the decline phase than in the other three biochar reactors (**Figure 1a**). This may have been due to a lower conversion of the soluble product into methane in the former reactors. The total COD concentration in the different biochar and dosing

reactors was also investigated, as shown in **Figure 1b**. As expected, tCOD degradation decreased under different biochar dosing conditions. In terms of total COD removal efficiency, the highest values of T1 (27.55%), T2 (21.35%), T3 (16.0%), T4 (20.8%), T5 (16.85%), and T6 (8.5%) were obtained for RHB550 and PBT400 biochar amended in the control reactors. The kinetics of tCOD from FW with the addition of biochar using the organic matter removal process in this study followed the reaction trend of the first-order kinetics model. The results of the first-order kinetics variables for the percentage of COD removal are presented in **Table S1**. Efficiency ultimately affects the development of methane under different biochar-modified digestion conditions [8]. In the digester, the degree of organic COD degradation and its assimilation are indicators of effective microbial behaviour. Wang et al. (2018) and Kaur et al. (2020) [22, 8] reported the same finding, which showed a more stable reduction in organic matter in the COD removal rate for intermediate reactors. As such, biochar could enhance the degradation of tCOD, thereby providing more biodegradable material for the growth of methanogenic archaea during the co-digestion of FW and SS. Therefore, a high COD removal efficiency was obtained for all the biochar addition groups. The 5 g/L addition of RH550 increased the removal of COD by 27.5% compared with the control, which could have beneficial consequences for the generation of methane by the amended RHB550 co-digestion (see Section 3.3 for the results of methane production).

The total soluble products included VFAs and alcohols which were developed during acidogenesis as well as hydrolysis in the FW AD system. They were tracked throughout the AD cycle as an essential intermediate for methanogenesis. The tVFA fluctuated similarly in all reactors throughout the experiment, increasing initially and then gradually decreasing, suggesting that tVFAs were produced and accumulated through the hydrolysis-acidification of FW and then used as the substrate for the subsequent methanogenesis. It is noteworthy that an obvious accumulation

of VFAs was observed owing to the increased biochar concentration. All reactors showed strong degradation due to increased tVFA concentration on days 2–8 (**Table 3**), when the highest tVFA concentration was reached. The highest tVFA concentrations were T1:  $10.5 \pm 0.14$  g/L, T2:  $11.0 \pm 0.19$  g/L, T3:  $11.7 \pm 0.35$  g/L, T4:  $9.6 \pm 0.28$  g/L, T5:  $11.5 \pm 0.18$  g/L, T6:  $13.3 \pm 0.39$  g/L, and control without biochar addition:  $13.33 \pm 0.26$  g/L. On day 7, VFAs accumulated to 13.33 g/L in the control group, resulting in a pH drop (from 7.32 to 6.21), which exerted a slight inhibition of microbial activity. Subsequently, the tVFA contents in all reactors decreased significantly as the removal efficiency of tVFAs was accelerated, indicating that the activity of microorganisms had recovered from various inhibitors. At the end of AD, a high tVFA concentration of 7.5 g/L was observed in the control reactor, implying that a high VFA concentration remained unconsumed in the control (without biochar addition), while it quickly dropped to a concentration of 2.8 g/L in the group supplemented with 5 g/L of RHP550 biochar. Furthermore, significant fluctuations were observed in the control group. After 20 days of digestion, the methane yield was lower than that of the control group during this period. Therefore, it was concluded that the addition of biochar could decrease VFA accumulation and stimulate methanogenic DIET microorganisms, which are crucial for the stability of the AD process and methane production [21].

### ***3.3. Performance of biochar amended digestion systems in methane production***

The use of biochar has been reported in classified co-culture (for example, *Geobacter* sp. and *Methanosarcina* sp.) or simple substrate cultures [23,21]. Only a few recent reports have investigated the suitability of biochar in meso- and thermophilic co-digestion of FW [15,24]. **Figure 2** illustrates the effect of different dosages of RHB550 and PTB400 on methane yield in FW AD. The maximum methane production rate acquired in biochar amended digesters was  $50.66 \pm 2.48$  mL/gVS/d at day 6,  $48.78 \pm 2.8$  mL/gVS/d at day 6,  $48.8 \pm 2.1$  mL/gVS/d at day 8,  $47.8 \pm$

0.25 mL/gVS/d at day 9,  $42.0 \pm 0.87$  mL/gVS/d at day 8,  $48.0 \pm 1.25$  mL/gVS/d at day 9, and  $33.7 \pm 1.5$  mL/gVS/d in the control at 8 days in T1, T2, T3 T4, T5, T6, and the control group, respectively. These results indicated that the methane production rate of biochars was significantly higher than that of the control group without biochar addition. The alkalinity of biochar can relieve the acid inhibition produced by acidification [8]. The addition of RHB550 helped to reduce the lag phase, while the control had a longer log phase. The maximum daily methane production in the T1 treatment group was 50 % higher than that in the control group. Pan et al. and Zhang et al. [15, 25] investigated the inhibitory effects leading to lower production of methane in large biochar supplementations. This is primarily because a small addition of biochar will efficiently alleviate VFA accumulation, leading to higher levels of methanogenic activity, whereas a higher concentration of biochar in the digesters of AD will contribute to a greater acid accumulation.

The cumulative methane production is illustrated in **Figure 3**. Different log lengths between test conditions were observed; however, the overall cumulative methane production increased positively from  $455.83 \pm 3.4$ , (T1),  $397.51 \pm 7.9$  (T2),  $365.65 \pm 7.1$  (T3),  $397.19 \pm 9.4$  (T4),  $366.63 \pm 2.8$ , (T5),  $332.41 \pm 6.2$  (T6) and  $333.33 \pm 6.4$  mL CH<sub>4</sub>/gVS<sub>added</sub>, respectively, which increased by  $36.75 \pm 1.65$  %,  $19.25 \pm 0.86$  %,  $9.6 \pm 0.38$ %,  $19.15 \pm 0.76$  %,  $9.9 \pm 0.49$ %, and  $-0.27 \pm 0.38$ % in RHB550 and PTB400 addition T1 (5 g/L), T2 (10 g/L), and T3 (15 g/L) respectively, as compared with control groups ( $p < 0.05$ ). It is worth noting that the cumulative methane production was 455.8 mL/gVS at 5 g/L RHB550, which enhanced the effect of methane production. At the same dosage of 5 g/L, the cumulative methane produced by the PTB400 addition was lower than that of RHB550 biochar. This phenomenon may indicate that biochar has alkaline groups on its surface, which can neutralise large amounts of organic matter generated in the early stage of AD and alleviate acid inhibition in the system. The specific surface area of

RHB550 was higher than that of PTB400, and a larger specific surface area was suitable for the metabolism and growth activities of methanogens and other microorganisms. The properties of biochar can be affected by different types of feedstocks and pyrolysis temperatures. Previous studies have also shown that surface area, porosity, and biochar production methods can influence the impact of biochar on AD [25]. The short lag phase (T1) of the group with 5 g/L RHB550 addition is associated with the elevated pH, surface area, and quicker rate of COD removal efficiency, in comparison to the control (Co) (see Sections 3.2 and 3.4 for details). Finally, RHB550 may contain nutrients that can be utilised by methanogens to promote their activity and increase the conversion efficiency of VFAs.

In addition to biochar type, biochar dosage can also influence VFA and cumulative methane. Therefore, the effects of biochar dosage on the cumulative methane yield were also investigated. Compared with the control, the cumulative methane yield increased by  $36.75 \pm 1.65 \%$ ,  $19.25 \pm 0.86 \%$ ,  $9.6 \pm 0.38\%$ ,  $19.15 \pm 0.76 \%$ ,  $9.9\% \pm 0.49\%$ , and  $-0.27 \pm 0.38\%$  in RHB550 and PTB400 addition T1 (5 g/L), T2 (10 g/L), and T3 (15 g/L), respectively. However, a further 5 g/L increase in the biochar amount caused a significant decrease in cumulative methane yields, which were 20% and 24% lower than the digester with 15 g/L of RHB550 and PTB400 biochar additions, respectively. A similar study was also conducted with biochar derived from corn [26]. The results showed that a significant inhibition effect occurred with a higher addition of biochar, resulting in lower methane production. However, a further increase in the amount of biochar resulted in a substantial decrease in the total methane production, which was 24% lower than that of the 15 g/L digesters. Zhang et al. [25] suggested that biochar extracted from sewage sludge to an AD system and, consequently, biochar incorporation not only greatly enhanced the total methane production but also increased the reaction rate. Similarly, they also found that the

optimal amount of biochar was 10 g/L, and greater amounts decreased the cumulative methane yield. This was mainly because the moderate biochar addition could effectively alleviate VFA accumulation. Supplementation of biochar at increasing dosages resulted in the accelerated degradation of COD to methanogenically favourable substrates and thus increased specific methane production yields [27]. RH550 had a larger precise surface area than that of PBT400, which may be ideal for the development of microorganisms and would promote the release of more VFA in the AD process for decomposing organic matter. Considering that biochar addition incurs a cost to the overall digestion process, it would be desirable to use a lower concentration without comparing the digestion performance. In these cases, 5 g/L biochar addition was proven to provide the desired positive impact on improving the digestion efficiency.

#### ***3.4. Effect of different biochars on pH, EC, and ammoniacal-nitrogen***

During the AD process, the pH of each treatment declined rapidly over one and two days due to the rapid acidification of the carbohydrates contained in the FW (**Table 3**). In the control treatment, the pH declined from 7.33 to 6.21, whereas due to the organic alkali functional groups found in the biochar, the decline in pH level was relatively low [28]. The pH of the anaerobic digesters was affected by several factors, including alkalinity, temperature,  $\text{NH}_4^+\text{-N}$ , and VFAs [29]. During the stable period, the pH value showed an increasing tendency with the increase in biochar dosage (5 to 15 g/L) and reached 7.5 to 7.8, respectively, at the end of the digestion (**Figure 4a**). The methanogenic activities of  $\text{C}_0$  were significantly inhibited at a pH lower than 6.5 to 7.01, thus maintaining a low yield of methane. For the biochar-amended digesters, pH varied in a slightly alkaline range (7.88 to 7.45), which was significantly higher than that of the control groups (6.31 to 7.01). This result indicates that the buffering capacity of biochar maintained a more appropriate range for microbial activity. Biochar RHB550 has a vital function in enhancing reactor stability



by increasing the degradation of organic matter in the digester. As shown in **Figure 4b**, the electrical conductivity (EC) of the initial FW AD of T1, T2, T3, T4, T5, and T6 was 15–18 uS/cm. The biochar addition groups in EC during the hydrolysis were 30 percent greater than the control, which is close to the earlier study on the addition of granular activated carbon during FW hydrolysis, which found that WSP550 increased conductivity by 60% through DIET participation [6, 8]. However, EC differed significantly between the different biochar additions and controls during methanogenesis.

The effects of different biochar types on the variation in  $\text{NH}_4^+$ -N concentration were investigated, and the results are shown in **Tables 3 and S1**. The results showed that the average  $\text{NH}_4^+$ -N concentrations of T1, T2, T3, T4, T5, and T6 were  $712 \pm 21.52$ ,  $793 \pm 30.1$ ,  $891 \pm 24.1$ ,  $841 \pm 33$ ,  $910 \pm 45$ ,  $976 \pm 29.5$ , and  $1088 \pm 5.4$  mg/L, respectively, indicating that the average  $\text{NH}_4^+$ -N concentration increased with increasing biochar dosage. Ammonium nitrogen inhibition is considered one of the primary causes of the AD process failure [30]. Notably, supplementation with RH550 biochar significantly lowered the  $\text{NH}_4^+$ -N concentration at the three dosage concentrations. This may be related to the adsorption of biochar [31]. In contrast to the control group, the concentrations of  $\text{NH}_4^+$ -N in the biochar-supplemented group were significantly lower than those in the digesters without biochar supplementation during the entire digestion period. These results suggest that biochar could effectively alleviate  $\text{NH}_4^+$ -N inhibition and create a suitable environment for methanogen growth.

**Table 3** shows the VS removal efficiencies of anaerobic digestion at various biochars and dosage concentrations. The VS removal efficiencies of biochar amended digesters T1, T2, T3, T4, T5, and T6 were  $45.48 \pm 1.43\%$ ,  $42.03 \pm 0.84\%$ ,  $46.76 \pm 1.2\%$ ,  $42.25 \pm 1.69\%$ ,  $40.91 \pm 0.6\%$ ,

38.16 ± 0.57%, and 37.85 ± 1.51%, respectively. Higher VS reduction values were obtained for the digesters with higher biogas production. The highest methane production and VS reduction were observed for different biochars and dosages of RHB550. In the biochar amended digesters, with increasing dosage concentration from 5 to 20 g/L, the VS reduction decreased from 20.15%, 11.04%, 7.6%, 11.6%, 8.01%, and 0.86%. The lower methane yield but comparable VS reduction at 15 g/L may be due to the conversion of VS into intermediate products such as VFAs [32].

#### **4. Conclusion**

The effects of the addition of different types of biochar to enhance methane production efficiency in mesophilic FW digestion were elucidated. Biochar addition could aid the AD process by facilitating rapid VFA digestion as well as by reducing the inhibitory effects of high  $\text{NH}_4^+\text{-N}$  concentrations. Biochar treatment under the same conditions boosted the cumulative methane yield to varying degrees compared with the control. Compared to the control, RH500 produced a 36.75% higher cumulative methane yield. RHB550 was also beneficial because of its physicochemical properties and decreased concentrations of VFA and  $\text{NH}_4^+\text{-N}$  during the AD process. Biochar addition led to syntrophic VFA oxidation and AD, whereas hydrogenotrophic methanogenesis occurred in the high-solid FW digesters. Thus, biochar addition is a useful mechanism for the generation of methane in AD.

### **List of Figure captions**

Figure 1. Effect of (a) sCOD and (b) total COD change in during biochar addition in food waste anaerobic digestion.

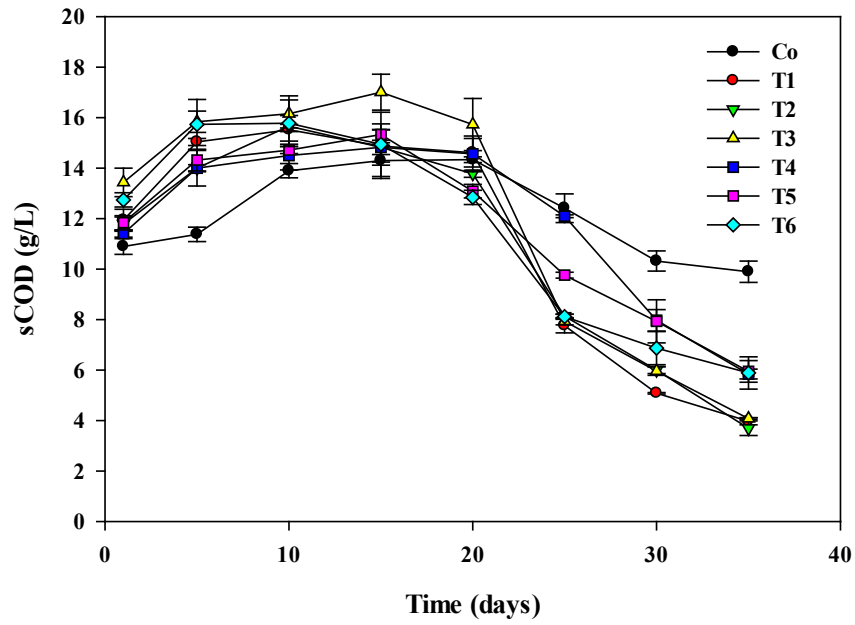
Figure 2. Influence of biochar addition on daily methane production profile during AD in biochar amended reactors and control reactor with no biochar addition.

Figure 3. Effect of cumulative methane production under different biochar dosage in food waste/sludge anaerobic digestion.

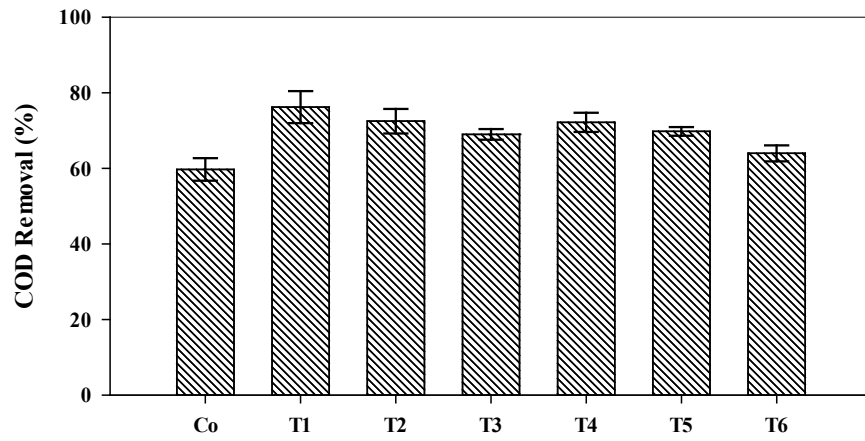
Figure 4. Effects of biochar additions on changes of pH (a) and electrical conductivity (EC) (b) after anaerobic digestion.

### **Supplementary file**

**S1.** Effect of  $\text{NH}_4^+$ -N concentration change in during biochar addition in food waste/sludge anaerobic digestion.



(a)



(b)

Fig 1. Effect of (a) sCOD and (b) total COD change in during biochar addition in food waste anaerobic digestion.

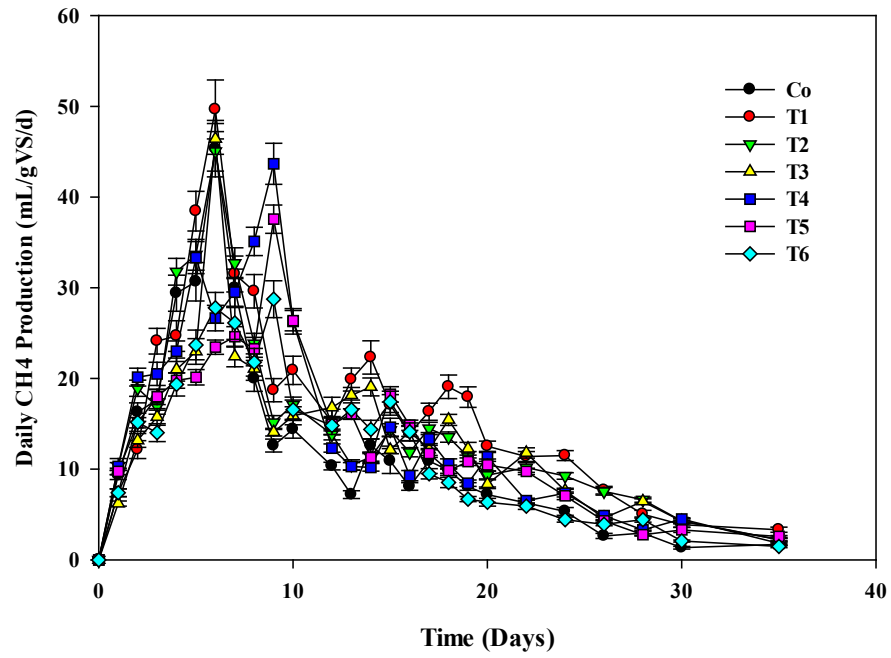


Fig 2. Influence of biochar addition on daily methane production profile during AD in biochar amended reactors and control reactor with no biochar addition.

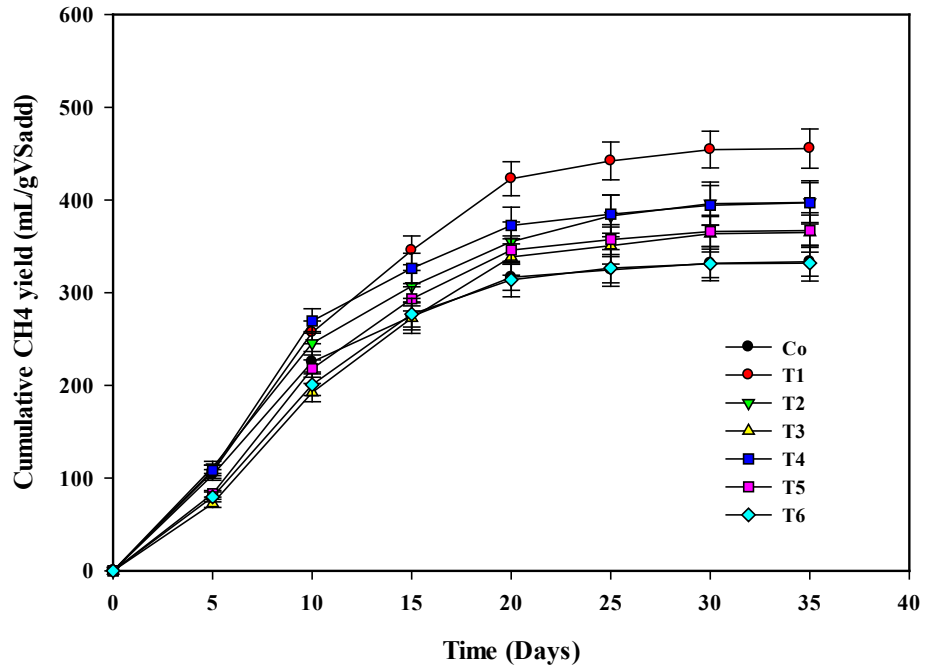
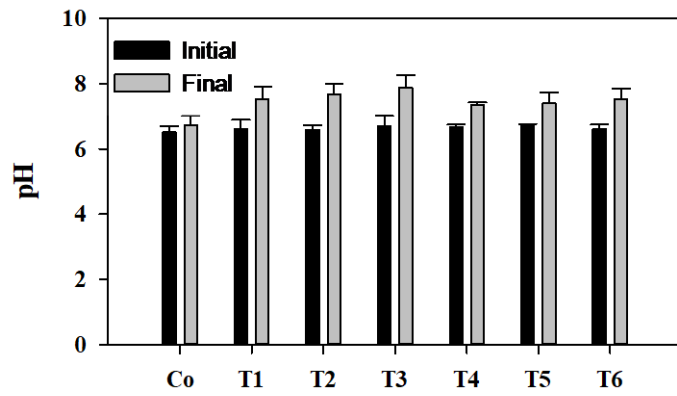
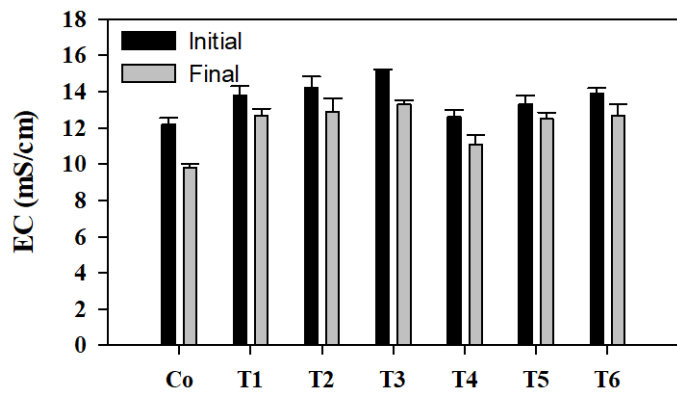


Fig 3. Effect of cumulative methane production under different biochar dosage in food waste/sludge anaerobic digestion.

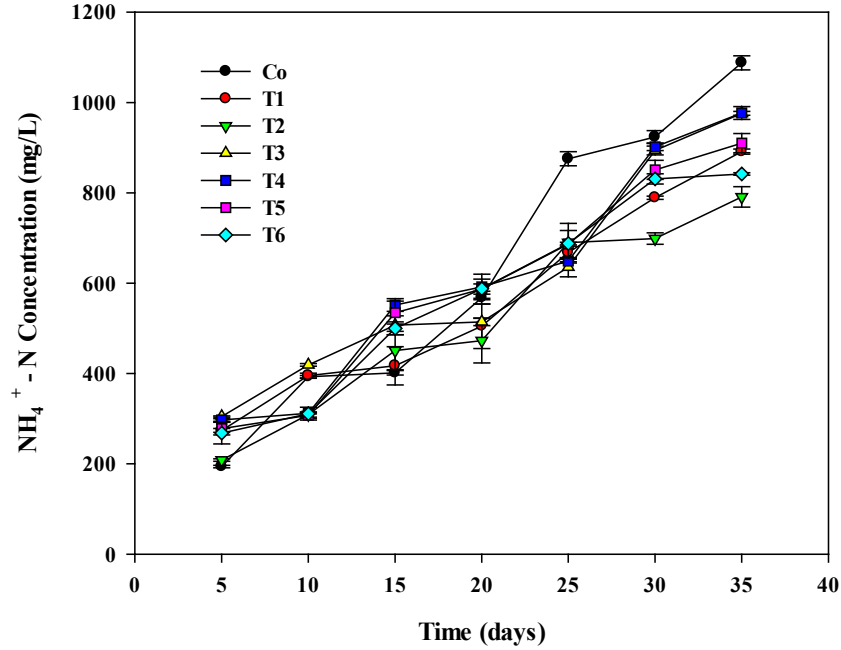


(a)



(b)

Fig 4. Effects of biochar additions on changes of pH (a) and electrical conductivity (EC) (b) after anaerobic digestion



**S1.** Effect of NH<sub>4</sub><sup>+</sup>-N concentration change in during biochar addition in food waste/sludge anaerobic digestion.

Table 1. Characteristics of food waste and sludge(s) used in this study

Parameter	Unit	Anaerobic Sludge	Food waste
		18924 ±662	61281 ±1838
VS	mg/L	8767 ±306	53879 ±2155
VS/TS	—		
sCOD	mg/L	TS	mg/L
EC	mS/cm	ND	9.27
Total N	mg/L	480 ±14	3150 ±110
Total P	mg/L	160 ±4	1613 ±56



pH	—	7.92	4.5
TOC	%	—	44.65± 0.07
TKN	%	—	3.497± 0.05

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Notes: sCOD: Soluble Chemical Oxygen Demand; TS: Total Solids; VS: Volatile Solids; TOC: Total organic Carbon; EC: Electric Conductivity; N: Nitrogen; P: Phosphorus

Table 2. Physicochemical characteristic of different types of Biochar

<b>Parameters</b>	<b>Rice husk biochar</b>	<b>Palm tree biochar</b>
pH	9.7	7.96
EC (dS/cm)	0.59	0.35
O wt%	32.1	14.44
CEC (cmol/kg)	16.54	23.03
O/C (mol/mol)	0.78	0.12
H/C (mol/mol)	0.21	0.61
C wt%	42.2	51.87
N wt%	1.53	0.45
P wt%	0.89	0.69
K wt%	1.35	2.17
SA (m <sup>2</sup> /g)	19.8	12.7
AS (mg/kg dry wt)	0.33	0.17
Cd (mg/kg dry wt)	4.41	2.17
Co (mg/kg dry wt)	0.64	1.1
Cu (mg/kg dry wt)	3.72	2.31
Mo (mg/kg dry wt)	2.06	1.57
Ni (mg/kg dry wt)	3.00	0.42
Zn (mg/kg dry wt)	31.1	23.2

Table 3. Summary of performance of biochar-assisted anaerobic digestion treatments relative to control.

Parameters	T1	T2	T3	T4	T5	T6	Co
VS removal efficiency (%)	45.48 ±1.43	42.03 ±0.84	46.76 ±1.2	42.25 ±1.69	40.91 ±0.6	38.16 ±0.57	37.85 ±1.51
Maximum CH <sub>4</sub> content (%)	65.4 ± 0.1	63.6 ±0.5	60.5 ±2.1	62.7 ±0.3	61.1 ±1.1	60.3 ±1.7	54.7 ±2.4
Cumulative CH <sub>4</sub> production(mL/g VS)	455.83±17 .9	397.51±7 .9	365.65±7 .1	379.19±9 .4	366.63±2 .8	332.41±6 .2	333.33±14 .9
Final day pH	7.52±0.39	7.67±0.3 2	7.88±0.3 8	7.34±0.0 84	7.39±0.2 5	7.51±0.3 3	7.01±0.29
CH <sub>4</sub> increase respect to control (%)	36.75 ±2.9	19.25 ±0.86	9.69 ±0.38	19.15 ±0.76	9.99 ±0.49	--	—
tVFAs max (g/L)	10.5± 0.14	11.0± 0.19	11.7± 0.35	9.6± 0.28	11.5± 0.18	13.3±0.3 9	13.33± 0.26
NH <sub>4</sub> <sup>+</sup> -N (mg/L)	891±21.52	791±30.1 1	712 ±24.27	976 ±33.41	910 ±45.10	841±29.2 8	1088 ±54.40

## **Highlights**

Biochar affected the anaerobic digestion of food waste/sludge at mesophilic temperature.

Biochar dosage increased specific methane yield while not influencing biogas yield.

Increasing biochar dosage improved reduction capacity of tVFA and ammoniacal nitrogen