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Research Paper

Integral multi-valorization of agro-industrial wastes: A review

Inés Prado-Acebo [a], Juan Cubero-Cardoso [a,b,*], Thelmo A. Lu-Chau [a], Gemma Eibes [a]

[a] CRETUS, Department of Chemical Engineering, University of Santiago de Compostela, Santiago de Compostela 15782, Spain

[b] Laboratory of Sustainable and Circular Technology, CIDERTA and Chemistry Department, Faculty of Experimental Sciences, Campus de "El Carmen", University of Huelva, 21071 Huelva, Spain

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ABSTRACT

Agriculture and industries related to the agriculture sector generate a large amount of waste each year. These wastes are usually burned or dumped, causing damage to the environment, the economy and society. Due to their composition, they have great potential for obtaining high value-added products in biorefineries. This fact, added to the growing demand for energy and chemicals from fossil resources, is driving the interest of the scientific community in them.

Biorefinery processes are hardly profitable when applied individually, so a better alternative is to develop integrated multi-feedstock and multi-product biorefinery schemes using all biomass fractions in a zero-waste approach. However, for industrial scale application, extensive research, scale-up studies, and techno-economic and environmental feasibility analyses are needed. This review compiles information on integrated multi-biorefinery processes from agro-industrial wastes to shed light on the path towards sustainable development and circular bioeconomy.

1. Introduction

One of the points highlighted by the United Nations in its 2030 Agenda for Sustainable Development is to ensure sustainable consumption and production patterns. Specifically, to reduce waste generation, they recommend circular economy strategies such as prevention, reduction, recycling and reuse (United Nations, 2015). To this end, the European Commission adopted in March 2020 a new Circular Economy Action Plan, which sets out initiatives to act throughout the entire product life cycle and identifies the food sector as one of the areas with the greatest potential to make the circular economy a reality. Other official documents that share the same objective are the EU Bioeconomy Strategy (European Comission, 2018) and the EU Green New Deal (European Comission, 2019). Zero Hunger is also one of the objectives included in the United Nations 2030 Agenda (United Nations, 2015).

Agricultural-based industries annually generate around 1.3 billion tons of waste (Kour et al., 2023; Sadh et al., 2018). Unfortunately, most of this waste is currently disposed of by incineration or landfill, which poses significant environmental, economic, and social challenges. Furthermore, agro-industrial wastes become a source of proliferation for microorganisms, which, while crucial for decomposition, can become problematic on a larger scale. This proliferation is closely linked to the production of greenhouse gases, the release of toxic degradation byproducts, and the growth of pathogenic bacteria and fungi (Freitas et al., 2021). As highlighted by Capanoglu et al. (2022), the carbon footprint associated with these agro-industrial wastes is substantial, equivalent to an annual accumulation of 3.3 billion tons of CO_2 in the atmosphere. Moreover, the improper disposal of this waste contributes to the release of pollutants into the atmosphere, which in turn, degrades air quality and triggers respiratory problems (Yaashikaa et al., 2022). The economic and social impacts are equally alarming. The cost of food waste alone amounts to 1000 billion dollars annually, a sum that could potentially cover the nutritional needs of millions of people (Capanoglu et al., 2022). This overabundance of agro-industrial waste particularly affects developing countries, where viable alternatives to recover and reuse these valuable resources remain scarce (Freitas et al., 2021). Nevertheless, these agro-industrial wastes, due to their composition, have an untapped potential for multi-valorization. They can be harnessed to produce a wide array of valuable products, including biofuels, biopolymers, biofertilizers, enzymes, nutraceuticals, biogas, and more (Sadh et al., 2018).

For all these reasons, it is necessary to develop a sustainable agro-industrial waste management system. The biorefinery concept is an approach to treat biowaste that falls within the framework of the

* Corresponding author at: CRETUS, Department of Chemical Engineering, University of Santiago de Compostela, Santiago de Compostela 15782, Spain.
E-mail addresses: Juan.cubero@usc.es, j.cubero@dqcm.uhu.es (J. Cubero-Cardoso).

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circular economy and plays a key role in the transition towards sustainable development ("National Renewable Energy Laboratory (NREL) Home Page," n.d.). The NREL defines a biorefinery as "a facility that integrates processes and equipment for the conversion of biomass fuels, energy, and chemicals. The biorefinery concept is analogous to today's oil refineries, which produce multiple fuels and products from petroleum. Industrial biorefineries have been identified as the most promising route to the creation of a new biomass-based domestic industry." In a biorefinery concept, it is desirable that the feedstock can be transformed into high-value products such as pharmaceuticals, food ingredients, enzymes and nutrients; and into low-value products such as biofuels, energy or soil conditioners (Madeddu et al., 2021).

Biorefineries can be classified according to their feedstock. First generation biorefineries use crops, second generation biorefineries use residues, agro-industrial wastes, and non-edible crops, and third generation biorefineries use algae (Moncada et al., 2014). Due to the growing concern about the "food-fuel conflict" and the mandatory inclusion of the sustainability goals into the biorefinery concept, biorefineries based on agro-industrial wastes are gaining popularity (Hiloidhari et al., 2020). However, the limited commercialization of processes carried out in biorefineries involving the chemical bioconversion of agro-industrial wastes indicates that research in this field is far from complete.

Although there are numerous papers on biorefinery processes from different types of biomasses, there is not much literature available on multi-biorefinery processes. To the best of our knowledge, the only work that reviews multi-product biorefineries is that of Clauser et al. (2021), which analyzed the different stages in the design of integrated multi-product biorefineries from agricultural and forestry residues. The present review focuses exclusively on agro-industrial wastes and processes in which more than one product is obtained. Furthermore, the degree of integration in terms of feedstock, product and technology was evaluated. Finally, the limitations and future challenges to which future innovative work should be oriented were addressed.

2. Types and composition of agro-industrial wastes

Agro-industrial wastes can be classified into on-farm (agricultural residues) and off-farm (industrial wastes) (Fig. 1). In the first group we can differentiate between field residues and process residues. Field residues (seeds, leaves, stems, bunches) are those left in the field after harvesting, meanwhile process residues (husks, roots, seeds) are crop wastes generated after the processing of raw products (Singh et al., 2021). Those wastes generated in food processing industries, such as peels, oil, pomace, grounds and molasses belong to the industrial wastes group.

Agro-industrial waste consists mainly of lignocellulosic material, i.e. cellulose and hemicellulose interconnected by lignin and other constituents such as proteins, ash, organic acids, and other non-structural material. Cellulose is a major structural component of lignocellulosic material, consisting of a disaccharide of D-glucopyranose linked by β-(1–4) glycosidic bonds called cellobiose. The structure of cellulose forms fibers which can be in crystalline or amorphous configuration. Hemicelluloses are short-chain polysaccharides composed of 500–3000 units of two or more monosaccharides. The monosaccharides present in hemicellulose are pentoses such as xylose, rhamnose, and arabinose, and hexoses such as glucose, mannose, and galactose, generating a random, amorphous and weak structure. Lignin functions as a binding substance that links cellulose and hemicellulose, creating the robust three-dimensional framework of the plant cell wall. The structure of lignin consists of monolignols connected by aromatic rings, typically up to 10,000 units (Arpit Singh et al., 2022; Phan et al., 2023).

The different structure and composition of agro-industrial wastes may require individualized treatment approaches (Awogbemi and Kallon, 2022). For instance, materials such as stems or leaves require severe pre-treatment to enhance access to their simpler structures (Velvizhi et al., 2023). Meanwhile, certain feedstocks such as pomace undergo pre-processing during manufacturing, which alters the structure of the lignocellulosic material. As a result, it is crucial to tailor the biorefinery process to the unique characteristics of each raw material (Langsdorf et al., 2021).

At present, most research on various biorefinery processes for the agricultural residues mentioned earlier indicates the requirement of a cascade process. This approach aims to extract as much value as possible from the waste while minimizing its environmental footprint and maximizing its usefulness. Tables 1 and 2 presents predominantly an initial phase aimed at breaking the structure of lignocellulosic material, known as pre-treatment. Typically, pre-treatment involves autohydrolysis (heat treatment with water) or other variety of acidic, alkaline, or organic solvents (Ashokkumar et al., 2022). While this treatment is

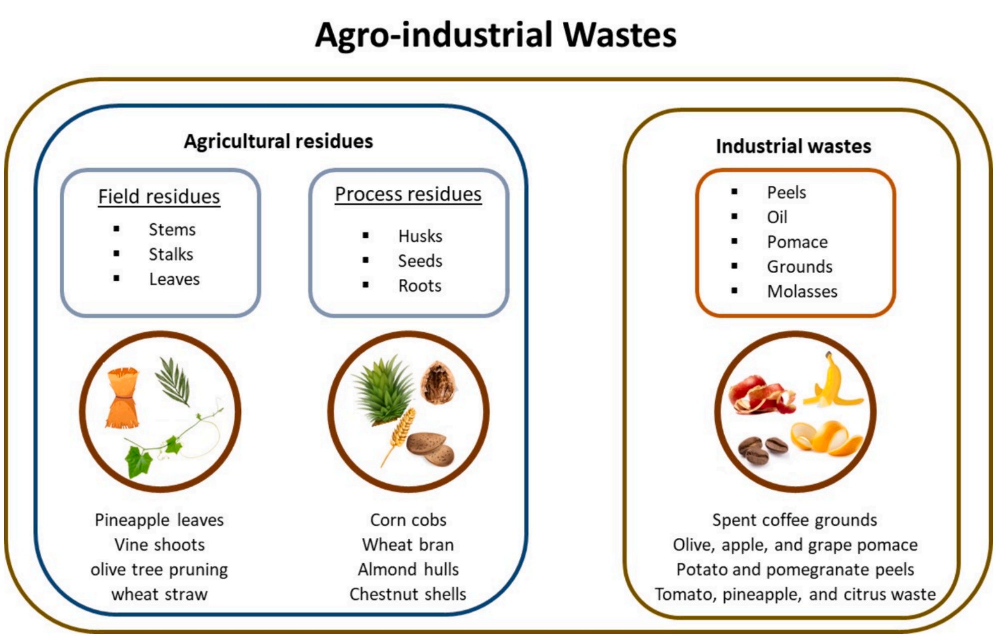

Fig. 1. Classification of agro-industrial wastes.

Table 1
Sequential multivalorization strategies for agricultural residues.

Crop	Waste	Worldwide estimated production (MTPA*)	Products	Technologies	References
Almond	Almond hull	1.4–2.5	Pectin, phenolics, pullulan, single cell protein	Acid extraction, ultrasonic, enzymatic hydrolysis	(Najari et al., 2022)
	Almond shell	0.8–1.7	Xylooligosaccharides, lignin, cellulose nanocrystals, glucose	Thermal treatment, alkaline and organic delignification, enzymatic hydrolysis	(Morales et al., 2020; Pirayesh and Khazaeian, 2012)
Chestnut	Chestnut shells	0.2	Lignin, bioethanol	Solve-thermal treatment, SSF	(He et al., 2016; Morales et al., 2018)
Rye	Rye straw	3.3	Xylitol, bioethanol	(Simulation) Thermal treatment, enzymatic hydrolysis, fermentation	(Franceschin et al., 2011)
Pineapple	Pineapple leaves	3.8	Bioethanol, biomanure	Enzymatic hydrolysis, SSF, fermentation	(Banerjee et al., 2018; Chintagunta et al., 2017)
Corn	Corn cobs	880	Xylanases, cellulases, glucose, lignin	Autohydrolysis, enzymatic hydrolysis, fermentation	(Guo and Lü, 2021; Michelin et al., 2018)
			Lignin, furfural, bioethanol	Acid treatment, enzymatic hydrolysis, fermentation	(Qiao et al., 2022)
			5-Hydroxymethylfurfural, cellulose, lignin	Acid treatment, microwave reaction, enzymatic hydrolysis	(Jin et al., 2020)
Wheat grain	Wheat bran	125.4	Proteins, fermentable sugars, lignin	Autohydrolysis, enzymatic hydrolysis	(Celiktas et al., 2014; Tirpanalan et al., 2015)
	Wheat straw	1140	Bioethanol, biodiesel	Acid treatment, fermentation, enzymatic transesterification, SSF	(Deka et al., 2023; Morikawa et al., 2014)
			Bioethanol, furfural, lignin	Acid treatment, SSF	(Zhao and Liu, 2019)

* MTPA: Million tonnes per annum; this value was estimated according to data of crop production from FAOSTAT (2021) and the mass fraction of the waste.

considered conventional, ongoing research is exploring environmentally sustainable alternatives, including the application of methods such as microwaving, ultrasound, or supercritical liquid treatments (Cubero-Cardoso et al., 2020). The second phase primarily involves enzymatic hydrolysis, which facilitates the dissolution of carbohydrates from cellulose and hemicellulose. Ultimately, a final fermentation stage is carried out, in whichseveral types of cultures are used to yield various high-value compounds, such as biogas, bioethanol, biodiesel, and various intermediate products resulting from different fermentation processes (Rodionova et al., 2022).

2.1. Agricultural residues

Agricultural residues can contribute to the bioeconomy, as they are sources of valuable compounds (Capanoglu et al., 2022). Table 1 summarises the main sequential multivalorization strategies for this type of waste found in the literature. Corn, wheat, and sugarcane generate the largest volumes of agricultural residues globally, exceeding 100 million tonnes annually (MTPA). This underscores the importance of exploring biorefinery models to maximize potential gains. Agricultural residues consist primarily of lignocellulosic material, which necessitates complex treatment and yields predominantly fermentable carbohydrates, enabling the production of various compounds. An example of a sugarcane biorefinery is the proposal by Nieder-Heitmann et al. (2019) who succeed in separate the three fractions by pre-treatment with formic acid followed by lignin precipitation. From sugarcane bagasse and trash lignocelluloses, the aim is to obtain succinic acid from glucose and hemicelluloses, polyhydroxybutyrate (PHB) from glucose and electricity from lignin. Biorefinery from wheat straw described by Zhao and Liu (2019) applies formic acid pre-treatment and simultaneous saccharification and fermentation (SSF) to obtain high purity ethanol, furfural and lignin. Lignin is burned along with other secondary streams of the process to generate steam.

2.2. Industrial wastes

According to Sadh et al. (2018), agricultural-based industrial waste is all organic residues and related effluents generated in food processing industries. Table 2 summarises the main integrated multi-valorization strategies for this type of waste found in the literature. Grape, potato, and barley generate the largest volumes of agricultural waste, exceeding 50 MTPA. Industrial waste generates a smaller volume of waste compared to agricultural residues, yet this biomass contains higher value-added compounds such as phenolics, oils, proteins, and other valuable constituents. Mussatto et al. (2013) exemplified a biorefinery system for barley, focusing on the comprehensive multi-valorization of brewer's spent grains. Their process yields xylitol, lactic acid, activated carbon, and phenolic acids as key products. Martinez et al. (2015) introduced a versatile four-step cascading biorefinery approach for the multi-valorization of grape pomace. The successive stages were designated for distinct purposes: the extraction of phenolics utilizing supercritical CO_2, the generation of volatile fatty acids (VFAs) via anaerobic acidogenic digestion, the utilization of these VFAs as feedstock for the biotechnological synthesis of polyhydroxyalkanoates (PHAs), and the production of methane-enriched biogas through anaerobic digestion of solid residues originating from the acidogenic process.

3. Integrated biorefineries

A crucial step in the development of profitable biorefineries is to enable scalable and integrated biorefineries that can efficiently transform a broad range of biomass feedstock into commercially feasible biofuels and bioproducts. In this sense, integration emphasizes on the maximum possible use of resources within the same plant. This integration can be of feedstocks, products and technologies, as shown in Fig. 2.

Feedstock and product integration are interrelated, since one

Table 2
Sequential multivalorization strategies for industrial wastes.

Crop	Waste	Worldwide estimated waste production (MTPA*)	Products	Technologies	References
Coffee	Spent coffee grounds	6	Biodiesel, biogas, fermentable sugars	Soxhlet extraction, transesterification, SSF, anaerobic digestion	(Battista et al., 2021)
Sugarcane	Sugarcane bagasse	514	Furfural, xylitol, energy	(Simulation) Autohydrolysis treatment, medium-density fibreboard,	(Clauser et al., 2016)
			Polyhydroxybutyrate (PHB), succinic acid, electricity	(Simulation) Enzymatic hydrolysis, fermentation	(Nieder-Heitmann et al., 2019)
Banana	Residual banana	30	Ethanol, PHB, fertilizer	(Simulation) Autohydrolysis, fermentations	(Naranjo et al., 2014)
Potato	Potato waste	97.6	Bioethanol, biomanure	Fermentation	(Chintagunta et al., 2016)
Tomato	Skin, seeds	9.3	Oil, carotenoids, proteins, carbohydrates, cellulose and lignin	Supercritical CO_2 extraction, autohydrolysis	(Kehili et al., 2016)
Pineapple	Skin, core	9.87	Bioethanol, bromelain, residual biomass	SSF, membrane separation, fermentation	(Seguí Gil and Fito Maupoey, 2018)
Pomegranate	Skin	4	Pectin, phenolics, bioethanol	Autohydrolysis, enzymatic hydrolysis,fFermentation	(Talekar et al., 2018)
Citrus	Citrus waste	15–25	Limonene, pectin, biogas, bioethanol	Acidic-thermal treatment, fermentation, anaerobic digestion	(Pourbafrani et al., 2010)
			Essential oils, pectin, succinic acid	Acidic-thermal treatment, enzymatic hydrolysis, fermentation	(Patsalou et al., 2017)
Olive	Olive pomace	7.05–8.23	Phenolics, antioxidants lignin, carbohydrates	Autohydrolysis and solvent treatment, enzymatic hydrolysis	(Fathy et al., 2018; Gómez-Cruz et al., 2021)
			Antioxidants, purified oligosaccharides, lactic acid, cellulose nanocrystals	Acidic-thermal treatment, fermentation	(Gullón et al., 2020)
Blackberry	Spent pulp	0.03	Phenolics, ethanol, xilitol, bioenergy	(Simulation) Acid-thermal hydrolysis, enzymatic hydrolysis, fermentation	(Dávila et al., 2017a)
Sugar beet	Sugar beet pulp	35.28	Pectin, phenolics, proteins, succinic acid	Solvent extraction, enzymatic hydrolysis, fermentation	(Alexandri et al., 2019; Usmani et al., 2022)
Grape	Vine shoots	176.7	Lignin, bioethanol	Autohydrolysis, enzymatic hydrolysis, fermentation, SSF	(Dávila et al., 2019; Filippi et al., 2022; Garita-Cambronero et al., 2021)
	Grape pomace	190	Phenolics, polyhydroxyalkanoates (PHA), biogas	Supercritical CO_2 extraction, anaerobic and aerobic fermentation, anaerobic digestion	(Martinez et al., 2015)
			Phenolics, Biocomposites	Pressurised liquid extraction, solvent extraction	(Monari et al., 2020)
	Grape pomace, stalks, wine lees	266	Oil, phenolics, bioethanol, tartaric acid, condensed tannins, bacterial cellulose, succinic acid	Solvent extraction, enzymatic hydrolysis, fermentation	(Ahmad et al., 2020; Filippi et al., 2022)
Avocado	Skin and seeds	2.04	Oils, phenolics, pectin, oligosaccharides, aromatic compounds, nanocristalline cellulose, ethanol, xylitol, succinic acid	(Review) Extraction techniques	(Dávila et al., 2017b; Del Castillo-Llamosas et al., 2021)
			Phenolics, oligosaccharides, succinic acid	Autohydrolysis treatment, SSF	(Rodríguez-Martínez et al., 2022)
	Pulp, skin and seeds	7.41	Oil, phenolics, ethanol, xylitol	Enhanced-fluidity liquid extraction, enzymatic hydrolysis, fermentation	(Dávila et al., 2017b)
Barley	Brewer's spent grains	48.67	Biodiesel, bioethanol, biogas	Solvent extraction, enzymatic hydrolysis, fermentation, anaerobic digestion	(Kavalopoulos et al., 2021)
			Xylitol, phenolics, lactic acid, activated carbon	Solvent extraction and autohydrolysis, enzymatic hydrolysis, fermentation	(Mussatto et al., 2013)
Apple	Apple pomace	25.8	Antioxidants, nutraceuticals, pectin, biomaterial	Solvent extraction and autohydrolysis	(Yates et al., 2017)
			Ethanol, mycoproteins, chitin, chitosan, biomass	Solvent extraction, SSF, aerobic, and anaerobic digestion	(Borujeni et al., 2022)

* MTPA: Million tonnes per annum.

product (or by-product) can serve as feedstock for direct use in another process in the biorefinery. Feedstock integration also includes the use of multiple feedstocks in the same biorefinery. The integration of technologies can be of three types: mass (recovering process streams), energy (minimizing utility consumption) or process intensification (reducing stages in the process).

3.1. Feedstock integration

Flexibility towards diverse feedstocks is an important driver for biorefinery cost reduction (Budzianowski and Postawa, 2016). When various types of feedstock and processing technologies are used to produce a variety of energy and chemical products, the biorefinery is known as phase III and is the most advanced type of biorefinery (Clark and Deswarte, 2014). Multiple feedstocks can derive either from a particular industrial process (valorization of different by-products obtained in one facility) or from different raw materials (valorization of similar by-products obtained in different facilities) (Table 3). In the first scenario, different platforms should be probably considered for the full exploitation of the materials. In this sense, the different nature and/or

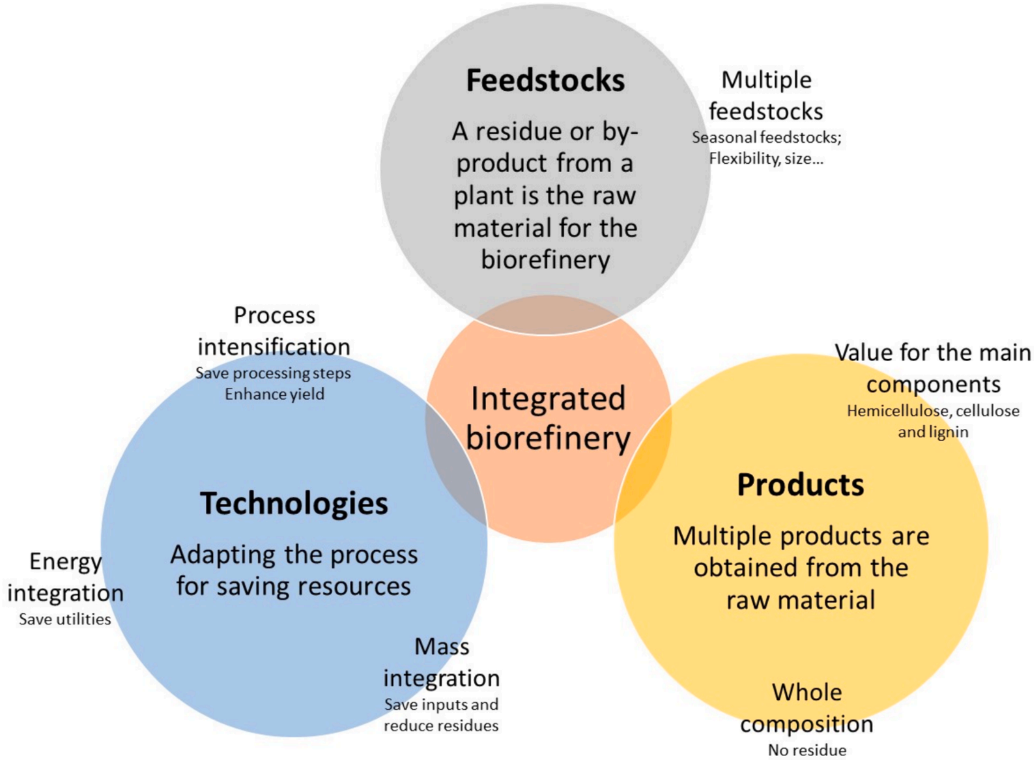

Fig. 2. Integrated biorefineries can include three different aspects: integration of feedstocks, products or technologies.

Table 3
Possibilities for the feedstock integration in a phase III biorefinery treating agro-industrial wastes.

Feedstock integration	Example	Characteristics	Challenges
Different by-products from the same raw material	Winery wastes, containing grape pomace, stalks and wine lees	Different platforms considered for the full exploitation of the materials	Effective pretreatment to enlarge feedstock base
Similar by-products from different raw materials and from different facilities/sites	Agricultural residues	Same platform but different operational conditions	Logistics due to material transportation

composition of the feedstocks makes it difficult to process them using the same biorefinery scheme. On the other hand, since the streams are produced in the same facility, feedstock logistics would be simplified.

Filippi et al. (2021) integrated winery waste components such as grape pomace, stalks, and wine lees into a biorefinery. They employed various processing methods, combining streams when possible. For instance, they combined grape pomace and stalks for pre-treatment, and sugars from solids with those from wine lees to produce succinic acid. This demonstrates the efficient use of winery waste. Gullón et al. (2020) demonstrated feedstock integration in cascade multi-biorefinery process focused on olive industry waste, encompassing by-products and wastewater. They initiated the process with the extraction of antioxidants from solid olive by-products. Subsequent autohydrolysis transformed the residue into a liquor rich in oligosaccharides and a solid phase abundant in cellulose and lignin. The liquid fraction, combined with olive processing wastewater, underwent membrane treatment, yielding purified oligosaccharides and antioxidants. This innovative approach, despite varying feedstock properties, underscores the versatility of autohydrolysis and its potential for expanding the scope of integrated biorefinery systems (Budzianowski and Postawa, 2016).

Effective pre-treatment may enlarge feedstock base, which would be one of the suitable approaches for integrating biorefinery systems in the total chain. However, maintaining identical processing conditions using different feedstocks can result in poor performance. In this framework, Zhang et al. (2018) conducted a comparative study involving 16 alternative feedstocks alongside the benchmark feedstock, wheat straw, using identical processing conditions. The outcomes varied significantly in terms of digestibility and subsequent potential for ethanol production, even when subjected to the same autohydrolysis. Another example considering feedstocks from different facilities, Piedrahita-Rodríguez et al. (2023) proposed a multi-feedstock biorefinery utilizing avocado and plantain residues to produce biogas, ethanol, and avocado seed oil. When combining feedstocks from different sources, logistical and geographical challenges can arise, particularly regarding raw material transportation. To address this, their study identified that the most optimal location for the biorefinery was proximity to the sources of avocado and plantain residues production and to the market for the traded products. This strategic location minimizes transportation issues.

An example of a semi-commercial phase III plant is Zea2 LLC (formerly ZeaChem), which developed its first commercial lignocellulose feedstock biorefinery in Oregon (US). Feedstocks processed include agricultural residues such as corn stover, wheat straw, sugarcane bagasse, energy cane, and sugar beets, with a processing capacity of 10 tonnes/day. The company can produce bio-based fuels, C2 chemicals (acetic acid, ethyl acetate, ethanol and ethylene) and C3 chemicals (propionic acid, propanol and propylene) using a hybrid process of biochemical and thermochemical processing (Clark and Deswarte, 2014).

3.2. Product integration

The zero-waste concept is directly associated with a decrease or elimination of waste streams, especially in the context of agro-industrial wastes. In this sense, the zero-waste concept encourages sustainable production and consumption, optimal recycling and resource recovery, while minimizing mass incineration and landfill disposal (ZWIA, 2018). Contrary to this approach, most biomass processing technologies focus on isolating a particular component of the lignocellulosic material, thus wasting the remaining components. Incomplete fractionation of lignocellulosic biomass produces impure isolated fractions, which may lead to lower yields of final products. As for instance, separation of hemicellulose and lignin is used to increase the accessibility of hydrolytic enzymes to cellulose, which enhances its digestibility and hence the final bioethanol production (Abraham et al., 2020). Therefore, by valorizing the main fractions of agro-industrial waste, which consists of approximately 90 % lignocellulose material, not only does it fulfill the zero-waste goal, but it also leads to the isolation of purer fractions (Fig. 3).

As evident from the data presented in Tables 1 and 2, the predominant outcome is ethanol. Traditional ethanol production methods primarily target the conversion of cellulose-derived hexoses into ethanol, while pentoses derived from hemicellulose are often lost during the pre-treatment process or are not used by the ethanol-fermenting strain. An alternative approach involves pre-treating the biomass to extract hemicellulose, yielding mainly pentoses that can be used to produce xylitol through fermentation (de Souza Queiroz et al., 2023; Franceschin et al., 2011). Since lignin is recognized as the lignocellulosic material least susceptibility to chemical and biological transformation techniques (Kohli et al., 2019), it is recommended to separate it early in the process to minimize the energy and material resources needed for its processing (Obydenkova et al., 2022). Nevertheless, lignin properties, such as thermal stability, phenolics composition, and molecular weight can vary depending on the stage of separation, as demonstrated by Dávila et al. (2019). Their research focused on a cascade biorefinery approach for vine shoots, aiming to produce bioethanol and lignin with characteristics suitable for value-added applications. Lignin has valuable potential applications in industries such as chemicals, food, pharmaceuticals, textiles, and cosmetics. In addition, researchers and industries are actively investigating lignin as an alternative to petroleum-based materials, particularly in the development of functional materials such as electrode components, separators, and electrolytes. This represents a promising avenue for lignin-based materials in energy-related applications (Beaucamp et al., 2022).

Few research papers have explored comprehensive zero-waste approaches for valorizing the three key components of lignocellulose material. One noteworthy example is the cascade biorefinery method of Michelin et al. (2018), which involved fractionation and multi-valorization of cellulose, hemicellulose, and lignin from corncobs. The process began by grinding and subjecting the raw material to autohydrolysis to disrupt its structure. This resulted in a hemicellulose-rich hydrolysate and a solid fraction comprising cellulose and lignin. The hydrolysate was employed in a fermentation process to generate xylanases, while the solid fraction underwent enzymatic hydrolysis, yielding lignin with antioxidant properties and glucose. Later, the glucose was used in a fermentation process to produce cellulase. Additional studies proposing zero-waste approaches for the utilization of agro-industrial products include the research of Gullón et al. (2020) focusing on by-products from the olive oil industry, and the work of Del Castillo-Llamosas et al. (2021) focused on avocado peels and seeds. An interesting approach to achieve a zero-waste objective is to utilize the residual solids remaining after carbohydrate fermentation for biomanure production. Chintagunta et al., (2017, 2016) adopted this strategy for the comprehensive multi-valorization of both pineapple leaves and potato waste. In both instances, the residue left after bioethanol production via SSF was enriched with nutrients such as nitrogen, phosphorous and potassium. This enriched solid residue serves as biomanure and is recognized as a cost-effective alternative to chemical fertilizers.

3.3. Technology integration

The establishment of a viable and enduring biorefinery requires its seamless integration into the total chain. At the facility level, integration of energy and mass processes must be achieved to reduce the demand for auxiliary utilities and the acquisition of fresh resources (Budzianowski and Postawa, 2016) and the adoption of process intensification strategies is crucial to increase the efficiency (Table 4). Mass integration encompasses a spectrum of strategies that involve mitigating raw material demand, recycling process streams, consolidating unit operations, and intensifying process efficiency. Conversely, energy integration is oriented towards the development of a heat exchange network and the incorporation of power cycles to optimize the efficiency of each biorefinery stage (Solarte-Toro and Cardona Alzate, 2023).

The most common example of mass integration in biorefineries involves the recovery of solvents employed in the extraction of bioactive compounds or in the organosolv pre-treatment process, including solvents such as water (Gullón et al., 2020), chloroform (Zhao and Liu, 2019), ethanol (Franceschin et al., 2011; Morales et al., 2020), or formic

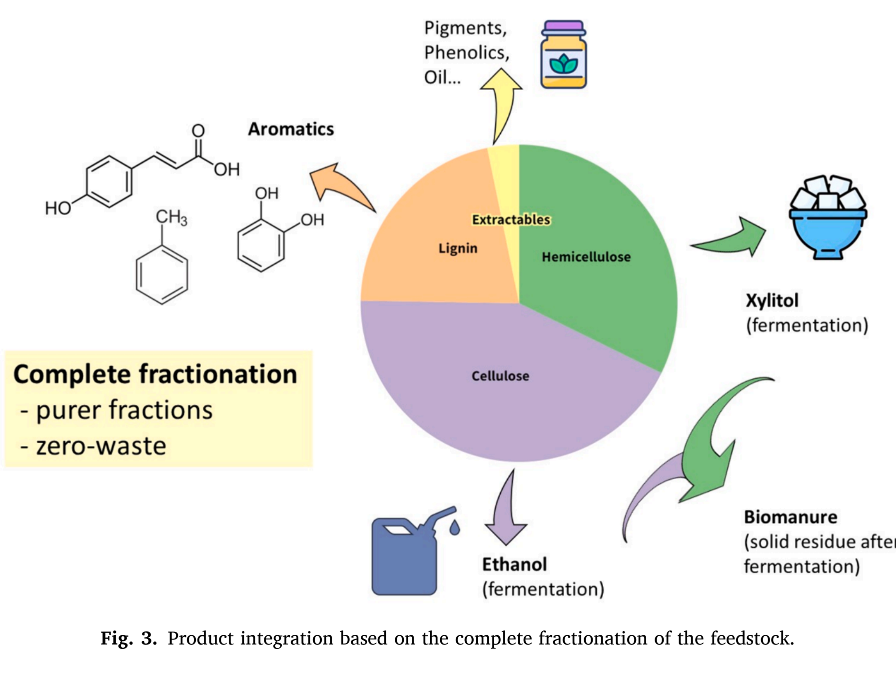

Fig. 3. Product integration based on the complete fractionation of the feedstock.

Table 4
The three main pillars of technology integration in biorefineries.

Mass integration	Mitigating raw material demand Recycling process streams: recovery of solvents used in extraction processes, recuperation of CO_2 from fermentations, recovery of water
Energy integration	Heat exchange network Power cycles Thermochemical use of solid residues Biogas from anaerobic digestion
Process intensification	Non-conventional extraction methods (microwave, ultrasounds…) Simultaneous saccharification and fermentation Consolidated bioprocessing Use of ionic liquids or reactive extraction in downstream

acid (Qiao et al., 2022). Furthermore, in instances where the facility produces bioethanol, a portion of the solvent generated may find utility in the pre-treatment or extraction of bioactive compounds, as exemplified in the study conducted by Borujeni et al. (2022). Dávila et al., (2017a) conducted a simulation of a model biorefinery, demonstrating that mass integration strategies resulted in a noteworthy 21 % reduction in the overall cost of raw materials used in the biorefinery. This reduction was particularly prominent in the case of ethanol, the primary product of the facility, which was also employed in the extraction of phenolics and the crystallization of xylitol. Other possibilities for mass integration in biorefineries encompass the recuperation of CO_2 generated during fermentations, which can be transformed into a carbon carrier, such as succinic acid, during acid fermentation. This approach not only facilitates the reduction of greenhouse emissions, but also enhances the overall sustainability of the process (Luo et al., 2010).

Leveraging energy integration via heat management is a critical opportunity for the cost-effective design of lignocellulosic biorefineries (Salvador et al., 2022). Energy-intensive operations, including distillation and pre-treatment, can be harmonized with steam generation through thermochemical utilization of solid residues (Balan, 2014). The pinch point method is applied to scrutinize available heat sources and sinks for effective heat integration analysis (Salvador et al., 2022; Wiranarongkorn et al., 2023). Naranjo et al. (2014) conducted simulations for both mass and energy integration in the biorefinery of residual banana plants. Implementing energy integration resulted in a remarkable 30.6 % reduction in overall energy consumption. Conversely, mass integration achieved a substantial 35 % reduction in water usage, as determined by the composite curves, which provide formation on the interactions between hot and cold streams throughout the entire biorefinery system. Mussatto et al. (2013) assessed a scenario in which water from various processing plants was recovered and reused, achieving mass and energy integration with the assistance of composite curves, particularly focusing on water management. Nieder-Heitmann et al. (2019) put forward a simulation for a biorefinery plant that incorporated a combined heat and power system, employing a condensing-extraction steam turbine. Furthermore, they suggested that surplus electricity generated could be fed into the grid for distribution. An alternative to energy integration is the utilization of biogas generated by anaerobic digestion, sourced from a biorefinery facility engaged in the extraction of biodiesel, bioethanol, and biogas from brewers' spent grain (Kavalopoulos et al., 2021).

A third key factor to ensure technology integration in biorefineries is process intensification (Stankiewicz and Moulijn, 2020). Process intensification can be defined as any effort aimed at achieving five key objectives by reducing equipment size, increasing production capacity, minimizing material inventory, reducing resource consumption, and improving overall performance for a given unit size (Ponce-Ortega et al., 2012). This approach has the potential to reduce costs and environmental impacts while improving operability and safety. Biorefineries can intensify their processes by incorporating state-of-the-art technologies such as microwave, ultrasound, pulse electric fields, and supercritical and subcritical fluid extraction. Process intensification is focused on the development of innovative, energy-efficient processes, reducing the number of unit operations, and enhancing resource efficiency through recovery and reuse. Additionally, it underscores the efficient handling of solvents and water, including rainwater recovery, as well as the design of more compact unit operations (Perino and Chemat, 2019; Segovia-Hernández and Sánchez-Ramírez, 2022).

The most process intensification, which features prominently in Tables 1 and 2, is simultaneous saccharification and fermentation (SSF), which stands out as a highly promising strategy for enhancing overall productivity. SSF is a bioconversion technique in which enzymatic hydrolysis (saccharification) and fermentation occur simultaneously in a single reactor. This approach finds extensive utility in the production of biofuels, particularly ethanol, from lignocellulosic biomass (Verardi et al., 2023). Recent research has introduced consolidated bioprocessing (CBP), which merges pre-treatment, saccharification, and fermentation in a single reactor, making it the focus of extensive study (Periyasamy et al., 2023). It is worth noting that while CBP offers the highest degree of intensification, it has not yet reached the level of industrial implementation due to certain challenges. These challenges include the need for reliability in well-established technologies, concerns about risks associated with lack of precedents, the expenses related to new equipment, safety and control issues, the absence of validated process intensification units, and the absence of performance evaluation metrics. Other intensification techniques in the field of liquid–liquid extraction include hybrid processes, reactive extraction, the use of ionic liquids, etc., which are applied in downstream processing of the fermentation products (Antony et al., 2021).

4. Perspectives and challenges of integrated biorefinery processes in the bioeconomy

Biorefineries play a fundamental role on sustainable development acting as a strategic mechanism for the realization of a circular bioeconomy. However, biorefineries and in particular agro-industrial biorefineries, still face significant hurdles prior to industrial scale-deployment. The challenges can be broadly categorised into the three main pillars of sustainability, and the economic and environmental challenges can be further subcategorized into those related to biomass or process (Fig. 4).

Most research on agro-industrial waste technologies is conducted at the laboratory scale at a low technology readiness level (TRL), resulting in a dearth of information on their feasibility and performance at the industrial scale (Rodríguez-Martínez et al., 2022). Consequently, the commercialization of these technologies is very limited. Factors limiting the commercialization of biorefinery plants of agro-industrial waste include the complex structure and seasonality of lignocellulosic biomass, the need to develop technologies that strike a balance between economic and industrial viability, the complexity of bioprocesses, transferring laboratory findings to a larger production scale, and financial constraints (Madeddu et al., 2021; Obydenkova et al., 2022; Yaashikaa et al., 2022).

According to Martinez et al. (2015) integrated biorefinery processes may not be economically feasible if implemented individually, and the profitability of bioprocesses is typically only guaranteed at high production capacities. To assess the viability of integrated biorefinery processes on an industrial scale, it is essential to conduct comprehensive laboratory and pilot plant experimental studies, as well as techno-economic and environmental impact analyses (Battista et al., 2021). Table 5 summarizes multi-biorefinery schemes that have been subjected to techno-economic analysis (TEA), environmental analysis (EA), and life cycle assessment (LCA). According to Table 4, most techno-economic and/or environmental analyses are based on laboratory-

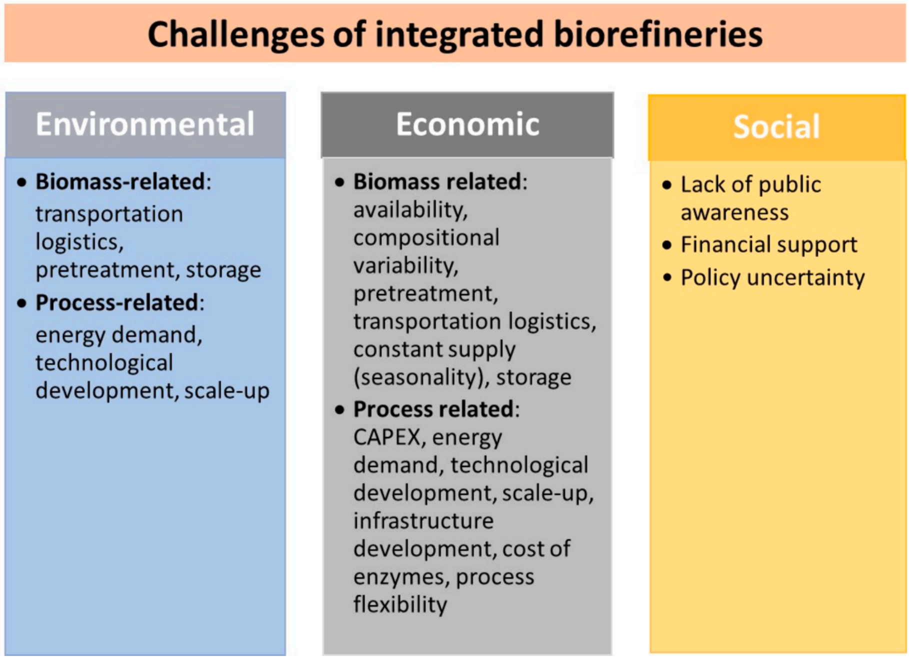

Fig. 4. Challenges in the sustainability of agro-industrial biorefineries.

scale results, despite the well-known challenges of scaling up bioprocesses while maintaining high yields. In fact, there is a limited number of pilot studies on lignocellulosic biomass biorefineries. Examples of larger laboratory scales include a study on the production and purification of succinic acid in a sugar beet pulp biorefinery, comparing results obtained at the laboratory scale (5 L) and pilot scale (50 L) (Alexandri et al., 2019), dilute acid hydrolysis of citrus residues in a pilot plant reactor equipped with an explosive drain (Pourbafrani et al., 2010), and extraction of phenolics from red grape pomace with supercritical CO_2 using a commercial pilot plant (Martinez et al., 2015).

Utilizing life cycle assessment (LCA) frequently reveals that products derived from lignocellulosic materials exhibit superior environmental performance compared to those sourced from fossil fuels (Moretti et al., 2021). This alternative presents a significant potential to mitigate overall climate impact. LCA study confirms that bio-based methanol production through sugarcane biorefineries exhibits superior technical efficiency compared to the traditional Fischer-Tropsch process (Farzad et al., 2017). An additional LCA study posits that procuring bio-based succinic acid from winery waste streams lacks profitability compared to conventional petroleum-based production. Nonetheless, it suggests that through its production into a biorefinery process alongside other co-products, a pioneering and sustainable bioeconomy business model may emerge, presenting novel market prospects and streamlined waste management practices (Ioannidou et al., 2022).

Research on lignocellulosic biorefineries has shown that biomass pre-treatment is the major economic and environmental bottleneck, followed by enzymatic hydrolysis, enzyme production and fermentation (Obydenkova et al., 2022). Pre-treatment is often essential for the economic viability of converting lignocellulosic biomass to high-added value compounds, and it can also impact the performance and cost of virtually all other downstream operations (Yang and Wyman, 2008). As shown in Tables 1 and 2, most studies have recommended a pre-treatment step to extract lignin, which facilitates the solubilization of hemicellulose for xylitol production and the use of cellulose for a fermentation process (Clauser et al., 2016; Franceschin et al., 2011; Obydenkova et al., 2022). Nieder-Heitmann et al. (2019) identified plant capacity as a key factor in the economic viability of a process, as it directly affects the product's selling price and the cost of equipment. For established products, high production capacities are desirable, while high-value-added products can be produced in smaller volumes alongside other low-value products, such as biofuel or electricity (Battista et al., 2021). Other authors have emphasized the importance of mass integration for reducing process intensification environmental impact, as it generates fewer waste streams (Dávila et al., 2017a; Ioannidou et al., 2022). Energy integration is also crucial, as it can reduce the total energy cost of a biorefinery by up to 70 % (Dávila et al., 2017b). Mass integration can also save large amounts of inputs, leading to greater economic incomes, lower environmental impacts, and reduced greenhouse gas emissions due to energy consumption (Mussatto et al., 2013). Process intensification approaches can significantly mitigate the limitations of unit processes by combining more than two unit operations in a single step (Muniasamy et al., 2023). Another important consideration, both economically and environmentally, is the integration of first, second, and even third generation biorefineries. This can improve the profitability of the process and reduce environmental impacts (Dávila et al., 2017b; Martinez et al., 2015).

The trend towards social awareness within the circular economy framework emphasizes the pivotal role of government policies in establishing biorefineries. Many countries have adopted national strategies to promote the commercialization of lignocellulosic biorefineries. These strategies typically include defined development pathways, financial subsidies, tax breaks, and incentives designed to accelerate biofuel production (Chen et al., 2023). According to Palgan and McCormick (2016), not only stable, long-term and targeted policies are critical for the development of biorefineries, but also collaboration between sectors and actors, as well as improving the alignment of visions to avoid contradictions in public perception.

5. Conclusions

Inappropriate management of agro-industrial waste causes problems for the environment, the economy and society. There are many proposals for biorefinery schemes to obtain high value-added compounds from these wastes, but most of them do not achieve zero-waste, or only achieved it some of the biomass fractions for energy production.

As the economic viability of individually applied biorefinery processes is difficult to achieve, an interesting strategy is the development of integrated multi-product biorefineries. With the aim of achieving this

Table 5
Techno-economic analysis (TEA) and environmental analysis (EA) of the proposed integrated multi-product biorefineries.

Agro-industrial Waste	Scale	Target compounds	TEA *	EA*	References
Chestnut shells	Lab	Lignin and bioethanol	−[1]		(Morales et al., 2018)
Rye straw	Simulation with data from lab and pilot plant (Ingram et al., 2009)	Ethanol and xylitol	+		(Franceschin et al., 2011)
Wheat straw	Simulation with data from lab (literature)	Bioethanol, lignin, and furfural	+		(Zhao and Liu, 2019)
Sugarcane bagasse	Simulation with data from lab (literature)	Xylose syrup and medium-density fibreboard	+		(Clauser et al., 2016)
	Simulation with data from lab (literature)	PHB, succinic acid and electricity	+		(Nieder-Heitmann et al., 2019)
	Simulation with data from lab (literature)	(Scenario 1: ethanol, lactic acid, and electricity; Scenario 2: Methanol and electricity)	+	+ (LCA)	(Farzad et al., 2017)
Spent coffee grounds	Lab	Biodiesel, biogas, and fermentable sugars	+[1]		(Battista et al., 2021)
Residual banana	Simulation with data from lab (literature)	Ethanol, PHB, fertilizer	+	+ (EIA)	(Naranjo et al., 2014)
Citrus waste	Lab	Essential oils, pectin, succinic acid and biomethane	−[1]		(Patsalou et al., 2017)
Blackberry spent pulp	Simulation with data from lab (literature)	Phenolics, ethanol, xylitol	+	+ (EIA)	(Dávila et al., 2017a)
Sugar beet pulp	Lab	Pectin, phenolics, proteins, and succinic acid	+[1]		(Alexandri et al., 2019)
Grape pomace, stalks, wine lees	Simulation with data from lab (Filippi et al., 2022)	Bacterial cellulose, oil, phenolic-rich extract, ethanol, crude tannin extract, tartaric acid, succinic acid	+	+ (LCA)	(Ioannidou et al., 2022)
Avocado pulp, skins and seeds	Simulation with data from lab (literature)	Oil, phenolics, ethanol, xylitol	−	+ (EIA)	(Dávila et al., 2017b)
Brewer's spent grains	Simulation with data from lab (literature)	Xylitol, phenolic acids, activated carbon and lactic acid	+	+ (EIA)	(Mussatto et al., 2013)

* + or − indicates that the outcome was positive or negative, respectively; Environmental impact assessment (EIA); Live cycle assessment (LCA).
[1] Preliminary economic analysis based on simple mass balances.

objective, various methods for processing agro-industrial waste and obtaining multiple products have been discussed in this comprehensive review.

For these proposals to become an industrial reality, techno-economic and environmental feasibility analyses are required. However, most of the available studies rely on simulations using laboratory-scale results, despite the difficulties of maintaining the yield when scaling up bioprocesses. Hence, testing the technology in a pilot plant, including process intensification and integration of mass and energy, is still a clear barrier in the deployment of biorefineries at industrial scale. Driving the large-scale deployment of sustainable biorefineries depends to a large extent on theintegration of interdisciplinary expertise, industry and stakeholder efforts and coordination with policy makers.

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CRediT authorship contribution statement

Inés Prado-Acebo: Writing – original draft, Methodology, Data curation, Conceptualization. **Juan Cubero-Cardoso:** Writing – review & editing, Writing – original draft, Methodology, Data curation, Conceptualization. **Thelmo A. Lu-Chau:** Writing – review & editing, Visualization, Conceptualization. **Gemma Eibes:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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