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Valuable compounds extraction, anaerobic digestion and composting: A leading biorefinery approach for agricultural wastes

Fernando G. Fermoso[1]*, Antonio Serrano[1,2], Bernabé Alonso-Fariñas[3], Juan Fernández-Bolaños[1], Rafael Borja[1], Guillermo Rodríguez-Gutiérrez[1]

[1]Instituto de Grasa, Spanish National Research Council (CSIC), Campus Universitario Pablo de Olavide – Ed. 46, Ctra. de Utrera, km. 1, Seville, Spain

[2]University of Queensland, School of Civil Engineering, Campus St Lucia, Ed. 49, 4072, QLD, Australia

[3]University of Seville, Higher Technical School of Engineering, Department of Chemical and Environmental Engineering, Camino de los Descubrimientos, s/n, Seville, Spain

ABSTRACT

In a society where the environmental conscience is gaining attention, it is necessary to evaluate the potential valorisation options for agricultural biomass to create a change in the perception of the waste agricultural biomass from waste to resource. In that sense, the biorefinery approach has been proposed as the roadway to increase profit of the agricultural sector and, at the same time, ensure the environmental sustainability. The biorefinery approach integrates biomass conversion processes to produce fuels, power, and chemicals from biomass. The present review is focused on the extraction of added-value compounds, anaerobic digestion and composting of agricultural waste as biorefinery approach. This biorefinery approach is, nevertheless, seen as a less innovative configuration compared to other biorefinery configurations as bioethanol production or white biotechnology. However, any of these

processes has been widely proposed as a single operation unit for agricultural waste valorization and a thoughtful review on possible single or joint application has not been available on literature up to know. The aim of this manuscript is to review the previous and current literature about the potential valorisation of agricultural waste biomass, focusing on valuable compounds extraction, anaerobic digestion and composting of agricultural waste, whether they are none, partially or fully integrated.

1. INTRODUCTION

Agricultural sector and agri-food industry are high volume generating sectors of organic waste, reaching up to 90 million tons per year in 2014 in the EU28 countries (Eurostat, 2017). Among these waste, 46 million tons correspond to lignocellulosic waste (Gil et al., 2015). Moreover, food losses and waste from agri-food sector are estimated at around 30% of global food production, therefore an adequate waste management is crucial in the context of global sustainability (European-Comission, 2015). The uncontrolled decomposition of organic waste can result in large-scale contamination of soil, water, and air. In fact, decomposition of one metric ton of organic solid waste can result in the release into the atmosphere of 50–110 m^3 of carbon dioxide and 90–140 m^3 of methane (Macias-Corral et al., 2008).

For a sustainable agricultural sector, it is necessary moving from the current linear, 'take, make, dispose (waste-creation)' model for resource-consumption, to the systemic, circular alternative of 'reduce, reuse, recycle, regenerate' (Rhodes, 2017). In that sense, the agricultural waste biomass should be considered as a sustainable resource, instead of a waste. To be considered a sustainable resource, it is necessary to develop management methods able to provide economic benefits and, at the same time, to ensure the environmental sustainability of the agricultural sector. In order to integrate the agricultural waste management in a circular system, extraction of high-value compounds, anaerobic digestion and composting is been proposed for a profitable and promising management of agricultural waste. Any of these processes has been widely proposed as single operation unit for agricultural waste valorization and a throutful review on posible joint application has not been available on literature up to know. Extraction of high-value compounds is very interesting due to the recovery of compounds with high economic interest, as well as the partial detoxification of the waste due to the removal of some compounds, which could be undesirables for subsequent biological post-treatments (Martín et al., 2010; Negro et al., 2017; Serrano et al., 2017a). However, recovery by extraction of the added value compounds from agricultural waste usually requires a previous separation of a liquid phase through carrying out a pretreatment process, with the consequent energy consumption (Rubio-Senent et al., 2013b). Anaerobic digestion presents the main advantage of generating methane from the organic waste, which could be used as an energy source due to its high calorific power (35,793 kJ/m^3, at 1 atm, 0 ºC) (Wheatley, 1990). Moreover, anaerobic digestion also allows the partial stabilization of the treated waste, although a post-treatment of the final effluent could be required for its use as an organic amendment (Bustamante et al., 2013). However, different compounds present into the organic waste could inhibit the process. The necessity of high investment costs has also been defined as an important limiting factor for the full-scale implementation of anaerobic digestion processes (Swindal et al., 2010). Composting has been proposed for a long time as a cheap option for agricultural waste management (Ernst, 1990; Gutiérrez et al., 2017). During composting, the organic matter is turned into stabilized humic substances through mineralization and humification. Moreover, pathogens are removed by the heat generated in the thermophilic phase (Dadhich et al., 2012). Unfortunately, although it is a sustainable way to returning the nutrients to the agricultural sector, the economic benefit derived from the compost is relatively low, i.e. 0-9 € per t (Evans and Wilkie, 2010).

Each of the described processes presents high advantages, but also some disadvantages which could limit its implementation. A biorefinery approach combining-value compounds extraction, anaerobic digestion and composting could be an attractive biorefinery approach for agricultural waste. In the extraction step, a high economic benefit could be obtained through the selling of the recovered compounds. The energy required for the whole biorefinery would be obtained from the anaerobic digestion in the second step. Finally, the composting process could allow the total stabilization of the organic matter and the nutrient recovery for the agricultural sector.

The aim of this manuscript is to review the previous and current literature about the potential of different agricultural waste biomasses in a sustainable system based on added-value compounds extraction, anaerobic digestion and composting of agricultural by-products, whether they are none, partially or fully integrated.

2. ADDED-VALUE COMPOUNDS FROM AGRICULTURE BY-PRODUCTS

Recovery of added-value compounds from agricultural waste biomass represents a market opportunity, with multiple applications in pharmacy, cosmetics and food industry. The interest of the different added-value compounds derives from their multiple health benefits, including antioxidant, cardiovascular, antihypertensive, and antiproliferative effects. The economic interest of recovering added-value compounds from agricultural biomass will vary in accordance with the kind of agricultural waste, generation volume, the concentration of desirable compounds, etc. An overview of the potential opportunities for each kind of agricultural waste is reviewed in the present manuscript. The different agriculture wastes or by-products have been clustered in three main groups with similar characteristics, i.e. lignocellulosic agricultural by-products, vegetable by-products, and fruit by-products. Most of reviewed by-products are coming from some of the main crops produced worldwide. The present review is also giving a general idea of possible biorefinery application of any other by-product not reviewed but included in the any of the three clusters.

2.1. Lignocellulosic agricultural by-products

Examples of lignocellulosic agricultural by-products are wheat straw, rice straw, maize stalks, corn straw, ensiled sorghum forage, barley, sunflower or cotton gin waste. These lignocellulosic agricultural by-products contain variable amounts of cellulose, hemicellulose, lignin and small amounts of protein, pectin, wax, and inorganic compounds. The use of lignocellulosic agricultural by-products has been gaining interest. From lignocellulosic agricultural by-products, several products as biofuels, chemicals and other biomass-derived with high added value product can be obtained through the integration of clean processes (Abraham et al., 2016).

Depending on the refinery system, these feedstocks could undergo various chemical or mechanical pretreatment in order to facilitate valuable products extraction. Among the different pretreatments, autohydrolysis or hydrothermal process is an interesting and well-established eco-friendly process in which lignocellulosic material is pretreated with high-pressure steam. After the hydrothermal pretreatment, a liquid phase and a solid phase are generated. Through the hydrothermal process, hemicellulose is extracted into the liquid phase. The acidic groups bonded to the hemicellulose are released at high temperature. These acids, mainly acetic acid and hydronium ions coming from water auto-ionization, enhance the hydrolysis of the solid lignocellulosic material that leads to the further solubilization of hemicellulose. This process also facilitates hydrolysis of cellulose. The obtained liquid phase is rich in products of degradation of cellulose, hemicellulose, and lignin. The introduction of a detoxification step or removal of these dissolved compounds is highly recommended if a subsequent bioprocess wants to be implemented (Fernández-Bolaños et al., 1998; Yu and Christopher, 2017). The liquid phase (hydrolysate) contains monomeric and oligomeric hemicellulose sugars, sugar degradation products, acetic acid, extractives and phenolics compounds derived from the acid-soluble lignin. These last compounds can be used for the production of compounds of interest for the health, cosmetic and food industries (Moure et al., 2006). The antioxidant and antimicrobial potential of depolymerized lignin fraction produced by mild acid hydrolysis of lignocellulosic material has been reported (Cruz et al., 2007). This type of compounds that inhibit growth and metabolism of microorganism can be selectively removed by solvent extraction (Fernández-Bolaños et al., 1998). The utilization of crude extracts as antioxidants, instead of a pure compound or purified fraction, is a frequent approach. It is an alternative more favorable from an economic point of view and, even in some case, the crude extract has presented higher antioxidant capacity than the mixture of main component (Rubio-Senent et al., 2014).

The hemicellulose fraction can be used as hydrogel being considered as an alternative for the development of new polymeric blends for food packaging (Ruiz et al., 2013). The oligomeric form can be used as functional food ingredients (Mäkeläinen et al., 2010) and the monomeric form can be fermented to ethanol or xylitol (Avanthi et al., 2017). Xylooligomers with prebiotic activity in addition of ingredients for food by stimulating the growth of bacteria in the colon and improving the host´s health, are also currently considered interesting in cosmetic for skin care, in pharmaceutical industries by its multiple biological properties (anticarcinogenic, immunomodulatory, antimicrobial activity or by their ability to decrease glucose and cholesterol) and in agriculture as ripening agents (Álvarez et al., 2017).

The remaining solid phase is enriched in both cellulose and lignin (Rodríguez-Gutiérrez et al., 2008; Rodríguez-Gutiérrez et al., 2014; Ruiz et al., 2013). Lignin is an amorphous polyphenol with high molecular weight, which can vary in structure according to the extraction method and plant source (Fernández-Bolaños et al., 1999; Rodríguez-Gutiérrez et al., 2014). Nowadays, the major lignin use is as fuel, and only a few products like vanillin are produced (Singh and Ghatak, 2017). However, lignin is becoming a high interesting product. For example given to its radical scavenging properties for the formulation of plastic and cosmetic (Guilhen et al., 2017; Morganti, 2016). Production of low-molecular weight compounds with potential uses as surfactants or additive for liquid fuels can be obtained after lignin fractionation with alcohols (Cabrera et al., 2016). Furthermore, lignin represents a removable low-cost alternative to natural antioxidants and could be used as a component of polymer composites or of the polysaccharides hydrogel films as antioxidant carriers or active packaging (Aguié-Béghin et al., 2015). Lignin is an important component of dietary fiber, being nontoxic and biocompatible, which undergoes minimal changes in the body (non-fermentable). Lignin has been proven to bind various bile acids and detoxify harmful metabolites inhibiting colonic carcinogenesis (Camire and Dougherty, 2003). Concretely, a fraction containing lignin from olive stones bound significantly more bile acid than any other fraction and an amount similar to that bound by cholestyramine (a cholesterol-lowering, bile acid-binding drug). Therefore, this lignin fraction from olive stone could contribute to the reduction of serum cholesterol levels and a decreased risk of bowel cancer (Rodríguez-Gutiérrez et al., 2014).

Although all agricultural by-products have approximately the same composition, the yield in hemicellulosic sugar or the development of valuable-added products differs, when applying the same conditions or type of pretreatment. It is thus important to study each raw material to determine the appropriate production conditions. For example, in the case of maize bran, which is a rich source of dietary fiber and phenolic antioxidants, the hemicellulose fraction is a complex heteroxylan consisting mainly of xylan backbone with arabinosyl side. The production of feruloylated arabinoxylan-oligosaccharides from maize bran by autohydrolysis may provide health benefits, including prebiotic effect and prevention of detrimental oxidation reaction (Rose and Inglett, 2010).

2.2. Vegetable by-products

In general, by-products from handily and commercialization of vegetables such as tomato, onion, potato or carrot, have been traditionally used as animal feedstuffs, for dietary fiber production and fuel production. Most of the vegetable by-products contain the same bioactive compounds than the vegetable itself (Table 1). Due to this, most of the vegetables by-products have been shown to present similar health benefits than the vegetable (Table 2). Therefore, an interesting approach is their use as source of phytochemicals and bioactive compounds.

2.2.1. Tomato by-products

The by-products resulting from tomato processing are mainly peel and seeds (Gharbi et al., 2017). Tomato seed oil has attracted interest by its high content of unsaturated fatty acid with over 50% linoleic acid (da Silva and Jorge, 2017). Tomatoes have been associated with reduced risk of some types of cancer and other diseases. These beneficial effects have been linked to the content of lycopene and other carotenoids as β–carotene and lutein (Fattore et al., 2016). Other bioactive compounds, particularly polyphenols, has been shown to contribute to the antioxidant effect. The major phenol compounds of tomatoes are the flavanones, naringenin glycosylated derivatives and flavanols, quercetin, rutin and kaempferol glycoside derivatives (Kelebek et al., 2017). Gharbi et al. (2017) showed that the antioxidant composition of tomato peels and tomato seeds Lycopene is mostly associated with the water insoluble fraction of the peel. Furthermore, tomato seed has been shown to be a source of pectin (Morales-Contreras et al., 2017) and protein of high-quality (Moayedi et al., 2016). At the industrial scale, the tomato waste is been using for animal feed or fertilizer, however, new techniques are been studied in order to extract pectin and/or bioactive components. These techniques include solvent extraction by stirring and heating, heat refluxing extraction, microwave or ultrasonic in combination with subcritical water (Grassino et al., 2016).

2.2.2. Onion by-products

The major by-product resulting from industrial peeling of onion bulbs is the brown skin, the outer two fleshy leaves and the top and bottom bulbs. Onions, as well as its by-products, are an important source of several phytonutrients as flavonoids, fructooligosaccharides (FOS) and thiosulfonates and other sulfur compounds (Liguori et al., 2017). Quercetin and kaempferol glycosides are the predominant polyphenols found in onions. They are present in higher concentration (280-400 mg/kg) than in other vegetables (Sharma et al., 2015). Anthocyanins are also present in red onions (Wiltshire et al., 2017). Onion by-products are also a source of inulin, a polysaccharide known as fructan, which is used as a dietary fiber with prebiotic effect (Smith et al., 2015) or as an energy source (Hughes et al., 2017). The onion waste is not suitable for food animal or for organic fertilizer, the only option is landfill, with high economic costs, and adverse environmental impact. The pretreatment methods studied for its valorization are based on technologies like organic extraction, supercritical carbon dioxide, supercritical water treatment, microwave, microwave assisted, hydro diffusion and gravity or high-pressure processing (Sharma et al., 2016).

2.2.3. Carrot by-products

Carrot is a rather inexpensive and highly nutrition vegetable. It contains natural antioxidants, including phenolic compounds and carotenoids. Carrot pomace, a major by-product of carrot juice processing, represents a rich source of bioactive compounds with antioxidants activities (Jabbar et al., 2015). Carrot pomace represents a valuable natural source of α– and β–carotene, with a total carotene content up to 2 g per kg dry matter. The pomace has been used for the production of antioxidant dietary fiber powder and for the extraction of pectin (Jabbar et al., 2015; Jafari et al., 2017).

The phytochemical profile of carrot by-product is composed of hydroxycinnamic acid derivatives, particularly four chlorogenic acids (5-O-caffeoylquinic) and six derivatives dicaffeoylquinic acids (Sánchez-Rangel et al., 2016) with many biological functions including antioxidant, antiviral, antimutagenic, anti-inflammatory, cardioprotective, antiobesity and therapy on wound healing (Akhtar et al., 2017; Bagdas et al., 2014).

After the carrot juice processing, around the 50 % of the raw material is discarded as a waste or use for feed animal purpose. Several pretreatments have been studied to extract the bioactive compounds like refluxing, boiling and heating, but some of the more promising technique is based on the ultrasound-assisted extraction up to 60 ºC (Jabbar et al., 2015), or the aqueous two-phase system extraction (Sánchez-Rangel et al., 2016).

2.2.4. Potato by-products

Potato peels, a by-product of potato processing, are available in large amounts, and, since peels have much more phenolic compounds compared to tubers, these phenolic compounds have a potential application in food and non-food applications. Aqueous peel extracts were shown to be used as an antioxidant on different oils (Amado et al., 2014), in minced horse mackerel (Sabeena Farvin et al., 2012) or in processed lamb meat (Kanatt et al., 2005). Also, this extract has been used as a source of phenolic compounds. Potato peels have relatively high content in phenolic acids, especially chlorogenic acid. In addition, the potato peels are also a source of water-soluble polysaccharides (Jeddou et al., 2016). Beneficial anticarcinogenic properties of potato glycoalkaloids have been recently reported (Friedman et al., 2017).

Nevertheless, potato glycoalkaloids at high concentration might be potentially harmful to human. Recently, the potato peel by-products have also been used as an effective biosorbent for removal of toxic metal agents from water (Azmat et al., 2016). Commonly used techniques to obtain antioxidant extracts from potato by-products are organic solvents, like ethanol, or pressurized liquid extraction, microwave-assisted extraction, and subcritical water extraction or even thermal treatment at 121 ºC which allows the application of the further biorefinery approach (Pathak et al., 2017).

2.3. Fruit by-products

Similar to vegetable by-products, most of the fruit by-products contain the same bioactive compounds than the fruit itself (Table 1). Due to this, most of the fruit by-products have been shown to present similar health benefits than the fruit (Table 2).

2.3.1. Grape by-products

The main by-product of the wine industry is known as grape pomace and consists mainly of skin, seeds, stems and remaining pulp. Its composition varies considerably depending on grape variety and technology of wine making. The moisture percentage varies from 50-72%. The insoluble residues from this material have a lignin content ranging from 17-24%, cellulose varying from 27-37% and protein content is lower than 4% (Teixeira et al., 2014). Grape pomace has been used as soil conditioner, as a source of fibers and energy by methanization (Barba et al., 2016). Also it has been used for seed oil extraction, as a source of tannins (Bindon et al., 2017) and as a source of protein for animal feed (Brenes et al., 2016). During the winemaking process part of the phenolic compounds in grape are transferred to the wine, but a high proportion still remains in the waste, especially in the grape pomace (Ribeiro et al., 2015). Numerous studies have demonstrated that these phenolic compounds exhibit health-promoting effect ascribed with cardioprotective, neuroprotective, anti-inflammatory, anticarcinogenic and other health benefits (Panzella and Napolitano, 2017) and are active against pathogenic bacteria, virus, and fungi (Friedman, 2014).

Grape contains a large amount of different phenolic compounds distributed in pulp (10%), in seeds (60-70%) and in the skin (28-35%). The most predominant polyphenols found in grape pomace are a) phenolic acids- caffeic, gallic, protocatechuic, 4-hydroxybenzoic and syringic acid, b) phenolic alcohols- hydroxytyrosol, c) flavonoids- (+)-catechin, catechin dimer, (-)-epicatechin, epicatechin gallate trimer, procyanidin B1 and B2, quercetin-3-O-rhamnoside, luteolin, d) stilbenes, with the presence of trans-resveratrol e) proanthocyanidins, also known as condensed tannins, complex phenols of high molecular weight and anthocyanins (Teixeira et al., 2014). These grape pomaces are currently used as a source of resveratrol and flavonoids, which are used as food supplements and for the isolation of anthocyanins for use as a substitute of synthetic colorants and for preparation of dietary fiber and polyphenol-rich extracts (Zhang et al., 2017). Moreover, grape seeds are a rich source of other antioxidant compounds such as vitamin E, which constitutes a family of lipid-soluble antioxidant compounds, containing a saturated (tocopherol) or unsaturated (tocotrienols) isoprenoid side chain (Barba et al., 2016). Grape seeds are also a good source of polyunsaturated fatty acids (PUFA), mainly linoleic acid, followed of α–linolenic and oleic acid (Ribeiro et al., 2015).

Besides the conventional methods based on the heating process and/or solid liquid extraction industrially used for antioxidant extracts, new techniques are been studied to improve the extraction and to diminish the thermal degradation of phenols, like pulsed electric fields, high voltage electrical discharges, pulsed ohmic heating, ultrasounds, microwave assisted, sub- and supercritical fluid extraction, high pressure, accelerated solvent or extraction assisted by hydrotropic solvents (Barba et al., 2016). The extraction method commonly used to produce commercial extracts is done by hydroalcoholic solvents, mainly ethanol (Teixeira et al., 2014).

2.3.2. Orange and lemon by-products

The residue from orange and lemon juice extractions industries are a good source of bioactive ingredients such as essential oil, which consist mainly in monoterpene (limonene) and triterpenoids as limonoids. Other bioactive compounds are dietary fiber (DF); pectin; ascorbic acid; phenols (coumaric, caffeic and ferulic acids); and flavonoids, mainly flavanones glycosides (hesperidin, naringin and narirestin), flavones (hesperetin, naringenin), flavones aglycon (luteolin) and polymethoxylated flavones (tangeretin) ((M'hiri et al., 2017). Flavonoids concentration in citrus peels is higher than in juice and seeds (Tao et al., 2014). These compounds inhibit the cell growth of a large group of microorganisms and may be useful as antiviral, antifungal and antibacterial agent (Damian-Reyna et al., 2016). In recent years has been accumulating evidence of the cancer-preventive effect of limonene (Mitropoulou et al., 2017). Also, D-limonene has been clinically used to dissolve cholesterol-containing gallstones. Because of its gastric acid neutralizing effect and its support of normal peristalsis, it has also been used for relief of heartburn (Sun, 2007). Another important compound recovered from the peel is pectin, which is usually extracted with hot dilute acid and used as thickening agent, gelling agent, and stabilizer in the food industry (John et al., 2017). Citrus limonoids (CLs) are a group of highly oxygenated terpenoid secondary metabolites found mostly in the seeds, fruits and peel tissues of citrus fruits. Represented by limonin, the aglycones and glycosides of CLs have shown to display numerous pharmacological activities including anticancer, antimicrobial, antioxidant, antidiabetic and insecticidal among others (Gualdani et al., 2016).

2.3.3 Apple by-products

The potential use of apple by-products to isolate specific phytochemicals for application in food or dietary supplements contributes to the recovery of these by-products. The by-products resulting from processing of apple fruit represent approximately 25-30 % of fruit weight (Schieber et al., 2003) and are made of peels, seeds and flesh. Although, normally is used as an animal feed or wasted, the apple pomace is utilized for the recovery of valuable compounds, such as dietary fiber and polyphenols (Sudhaa et al., 2007; Parra et al., 2015). The production of pectin from apple, 10-15% of apple pomace, on a dry weight basis, have been established even an industrial scale. Apple pomace is also consider a good source of natural antioxidants with important properties that include antimicrobian, anticancer and cardiovascular-protective activities (Eberhardt et al., 2000) (Sesso et al., 2003). The major antioxidants isolated and identified include the flavanols quercetin glycosides, kaempherol, catechin, and procyanidins

(Kammerer et al., 2011). A method for the combined recovery of pectin and polyphenol was established (Schieber et al., 2003). Apple seeds has been used for the recovery of oil and bioactive compounds. The oil extractable from seed of apple stood out for its high content in α- and γ-tocopherol and β-sitosterol (Da Silva & Jorge, 2017) and is interesting for their use in cosmetic, food and pharmaceutical application (Walia et al., 2014).

2.3.4. Mango by-products

Mango processing by-products (peel, kernel, and seed) comprise 35-60% of the total fruit weight thus representing a potentially high volume resource of exploitable biobased chemicals and material within the context of biorefinery (Matharu et al., 2016).

The analysis of the oil extracted from fruit seeds verifies the presence of bioactive compounds, such as phytosterols, as well as phenolic compounds (gallic acid, salicylic acid, epicatechin, and quercetin) and tocopherols which present antioxidant capacity (da Silva and Jorge, 2017). The kernel contains high amount of fiber, starch and hydrolysable tannins, which possess antimicrobial activity (Jahurul et al., 2015). The mango peels have high amount of extractable polyphenols, hydrolyzable tannins, flavonoids, xanthones and anthocyanins (Dorta et al., 2014) and a high antioxidant activity (Sabino et al., 2015). Mango by-product is also a rich source of highly esterified pectin (Matharu et al., 2016). For the industrial valorization of mango by-products it is necessary to use heating but in combination with additional techniques to reduce the volume of effluent generated, like microwaves, pulsed electric energy or ultrasound (Matharu et al., 2016).

2.3.5. Papaya by-products

The papaya peel flour has a high amount of ascorbic acid and lycopene (Sabino et al., 2015). The carotenoid content, including lutein, cryptoxanthin, and β-carotene, are also significant in papaya by-product (de Moraes Crizel et al., 2016). It was recently found the presence of terpenoids in papaya extracts with important antibacterial activity (Lawrence et al., 2015). The seed oil is rich in oleic acid, α-tocopherol, carotenoids and a significant source of phytochemical (da Silva and Jorge, 2017). Oil is obtained from the papaya seed by ultrasound-assisted extraction, extrusion-expelling processes and solvent and aqueous enzymatic extraction (Cheok (Cheok et al., 2017) et al., 2016). Papain, a proteolytic enzyme used in food industries as a meat tenderizer, for stabilizing and chill-proofing beer and in baking processes, is recovered from the latex of papaya fruit (Rocha et al., 2016).

Conventional treatment based on grinding and heating is commonly used to revalorized the papaya by-products, but new technologies are been also studied to improve the extractions, diminishing the time and the degradation products, like Pulsed Electric Fields (Parniakov et al., 2016).

2.3.6. Banana by-products

Banana peel is a waste produced in large volume annually by food-processing industries. For 1 ton bone-dry banana peel could be obtained 430 kg of protein or 170 kg of citric acid, 170 kg of pectin, 325 m^3 of ethanol, and 220 m^3 of methane (Pathak et al., 2017). Banana by-products are also a good source of bioactive compounds with high added value (Singh et al., 2016). Banana by-products have a high phytochemicals concentration that consist mainly of phenolic compounds, flavonoids, proanthocyanidins (Vu et al., 2016), carotenoids (α - and β–carotene and xanthophyll) (Davey et al., 2009), sterols and triterpenes (Hernández-Carranza et al., 2016) and saponin (Siddique et al., 2017). Banana peel contained large amounts of dopamine and L-dopa, catecholamines with a significant antioxidant activity (González-Montelongo et al., 2010). For the extraction of the bioactive compounds, techniques like the use of solid-liquid with hot organic solvents, alkaline, enzyme-assisted, ultrasound, and supercritical fluid extraction, have been studied (Hernández-Carranza et al., 2016).

2.3.7. Pineapple by-product

Pineapple processing generates large quantities of solid and liquid waste. The peel represents the largest portion (30-42% w/w), followed by the core (9-10%) and stem (25%). These by-products could be a potential source of sucrose, glucose, fructose, saccharose, fiber, bromelain and phenolic compounds (Dorta and Sogi, 2016).

Bromelain, a protease with a wide range of industrial application in food, beverage and cosmetic, may be recovered from pineapple by-products including stem, fruit, leaves, and peel. The high demand for bromelain has led to the need for a high purified bromelain production at low cost (Ramli et al., 2017) like membrane systems (Nor et al., 2017) or aqueous two-phase system (Upadhyay et al., 2010).

Pineapple by-products may be utilized for the recovery of phenolic compounds. Gallic acid, catechin, epicatechin and ferulic acid were found to be the main polyphenolic in pineapple peels (Li et al., 2014). Also, a high content of anthocyanin, α and β–carotene were also found (Da Silva et al., 2014). The most common system used to extract them are based on solid-liquid extractions, using water or organic solvents (Upadhyay et al., 2010).

Pineapple pomace containing high amount of dietary fiber was used for fortificant extruded products (Selani et al., 2014). Peel flour, which presents a good prebiotic potential, could be used to support probiotic bacteria in the gut (Dorta and Sogi, 2016).

2.3.8. Olive oil by-products

The olive oil industry generates by-products from the three olive oil extraction system, the liquid (alpechin) and the solid (orujo) with a 45-55% of humidity, and from the two olive oil extraction system, the solid with high humidity (70-80%) called alperujo. The presence of bioactives compounds in all these by-products are similar, being concentrated in the solid phases, hence it is necessary to apply a pretreatment to extract them and to facilitate the solid/liquid separation. Despite many studies have been carried out to treat the alperujo and the orujo (chemicals, physicals, biological or mix systems) only the thermal treatments are been industrially used (Fernández-Bolaños et al., 2002). Two kinds of thermal treatments are been used, a malaxation at 55-65 ºC and a steam treatment at 150-170 ºC. The effects of the thermal treatment are based on the solubilization of sugars and phenols, the easier separation of the phases, the final solid with lower humidity (30-55%) and the oil richer in minor components and concentrated two or three times in the final solid (Lama-Muñoz et al., 2011).

The bioactive compounds present in the olive oil industry are mainly the phenols and sugars. Hydroxytyrosol is one of the most actives and remarkable phenol in the olive because of its high anti-oxidant, anti-inflammatory and anti-platelet potency in humans (Fernández-Bolaños et al., 2008). In addition to hydroxytyrosol, there are other interesting phenols in the by-products such as 3,4-dihydroxyphenylglycol, triterpenic acids, lignans and secoiridoids, all of them with potential anti-inflammatory and anti-oxidant activity (Rubio-Senent et al., 2012). The application of solid/liquid extraction with an organic solvent or thermal pretreatment following by chromatographic or liquid/liquid extraction leads to produce a commercial extract rich in phenols being the hydroxytyrosol the majority (Rubio-Senent et al., 2013b). Beside the phenols, the carbohydrate fraction has been shown biological activities, like oligosaccharides with low or high molecular weight, pectins or phenolic glucosides, with antioxidant, antiproliferative or probiotic activities (Rubio-Senent et al., 2015b).

3. ANAEROBIC DIGESTION

Anaerobic digestion of agricultural waste has been widely reported in literature. Biomethanization of agricultural waste not only delivers directly usable energy, as biogas but also retains the nutrients contained in the biomass (Braun, 2007). The obtained energy could be used for the energy demands of the extraction of added-value compounds in a biorefinery system, but also to supply to other energy requirements such as water pumping. The three groups defined in the previous section has been followed. The main considerations for the biomethanization of each group are described below.

3.3. Lignocellulosic agricultural by-products

Anaerobic digestion of lignocellulosic by-products is usually characterized by a low biodegradability, especially due to the lignin content of this kind of by-products. For example, winter harvested switchgrass showed a much lower biodegradability than the fresh summer harvested switchgrass, probably due to a much lower lignin fraction than the winter harvested switchgrass (Frigon et al., 2012; Table 3). In order to enhance biodegradability, trials applying strong pretreatments such as steam explosion and/or alkaline dosage are usually done in order to mainly breakdown the lignin fraction (Table 3) (Bauer et al., 2009; Estevez et al., 2012; Frigon et al., 2012; Monlau et al., 2012; Rodriguez et al., 2017; Sambusiti et al., 2013; Xie et al., 2011). Other authors have proposed the addition of acid compounds instead (Hassan et al., 2016; Monlau et al., 2012). The selection of a pretreatment must be in accordance with the composition of the agricultural waste to be treated. Acidic pretreatments removed more than 90% of hemicelluloses and uronic acids whereas alkaline and oxidative pretreatments were more effective in dissolving lignin (Monlau et al., 2012). For example, Xie et al. (2011) reported that a thermal pretreatment (100ºC) with NaOH addition allowed the solubilization of 65.6% of lignin, 36.1% of hemicellulose and 21.2% of cellulose.

As an alternative to the chemical addition in thermal pretreatments, steam explosion could pretreat the lignocellulosic waste without the addition of chemicals and with minimal sample handling (Estevez et al., 2012). Steam explosion could increase the methane yields up to 50%, (Monlau et al., 2012). Hemicelluloses solubilization debilitates the lignocellulosic structure of the substrate and facilities thus, its microbiological degradation. However, the overproduced methane due to the pretreatment is not necessarily enough to cover the energy requirements of the applied pretreatment (Serrano et al., 2017b).

It is not so unusual to observe a decrease of methane production after using a high-temperature pretreatment. The decrease of methane yield when high temperatures are applied is usually attributed to the formation of substances inhibiting the microorganisms responsible for the anaerobic digestion process (e.g. phenolic compounds or furan derivatives) as well as to the loss of sugars due to pseudo-lignin formation (Ghasimi et al., 2016; Monlau et al., 2012; Rodriguez et al., 2017). The inclusion of an extraction step of added values after the pretreatment and before the digester reduce this drawback (Serrano et al., 2017a). During the extraction step, the concentration of some inhibitors can be reduced in the anaerobic influent, whereas the benefit derived from the recovery of high added-value compounds could compensate the extra energy requirements of the pretreatments.

3.4. Vegetable by-products

Anaerobic digestion of vegetable by-products is mainly characterized by high fiber content, mainly hemicellulose and, to a lesser extent, cellulose (Ji et al., 2017). Vegetable by-products present higher methane yields and biodegradability values than the obtained for lignocellulosic by-products, probably due to a lower content of lignin (Table 3 and Table 4). Reported values of methane yield for vegetable by-products can reach really high values, e.g. 390 mL CH_4/g VS (Volatile Solids) for onion waste (Ji et al., 2017; Menardo and Balsari, 2012), 320 mL CH_4/g VS for potato waste (Parawira et al., 2004), or 198 mL CH_4/g VS for carrot waste pomace (Garcia et al., 2011). Moreover, biodegradability values were also higher than the obtained for lignocellulosic by-products (Table 3 and Table 4). Typical biodegradability values for vegetable by-products have been reported in a range from 50 to 70%, in VS (Garcia et al., 2011; Ji et al., 2017; Menardo and Balsari, 2012). The high methane yield and biodegradability values advise against the implementation of pretreatments methods to enhance the biomethanization step, although it would be necessaries for some previous extraction procedures.

Unfortunately, some drawbacks have been reported for the anaerobic digestion of vegetable by-products. Several vegetable wastes have rather low pH values, e.g. a pH of

3.2 for onion, a pH of 5.6 for Lettuce, or a pH of 5.06 for pepper (Ji et al., 2017; Menardo and Balsari, 2012), which can cause the acidification of the anaerobic process. In that sense, Lubberding et al. (1988) described the accumulation of butyric and valeric acid during the biomethanization of onion waste. Moreover, although biodegradability during the biomethanization is relatively high, a post-treatment would be desirable for the total stabilization of the organic matter. The employment of alkaline pretreatments in the extraction process could compensate the low initial pH of the vegetable waste, whereas the subsequent composting process could mineralize the anaerobic effluent.

3.5. Fruit by-products

Anaerobic digestion of fruit by-products is mainly characterized by high sugar and fiber content, mainly hemicellulose (Bouallagui et al., 2005; Ji et al., 2017). As in vegetable by-products, lignin fraction is lower than the reported for lignocellulosic by-products. However, the presence of husks in some fruit by-products, such as olive husks or strawberry achenes, could entail significant lignin concentrations (Serrano et al., 2017a; Serrano et al., 2017b; Siles et al., 2013). Reported values of methane yield for fruit by-products widely vary according to the treated fruit, or if the biomass is composed of the whole fruit or just the peel of the fruit (Table 5). The highest reported methane yields were reached for pineapple waste, i.e. 413 mL CH_4/g VS, kiwi waste, i.e. 371 mL CH_4/g VS (Menardo and Balsari, 2012), and two-phase Olive Mill Solid Waste (OMSW), i.e. 373 mL CH_4/g VS (Rincón et al., 2013). However, significant lower methane yield values were reported for fluted pumpkin peels and banana peels, i.e. 164 mL CH_4/g VS and 188 mL CH_4/g VS, respectively (Bardiya et al., 1996; Dahunsi et al., 2016) (Table 5). The marked difference in the methane yield could be due to the carbohydrates percentage into the waste and/or the different fiber composition (Bardiya et al., 1996; Ji et al., 2017). Biodegradability values of fruit by-products could reach values higher than the reported for vegetable and lignocellulosic by-products. In fact, biodegradability values up to 90%, in VS, for orange peel (Siles et al., 2016), 90%, in VS, for strawberry extrudate (Siles et al., 2013), or 73.8%, in VS, for pineapple pulp (Namsree et al., 2012) (Table 5) have been reported.

In order to improve the biomethanization of fruit by-products, several kinds of pretreatments have been proposed. Pretreatments focus on the removal or reduction of potential inhibitors, this results in an improvement of the process stability and/or the methane yield. For example, removal of D-limonene, which is an economically interesting compound, was reported as necessary to avoid the inhibition in the biomethanization of orange peel (Martín et al., 2010; Siles et al., 2016). The recovery of phenolic compounds also reduces the inhibition risk on the biomethanization of waste such as OMSW (Serrano et al., 2017a). Additionally, other authors have reported improvements in the methane from 230 to 312 mL CH_4/g VS, i.e. 36% higher, in the biomethanization of strawberry extrudate due to the reduction of the lignin content through the removal of the achenes from the strawberry extrudate (Siles et al., 2013). However, the application of thermal pretreatments for increase the solubilization of the waste does not usually result in significant methane yield improvements (Dahunsi et al., 2016; Rincón et al., 2013). The low reported improvements could be a consequence of the low cellulose content of fruit waste, which means that the hydrolytic process during anaerobic digestion is not the rate-limiting step (Ji et al., 2017). Therefore, the energy requirements of some pretreatments only could be compensated if the extraction of high added-value compounds is implemented prior the biomethanization (Serrano et al., 2017b).

The anaerobic digestion of fruit by-products could present some problems if the process is not correctly monitored. For example, the low pH values of fruit by-products can result in the acidification of the anaerobic process. Moreover, an additional nitrogen source could be required due to the low nitrogen content of some fruit waste (Belhadj et al., 2014; Gil et al., 2015). To avoid these inconveniences, several authors have proposed the addition of co-substrates to provide buffering capacity, to stabilize the pH, and/or to optimize the nutrient balance (Belhadj et al., 2014; Suryawanshi et al., 2013).

4. COMPOSTING OF DIGESTATE

After anaerobic digestion process, a wet residue called digestate is produced. The digestate is a mixture of partially stabilized organic matter, microbial biomass and inorganic compounds (Serrano et al., 2014b). Due to its content on carbon, nitrogen and phosphorus, several authors have proposed that the digestate could be employed as nutrient source for agriculture (Alburquerque, J. A. et al., 2012; Kaparaju et al., 2012; Vaneeckhaute et al., 2017). Therefore, the nutrient cycle would be closed by returning the nutrients contained in the agricultural wastes to the soil. Direct application of digestate to the soil has been widely proposed (Alburquerque, J. A. et al., 2012; Gómez-Brandón et al., 2016).

However, the application of the digestate without a post-treatment could not be always suitable or safe (Monfet et al., 2017). By one hand, direct application of digestate to soil presents operational difficulties derived from its viscosity, odor, and high humidity, which could complicate its handling (Bustamante et al., 2012; Walker et al., 2009). By another hand, digestate composition includes a high content of potentially phytotoxic compounds such as volatile fatty acids and ammonia (Hanajima et al., 2007; Kaparaju et al., 2012). These compounds can cause harmful effects on seed germination, and plant growth and development (Kaparaju et al., 2012; Tiquia et al., 1996). Other minority compounds of the digestate, such as heavy metals, phenolic compounds or salts can also produce phytotoxicity if good agricultural practices are not ensured (Alburquerque, José Antonio et al., 2012; Bustamante et al., 2012; Kaparaju et al., 2012). In addition, although anaerobic digestion usually reduces the pathogenic load respect the untreated substrates, prions and spore-forming bacteria could be present in the digestate (Gómez-Brandón et al., 2016). So, direct application of digestate to the soil could entail the spread of pathogens in the environment, such as *Salmonella, Campylobacter, Yersinia enterocolitica,* and *Cryptosporidium* (Bustamante et al., 2012; Vaneeckhaute et al., 2017).

Due to the potential risk for the agricultural soil derived from the direct application of the digestate, full biological stabilization of the organic matter from digestate can be ensured by aerobic maturation or complete composting (Monfet et al., 2017). During composting, the organic matter is turned into stabilized humic substances through mineralization and humification, resulting in a significant decrease in volume (Gutiérrez et al., 2017). In addition, pathogens are mainly removed by the heat generated in the thermophilic phase (Bustamante et al., 2013; Gutiérrez et al., 2017). Moreover, the compost has been proposed for soil remediation processes such as in situ heavy metal removal, immobilization of pesticides, and removal of emerging pollutants (Cerda et al., 2017; Kuppusamy et al., 2017; Zhou et al., 2017). Therefore, composting as final biorefinery step is a win-win strategy for the agricultural sector, which allows the recovery of the nutrients presented in the digestate and to minimize the environmental pollution risk.

Composting of digestate has been widely proposed as post-treatment of anaerobic digestion of animal manures and/or sewage sludge (Bustamante et al., 2014; Spencer, 2007; Torres-Climent et al., 2015). However, composting of digestate from agricultural waste has not been extensively reported in literature. One of the main challenges for the application of composting processes to digestate is the high humidity content of the digestate. Due to its low cost, one reported strategy to compensate the humidity content of the digestate is the employment of vegetables as a bulking agent during the composting process (Bustamante et al., 2012; Bustamante et al., 2013). As an example, Bustamante et al. (2012) reported the employment of vine shoot prunings as a bulking agent for the composting of digestate obtained from the anaerobic co-digestion of cattle slurry and silage. According to these authors, the use of vine shoot prunings as bulking agent reduced the temperature of the process, the electrical conductivity, and N losses during the composting process. Other agricultural wastes proposed as bulking agents for composting digestate were wheat straw, exhausted grape marc or pepper plant prunings (Bustamante et al., 2013). Moreover, the use of agricultural waste as a bulking agent could dilute a possible excess of pollutants, such as metals, which could be found in large concentration in digestates not only from agricultural waste but also for sewage sludge or animal slurries (Moral et al., 2008).

Other reported strategy to allow the composting of digestate is the separation and further composting of the solid fraction of the digestate (Holm-Nielsen et al., 2009). This separation reduces the humidity content of the substrate to be composted (Bustamante et al., 2012). Moreover, composting only the solid fraction of digestate can improve the quality of the obtained compost, contributing to the elimination of pathogens, and to reduce the odor emission by decreasing the concentration of volatile compounds, which could also reduce the potential phytotoxicity (Smet et al., 1999; Tchobanoglous and Kreith, 2002). Tambone et al. (2015) studied the solid fraction of digestates obtained from anaerobic digestion of pig slurry, energy crops and agro-industrial residues. For the composting of the solid fraction of these digestates, the employment of a lignocellulosic bulking agent did not give remarkably different results at the final product (Tambone et al., 2015).

A liquid fraction of digestate is also obtained through the separation processes. Usually, liquid fraction of digestate is rich in nitrogen but poor in phosphorous. Therefore, it could be applied as fertilizer due to its high N/P ratio and relatively high ammonia content might be of particular interest in P saturated areas (Sigurnjak et al., 2017; Vaneeckhaute et al., 2013). The fertilizer potential of the liquid fraction of an anaerobic co-digestion plant with an input feed consisting of 30% pig manure, 30% energy maize and 40% organic waste originating from the food industry was studied by Sigurnjak et al. (2017). According to these authors, the employment of the liquid fraction of digestate did not show significant differences in crop yield and soil quality at harvest in comparison to the employment of inorganic fertilizers. Nevertheless, the same authors advised that experiments on longer-term are required to fully evaluate the effects of the continuous application of liquid fraction of digestate on crop growth and soil fertility (Sigurnjak et al., 2017).

Other authors proposed the obtaining of magnesium ammonium phosphate hexahydrate, i.e., struvite, from the liquid fraction of digestate (Akhiar et al., 2017; Estevez et al., 2014). Struvite crystallization can help control the N/P ratio while simultaneously producing a slow release phosphate (Sheets et al., 2015). Unfortunately, struvite crystallization of liquid fraction of digestate from agricultural waste present some disadvantages. By one hand, the dissolved phosphate content in digested agricultural feedstocks may be lower than the requirement for a proper struvite crystallization (Sheets et al., 2015). Therefore, external phosphate may be needed, with the consequent increases costs. By the other hand, the liquid fraction of digestate from agricultural waste usually presents high levels of calcium. The calcium competes with magnesium to form calcium phosphate precipitates, reducing the precipitation of struvite (Martí et al., 2010). The suitability of each strategy for recovering the nutrients for the soil will depend on the actual agricultural system, i.e. soil requirements of nutrients, concentration of nutrients in the wastes, generated volume of biomass or the investment capacity of the agricultural sector.

5. ECONOMIC ASPECTS

The sustainability of a biorefinery approach should consider not only the environmental aspects but also the economic interest for the agricultural sector. The most significate aspects that should be considered for the integrated evaluation of the processes involved in the biorefinery approach are described below.

5.1. Valuable compounds extraction

The extraction section (excluding pretreatment) is most likely to involve the highest costs of both inversion and operation of the biorefinery approach (Serrano et al., 2017a; Serrano et al., 2017b). These costs depend on the selected extraction process: solid-liquid extraction, ultrasound, supercritical fluid extraction, biochemical, etc. The inversion cost of a steam explosion facility treating 100000 t/year of pinewood was reported as 4.2 M€ (Shafiei et al.,

2014) and the price of a thermal hydrolysis system with 170 ºC steam for treating 30000 t/year of different organic waste was reported as 1.0 M€ (Cano et al., 2014). These inversion costs can be taken as reference for a preliminary economic assessment (Aden et al., 2002).

Due to the high price of the extracted compounds, the highest economical incomings also come from this section, warranting a high profitability for the biorefinery. Nowadays the price of potential extracted products reaches values from 0.9-1.2 €/kg for lignin (Purelignin, 2017) to 520 €/kg for hydroxytyrosol (solution 10 % wt). However, these prices can fluctuate according to the market changes, affecting the result of an economic assessment. In the case of pectin, the global demand grown from 30000 t/year in 2009 to 60000 in 2015 with a price increment from 9.2-10.9 €/kg to 12.6 €/kg in the same period (Ciriminna et al., 2016). In other cases, the apparition of a new process could increase the offer reducing the price. In this sense, in order to study the profitability of the biorefinery, a sensibility analysis of the influence of the extract price must be done.

5.2. Anaerobic Digestion

In this stage, the final use of the methane generated involves different costs, inversion and operational, and revenues possibilities. The alternatives for the use of the methane are listed below:

- Direct use (without purification). The gas is conducted to a close consumer (0.6-15 km) for use in boilers or ovens as fuel. The profitability may be compromised if the distance to the consumer is higher than 5 km.
- Direct use (after purification). The gas, after being treated for purification, is conducted to the natural gas grill. Due to the high cost of the technology, this alternative can be only applied to great scale projects.
- Electricity generation. Capacity, energy efficiency, and costs are a function of the generation technology, i.e. internal combustion generator (350 kW-3MW), turbine (1-6 MW) or micro-turbines (30-200 kW).

- Electricity and heat generation. Simultaneous electricity generation with the recovery of useful heat. Higher capital costs than only power generation due to the heat recovery system, but higher global energy efficiency of the generation system. Efficiencies of cogeneration systems can be assumed 33 % (electricity) and 55 % (heat) (Cano et al., 2014).

The election process of one of these alternatives must be carried out taking into consideration different aspects as:

- Potential methane or heat consumers around the plant. The profitability of a project with the direct use of methane or cogeneration are conditioned to the existence of potential consumers of the generated methane or heat depending on the case.

- Local electricity market regulation. The price of the electricity can vary depending on the electricity market regulation of each country or region. The production of "green energy" can be incentivized by increasing the sale price of the electricity generated from biogas respect to that generated from fossil resources (Ruffino et al., 2015). Also, projects including cogeneration systems can receive economic incentives due to the higher energy efficiency (Real_Decreto_436, 2004).

- Waste availability. The capacity of waste supply to the biorefinery or the seasonal variation of this capacity are key parameters on the project design. Some power generation systems as gas turbines have a very low flexibility to changes in the gas charge. On another hand, the high cost of a cogeneration system makes this technology only appropriate for high capacities due to the economy of scale.

- Internal energy requirement of the biorefinery. An exhaustive study, taking also into consideration the parameters describes before, must be done to determine the optimal energy integration level of the anaerobic digestion system in the biorefinery. It can be noticed that, due to the existence of economic incentives given for the production of energy from removable sources, an energy self-sufficient design doesn't have to be the best option from an economic point of view. A higher integration level involves high inversion costs, a more complex operation and reduces the revenues for methane or energy sale. On another hand, the energy integrations reduce the operational cost derived from the use of electricity and/or fuels. Regarding to the energy requirement of the anaerobic digestion, in a cogeneration system, between the 30 to 50 % of the heat produced can be consumed, to keep the optimal temperature in the digester (Cano et al., 2014; Serrano et al., 2014a), and around 15 % of the electricity (Serrano et al., 2014a). The thermal energy requirement drastically increases when a thermal pretreatment is necessary (Aden et al., 2002; Shafiei et al., 2014). By energy integration, the methane generated by the anaerobic digestion can supply the energy necessary for the production of the steam employed in the pretreatment. In some cases, the methane production excess the pretreatment requirement but, in others, is necessary an additional fuel to cover the steam demand (Cano et al., 2014). On another hand, according to (Franchetti, 2013) up to 80 % of the thermal energy employed in the pretreatment with steam can be recovered as useful heat.

5.3. Composting

The landfill treatment and disposal of the digestate, with a cost in Spain around 25 € per tonne (excluding transport), have been reported as an important fraction of the anaerobic digestion costs (Cano et al., 2014). But "the digestate problem" can be turned into a revenue source by optimal recovery and recycling of nutrients. In UE, additional composting following digestion adds an additional cost up to 30 € per tonne of digested waste and the composted digestate generate prices between 0-50 €/t. The range of price is justified by the different demand depending on the region. I.e. regions with high manure offer, have a lower demand for compost. In order to evaluate the potential use of the compost for agriculture porpoises and to estimate the potential revenue associated with its sale, some critical aspects must be taken into consideration (Saveyn and Eder, 2014):

- Fertilisers market. As the compost is used as a substitute of industrial fertilizers, the price of these products can be taken as a reference to estimate the price of the compost. The reference fertilizer must be chosen in base on the compost composition and its nutrients concentration.

- Transport costs. The use is usually limited to areas sited in less than 100 km from the biorefinery. For higher distances, the transports cost would be higher than the value per tonne.
- Regulation of compost use. Depending on the country or region, the use of compost can be regulated in terms of composition-quality and/or maximum amount of compost that can be applied per year in the same area.

As the price of compost is usually lower than the composting costs, with the aim to obtain higher added values products, several alternatives previously cited in this review have been proposed in the literature for nutrients recovery from the digestate. All of them are based in the first step in which solid and liquid phase are separated (Monfet et al., 2017, Gutiérrez et al., 2017, Vaneeckhaute et al., 2017). The compost would be only obtained from the solid fraction, reducing the composting cost. But additionally, the solid-liquid separation process has an energy cost of 3.5 kWh/t (Tampio et al., 2016). The costs of the global nutrient recovery process and the obtained revenue will depend on the kind of final product selected.

5.4. Economic viability assessment

Table 6 shows the results of a preliminary economic viability assessment performed for the valorization of two different waste streams, two-phase olive mill solid waste (OMSW) and orange peel waste (OPW), within the bio-refinery concept: steam explosion as thermal pretreatment (SE) + valuable compound extraction (VCE) + anaerobic digestion with cogeneration (AD/CHP) + composting of digestate (CD). Net present value (NPV), internal rate of return (IRR) and payback period (PB) were used to evaluate the economic viability of the two bio-refinery approaches. IRR is calculated as the discount rate for which the Net Present Value (NPV) reaches a value of zero.

Technical and economic assumptions employed for the economic viability assessment are:

– Plant capacity of 50,000 t/y of waste. The bio-refinery is assumed to be located in Spain and to operate 8000 h along 12 months per year with a 25-year lifetime (IET/1045/2014).

– Steam Explosion and valuable compound extraction. OMSW (Serrano et al., 2017b): 4,200,000 € (SE) + 21,000,000 € (VCE) Investment; 2% of the investment (SE) + 25,000,000 €/y (VCE) operational and maintenance costs; electricity consumption 15 % of electricity production in AD; high-pressure steam requirement 905 MJ/t OMSW (SE); 12.7 kg of 10 wt% phenol extract per kg OMSW. OPW (Forgács et al., 2011): 2,600,000 € (SE+VCE); 2% of the investment operational and maintenance costs (SE+VCE) (general assumption); electricity consumption 1 kWh/t OPW (SE+VCE); high-pressure steam requirement 790 MJ/t OPW (SE+VCE); 7.1 kg/t OPW.

– Anaerobic digester and co-generation biogas engine. 39.95 l CH4/kg OMSW (Serrano et al., 2017b); 82.16 l CH4/kg OPW (Forgács et al., 2011; Calabrô and Panzera, 2017); 3398 € of construction cost per installed kWe (TrustEE, 2017); electric energy self-supply of the anaerobic digester 15% of the electricity generated by the co-generation biogas engine (Angelidaki et al., 2006); operational and maintenance costs 2% of the construction cost; complementary costs ought to be considered regarding feasibility studies and administrative and authorization requirements (30,000 €) (González-González and Cuadros, 2013). The methane production was calculated based on lab scale experiment by applying a scale up factor of 85%. In the case of OPW, the experimental methane yield obtained in (Forgács et al., 2011) was adjusted for a campaign of 8 month using fresh waste and 4 months using ensiled waste with a 30 % reduction of volatile solids Calabrò and Panzera, 2017)

– Energy Integration. The efficiency in the energy obtained through a cogeneration biogas engine is considered 33% for electricity and 55% for thermal energy (30% in hot water and 25% in exhausted gas). Hot water is employed to keep the operation temperature of the AD reactor (Cano et al., 2014). The thermal energy contained in the exhausted gas from the biogas engine and an additional stream of natural are used to obtain the necessary high-pressure steam supply for the steam explosion treatment. Any excess electricity is fed into the grid. Composting penalizes the electricity sale by 3,5 kWh per ton of digestate (Tampio et al., 2016).

– Prices. 42.67 €/MWh of electricity (creara.es, 2018), 0.04 €/kWh of consumed Natural gas (endesaonline.es, 2018), 520 €/kg of 10% phenol extract (Ciriminna et al., 2016), 1 €/kg of

Limonene (Negro et al., 2017b). No profit has been considered from the sale of compost. The composting cost is assumed to be equal to the incoming from the sale of it. Under the economical point of view, composting would be justified by the aim to avoid the cost of bio-waste dumping.

– Economic outlook. The value of the discount rate is assumed to be equal to the Spanish ten-year bond yield. The industrial profit taxes are fixed at 25%. Additionally, a minimum admissible price for the extracted valuable product has been calculated by imposing a NPV equal to zero.

Both wastes show promising results with positive NPV and IRR higher than 7.5%, which is an adequate IRR value according to the Spanish regulation for small scale AD plants (Biogas, 2014). In the case of OPW, a payback period of more than 5 years would be an inconvenience in order to face the inversion. A reduction of the price of the valuable extracted over 91% (phenol extract) and 70% (D-limonene) would turn this bio-refinery pathway as unprofitable for OMSW and OPW respectively.

The profitability of the inversion would be higher if the electricity excess is used to supply the electricity requirement in the olive oil production plant or orange juice plant respectively. In this case, the excess of electricity, of a portion of it, could be assumed as a save cost, with a price of 123,4 €/MWh (EUROSTAT, 2018), higher than the sale price of 42.67 €/MWh.

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Table 1. Bioactive compounds in fruit and vegetables by-products.

By-product	Phenolic	Flavonoids	Antocyanin	Carotenoids	Phytosterols	References
Citrus	Cumaric, cafeic, ferulic acids	Hesperidin, hesperetin Narigin, naringenin, Luteolin, tangeretin	cyanidin 3-glucoside and cyanidin 3-(6″-malonyl-glucoside)		In oil mixture peel/seed	(Fabroni et al., 2016; M'hiri et al., 2017; Ndayishimiye et al., 2017)
Tomato		Quercetin, rutin, kaempferol, narigenin		β carotene, lutein, lycopene		(Fattore et al., 2016; Gharbi et al., 2017)
Onion		Quercetin and kaempherol glycosides	In red onion	α- and β carotene		(Sharma et al., 2015; Wiltshire et al., 2017)
Carrot	chlorogenic acids, dicaffeoylquinic acids			α– and β–carotene		(Sánchez-Rangel et al., 2016)
Potato	chlorogenic , caffeic, protocatechuic and ferulic acid	Catechin, quercetin and kaempherenol rutinoside	anthocyanidins			(Akyol et al., 2016)
Grape	caffeic, gallic, protocatechuic, 4-hydroxybenzoic and syringic acid, hydroxytyrosol,	(+)-catechin, catechin dimer and trimer, (-)-epicatechin, procyanidin B1 and B2, quercetin-3-O-rhamnoside, luteolin, reverastrol (stilbene)	Proanthocyanidins and anthocyanins		tocopherol tocotrienols	(Barba et al., 2016; Teixeira et al., 2014)
Mango	gallic acid, salicylic acid	Epicatechin, quercetin	hydrolysable tannins, anthocyanins			(da Silva and Jorge, 2017; Dorta and Sogi, 2016)
Papaya	Gallic, chlorogenic, caffeic , protocatechuic acid	quercetin, kaempherol		lutein, lycopene cryptoxanthin β-carotene	α-tocopherol	(de Moraes Crizel et al., 2016)
Banana	Syringic, tannic, gallic, cinnamic, p-coumaric, ferulic	Gallocatechin gallate. quercitin	proanthocyanidins	α - and β–carotene, xanthophyll	B-sitosterol, campesterol, stigmasterol	(Singh et al., 2016; Vu et al., 2016)

	acid.					
Pineapple	Gallic acid, ferulic acid	catechin, epicatechin	anthocyanins	α and β–carotene		(Da Silva et al., 2014; Li et al., 2014)
Olive oil	Hydroxytyrosol, 3,4-dihydroxyfenylglicol, secoiridoides.	Luteonin, apigenin.				(Fernández-Bolaños et al., 2002; Rubio-Senent et al., 2013b)

Table 2. Health benefits from compounds of fruits and vegetables by-products.

By-product	Compound	Uses and health benefits	Reference
Citrus	Pectin	Thicking agent, gelling, stabilizer	(John et al., 2017)
	Dietary fiber	Regulation of intestinal transits, prevention of diabetes, hypertension, cardiovascular disease and colon cancer.	
Tomato	Pectin	Thicking agent, gelling, stabilizer	(Morales-Contreras et al., 2017)
Onion	Fructooligosaccharides	Prebiotic effect	(Smith et al., 2015)
Carrot	Pectin	Thicking agent, gelling, stabilizer	(Jabbar et al., 2015; Jafari et al., 2017)
	Antioxidant dietary fiber	Benefits from fiber together with antioxidant activity.	
Potato	Water-soluble polysaccharides	Additive in food, pharmaceutical and cosmetic preparation.	(Jeddou et al., 2016)
Grape	Antioxidant dietary fiber	Benefits from fiber together with antioxidant activity	(Zhang et al., 2017)
Mango	Antioxidant dietary fiber	Benefits from fiber together with antioxidant activity	(Matharu et al., 2016)
	Pectin (highly-esterified)	Used in marmalade, dairy and meat preparations	
	Soluble dietary fiber	Control diabetes	
Pineapple	Dietary fiber	Fortifying extruded products. Prebiotic effect.	(Selani et al., 2014)
Olive oil	Oligosacarides, pectins, fenolic glucosides	Prebiotic agents, antioxidant, antiproliferatives	(Rubio-Senent et al., 2015a; Rubio-Senent et al., 2013a)

Table 3. Methane yield and biodegradability of different lignocellulosic by-products

Waste	Pretreatment	Methane yield (mL CH_4/g VS)	Biodegradability (in VS)	Reference
Switchgrass (winter and fresh summer harvested)	Pretreatment: grinding + alkalinisation + autoclaving.	140 (winter harvested) 298 (fresh summer harvested)	No available	(Frigon et al., 2012)
Grass (*Pennisetum hybrid)* Hay	*Grass* pretreatment: 30 min with water vapour. Hay *pretreatment:* Steam-explosion at 175 ºC for 10 min.	190 (raw grass) 236 (raw hay) 198 (pretreated grass) 281 (pretreated Hay)	No available	(Rodriguez et al., 2017)
Dried grass silage	Pretreatments at 100ºC and NaOH loading rates of 1%, 2.5%, 5% and 7.5% by VS mass in grass silage	360 (NaOH 1%) 402 (NaOH 2.5%) 450 (NaOH 5%) 453 (NaOH 7.5%)	76.9% (NaOH 1.0%) 85.3% (NaOH 2.5%) 95.2% (NaOH 5.0%) 96.7% (NaOH 7.5%)	(Xie et al., 2011)
Salix biomass	Steam-explosion pretreatment at 170-230 ºC, during 5-15 min.	161 (raw Salix) 234 (pretetad Salix, 230 ºC, 10 min)	No available	Estevez (Estevez et al., 2012)
Sunflower stalks	Two thermal (55 and 170 ºC) and five thermo-chemical pretreatments (NaOH, H_2O_2, $Ca(OH)_2$, HCl and $FeCl_3$)	192 (raw sunflower stalks) 259 (pretreated sunflower stalks, 55 ºC with 4% NaOH for 24 h)	No available	(Monlau et al., 2012)
Wheat Straw	Steam-explosion pretreatment: 160-200 ºC, 10-20 min.	276 (raw wheat straw) 331 (pretreated wheat straw, 180 ºC, 15 min)	Maximum VS removal efficiency: 46.3% (200 ºC, 10 min).	(Bauer et al., 2009)
Wheat Straw	Thermal pretreatment at 80 ºC, 10 min + H_2O_2 (7.5%).	188 (raw wheat straw) 274 (pretreated wheat straw)	41.1% (raw wheat straw)	(Hassan et al., 2016)
Barley Straw (BS) Wheat Straw (WS) Rice Straw (RS)	Thermal pretreatment in autoclave at 90 and 120 ºC during 30 min.	240 (raw BS) 197 (raw WS) 182 (raw RS) 338 (pretreated BS, 120ºC) 299 (pretreated WS, 120ºC)	No available	(Menardo et al., 2012)

		261(pretreated RS, 120ºC		
Peanut hull (*Arachis hypogaea)*	Mechanical (grinding into < 20 mm) + thermal (80 ºC, 70 min) + 3 g NaoH/100 g TS (24 h, at 55 ºC).	112 (raw peanut hull) 182 (pretreated peanut hull)	26% (raw peanut hull) 47% (pretreated peanut hull)	(Dahunsi et al., 2017)
Ensiled sorghum forage (ESF) Wheat straw (WS)	Thermal and thermo-alkaline pretreatments at 100 ºC, and 160 ºC for 30 min, without and with the addition of NaOH solutions at 1% and 10% g NaOH/g TS.	269 (raw ESF) 204 (raw WS). 361 (pretreated ESF, 10% NaOH, 100 ºC) 341 (pretreated WS, 10% NaOH, 100 ºC)	84-85% (pretreated ESF and WS, 10% NaOH, 100 ºC)	(Sambusiti et al., 2013)

Table 4. Methane yield and biodegradability of different vegetable by-products

Waste	Pretreatment	Methane yield (mL CH_4/g VS)	Biodegradability (in VS)	Reference
Onion skin Cauliflower stems Potato skin Carrot petioles Beet leaves	No pretreatments	400 (Onion skin) 331 (Cauliflower stems) 267 (Potato skin) 309 (Carrot petioles) 231 (Beet leaves)	No available	(Ji et al., 2017)
Onion waste	No pretreatments	340	50–70%	(Lubberding et al., 1988)
Onion waste	Mechanical pretreatment (pieced to 1-2 cm size)	378	61%	(Menardo and Balsari, 2012)
Carrot waste pomace	No pretreatments.	198	56% COD removal	(Garcia et al., 2011)
Potato waste	No pretreatments.	320 mL CH_4/g VS	No available	(Parawira et al., 2004)
Mixture Tomato waste: sewage sludge (5:95)	No pretreatment	159	95%	(Belhadj et al., 2014)
Tomato waste Cucumber waste	No pretreatment	299 (Tomato waste) 177 (Cucumber waste)	No available	(Gil et al., 2015)
Tomato waste Cucumber waste	No pretreatment	320 (Tomato waste) 260 (Cucumber waste)	79% (Tomato waste) 43% (Cucumber waste)	(Jagadabhi et al., 2011)

Table 5. Methane yield and biodegradability of different fruit by-products

Waste	Operational conditions	Methane yield (mL CH_4/g VS)	Biodegradability (in VS)	Reference
Banana peel Pineapple waste	No pretreatment	188-240 (Banana peel) 413 (Pineapple waste)	36% (Banana peel) 58% (Pineapple waste)	(Bardiya et al., 1996)
Peels of Fluted pumpkin (*Telfaria occidentalis)*	Combination of mechanical (grinding into < 20 mm), thermal and chemical (NaoH) (3 g NaoH/100 g TS for 24 h at 55 ºC)	164 (mechanical, thermal and chemical pretreatment) 161 (mechanical and chemical pretreatment)	67.2% COD reduction	(Dahunsi et al., 2016)
Pineapple pulp and peel	No pretreatment	111-430	56.0-73.8%	(Namsree et al., 2012)
Mango Peel waste and Pomegranate seed de-oiled cake (PSDC)	No pretreatment	300-410 (Only mango peel waste) 610 (Mango peel waste and PSDC)	23.4% (Only mango peel waste) 25.8% (Mango peel waste and PSDC)	(Suryawanshi et al., 2013)
Banana Skin Tomato waste	No pretreatment	277 (Banana skin) 298 (Tomato waste)	74% (Banana skin) 77% (Tomato waste)	(Ji et al., 2017)
Orange peel	Steam distillation (6 h) for 70% Limonene removal	230-290	84-90% COD removal	(Martín et al., 2010)
Orange peel	Steam distillation (6 h) for 70% Limonene removal	332 (thermophilic conditions) 230 (mesophilic conditions)	84-90% COD removal	(Siles et al., 2016)
Kiwi waste	No pretreatment	371	58% in VS	(Menardo and Balsari, 2012)
Strawberry extrudate	Thermal pretreatment (120 ºC, 2 atm, 5-60 min).	285 (raw strawberry extrudate) 339 (pretreated strawberry extrudate)	No available	(Serrano et al., 2015)
Strawberry extrudate	Mechanical sieving (1 mm mesh size).	230 (raw strawberry extrudate) 312 (pretreated strawberry extrudate)	90% in VS	(Siles et al., 2013)
Winery waste	No pretreatment	250	79% in COD	(Siles et al., 2011)
Two-phase OMSW	Thermal pretreatment at 100, 120, 160 and 180 ºC during 60, 120 and 180 min for each temperature studied.	373 (raw OMSW) 392 (pretreated OMSW, at 120ºC, 180 min)	No available	(Rincón et al., 2013)

		380 (pretreated OMSW, at 180ºC, 180 min)		
Two phase OMSW	High temperature thermal pretreatment (170 ºC for 60 min) and phenol extraction	261 (raw OMSW) 290 (OMSW, thermal pretreatment) 350 (OMSW, thermal pretreatment and phenol extraction)	57.3% (raw OMSW) 63.4% (OMSW, thermal pretreatment) 75.3 (OMSW, thermal pretreatment and phenol extraction)	(Serrano et al., 2017a)
Two phase OMSW	Steam–explosion pretreatment: 200 ºC, 5 min (pressure: 42 kg/cm^2) and phenol extraction	280 (raw OMSW) 294 (OMSW, steam-explosion) 261 (OMSW, steam-explosion and phenol extraction)	66.4% (raw OMSW) 65.0% (OMSW, thermal pretreatment) 60.5 (OMSW, thermal pretreatment and phenol extraction)	(Serrano et al., 2017b)

Table 6. Economic viability assessment of OMSW and OPW AD-biorefineries

Electricity to the grid (GWh/year)	Extract[a] production (t/year)	Initial investment cost (€)	Incoming (€/year)	Annual costs (€/year)	NPV (M€)	IRR (%)	Payback period (years)	Limit extract[a] price (€/kg)[b]
4.0	633	28,013,747	329,354,041	25,444,066	4.7·103	815	<1	42.0
11.3	353	8,355,042	834,871	196,947	3.86	5	17	0.3

a Extract: 10 %w phenol extract for OMSW and D-limonene for OPW
b NPV = 0;

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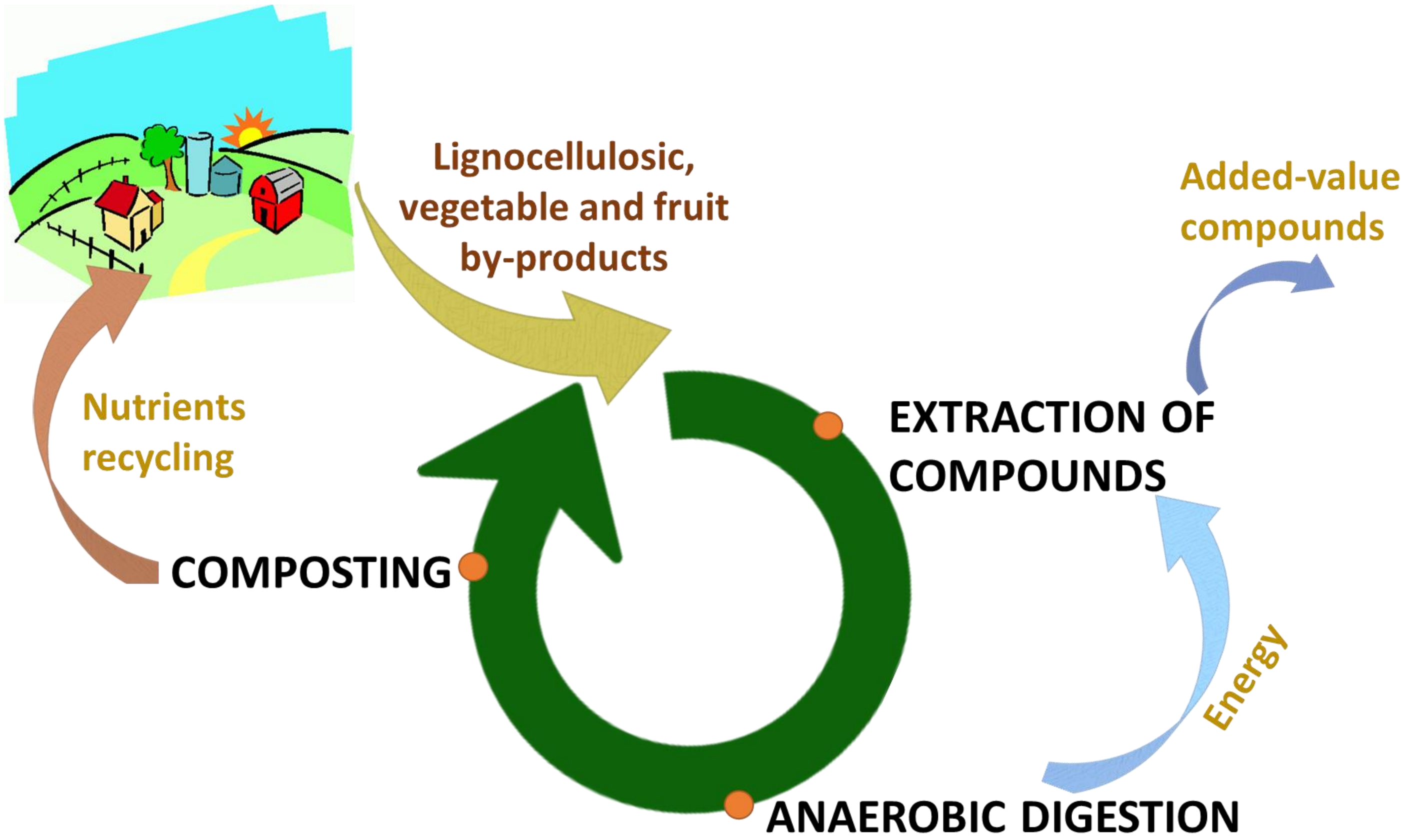

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