

Minimization of Agricultural Waste through Energy Recovery. Evaluation of the Production of Green Biofuels Using Theoretical Models

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Abstract

The transition to a circular economy requires the search of alternative techniques to manage residues through the priority actions established in the waste hierarchy defined by the European Commission. Intensive greenhouse agriculture produces around 1.2 million tons of horticultural waste per year in the so-called European orchard or Spanish plastic sea in Almeria (Spain). These residues come from the main greenhouse crops grown in this region and currently are re-used as animal feed and recycled as organic fertilizer. However, these actions are not enough to avoid their accumulation in the environment. To minimize the negative impact that it means, it is necessary to look for a complementary solution focused on energy recovery actions. In this context, the anaerobic digestion process is proposed as an adequate management tool to address this problem and obtain a green biofuel that supplies an energy demand. The evaluation of the theoretical biochemical methane potential (TBMP) of this waste and the estimation of the physicochemical quality of the biogas produced are carried out by adapting of a theoretical model based on the Buswell-Mueller equation. The results show that the anaerobic digestion of the studied waste can produce a green gas with an intermediate quality (46.38% vol. CH₄, 48.34% vol. CO₂, 5.28% vol. NH₃), an adequate energy potential (15,185 kJ/ Nm³) and an appropriate value of TBMP (0.30 m³ of CH₄ per kilogram of volatile solids), thus reducing the cumulative excess cumulative at a rate of 2.88 Megajoules per kilogram of recovered waste.

Keywords: Waste-To-Energy, Waste Management, Agricultural Residues, Waste Minimization, Anaerobic Digestion

Introduction

In recent decades, waste control has become especially relevant in our society because of the enormous volume of waste generated and the diversity that characterizes it. The volume of production and the type of waste generated make it difficult to manage and remove [1]. The transition from a liberal economy based on the production, consumption and disposal of waste to a circular economy founded on effective management, makes necessary the search for new techniques for recovering residues and transform them into environmentally friendly resources [2,3].

In 2008, the European Commission published Directive 2008/98/EC where was laid down some basic waste management principles. In this scenery, the legislation and policy of EU Member States apply the actions listed in the waste management hierarchy as a priority (Figure 1). In this hierarchy, the EU gives greater priority to waste prevention, followed by preparing for reuse, recycling, or other management techniques, in which energy recovery is included [4].

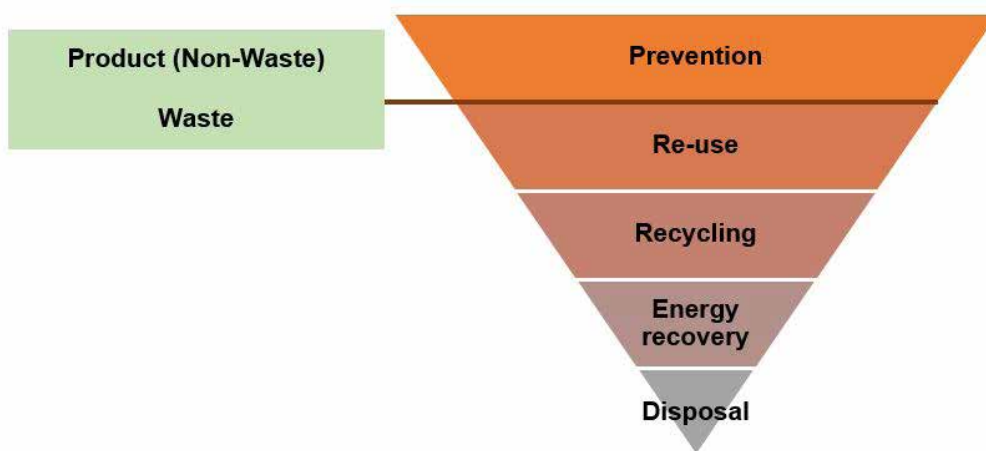


Figure 1: Waste hierarchy to waste management

The application of these techniques allows the important reduction of residues that are disposed of in landfill and the recovery of energy from a finite resource to produce, for example, bioenergy for industrial consumption. In this latter case, anaerobic digestion systems are increasingly attracting attention as a waste management technique [5-8]. On the one hand, this biological process provides a biogas that can be used as an energy source to satisfy an energy demand and, on the other hand, a residual material, called digestate, that can be recycled as a valuable nutrient source of nutrients and soil conditioner [9].

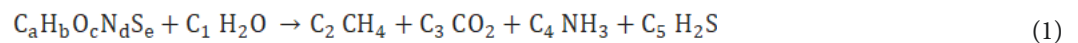
Anaerobic digestion is a biodegradable process by phases in which a series of metabolic reactions, such as hydrolysis, acidogenesis, acetogenesis, and methanogenesis, occur in the absence of oxygen, to biologically transform organic wastes into other recoverable materials [10]. The quality of these materials, biogas and digestate, depends on the physical and chemical characteristics of the substrate fed to the system, such as moisture, volatile matter and the C-N ratio, and operating conditions established in the process, such as temperature, pH and the residence time of the substrate in the anaerobic reactor [11].

The application of this technology is not a novelty. There are many studies regarding the anaerobic digestion of a wide range of different waste, for example, municipal, food, agricultural and industrial residues [12-16]. In fact, anaerobic digestion of horticultural waste is one of the most developed studies in the last decade, together with the evaluation of the digestion of activated sludge, meat waste residues, and pig slurry [17-19]. However, in this research, the knowledge gap is in the evaluation of the anaerobic digestion potential associated with the horticultural waste managed in this study and, in this sense, this work has focused on. These biomass residues come from a waste treatment plant located in the Almeria region (Spain), which receives more than 570,000 tons of waste per year from horticultural greenhouses placed in the southwest of Almeria [20,21]. Here, they are stored and processed to make vegetable compost. The residues collected in this plant are characterized by being a mixture of stems, leaves and unsuitable vegetables for sale derived from the main greenhouse crops in this area, such as tomatoes, peppers, cucumbers, eggplants, zucchinis, watermel-

ons and melons, which are harvested before senescence, providing them a high content of moisture and salt, and high level of biodegradability [22]. In addition, the heterogeneity and seasonality associated with the production method of these studied bioresidues, both in production volume and in composition, make the difference between them and other waste biomass residues analyzed in anaerobic digestion processes so far.

In 2015, approximately 83% of the horticultural waste produced in Almeria was managed to be reused and recycled. Around 75% were treated via composting and the rest were prepared to be used as animal feed [22]. However, the low-quality compost, which causes a fall in sales and, hence, in compost production, and the huge volume of residues produced in this region, about 1.2 million tons per year, make necessary the search of waste management techniques beyond conventional actions to avoid their accumulation in the natural environmental or in the landfills.

In this context, this research work evaluates the anaerobic digestion potential of Almeria greenhouse agricultural waste, hereinafter called GAW, from the evaluation of the biochemical methane potential (BMP) using theoretical models. The application of theoretical studies is useful when access to laboratory facilities is limited or when fast prediction of BMP from a new biomass is required [23]. There is a wide variety of the theoretical models, ranging from complex dynamic models [24,25] to simpler models that required obtaining relevant data from the substrates, such as elemental composition (C, H, O, S and N), component composition (lignin, cellulose, hemicelluloses, starch, total soluble sugars, proteins, and lipids) or chemical oxygen demand [26,27]. Specifically, theoretical models based on the Buswell and Mueller formula modified by Boyle (1) estimate the BMP providing proximate composition data of biomass or waste biomass to digest ($C_aH_bO_cN_dS_e$), considering their total degradation during the process of producing a biogas with high content of methane (CH_4), carbon dioxide (CO_2), and low content in of ammonia (NH_3) and hydrogen sulfide (H_2S).



An important factor to consider in this evaluation when dealing with lignocellulosic substrates is the recalcitrance of the biomass feedstock. This aspect is related to the lignocellulose content in the digested substrate, which in turn directly correlated with the biodegradability of the substrate [28]. The theoretical model applied in this research takes into account the above, adapting the semiempirical model developed by Santolaria Capdevila in 2014 (2) to the waste study of the digestion process from Almeria.

$$TBMP (Nm^3 / kg VS) = C_{VS} \cdot BD \cdot SMY \cdot (TS / VS) \cdot 1.867E^{-2} \quad (2)$$

From this equation, it is possible to estimate the theoretical biochemical methane potential (TBMP) of organic waste, the known percentage of carbon content based on volatile matter (C_{VS}), the biodegradability associated with substrates (BD), the maximum percentage of methane in biogas produced, named specific methane yield (SMY) and the percentage of total solids (TS) and volatile solids (VS) present in the waste to be treated.

In this case, the elemental composition of the GAW has been determined in the laboratory of the School of Engineering of Seville, together with other parameters such a volatile solids (VS), total solids (TS), moisture content (W), and ash content (Ash).

The biodegradability associated with GAW has been estimated from the application of the log-linear relationship provided by Van Soest in 1996 (23), and the assumption of the lignin content (X) of these wastes after an exhaustive review of existing studies which use waste biomass with similar properties.

$$BD (\%) = 100 - 5.41 \cdot X (\%)^{0.76} \quad (3)$$

Once the GAW digestion is known, it is possible to estimate the low calorific value (LCV) from the composition of the biogas produced and the reduction potential associated with the waste energy recovery of the studied waste from the definition of a specific reduction indicator (SRI). This indicator will be an indirect measure of the capacity for minimizing of the GAW accumulated in the environment that this technology may offer as a complementary management tool for these residues.

Materials and Methods

The methodology established to evaluate the potential of the anaerobic digestion (AD) of GAW is shown in Figure 2.

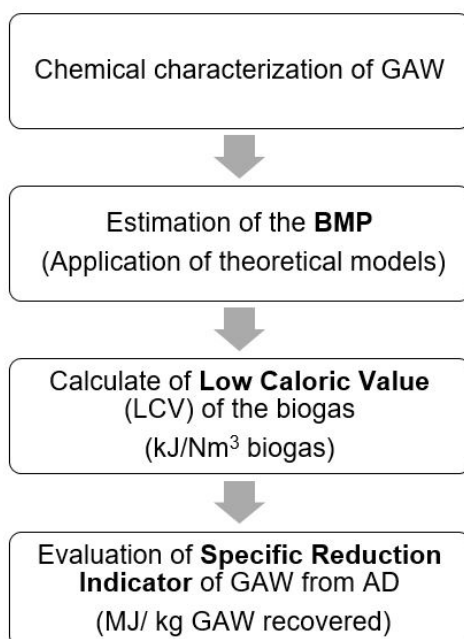


Figure 2: Methodology for evaluating the anaerobic digestion potential of GAW

The chemical characterization of GAW was carried out in the laboratory of the School of Engineering of Seville. Eight waste samples were received throughout the year following a previously defined sampling calendar to measure the heterogeneity and seasonality of these waste. Each sample was collected in the waste treatment plant located in Almeria according to the standard “UNE-EN 14899:2007: Characterization of waste - Sampling of waste materials - Framework for the preparation and application of a sampling plan” [31], and immediately sent to the laboratory for evaluation (Figure 3).



Figure 3: GAW sample collection in the waste treatment plant

Once there, each sample was prepared for analysis (figure 4) following the standard “UNE-EN ISO 14780: 2018: Solid biofuels. Sample preparation” [32].



Figure 4: Sample for analysis. Particle size less than 1 mm

The elemental composition (C, H, N, O, and S) of the samples was determined using a LECO elemental analyzer according with the standards UNE-EN ISO 16948: 2015 [33] and UNE-EN ISO 16994:2015 [34].

The moisture and ash content were determined by thermogravimetric analysis (TGA). For it, the standards UNE-EN 18134 2: 2016 and UNE-EN 18134 3: 2016 [35], [36], and the UNE-EN ISO 18122: 2016 Standard [37], were considered.

The volatile content was quantified using the analytical method defined in the standard ‘UNE-EN 15448:2010. Solid biofuels. Determination of the content of volatile matter’ [38]. The total solid content was calculated from the value of the moisture content associated with each sample analyzed.

Based on these results, the equation 2 was applied to obtain a value of TBMP associated to the studied waste. Previously, it was necessary to estimate the biodegradability (BD) of GAW and the SMY or CH_4 content in the biogas produced in the digestion process. BD was evaluated from the equation 3, assuming a percentage of lignin in the residues and considering the results obtained in other experimental tests for similar biomass waste [28]. The SMY was calculated from Equation 1, where the expression $\text{C}_a\text{H}_b\text{O}_c\text{N}_d\text{S}_e$ define the empirical formula of the substrate fed to the digestion process. In this case, the formula was obtained from the chemical characterization of the GAW samples. Thus, it was possible to calculate the molar constants a, b, c, d, e, which define the chemical Equation, using the following expressions:

$$a = \frac{C}{12.0107}$$

$$b = \frac{H}{1.0079}$$

$$c = \frac{O}{15.999}$$

$$d = \frac{N}{14.0067}$$

$$e = \frac{S}{32.065}$$

The coefficients C_1 , C_2 , C_3 , C_4 and C_5 described in equation 1 were calculated from these mathematical expressions:

$$C_1 = a - b/4 - c/2 + 3 \cdot d/4 + e/2$$

$$C_2 = a/2 + b/8 - c/4 - 3 \cdot d/8 - e/4$$

$$C_3 = a/2 - b/8 + c/4 + 3 \cdot d/8 + e/4$$

$$C_4 = d$$

$$C_5 = e$$

From the value of these coefficients, the SMY parameter to be used in calculating the calculate of TBMP value. This value was compared with the BMP associated with other well-known waste biomass which is commonly used in anaerobic digestion process. The LCV was calculated from the estimated CH_4 content in the biogas and the LCV of this chemical compound. Furthermore, the density of biogas and the yield of the biogas expressed in Nm^3 biogas per kilogram of GAW, were assessed from the biogas composition.

The viability of anaerobic digestion as management tool of the GAW was evaluated using the definition of a Specific Reduction Indicator or SRI (4).

$$SRI \text{ (MJ/kg GAW)} = \text{Yield}_{\text{GAW Biogas}} \text{ (Nm}^3\text{/kg GAW)} \cdot \text{LCV}_{\text{GAW Biogas}} \text{ (kJ/Nm}^3\text{)} \cdot 0.001 \quad (4)$$

This indicator allows assessing the fraction of energy in the biogas produced that could be recovered per kilogram of digested or, in other words, no deposited in landfills or in the environment. Thus, the SRI value is useful to show the capacity of the anaerobic digestion technology for minimizing the accumulated GAW in the environment.

Results and Discussion

Following the methodology described for the evaluation of the anaerobic digestion potential of GAW, the results obtained in this research are shown below.

Table 1 shows the average chemical composition of the GAW studied. These values were obtained from chemical analysis of the eight waste samples received in the laboratory over a year.

| Parameters | Composition (wt. %) |
|-------------------------------------|---------------------|
| Carbon (C) ^(a) | 36.59 ± 5.36 |
| Hydrogen (H) ^(a) | 4.06 ± 0.92 |
| Nitrogen (N) ^(a) | 2.38 ± 0.25 |
| Oxygen (O) ^(a) | 29.92 ± 5.58 |
| Sulphur (S) ^(a) | 0.48 ± 0.18 |
| Volatile solids (VS) ^(b) | 63.20 ± 6.26 |
| Ash content (Ash) ^(b) | 26.57 ± 11.28 |
| Moisture (W) ^(c) | 54.51 ± 14.39 |
| Total solids (ST) ^(c) | 45.49 ± 14.39 |

wt. % (db): weight percentage on a dry base; ^(b) wt. % (db as % TS): weight percentage in dry base expressed as total solids; ^(c) wt. % (ar): weight percentage in the base as received

Table 1: Chemical composition of GAW from Almeria (Spain)

The samples showed a heterogeneous aspect when received in laboratory (Figure 5). However, these differences were associated with the physical aspect rather than with the chemical composition. Carbon, hydrogen, nitrogen, oxygen and sulfur content resulted in similar values between different waste samples, and the volatile matter and total solids resulted between 63 wt. and 45.5 wt.%, respectively. Regarding the moisture content, its value was changing over the course of a year, but it was higher than 50 wt.% in all cases. However, this measure may be considered a low value if compared with the optimal value defined to carry out an adequate anaerobic digestion process [26]. In this case, the moisture content should be externally controlled to achieve at least the 70 wt.% moisture. In this way, the GAW may be assessed as a suitable substrate for the digestion process.



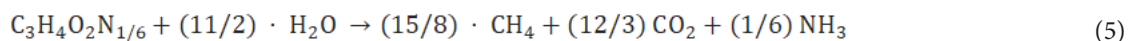
Figure 5: Sample of GAW received in the laboratory before its analysis

TBMP was calculated from the chemical composition results by applying equation 2. Previously, it was necessary to estimate the SMY from the Buswell and Mueller formula, and the BD from equation 3 considering a content of lignin in the GAW of 5.36%, calculated as the average value from the percentage of lignin measured in some feedstocks with the same nature of the wastes assessed in this work (Table 2).

| Feedstocks | X (%) |
|----------------------------|-------------|
| Young garlic shoot | 3.86 |
| Coriander | 5.96 |
| Broccoli | 3.90 |
| Spinach | 9.43 |
| Cauliflower | 3.87 |
| Lettuce | 6.16 |
| Leek | 4.81 |
| Romaine lettuce | 4.88 |
| GAW – Average value | 5.36 |

Table 2: Evaluation of the lignin content of some feedstocks of a similar nature to the GAW [39]

For estimating SMY it was necessary to define the molar constants a, b, c, d, and e, which represent the chemical formula of the substrate in the Buswell and Mueller formula modified ($C_aH_bO_cN_dS_e$), and the stoichiometric coefficients C_1 , C_2 , C_3 , C_4 , and C_5 of this chemical reaction. These coefficients are expressed according to the above-named molar constants. The Buswell and Mueller formula modified particularized for the GAW is shown in Equation 5, where sulphur disappears in the equation because it is so low in the residue evaluated.



From this expression, it was possible to obtain the chemical composition associated with the biogas produced in the digestion of GAW, and hence, the SMY value of these studied residues. Table 3 shows these results. In this case, the methane SMY or the content in the biogas was 46.38 %, resulting in a low value bearing in mind the methane content of the biogas produced from digestion of other substrates ($[CH_4]_{GAW \text{ Biogas}} = 46.4 \text{ vol.}$ vs. $[CH_4]_{Common \text{ Biogas}} = 50 - 80 \text{ vol.}$).

| Compounds | Biogas (vol. %) |
|-----------------|-----------------|
| CH ₄ | 46.38 |
| CO ₂ | 48.34 |
| NH ₃ | 5.28 |

Table 3: Chemical composition of biogas produced in the digestion of GAW

Concerning the calculate of the BD, the application of equation 3 resulted in a value of 80.6% of biodegradability for a lignin content of 5.36% in the GAW.

From these estimations and considering Equation 2, the TBMP value of 0,300 Nm³ of CH₄ per kilogram of volatile solids fed into the digestion process was obtained.

Figure 6 shows a comparison between the biomethane potentials associated with other well-known substrates and the TBMP value of the residues evaluated in this work [40-45].

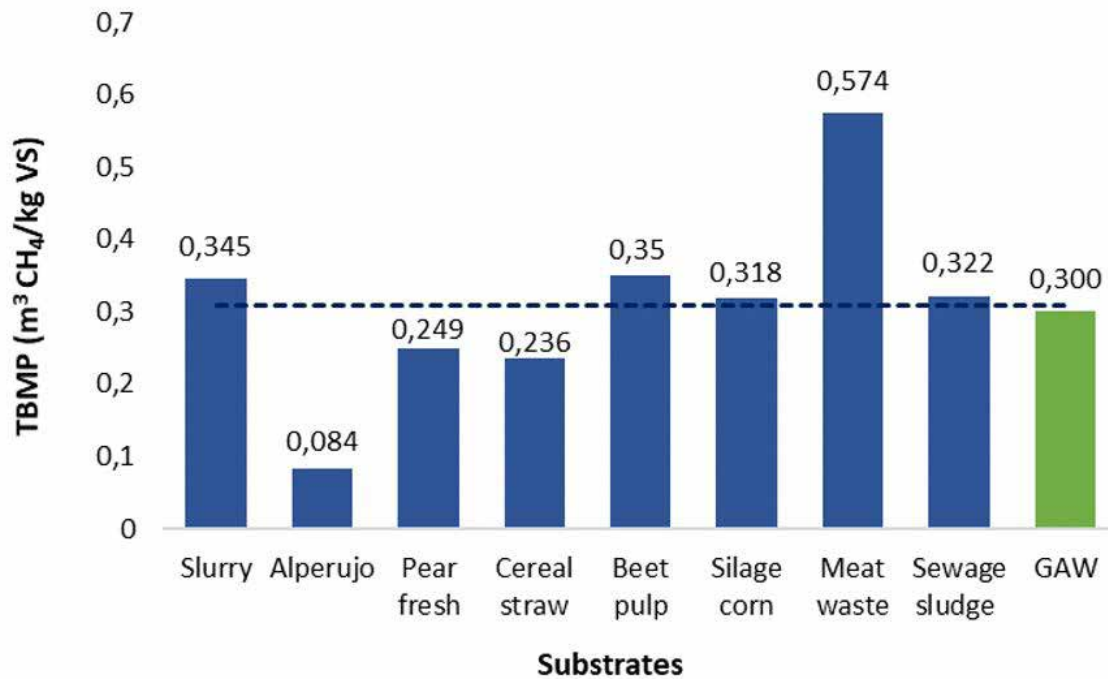


Figure 6: Comparison of the biomethane potential values of GAW and other well-known substrates

As seen in this figure, the GAW TBMP is above the values of the biomethane potential of alperujo, fresh pear fresh and cereal straw, and slightly below the mean value calculated from the potential values of all substrates considered in this evaluation. However, the GAW shows a TBMP value around the values of wastes such as sewage sludge, silage corn, slurry, and beet pulp, which are frequently used in the digestion process. Meat waste has a high value of TBMP. For this reason, both are usually employed together with substrates with low TBMP values in the codigestion process. In this case, considering the moderate biomethane potential value of the GAW and the gap between the moisture content in these residues and the optimal value, the waste codigestion of the studied with meat waste residues could be an option to increase the production of a high-quality biogas.

From the estimated composition for the GAW, the biogas low calorific value of biogas, $LCV_{GAW\ Biogas}$ was estimated. In this case, considering the methane content in the biogas ($[CH_4]_{GAW\ Biogas} = 46.4\ vol.\%$) and the LCV of this compound ($LCV_{CH_4} = 32,740\ kJ/Nm^3$), the result of $LCV_{GAW\ Biogas}$ was $15,185\ kJ/Nm^3$.

Biogas density ($\rho_{\text{GAW Biogas}}$) and biogas production yield ($\text{Yield}_{\text{GAW Biogas}}$) were also assessed from the biogas composition and the value of TBMP_{GAW} . The values obtained for each parameter were 1.31 kg/Nm^3 and 0.19 Nm^3 biogas per kilogram of GAW, respectively.

The specific reduction indicator or SRI was calculated using the equation 4. In this case, considering the values obtained for $\text{LCV}_{\text{GAW Biogas}}$ and the $\text{Yield}_{\text{GAW biogas}}$, the result achieved for this indicator was $2,880 \text{ kJ/kg GAW}$. This value means that it might be possible to produce 2.88 MJ of energy from biogas produced by the digestion of one kilogram of GAW that is currently accumulated in the environment or in landfills.

According to these results, with the aim of clarifying the importance of considering this process as a management tool for the residues studied, consider a scenery in which the accumulation of 17% of the GAW produced in the Almeria region is avoided. This situation would mean the recovery of around 204,000 tons per year to be transformed into suitable biogas for consumption. If the use of anaerobic digestion is proposed to minimize this amount of wastes, the energy recovery from the GAW biogas with the properties estimated in this work may provide approximately 581 TJ/year .

This amount of energy may be recovered in the industrial process, reducing its external energy consume and its operating costs. In this case, the production of 581 TJ/year can replace, for example, about 26% of the thermal energy consumed by a cement plant that produces 700,000 tons of clinker per year.

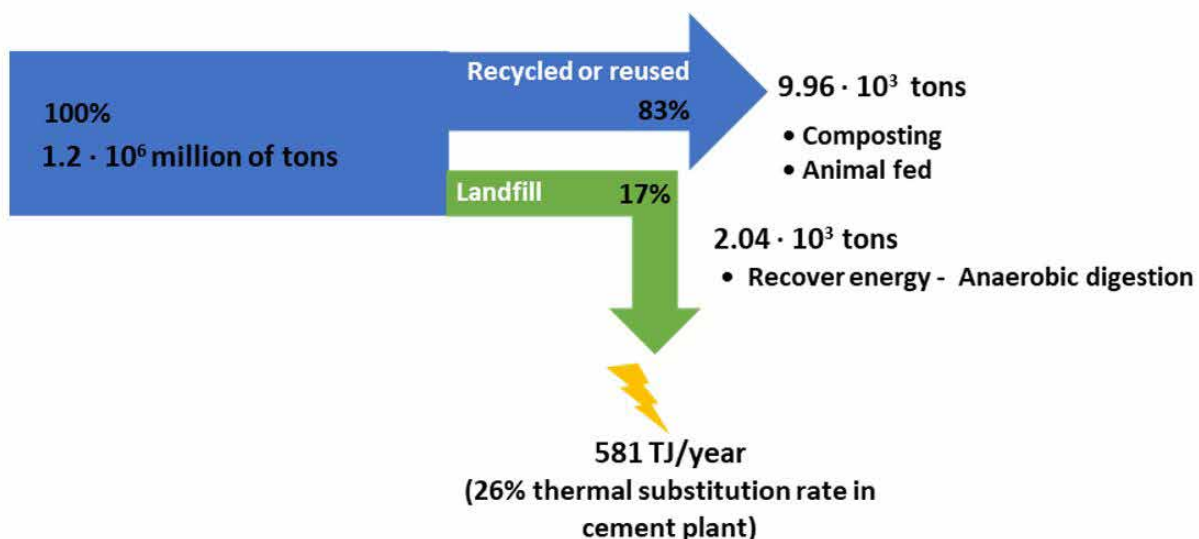


Figure 7: Evaluation of the GAW produced in the region of Almeria. Proposal to minimize the non-management wastes

Conclusion

The Almeria region produces a high volume of horticultural waste from intensive greenhouse agriculture. The 17% of these residues are accumulated in environmental or landfills withing any incentive to be recovered beyond their reuse and recycled as a composting for agriculture use or animal feed. The energy recovery of this fraction of waste from the anaerobic digestion process appears to be a good option to reduce the negative impact of the accumulation of this waste in this region. In this work, the application and adaptation of theoretical models to the GAW properties has allowed one to estimate the composition of the biogas produced from the digestion process of GAW and to evaluate the potential of this technique to use it as a complementary waste management tool.

The digestion of these residues may predict the composition of a biogas of 46.38 vol.% of CH_4 , 48.34 vol.% of CO_2 and 5.38 vol.% of NH_3 , and a biochemical methane potential of $0.300 \text{ Nm}^3 \text{ CH}_4/\text{kg VS}$. The low value of BMP predicted and the moisture content of these wastes, below the optimal moisture content, suggest that, for example, the co-digestion of the GAW with meat residues may provide a biogas with properties better than the biogas generated in a monosubstrate digestion, although this proposal applied to

this particular case would need to be studied in more detail. However, the energy recovery of the biogas produced in mono-substrate digestion may produce around 2.88 MJ per kilogram of GAW treated, which is about 26% of the substitution of thermal energy consumed, for example, in a cement plant that produces 700,000 tons of clinker per year.

The application of theoretical models may be considered a good approximation to evaluate the potential interest for using the digestion process to produce energy from novel waste or when access to pilot scale tests is limited. In this case, it is concluded that GAW may be digested to produce a biogas with adequate properties to be recovered. However, a co-digestion with meat waste residues could provide a biogas with a higher energy potential.

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