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Review

Review of methodological decisions in life cycle assessment (LCA) of biorefinery systems across feedstock categories

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ABSTRACT

The application of life cycle assessment (LCA) to biorefineries is a necessary step to estimate their environmental sustainability. This review explores contemporary LCA biorefinery studies, across different feedstock categories, to understand approaches in dealing with key methodological decisions which arise, including system boundaries, consequential or attributional approach, allocation, inventory data, land use changes, product end-of-life (EOL), biogenic carbon storage, impact assessment and use of uncertainty analysis. From an initial collection of 81 studies, 59 were included within the final analysis, comprising 22 studies which involved dedicated feedstocks, 34 which involved residue feedstocks (including by-products and wastes), and a further 3 studies which involved multiple feedstocks derived from both dedicated and secondary sources. Many studies do not provide a comprehensive LCA assessment, often lacking detail on decisions taken, omitting key parts of the value chain, using generic data without uncertainty analyses, or omitting important impact categories. Only 28% of studies included some level of primary data, while 39% of studies did not undertake an uncertainty or sensitivity analysis. Just 8% of studies included data related to dLUC with a further 8% including iLUC, and only 14% of studies considering product end of life within their scope. The authors recommend more transparency in biorefinery LCA, with justification of key methodological decisions. A full value-chain approach should be adopted, to fully assess burdens and opportunities for biogenic carbon storage. We also propose a more prospective approach, taking into account future use of renewable energy sources, and opportunities for increasing circularity within bio-based value chains.

1. Introduction

Earth is currently at a critical juncture in time, facing many sustainability challenges. Population has been increasing exponentially, growing from 626 million in 1700 to 2 billion by 1930, on to a projected 10 billion inhabitants by 2050 (Goldewijk, 2005; Searchinger et al., 2019). This growing population brings with it a growing middle-class and a growing demand for protein, materials and energy. At the same time, Earth faces a climate crisis, reflected in the Paris Climate Accord target limiting global warming to well below 2, preferably to 1.5 °C, compared to pre-industrial levels. Climate change is one of four earth system processes, alongside biosphere integrity, biogeochemical flows, and land-system change, which exceed the proposed planetary boundaries, defining the environmental limits within which humanity can safely operate (Steffen et al., 2015). Loss of biosphere integrity is also represented in the widely acknowledged biodiversity crisis, brought about in part, by practices and policies aimed at ensuring that land- and resource-intensive production systems meet increasing demands from consumers (Crenna et al., 2019). At a global level many of the wider sustainability challenges we face are highlighted within the United Nations Sustainable Development Goals (SDGs).

Development of a sustainable bioeconomy, defined as an economy that relies on renewable natural resources to produce food, energy, products and services, may be one strategy which can help humanity to meet many of the challenges that it currently faces (Barrett et al., 2021). Whereas various specific technologies, such as renewable energy technologies, may help to address one or two of the challenges we face, the development of the bioeconomy and bio-based technologies may help to address multiple challenges, such as increasing renewable energy and materials, ensuring sufficient food production and supporting climate

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mitigation, simultaneously. According to Lokesh et al. (2018), a successful bioeconomy aligns with 11 of 17 UN SDGs, while a successful circular economy – defined by Geissdoerfer et al. (2017) as a regenerative system in which resource input and waste, emission, and energy leakage are minimised by slowing, closing, and narrowing material and energy loops – may help to meet 10 UN SDGs. The Circular Bio-Society 2050 Vision describes a scenario in which food, renewable products and energy for our communities are provided in a sustainable manner through circular bioeconomy principles (Bio-based Industries Consortium, 2018). However, a bioeconomy, and the use of its biological raw materials, can also be unsustainable, and this will be addressed later.

Biorefineries represent key enabling technologies for the widespread and successful implementation of the bioeconomy concept. A biorefinery is defined by IEA Task 42 Biorefinery as the sustainable processing of biomass into a spectrum of marketable products and energy (Cherubini et al., 2009b). In a biorefinery, a combination of processes (mechanical, biological, chemical and thermochemical) may be applied to convert biomass into a range of end products including fertilisers, chemicals building blocks, polymers and resins, food, animal feed and biomaterials as well as energy (Cherubini et al., 2009b). According to Platt et al. (2021) approximately 300 chemical and material driven biorefineries operating at commercial or demonstration scale have been identified across the European Union (EU). Meanwhile it is estimated that there is the potential for up to 300 new commercial biorefineries to be operational by 2030 (European Commission, 2018). The further development of the bioeconomy and biorefineries have been promoted through the EU bioeconomy strategy, research and demonstration initiatives such as Circular Biobased Europe Joint Undertaking (CBE JU), and through national level policies such as Italy's new Bioeconomy in Italy (BIT II) Strategy and the National Bioeconomy Strategy of Germany (National Bioeconomy Task Force, 2019; The Federal Government, 2020). Aside from facilitating resource-efficient and high-value uses of biomass, through cascading use of resources, biorefineries can produce materials which incur reduced emissions compared to prevailing conventional products on the market. The carbon footprint of the drop-in biopolymer bio-polyethylene (bio-PE), for example, has been found to be −3.9 CO_2eq per kilogram of bio-based bio-PE produced compared to 1.8 CO_2eq per kilogram of fossil-based bio-PE produced, with CO_2 uptake during sugarcane production making the largest contribution to CO_2 emission mitigation (Ziem et al., 2013). Other environmental benefits may include a reduction in plastic waste accumulation through use of recyclable and compostable bioplastics (World Economic Forum, 2016).

At the same time, the sustainability of the bioeconomy is not a given, and there are many pitfalls which could derail its potential. According to Platt et al. (2021), sustainability considerations could represent a barrier to implementation of biorefineries in Europe, as there remains a lack of evidence on the sustainability of bio-based products, with some bio-based products offering low greenhouse gas (GHG) savings, or even GHG increases, compared with fossil products, depending on the feedstocks and pathways used. A number of studies debate the actual emission-saving benefits of biofuels and biomaterials, particularly when considered over the full life cycle (Piemonte and Gironi, 2011; Smith and Searchinger, 2012). In addition, the developing bioeconomy may impact negatively on other aspects of our environment, like biodiversity, ecosystem functioning and water quality. A major component of the EU Commission's 2018 Bioeconomy Strategy Update, was to better understand the ecological boundaries of the bioeconomy (European Commission, 2018). Recently, attention has also shifted to ensuring better utilisation of bio-waste and by-products within the bioeconomy, with the 2018 Strategy Update, noting this as a central component of the bioeconomy's contribution to a circular economy (European Commission, 2018). Competition for land use and the "food versus fuel" controversy of using food for material or energy applications, have also contributed to a shift in focus towards residue feedstocks (Hassan et al., 2019). Adding to this are concerns around the actual environmental benefits that dedicated feedstocks can deliver. For example, a previous LCA study found that the global warming potential (GWP) of corn ethanol and soya bean biodiesel in China were 40% and 20% higher than petrol and diesel, respectively, owing to the relatively high use of fertilizers, high process energy consumption and the coal-dominated energy mix of China (Jeswani et al., 2020).

LCA is a methodology commonly used to assess the environmental efficiency of biorefineries and bio-based products. Based on the ISO14040 series and ISO14044:2006, LCA provides a framework to quantify the environmental impact arising over an entire value chain, product or service. A number of previous studies have reviewed the application of LCA specifically to biorefineries, or to specific bio-based product categories. Ahlgren et al. (2015) reviewed 12 standards and guidelines relevant for LCA of biorefinery systems, providing recommendations on how to handle key methodological issues when performing LCA studies of biorefinery systems. A review study by Bernstad Saraiva (2016) of 38 biorefinery LCA studies, focused primarily on biorefinery feedstock provision as well as the influence of system boundary definition on LCA results. Using a hybrid approach, Liu et al. (2021) conducted a high-level review of LCA studies focused on waste-feedstock biorefineries, while also demonstrating an LCA case study model for a microalgal biorefinery. Katakojwala and Mohan (2021) conducted a critical review of the environmental sustainability of biorefinery systems, with a high-level exploration of some key LCA considerations for biorefineries including system boundary definition, impact assessment method selection, and representation of uncertainties. Talwar and Holden (2022) focused on the limitations of bioeconomy LCA studies for understanding the transition to sustainable bioeconomy, with a focus on goal, system boundary and impact assessment methods. Focusing on specific products which can be produced in biorefinery systems, Bishop et al. (2021) explored the critical methodological decisions for LCAs specifically comparing bioplastics with fossil-based plastics. Soleymani Angili et al. (2021), meanwhile, evaluated 48 papers describing LCA of bioethanol production, considering different methodological approaches, feedstock types and technology pathways, while Osman et al. (2021) reviewed LCA studies related to biomass to biofuel conversion more broadly, including 40 studies published between 2019–2021, focusing on a comparison between biochemical and thermochemical conversion routes, and exploring LCA methodological approaches. From the review of previous studies, there is a lack of studies which comprehensively compare LCA methodologies as applied to biorefineries of different feedstock types. This is a significant gap, given that feedstock selection, and how their associated burdens (and potential for biogenic carbon storage) are treated, can have a significant impact on the overall sustainability of the bio-based value chain, as evidenced by Ziem et al. (2013), above. Such a focus is also timely, given the current shift towards including more residual and by-product feedstocks. The paper also focuses on a broader set of LCA methodological decision which span across the entire value chain. The focus of this current study is to undertake a holistic review of key methodological decisions in recent LCA studies applied to biorefinery systems, with the following novelties: (i) a focus on (diverging) feedstock specificities; (ii) a focus on important gaps in studies from a whole value chain perspective regarding longitudinal issues such as allocation and biogenic carbon storage; (iii) consideration of wider (future) contexts in which biorefineries (will) operate; (iv) presentation of recommendations arising from the analysis, including future areas of research and development. Based on this review, recommendations are provided that will help guide future studies in this area.

2. Materials and methods

For the current study, a review was undertaken of recent peer-reviewed publications of LCA studies applied to biorefinery systems. Scopus and Web of Science were used to search the literature, ensuring a broad coverage of pertinent studies. The search included variations of the following keywords: “life cycle assessment”, “life cycle analysis” or “LCA” in combination with terms including “biorefinery”, “bio-refineries”, “bio-processing” and “biomass conversion”. To ensure a contemporary review of the literature, the search included studies which were published from January 2016 until March 2022. This focus on contemporary studies was to reflect state-of-the-art in LCA methodology, which is continuously evolving, but also to capture a growing focus on residue, waste or by-product feedstocks, in addition to dedicated feedstocks. Only peer-review publications in the English language were included within the analysis. In total, 81 studies were included within the initial screening. On initial inspection, seven of these studies were screened out, either because they were review-only papers, or because LCA methodology was not applied. Then, to ensure only inclusion of studies which aligned with the definition of biorefineries being systems which produce multiple products (Cherubini et al., 2009b), a further 15 studies relating to single material or energy product systems were screened out. This resulted in 59 studies being included within the final analysis. The screening procedure is summarised in Fig. 1 below.

Studies were categorised according to feedstock type and reviewed across a range of key LCA methodological considerations for biorefinery systems. The main considerations are presented in Fig. 2 below and apply to different or multiple points within the biorefinery system, from feedstock production right through to end use of products, along with system level considerations.

The methodological considerations include.

- Functional unit: The functional unit represented by #1 in Fig. 2, is the basis that enables alternative goods, or services, to be compared and analysed within LCA studies (Rebitzer et al., 2004).
- System boundary: In LCA, the system boundary, highlighted by #2 in Fig. 2 indicates the boundaries defined between the product or system under study and the surrounding systems (Khatri and Pandit, 2022).
- Attributional versus Consequential LCA: Two common modelling approaches used within LCA are attributional LCA (ALCA), which assesses the global impact share of a product’s life cycle, and consequential LCA (CLCA), which evaluates the consequential impact of a decision (Schaubroeck, 2023). This decision is displayed schematically at #3 in Fig. 2.
- Allocation: During LCA, allocation, represented by #4 at both feedstock and product points in Fig. 2, refers to the partitioning of the input or output flows of a process or a product system between the primary product under study and one or more other co-products (Pelletier et al., 2015).
- LCA Inventory data: Within LCA methodology, the step of collecting data is known as life cycle inventory (LCI), and involves creating an inventory of input and output flows for a system under study (Vaskan et al., 2018). This is highlighted by #5 in Fig. 2.
- Land use changes: This is highlighted by #6 in Fig. 2 and refers to changes in land use which can be subdivided into Direct Land-Use Changes (dLUC) which are changes in human use or management of land within the boundaries of the product system being assessed, while indirect Land-Use Changes (iLUC) are changes in the use or management of land which is a consequence of direct land use change, but which occurs outside the product system being assessed (De Rosa, 2018).
- Product use and end-of-life: This is highlighted by #7 in Fig. 2 and concerns the impact of the products produced within the biorefinery at the use and end-of-life (EOL) phase.
- Biogenic carbon storage: This is highlighted by #8 in Fig. 2 and refers to the carbon which is stored in feedstock of biological origin, or embedded in bio-based materials produced within biorefineries.
- Impact Assessment: Life cycle impact assessment (LCIA) is the phase of an LCA where the evaluation takes place of the potential environmental impacts stemming from the elementary flows (environmental resources and releases) obtained in the life cycle inventory (Nieuwlaar, 2013). It is highlighted at #9 in Fig. 2.
- Sensitivity and uncertainty analyses: Sensitivity and uncertainty analyses are recommended to support interpretation of LCA results (Cucurachi et al., 2021; Steen, 1997). In Fig. 2 these decisions are presented at #10.

3. Results and discussion

To begin with, studies were categorised based on the primary feedstock type used within the biorefinery. Feedstock type and origin are important factors that can determine how some environmental burdens are allocated. When it comes to classifying feedstocks for biorefineries, Cherubini et al. (2009b) makes a distinction between dedicated feedstocks, such as sugar and starch crops, grasses, lignocellulosic crops and marine biomass, and those which constitute residues, including crop residues, oil-based residues and organic residues. This distinction has become more relevant in recent years as the use of residues to develop bio-based materials and energy has become central to the concept of a circular bioeconomy. According to Cherubini et al. (2009a) the source of biomass has a significant impact on LCA outcomes, with notable differences between purpose grown biomass versus biomass residues and wastes. In this review of 59 studies, 22 studies involved dedicated feedstocks, while 34 involved residue feedstocks (including by-products and wastes), and a further 3 studies involved multiple feedstocks derived from both dedicated and secondary sources. In the literature biorefineries have also been classified as first, second, third or fourth-generation, depending on feedstock source (Almada et al., 2023). First-generation biorefineries depend on dedicated feedstocks cultivated

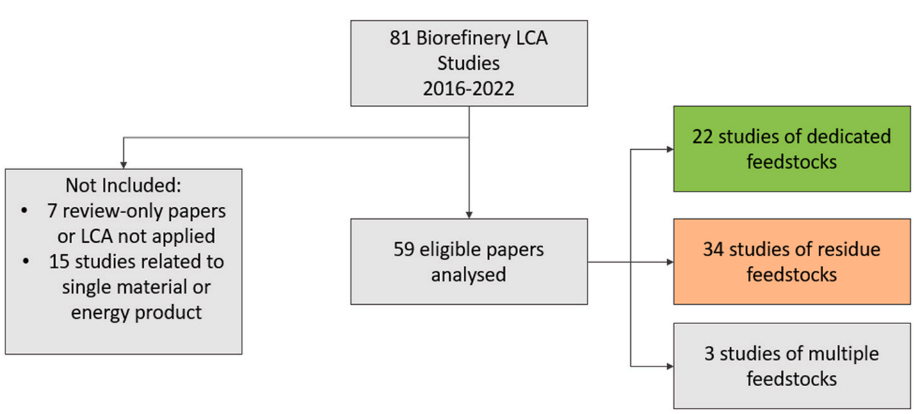

Fig. 1. Summary of the screening approach for LCA studies.

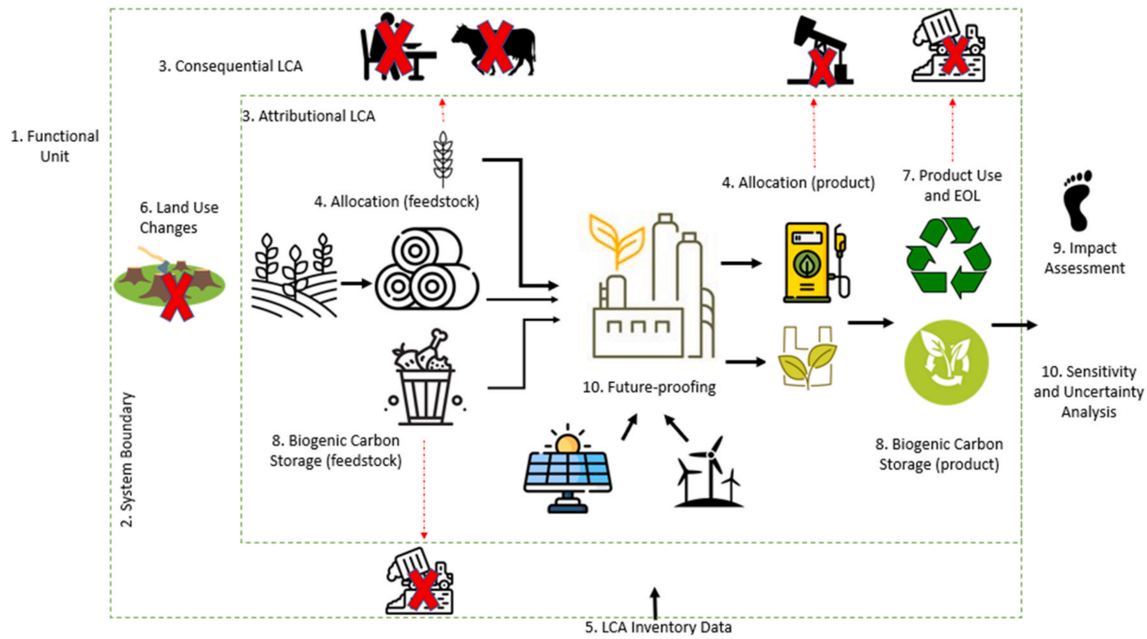

Fig. 2. Important methodological considerations (numbered) when applying LCA to biorefinery systems and associated value chains.

on lands, such as edible crops or forest plantations. Second-generation biorefineries employ by-products and residues such as lignocellulosic biomass from forestry, agricultural activities, or municipal wastes. Third-generation biorefineries rely on aquatic feedstocks such as algae, while fourth-generation biorefineries employ genetically modified algae, cyanobacteria, and crops. Under this classification, the following breakdown of biorefineries can be distinguished from the studies analysed. Under this classification 19 studies fall into the category of first-generation biorefineries, 34 fall into the category of second-generation, while 3 fall into third-generation, with no processes identified as fourth-generation. Three studies focusing on multiple feedstocks fall between first and second-generation processing. During this study, key methodological decisions for application of LCA to biorefinery systems were reviewed and the result of this analysis is further discussed below.

3.1. Functional unit

The selection of a functional unit is one of the earliest decisions when planning an LCA study. According to Ahlgren et al. (2015), the functional unit (FU) is a particularly important consideration for biorefineries, since these systems produce more than one product, and it may therefore be difficult to identify a single main function. Sills et al. (2019) states that a misleading FU can lead to incorrect results and conclusions about the environmental performance of a process under study, while a correctly defined FU is more likely to yield an "apples-to-apples" comparison across products and scenarios. Selection of an appropriate FU to form the basis for comparisons between new bio-based alternatives, and incumbent products, such as fossil-based materials, is critical, since many of these products are different in composition and properties. Even where a comparison is possible, it may not always be on the basis of a convenient 1:1 mass or volume replacement in a particular application. This has been seen in case of biofuels, such as ethanol, which often have a lower energy density than the replacement or blending product, such as petrol (Kralova and Sjöblom, 2010). Recent studies show that other biorefinery products such as sustainable protein feed or natural fibre insulation, likewise, do not always achieve a direct replacement of incumbent products (Franchi et al., 2020a,b).

According to Ahlgren et al. (2015) there are four main categories of FU: (i) mass (e.g. 1 kg) of feedstock which can be useful when assessing best management of waste; (ii) mass (e.g. 1 kg) of product which can be useful when communicating information about the product such as environmental labelling; (iii) function of a single product (e.g. 1 MJ electricity generation) which can help to make certain output products comparable (e.g. comparing different fuel types); (iv) multifunctional (e.g. 1 biorefinery or a portfolio of output products) which could be useful when identifying hotspots within a system as well as comparing multiple standalone systems with integrated systems. The results of the current review of FUs applied to biorefinery systems by feedstock category are presented in Fig. 3(a). The review found 27% of studies which used a feedstock-based FU, 49% of studies which used an FU based on a single product, 12 % of studies which used a FU based on the function of a single product, 8% of studies which used a multifunctional FU while 3% of studies did not specify a FU. Single product was the most likely FU to be selected in both dedicated feedstock as well as residue feedstock studies (in 55% and 41% of studies respectively). Feedstock based FUs were less likely to be used in dedicated feedstock studies than residue feedstock studies (22% as opposed to 32% studies). Given the multifunctional dimension of biorefinery systems, it may be surprising that only 8% of studies (5% of dedicated feedstock studies, and 12% of residue feedstock studies) chose a multifunctional FU. In dealing with multifunctionality, Gonzalez-Garcia et al. (2018) and Parajuli et al. (2018) selected a portfolio of biorefinery products, while González-García et al. (2016) and Gullón et al. (2018) chose an economic value and revenue-based FU respectively. Sills et al. (2019), applying the system expansion method, noted that various functional unit selections such as 1 MJ of fuel, 1 kg of animal feed, or 1 ha of cultivated land resulted in major differences in environmental burdens for two LCA indicators: ecosystem quality and non-renewable resources. Above all, Ahlgren et al. (2015) recommends that when selecting FUs for biorefineries, the FU should be closely related to the study aim.

3.2. System boundary

In an LCA, the system boundary indicates the boundaries defined between the product or system under study and the surrounding systems (Khatri and Pandit, 2022). According to Bernstad Saraiva (2016), LCA studies typically have the aim of being sufficiently comprehensive to be used as decision-making support, with comprehensiveness largely

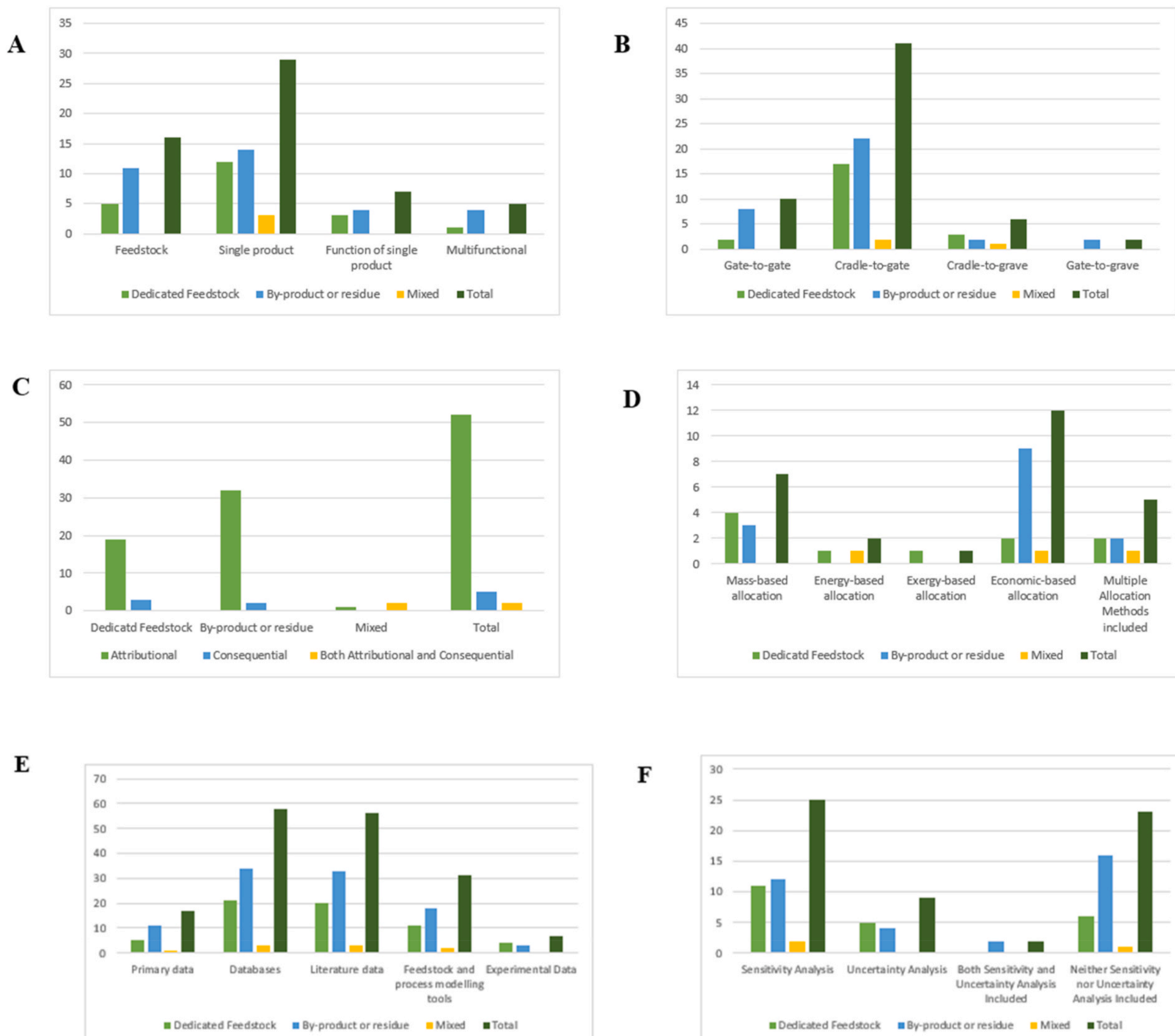

Fig. 3. Highlights of biorefinery LCA review findings according to feedstock inputs (a) Number of LCA studies for biorefineries categorised according to feedstock inputs applying different functional units (b) Number of LCA studies for biorefineries categorised according to feedstock inputs applying different system boundaries (c) Number of attributional and consequential LCA studies for biorefineries categorised by feedstock inputs (d) Number of LCA studies for biorefineries categorised according to feedstock inputs applying different methods of allocation (e) Number of LCA studies by inventory data source per feedstock category (f) Number of LCA studies by inclusion of uncertainty and/or sensitivity analysis classified by feedstock category.

determined by the setting of system boundaries, which will determine what is or is not to be considered in the assessment. In biorefinery systems reviewed within the present study, four main system boundaries were identified, namely; Cradle-to-gate, in which the study scope extends from feedstock production to production of end products within the biorefinery; Gate-to-gate, in which the study focuses only on the activities within the biorefinery; Gate-to-grave; which includes those activities within the biorefinery including the production of end products, as well as their subsequent use and disposal of products; and finally Cradle-to-grave which extends all the way from feedstock production and conversion to use and disposal of those end products. The review of biorefinery system boundary by feedstock category is presented in Fig. 3 (b). In total, the review found that 69% of studies constitute Cradle-to-gate studies, 17% were Gate-to-gate studies, 10% were Cradle-to-grave studies and 3% were Gate-to-grave studies. Cradle-to-gate was the most selected system boundary for both dedicated feedstocks and residue feedstocks (77% and 65% of studies respectively), however quite a large difference existed in the number of Gate-to-gate studies (9% for dedicated feedstocks and 24% for residue feedstocks). In addition, some residue feedstocks studies (Silva, 2021; Unrean et al., 2018) contained Gate-to-grave studies with no such studies identified for dedicated feedstocks.

Overall, a higher proportion of residue feedstock studies were likely to omit feedstock cultivation phase, implicitly treating residues as a “waste”. This is in line with findings from an earlier review of studies from 2011 to 2016 by Bernstad Saraiva (2016), which found that direct inputs and agriculture activities tended to be included in studies of dedicated biorefinery systems, while were likely to be omitted when feedstock was defined as residue. In the current review, there were no studies detected in which waste materials from biorefineries were used as feedstocks within new processes. As circularity becomes a more intrinsic component of the bioeconomy, there is likely to be a stronger emphasis on closing loops between biorefinery systems and other systems in which synergies of material, energy, water and waste gases such as CO_2, are further utilised or valorised downstream.

3.3. Attributional or consequential LCA approach

A further difference in applied LCA methodology is the choice of an attributional or consequential LCA approach, and this will largely depend on the goal and scope of the study, with selection having potential to dramatically impact the conclusions of the study. In ALCA, all relevant material and energy inputs are based on average supply data and this is most commonly used in the calculation of environmental "footprints" (Bishop et al., 2021). In this sense ALCA may be a useful and comparatively quick way to calculate, using generic data, the footprint of a bio-based product produced by a biorefinery, or to compare two similar products. CLCA, on the other hand, examines the environmental consequences of a system change, often using a market-oriented approach and considering future supply-demand shifts to model marginal effects (Ahlgren et al., 2015; Zamagni et al., 2012). CLCA may therefore be a more suitable method to estimate the environmental consequences of potential current or future changes between or within one or multiple product systems. Weidema et al. (2018) highlights the importance of taking responsibility for consequences, made possible through a CLCA approach, while Bishop et al. (2021) recommended the wider use of forward-looking CLCA built on plausible scenarios as arguably the most pertinent approach for assessing replacement of fossil-based materials with bio-based materials. In CLCA studies, aspects such as "lost opportunities" (potential diversion from existing applications) and indirect land use changes (iLUC) can become very relevant for modelling (Bernstad Saraiva, 2016). These two aspects will be discussed later in Section 3.4 and Section 3.6, and are highlighted in Fig. 1, as included with the CLCA.

The vast majority of reviewed studies (88% of studies) used an attributional LCA approach, with only 8% of studies undertaking a consequential approach (Khoshnevisan et al., 2020; Marami et al., 2022; Parajuli et al., 2018; Parsons et al., 2019; Seghetta et al., 2016), and 3% of further studies deploying both attributional and consequential approaches (Karka et al., 2017; Parajuli et al., 2017). The results are presented in Fig. 3(c) above, categorised by feedstock input. An earlier study from Alvarenga et al. (2013b) used ALCA to investigate the sustainability of bioethanol-based PVC compared with fossil-based PVC, but complementing this with CLCA, to evaluate environmental aspects that might emerge if bioethanol-based PVC induces an extra demand for bioethanol. Within the ALCA only the direct land use changes (dLUC) were accounted and based on measured data from the past (between 2003 and 2009), while in the CLCA approach land use changes (considering both dLUC and iLUC) were based on assumptions for the future (Alvarenga et al., 2013a, 2013b). Within this review, only Parajuli et al. (2017) applied ALCA and CLCA, using a common reference flow of 1 MJ EtOH + 1 kg LA. Regardless of the LCA approach adopted, the same biorefinery scenario performed better in most of the impact categories assessed, identifying similar hotspots and delivering similar recommendations. However, this should not create an assumption ALCA and CLCA will provide similar results in most cases, or that the choice is not likely to impact on outcomes. While limited comparative examples exist comparing ALCA and CLCA outcomes for biorefineries, other study areas provide warnings to this effect. A study from Styles et al. (2018) investigating dairy intensification showed wildly different results arising from ALCA and CLCA. Under ALCA a 10% reduction in the carbon footprint of milk was seen following intensification (from 1.02 to 0.92 kg CO_2eq kg^{-1}), while CLCA results varied massively by scenario, and ranged up to +2 kg CO_2eq kg^{-1} milk production shifting from the less intensive to the more intensive farm. Ahlgren et al. (2015) recommends that the choice between ALCA and CLCA be closely related to the research question and should be clearly justified by practitioners. However, few reviewed studies were so transparent, and in this regard, more clarity and justification of how the chosen approach relates to the goal should be included.

3.4. Allocation

As biorefinery systems are multifunctional systems, the issue of burden allocation is an important methodological consideration, with the issue potentially arising at both the feedstock supply and processing phases (Ahlgren et al., 2015). Ahlgren et al. (2015), in line with IS014040/14,044 guidelines, recommends that, where possible, allocation be avoided by increasing the level of detail, either using a sub-process approach or through a system expansion using substitution or system enlargement (Rebitzer et al., 2004). Where allocation is to be used, it is recommended to first allocate based on the physical relationship between products, and where this is not possible to allocate based on other relationships, with revenue as a first choice (Ahlgren et al., 2015; Rebitzer et al., 2004).

3.4.1. Feedstock allocation

During feedstock production, burdens associated with cultivation may be attributed to main feedstocks (e.g., grain) and/or to co-product feedstocks (e.g. straw). Allocating burdens to dedicated feedstocks should be relatively straight-forward, as these are generally purpose-grown feedstocks, and all of the input-related emissions associated with cultivation activities should therefore be allocated to such feedstocks (Ahlgren et al., 2015). In this review, 20 of the 22 dedicated feedstock studies include details on cultivation activities within their inventories, with a number of studies providing detailed inventory tables specifically covering feedstock cultivation (Chopra et al., 2020; Chrysikou et al., 2018; Espada et al., 2020; Ghani and Gheewala, 2018; Rahimi et al., 2018; Seghetta et al., 2016; Silalertruksa et al., 2017). Most were related to crop cultivation, with primary inputs and emissions accounted for (e.g., N_2O from fertiliser application or CO_2 from fossil inputs), with some studies (Budsberg et al., 2020; Larnaudie et al., 2021; van Schalkwyk et al., 2020) also considering carbon sequestration during the crop production phase. The evaluation of biogenic carbon will be discussed further in Section 3.8.

For residue feedstocks, a further distinction may be made between co-products or by-products which may have pre-existing uses, and waste streams which are unlikely to be further utilised, and instead may require management and disposal. While co-products or by-products may be allocated a share of the upstream environmental burdens, zero burdens are allocated to waste feedstocks, which would likely make certain feedstocks for biorefineries more environmentally sustainable from an LCA perspective (Patrizi et al., 2020). Thus, defining when and how the residue feedstock burdens or credits should be allocated can have a significant influence on the overall environmental sustainability determined for the system or its products.

While residue feedstocks are less likely to be a cause of direct input cultivation burdens, their use and diversion from existing activities, sometimes referred to by Bernstad Saraiva (2016) as "lost opportunities", may result in other environmental pressures, which can be explored using CLCA, such as reductions in soil carbon storage or depletion of soil nutrients (Bernstad Saraiva, 2016). In such instances, burdens in addition to avoided offsets and substitutions, caused by such use diversions should be attributed to the residue (Ahlgren et al., 2015).

In the current review a number of LCA practitioners used allocation in order to attribute partial burdens from upstream processes. Kachrimanidou et al. (2021) used mass allocation to distribute environmental burdens of sunflower cultivation to sunflower meal, produced during the processing of sunflower seeds into oil, and later converted into bioplastic and biodiesel. Secchi et al. (2019) likewise adopted a mass allocation approach. Other studies used economic allocation in order to attribute partial environmental burdens from upstream processes (Ali Mandegari et al., 2017; Farzad et al., 2017; Gezae Daful and Görgens, 2017; Kapanji et al., 2021; Vaskan et al., 2018; Zucaro et al., 2018). Zucaro et al. (2018) investigated the production of bioethanol from wheat straw, justifying economic allocation between the wheat grain and straw on the basis that feedstock needs to be purchased to satisfy the feedstock requirements of the biorefinery. The choice of allocation procedure may have a significant influence on the burdens allocated to the biorefinery feedstock (Ahlgren et al., 2015). According to Hierro et al. (2021), mass allocation resulted in system burden shares of 49.50% and 50.50% for barley grain and straw, respectively, while economic allocation resulted in allocation factors of 89.90% and 10.10% respectively. On the other hand, a number of study authors did not allocate burdens to residue feedstocks, instead considering the chosen feedstock as a waste – e.g. Joglekar et al. (2019) in the case of citrus peel, and Foulet et al. (2018) in the case of municipal biowaste. Several other studies adopted a similar approach of zero upstream burden allocation (Barbanera et al., 2021; González-García et al., 2016; Gullón et al., 2018; Unrean et al., 2018). However, it could be noted that several of these residues may, currently or in future, otherwise serve in valuable applications other than the one investigated within the specific LCA study, in which case the potential for diversion from an existing application should at least be discussed, and preferably explored in sensitivity analyses. For example, while excluding any burdens associated with grape marc on the basis of this being solely a waste product from bottled wine, Cortés et al. (2020), somewhat contradictorily notes in the introduction, that grape marc rape is commonly used to produce brandy spirits in a separate utilisation pathway. Zucaro et al. (2018) notes the inconsistency with which some practitioners have applied approaches to the same feedstock, with some taking a zero-burden approach and others an allocation approach, e.g., for wheat straw.

3.4.2. Product allocation

During biorefinery LCA, allocation can arise further along the value chain when assigning how to allocate burdens between products produced within the biorefinery. The current review evaluated the use of allocation methods for assigning burdens across multiple (co-) products, and the results are presented in Fig. 3(d). 47% of studies used some form of allocation between products, with the largest number using economic allocation (20% of studies), followed by mass-based allocation (12% of studies), energy-based allocation (3% of studies) and exergy-based allocation (2% of studies). A further 31% of studies avoided allocation, instead using a system expansion approach. In the case of system expansion, the main product takes all the burdens associated with material and energy inputs but also takes credits for all the burdens avoided by co-product substitution of conventional products often derived from fossil resources. For studies that used the allocation approach, the largest number of dedicated feedstock studies used mass-based allocation (18% of studies versus 9% in the case of residues), while the largest share of residue feedstock studies used economic allocation (26% versus 9% in the case of dedicated feedstocks). A further 8% of studies used a combination of different allocation methods for comparative purposes (Bartling et al., 2021; Budsberg et al., 2020; Karka et al., 2017; Secchi et al., 2019; Sreekumar et al., 2020). Budsberg et al. (2020) for example attributed 69.5% of biorefinery burdens to acetic acid, with 30.5% to the lignin co-product based on mass allocation, while scenarios using economic allocation attributed circa 95% of system burdens to acetic acid. Sreekumar et al. (2020), comparing the total GWP for 1 L of ethanol produced in a biorefinery process, which included co-products methanol, feed and recovered CO_2, found a significant difference in outcome depending on the allocation method applied, with 2.8 kg CO_2eq. per litre of ethanol for economic allocation and 1.5 CO_2eq. per litre of ethanol for mass allocation. Even within one specific allocation method itself, variation in the way in which allocation is applied may also yield different results. Obydenkova et al. (2021) noted the inconsistency of results obtained via two allocation methods, one based on the total mass and the second based on the dry mass allocation, assessing a biorefinery producing ethanol, soluble lignin oligomers and electricity. Where allocation is being applied both at the feedstock and product phase, Ahlgren et al. (2015) recommends consistent use of the same method for handling multifunctionality at both points in the study, with clear justification provided where a mixed approach is to be applied.

3.5. Life cycle inventory data quality

In order to have confidence in LCA results, it is important that the quality of data used within the LCI reflects real-world situations. As many biorefineries are still in the development phase, primary data from scaled processes can be scarce or deemed too commercially sensitive to use within published LCA studies. Modelling tools may be used to fill some of the information gaps. However, it should be recognized that a level of uncertainty exists in projecting scale-up calculations. Winickoff and Philp (2018) notes that many of the technical and supply chain scale-up challenges for a biorefinery only become fully apparent at demonstration scale.

The main sources for life cycle inventory data varied between the surveyed studies and are presented in Fig. 3 (e); 28% of studies included some primary data, 98% of studies used data obtained from databases, 95% of studies used secondary data obtained from the literature, 53% of studies used feedstock and process modelling tools to scale and build foreground scenarios, while a further 12% of studies included some experimental data. One study did not provide information regarding its life cycle inventory. There were no obvious differences noted in data source trends for studies involving dedicated feedstocks or those involving residue feedstocks. 57% of studies used no primary data, mainly relying on a mixture of literature data and modelling tools. Many studies used the process engineering software Aspen Plus, (Ali Mandegari et al., 2017; Farzad et al., 2017; Joglekar et al., 2019; Levasseur et al., 2017; Pachón et al., 2020; Poveda-Giraldo et al., 2021; Rahimi et al., 2018; Vaskan et al., 2018) to develop foreground systems for the biorefineries, while using LCA databases like ecoinvent for background data. In some cases, primary data is applied only to limited aspects of the biorefinery system, with secondary data supplementing the remaining system. For example, Ncube et al. (2021) utilised primary data only for feedstock production (grapeseed oil from grape pomace), using literature sources for remaining processes. Other studies provide primary data from biorefinery processes. Some of these processes are based on pilot (Barbanera et al., 2021; Cortés et al., 2020; Gullón et al., 2018; Sillero et al., 2021; Zucaro et al., 2018) or demonstration scale (Sreekumar et al., 2020) operations. A few studies also use primary data from industrial scale plants (Ghani and Gheewala, 2018; Seghetta et al., 2016; Silalertruksa et al., 2017). Meanwhile a number of studies attempt to use primary data for both feedstock production and conversion steps (Ghani and Gheewala, 2018; Papadaskalopoulou et al., 2019; Silalertruksa et al., 2017). Certain studies take an ex-ante and prospective approach aimed at identifying the most promising bio-based options and opportunities for value chain improvements. Ncube et al. (2021) for example, explored possibilities for upgrading wineries into future biorefineries, proposed on an existing Italian winery case study. Two by-product production chains were assessed, in addition to greater inclusion of circular practices. Improvements at the cultivation phase, through use of renewable energy and organic fertilisers, and at the vinification phase, by replacing electricity with steam derived from prunings were modelled, largely using baseline data from ecoinvent alongside data from previous literature studies. Other studies use modelling tools such as Aspen Plus to help undertake an ex-ante LCA study of biorefinery scenarios. Pachón et al. (2020) for example, undertook a prospective LCA to investigate scenarios for a biorefinery development based on vine shoots. The scenarios investigated potential value chain routes to produce lactic acid and energy via combined heat and power (CHP), in addition to lactic acid with the co-production of furfural and energy. To build the foreground system, Aspen Plus was used for process design and simulation, based largely on data arising from experimental analysis, including feedstock compositional analysis by an analytical laboratory, and using biorefinery process conditions and yields based on previous experimental work of the authors. The energy requirements in the simulated plant was optimized using Aspen Energy Analyzer V11. Both scenarios investigated offered environmental benefits versus the reference scenario. As the number of studies which did not include any primary data is still at quite a high level, it makes the importance of including sensitivity and uncertainty analyses (discussed further in Section 3.10) even more critical.

3.6. Land use changes

While the use of marine-based biomass is a growing area, it is still the case that using biomass for biorefinery applications usually involves some form of land appropriation. According to Bishop et al. (2021), land-use change can have considerable impacts on the global carbon cycle, causing significant GHG emissions by disturbing carbon stocks in soil and vegetation, and its inclusion within LCA studies can therefore have a significant impact on the outcome. From an LCA perspective, there is a distinction that can be made between land use (LU) or 'land occupation' and land-use change (LUC) or 'land transformation', with LUC being further subdivided into direct land-use change (dLUC) and indirect land-use change (iLUC) (Ahlgren et al., 2015). dLUC changes concern the recent change of use of land on which the specific feedstock is produced (Ahlgren et al., 2015). In certain circumstances, such changes can have a significant impact on the full life cycle sustainability of the value chain. Piemonte and Gironi (2011), for example, estimated that converting rainforests, peatlands, savannas, or grasslands to produce food and other bio-based products would generate a "carbon debt" by releasing 9 to 170 times more CO_2 than the annual GHG reductions that these bio-based products could support through displacement of petroleum-based products. Indirect changes, meanwhile, are market-induced effects elsewhere due to changes in the feedstock production system. iLUC occurs when an increased product demand leads to displacement of conventional agricultural production. For example, if food crops are partly diverted to develop a biorefinery supply chain, the resulting food supply deficit may be partly filled by the expansion of cropland around the world, and partly by intensification of crop production elsewhere, and the resulting GHG emissions are therefore an iLUC effect (Overmars et al., 2015). The need to mitigate impacts from LUC and iLUC are recognized within the EU 2018 Bioeconomy Strategy Update and is also a key component of the recast EU Renewable Energy Directive published in 2019 which mandates to decrease use of high iLUC-risk fuels to zero by 2030 (European Parliament, 2019).

Despite the importance of land use, only 8% of studies included information on dLUC within their calculations (Barbanera et al., 2021; Budsberg et al., 2020; Rahimi et al., 2018; Secchi et al., 2019; Vaskan et al., 2018). Rahimi et al. (2018) and Barbanera et al. (2021), considered LUC associated with the conversion of marginal and set-aside land for cultivation of Eruca Sativa and cardoon respectively, both utilised as feedstocks for the production of biodiesel and various co-products. Both studies used an average carbon sequestration rate of 0.6 t C ha^{-1} yr^{-1} for the conversion of set-aside land to crop land. When it comes to dLUC, Ahlgren et al. (2015) notes a major uncertainty in how burdens shall be allocated over time, for example, the number of years over which the emissions should be distributed after an area of land is converted. Rahimi et al. (2018) considered LUC effects in accordance with the default 20-year transition applied in IPCC national inventory guidelines to annualise the flux associated with terrestrial carbon stock change from one land use to another (Watson et al., 2000). Based on a plant capacity of 105 kt feedstock and associated land requirements, Rahimi et al. (2018) estimated that an additional 55 kt CO_2eq/yr could potentially be sequestered annually in the soil pool, when converting marginal and set aside lands for the cultivation of Eruca Sativa. Investigating the life cycle impact of a biorefinery process producing ethanol and co-products from palm fruit branches (lignocellulose), Vaskan et al. (2018) allocated LUC to only the first generation of plantation use and considered a 25-year time horizon, with change in carbon stock included in the climate change impact category and evaluated according to IPCC methodology (Institute for Global Environmental Strategies, 2006). Ahlgren et al. (2015) also notes that uncertainties may exist in establishing the status of land before and after the change along with uncertainties in quantification of carbon stocks (and thus changes), which can be highly variable and site-specific.

Another 8% of studies included information on iLUC within their calculations (Corona et al., 2018; Khoshnevisan et al., 2018, 2020; Marami et al., 2022; Parajuli et al., 2017, 2018). Quantification of environmental burdens due to iLUC is different from those resulting from dLUC, as it is based on expected market reactions to increasing demand for a product, and is therefore only quantifiable by models (Ahlgren et al., 2015). Within the current review, inclusion of iLUC was prevalent in consequential LCA studies, with 80% of studies considering iLUC being CLCA studies. This is not unexpected, as the economic approach used to quantify iLUC is similar to that employed more widely within CLCA to identify marginal activity responses to market signals (Ahlgren et al., 2015). Khoshnevisan et al. (2020) and Marami et al. (2022) considered the iLUC burdens from avoided soybean meal and barely production through displacement with a protein feed co-product produced from a municipal waste and wastewater biorefinery, respectively – balancing this against additional palm oil production associated with displaced soybean oil. Investigating biorefinery systems which utilised straw and grass clover, respectively, Parajuli et al. (2017, 2018) included iLUC to investigate induced GHG emissions resulting from the use of productive land to produce clover for biorefinery feedstock, as well as avoided burdens resulting from biorefinery co-products, such as animal feed, which were assumed to displace corresponding agricultural commodities and associated iLUC. For the first aspect, an iLUC factor of 1.73 t CO_2eq $ha^{-1}y^{-1}$ (Schmidt and Muños, 2014) was applied, based on a global average of GHG emissions for occupation of 1ha of arable land. For the second aspect, avoided iLUC was considered whenever the co-products displaced alternative agricultural products. Avoided burdens were calculated for the substitutions of Brazilian soymeal and Ukrainian barley, which were assumed to be displaced by feed protein and fodder silage, respectively, produced through a local biorefinery approach. In another grass biorefinery LCA, Corona et al. (2018) included iLUC resulting from the occupation of arable land for the production of the biomass, and iLUC avoided by the displaced conventional products, including grass biorefinery protein replacing soymeal and composite from press-pulp replacing rockwool insulation material. Corona et al. (2018) used a different iLUC factor of 1.43 t CO_2eq ha of agricultural land used, derived from Schmidt et al. (2015), in which the share of global annual GHG emissions from land use change that is caused by agriculture is evenly distributed across all agricultural lands on a per hectare basis. As bio-based industries generally intersect with other land use applications and are often dependent on land use for supply chain development, the importance dLUC and iLUC should be more strongly reflected in reviewed studies in this area, to better understand and contextualize the overall environmental benefits or impacts.

3.7. Product use and end-of-life phase

Including this step within the LCA is key, as several factors impacting the comparative environmental performance of the full biorefinery value chain and the products it produces, may only become clear during the product use and/or EOL phase. For example, the inclusion of biocomposites within interior parts of automotive vehicles can reduce the overall weight, leading to a reduced fuel consumption for the vehicle, a benefit that will only be fully observed during the use phase of the car (Shaker et al., 2020). Research in Netherlands, Denmark, and Ireland, investigating the production and use of various green biorefinery feed products, has shown that there may be positive environmental benefits associated with these products, which are observed at the animal feeding phase (Damborg et al., 2019; Pijlman et al., 2018; Serra et al., 2023).

There is also a need for careful consideration of the EOL phase of bio-based products, as certain products may meet a different fate to incumbent products. Bishop et al. (2021) notes that understanding the EOL of bioplastics is essential in assessing their sustainability comparative to petrochemical plastics. Certain environmental benefits of bioplastics, for example associated with compostability, are only seen downstream, while other challenges, including inability to recycle certain bioplastics alongside petrochemical plastics, may create interim EOL challenges (Bishop et al., 2021).

Despite the importance of impacts from product use and EOL, only 14% of the reviewed studies extend the system boundary to the “grave” (either cradle to grave or gate to grave), with most studies ending at the biorefinery gate, indicating that the actual use and EOL phase has often been overlooked, or not fully quantified. Studies that extend the system boundary to include the use phase include Rahimi et al. (2018) and Khoshnevisan et al. (2018), which considered biodiesel in the form of B100 and B35 respectively, as the primary product produced in a biorefinery based on Eruca Sativa and castor feedstocks respectively, and considered the distribution of biodiesel and fossil-based diesel to filling stations and comparing the impacts from fuel combustion. Vaskan et al. (2018) considered the use phase of E85 ethanol blend produced from a biorefinery using palm empty fruit bunches, while Zucaro et al. (2018) considered the use phase of E10 and E85 wheat-straw derived bioethanol blends compared to conventional gasoline fuel use. Papadaskalopoulou et al. (2019) modelled the use phase of biowaste-derived biorefinery products including ethanol as transport fuel, biogas heat and electricity for plant and grid supply and biogas digestate as fertiliser. Rosa et al. (2020) included the EOL phase, assuming composting for a biodegradable plastic produced from black soldier fly protein for use in agricultural mulch film. Overall, the inclusion of product use and EOL phase within the studies is very low, and this is very likely to omit some of the prospective benefits and/or impacts of introducing biorefineries and their associated products.

3.8. Biogenic carbon storage

As biorefinery products are derived from biological resources and often replace traditional fossil-based products, there may be differences in biogenic carbon flows which may impact the overall GWP balance. Bernstad Saraiva (2016) states that one of the main reasons behind the production of biofuels is the difference between EOL emissions between short cycle biogenic CO_2-emissions and fossil CO_2-emissions. Similarly, there is potential for biogenic CO_2 storage within bio-material products such as natural fibre, with several insulation materials derived from biomass, such as hemp or wood waste, potentially offering carbon neutral or carbon negative opportunities (Cetiner and Shea, 2018; Jami et al., 2019). According to Ahlgren et al. (2015) carbon storage may also be a factor if some of the biorefinery biogenic waste ends up in a long-term landfill or if carbon capture and storage technology is used in the future. There are two main ways in which biogenic carbon is modelled within LCA studies: 1) temporary carbon storage and 2) carbon neutrality (Pawelzik et al., 2013). According to Bishop et al. (2021) climate neutrality is often assumed, in which the carbon that is sequestered by the growing biomass is released back into the environment over a short time period with no net climate forcing effect – in line with the argument that biogenic carbon storage should be excluded from LCA as bio-based products will almost always release the stored carbon at some point in the future (Bishop et al., 2021). The calculated benefits from modelling biogenic carbon storage are especially sensitive to the time horizon over which the GWP is considered. In a recent review of bioplastic LCA studies, Bishop et al. (2021) found a number of studies which attempted to measure the benefit of temporary biogenic carbon storage, with some examples of short-term storage treated as long-term storage. Ahlgren et al. (2015) recommends that if significant time elapses along value chains between CO_2 uptake and emissions from the system under study, this should at least be discussed within the study and efforts made to quantify the impact.

From our review of 59 biorefinery LCA studies, only 7% of studies attempt to quantify and model biogenic CO_2 fluxes explicitly, as opposed to assuming carbon neutrality (Foulet et al., 2018; Lan et al., 2020; Larnaudie et al., 2021; Zucaro et al., 2018). Lan et al. (2020) assessing a decentralized system for fast pyrolysis of pine residues and switchgrass, estimated CO_2 sequestration based on the mass of carbon contained aboveground in pine trees, with rotation age for pine plantations assumed at 25 years and found that in most scenarios the carbon sequestered was similar or greater than the combined biogenic and fossil based GHG emissions, implying a net CO_2 sink. Larnaudie et al. (2021) factored in the biogenic carbon at the cultivation, processing and product use phase, including that absorbed by the crop (switchgrass) through photosynthesis, and emitted during fermentation, wastewater treatment, lignin combustion, and ethanol combustion. Carbon sequestered by the crop was calculated by considering the base biomass composition of 1.63 $gCO_2/g_{dry\ switchgrass}$. Zucaro et al. (2018) included a complete carbon balance for both E10 and E85 wheat-based ethanol blends, to check biogenic carbon emissions along the whole ethanol supply-use chain. Building on recommendations from the ILCD Handbook to present both neutral and non-neutral biogenic CO_2 contributions to climate change impacts, Foulet et al. (2018) included biogenic carbon in a biorefinery model producing succinic acid from organic waste. A further 14% of studies explicitly expressed biogenic carbon as having a neutral GWP effect (Karka et al., 2017; Khoshnevisan et al., 2018; Liang et al., 2017; Marami et al., 2022; Papadaskalopoulou et al., 2019; Rahimi et al., 2018; Sreekumar et al., 2020; Unrean et al., 2018) with a default implicit assumption that remaining studies did not reference biogenic carbon as a GWP neutral approach was adopted. Some of these included studies which focused on the use of low- or no-burden feedstocks like mixed organic wastes and wastewater, being converted to fossil-displacing products like bioethanol and bioplastics (Chen et al., 2017; Silva, 2021), in which case accurate biogenic accounting may have highlighted additional environmental benefits.

Another important reason for explicit accounting of biogenic carbon flows in biorefinery LCA is the growing emphasis on CO_2 mitigation through Carbon Capture and Utilisation (CCU) technologies, and its potential integration within biorefineries. Lee et al. (2021) calculated that if CO_2 produced during the corn to ethanol production process could be converted into ethanol by CCU technologies, ethanol production could be increased by more than 37% without additional corn grain inputs. In addition, a variety of material products are now being produced using CCU technologies including concrete, carbonate aggregates, fuels, polymers, methanol and carbon monoxide (Zimmermann et al., 2020). Such technological advances and their potential integration within biorefineries, should offer even greater motivation for including biogenic carbon calculations within LCA.

3.9. Impact assessment

Biorefinery LCA studies often use different impact assessment methodologies and indicators to assess the impacts of the system and associated products. The way in which LCIA is applied varies from study to study. Some studies only focus on a few impact categories deemed most important by the authors, while neglecting others. This may often reflect time constraints, but could give misleading results, given that a biorefinery system may impact positively on one impact category but negatively on several others. This so-called “trade-off” has been highlighted by Miller et al. (2007), who assessed the environmental impact of a PLA, biodiesel and other bio-based products, indicating varying environmental profiles. While each bio-based product achieved lower GWPs relative to petroleum products, their eutrophication impact was higher in each scenario (Miller et al., 2007). A later study from Cherubini and Jungmeier (2009) found that while the use of switchgrass in an ethanol biorefinery offsets GHG emissions by 79% and reduces fossil energy demand by 80%, larger negative impacts were seen in the acidification and eutrophication categories.

In order to analyse the representation of different impact categories, and to take into account variations of mid-point categories proposed by different impact assessment methodologies, this study followed the approach of Bishop et al. (2021) in clustering impact categories for comparative purposes. From the current analysis of 59 studies, there was an average of eight impact categories considered per study, with a significant variation between studies, wherein certain studies considered a large number of impact categories, with others only considering a few impact categories and two studies considering only a single impact category. GWP, or derivatives thereof, was the most prevalent impact considered and was included in 97% of the studies. This outcome is not unexpected given the global focus on GHG emissions reduction and with several regions setting targets of net zero emissions in order to meet this objective (Rogelj et al., 2021). Apart from GWP, the most prevalent categories of impact assessment were eutrophication potential and derivatives (76% of studies), acidification potential (75% of studies), human toxicity (64% of studies), eco-toxicity (63% of studies), resource depletion (61% of studies), and ozone depletion (58% of studies). Despite, the importance of land use as a consideration in biorefinery developments, based on Section 3.6 above, the current review found that less than one third of studies (27% of studies) included any form of impact factor relating directly to land use. For those that do, impacts are related to land use/land occupation (Barbanera et al., 2021; Khoshnevisan et al., 2018; Khounani et al., 2021; Levasseur et al., 2017; Marami et al., 2022; Rahimi et al., 2018; Secchi et al., 2019; Surra et al., 2021), agricultural land occupation (Chopra et al., 2020; Corona et al., 2018; Nitkiewicz et al., 2020; Parajuli et al., 2017; Poveda-Giraldo et al., 2021; Prieler et al., 2019), and urban land occupation (Chopra et al., 2020; Nitkiewicz et al., 2020) (e.g. land-use, land occupation, agricultural/urban land occupation, natural land transformation), with land transformation included within only 3 studies (Chopra et al., 2020; Nitkiewicz et al., 2020; Zhang et al., 2018). Due to the trade-offs evident to date in biorefinery LCAs, it is important that a greater number of impact categories be included to take these into account. Little justification has been found within studies for omitting impact categories, and this is something that should be much more clearly justified.

3.10. Sensitivity and uncertainty analyses, and future-proofing of studies

From this current review, the majority of studies, 61% in total, have undertaken some form of sensitivity or uncertainty analyses of their findings. However, a relatively large number of studies, 39%, did not undertake any such analyses. In particular, a large number of residue feedstock-focused studies (47%), did not undertake either uncertainty or sensitivity analyses (Farzad et al., 2017; Foulet et al., 2018; Gezae Daful and Görgens, 2017; Joglekar et al., 2019; Kachrimanidou et al., 2021; Khounani et al., 2020; Liang et al., 2017; Sreekumar et al., 2020). Where included, the majority of studies focus on sensitivity analyses, with a smaller number including uncertainty analyses.

According to Laurent et al. (2020) the aim of a sensitivity analysis is to assess and enhance the robustness of the study's final results and conclusions, by determining how the conclusions of the study may be affected by uncertainties, such as those related to the LCI data, LCI modelling, LCIA methods, or to the calculation of category indicator results, to name a few. In this review, a small number of sensitivity analyses focused on the impact of the chosen method of allocation (Ali Mandegari et al., 2017; Parajuli et al., 2017; Santiago et al., 2020), while a larger cohort of studies consider aspects related to input and processing parameters, such as feedstock yields and composition (Larnaudie et al., 2021; Seghetta et al., 2016; Silalertruksa et al., 2017) and processing parameters and productivities (Budsberg et al., 2020; Gezae Daful and Görgens, 2017; Karka et al., 2017; Lan et al., 2020) as part of their sensitivity analyses. Other factors considered within sensitivity analyses included different avoided product scenarios (Parajuli et al., 2017), the inclusion or not of feedstock within the system boundary (Papadaskalopoulou et al., 2019), impact of varying levels of nitrogen fertiliser application (Khoshnevisan et al., 2018) and assumed product market value (Secchi et al., 2019). A number of studies used also sensitivity analyses to help future proof their results. Chen et al. (2017) included a sensitivity analysis to calculate the life-cycle environmental impacts of alternative materials and several proposed future improvement strategies, while a number of studies included future alternative renewable energy strategies within their sensitivity analyses (Barbanera et al., 2021; Chrysikou et al., 2018; Ncube et al., 2021; Sreekumar et al., 2020; Surra et al., 2021). Ncube et al. (2021) included the integration of fatty acid methyl esters (FAME) for energy use in the feedstock cultivation stage, replacing diesel, and included a 50% reduction of fertilizers assuming a shift towards organic fertilizers from exhausted pomace, along with an additional reduction in electricity consumption by replacing electricity with steam from pruning residue combustion in a future scenario. At the transportation phase, Surra et al. (2021) considered the option of substituting the fossil fuel of the OFMSW collection fleet with biomethane. While some studies did include alternative renewable energy mixes within their sensitivity analyses, no studies were found which aligned the renewable energy scenarios to specific renewable energy targets set out by governments. This could be an important step, in order to create accurate future scenarios for biorefinery systems.

Laurent et al. (2020) notes that many input parameters to an LCA study are associated with uncertainties which can be defined as "the discrepancy between a measured or calculated quantity and the true value of that quantity". Uncertainty analyses therefore focus on the influence of the totality of those individual uncertainties on the final results (Laurent et al., 2020). Monte-Carlo analysis was used by the small number of studies performing uncertainty analyses (Kapanji et al., 2021; Pachón et al., 2020; Parsons et al., 2019; Secchi et al., 2019; Silva, 2021; Zucaro et al., 2018). According to Sun and Ertz (2020) the Monte Carlo method can randomly sample the values of uncertain variables based on probabilistic analysis, and combine with the pre-determined impact assessment method in order to simulate the statistical distribution of outcomes, to obtain statistically significant environmental impact evaluation results. In this review, this procedure was mainly found to be used with input data and parameters from uncertain sources, such as secondary sources from literature (Kapanji et al., 2021; Pachón et al., 2020; Zucaro et al., 2018). Given the fact that biorefinery and bio-based product value chains are often in early stages of TRL, often requiring assumptions and uncertain information to analyse, sensitivity and uncertainty analysis should be integral to all studies, which is clearly not the case currently. As new biorefinery investments will need to perform through rapidly changing contexts, in particular decarbonising energy sectors, the authors recommend that practitioners also incorporate a prospective approach within these analyses. The breakdown of studies which included uncertainty and/or sensitivity analyses is presented by feedstock type in Fig. 3(f).

4. Conclusions

From the current review of biorefinery LCA studies focusing on different feedstock specificities, there are several limitations which have been identified. While the study finds a strong presence of residue feedstocks, in addition to dedicated feedstocks, and this is a welcome trend, there are fewer studies focused on third and fourth generation feedstocks. It is noteworthy that none of the studies comprehensively dealt with all of the considerations covered through section 3. There is a lack of justification regarding selected approaches of LCA studies, on areas such as system boundary, allocation and consequential versus attributional approaches. Studies focused on residue feedstocks were more likely to use a feedstock-based FU, less likely to include feedstock cultivation inputs, and more likely to include mass-based allocation, than dedicated feedstocks. A large number of studies do not include any primary data, raising concerns about the reliability of the studies, while a significant number of studies failed to include an uncertainty analysis, despite such a lack of primary data. Important issues which can impact the sustainability outcome of biorefinery LCAs such as land use changes and biogenic carbon storage are often omitted or not fully discussed. Meanwhile, product use phase and end of life, was frequently excluded, overlooking potential product displacement benefits, or consequences. In addition, too many studies apply a limited set of impact categories, despite the environmental trade-offs between different impact categories which can result from different biorefinery systems. Overall, a lack of prospective analysis was noted within the studies, with respect to circularity of products, future process innovations, renewable energy, or carbon capture and utilisation (CCU).

Based on the review, a number of recommendations can be made regarding LCA approach and future research in the area, to support a more comprehensive LCA evaluation of the environmental sustainability of biorefinery feedstocks, processes and products.

- More clarity and justification of the selected functional unit and methodologies (e.g. ALCA or CLCA, and allocation method) applied, should be detailed within future studies.
- Feedstock burdens should be accounted for appropriately, as their contribution to the overall impacts of the system can be significant. This includes accounting for the various energy and material inputs which occur at the cultivation phase, and appropriately allocating these – with particular justification for treating residues as zero-burden "wastes".
- In the case of CLCA analysis, where by-products or wastes serve as the feedstock, practitioners should evaluate the emissions resulting from lost opportunities, or equally the emissions which may be avoided as a result of the diversion. dLUC and iLUC caused by the introduction of the biorefinery system should be evaluated within the study, and where possible, accounted for within modelling.
- Including the use phase and EOL within the biorefinery system assessment is vital to understand the true sustainability impacts (positive or negative) of the various bio-based products produced, vis-à-vis their often fossil-based counterparts. Biorefinery material and energy products often have some important differences to the products they replace which should also be accounted for. Future research should better consider the intended application of the bio-based products and energy, including areas such as recycling which are often still evolving in the case of certain bio-based materials.
- Practitioners should note whether or not biogenic carbon storage may be a significant consideration for the feedstocks and products under study, and how this biogenic carbon is dealt with within the analysis should be clearly justified in future studies. If reliable data exists, then practitioners should model this carefully, otherwise a simplified approach in which biogenic carbon cycling is treated as GWP neutral may be adopted.
- A greater number of impact categories should be considered for LCIA, to capture the many trade-offs that exist for biorefinery LCAs. While GWP is a pertinent consideration for bio-based value chains in order to meet climate neutrality targets, the potential negative impacts of biomass feedstock cultivation on categories such as eutrophication and acidification, also need to be fully evaluated in many cases. Where impact categories are omitted, justification should be provided.
- As many biorefinery processes are developing and at an early stage, data uncertainties often exist, sensitivity and uncertainty analyses should therefore, be built into LCA assessments, as appropriate, to explore implications of major system assumptions and data limitations. As greater investments come on stream for pilot, demonstration and commercial biorefinery processes as well as their feedstock supply chains and end products, in future practitioners should attempt to integrate a higher level of primary inventory data to improve the quality of the LCA. Ex-ante LCA approaches, should be used as a tool to inform investment of such facilities, ensuring that investments are being targeted towards biorefinery infrastructure with the highest chances of delivering environmental benefits.
- Emerging biorefineries are not developing in isolation and should be modelled taking into account the evolving contexts in which they are likely to operate. Therefore, future ambitious renewable energy targets, and the increasing integration of systems innovations such as circular design and CCU technologies along with more sustainable land management practices are among some important future considerations which should be accounted for within developing biorefinery LCAs. Notably, biorefineries will, in many cases, need to compete with other decarbonising sectors in energy and materials, and will therefore need to consider the sustainability of their operations in current and future contexts, placing a high emphasis on the need to secure sustainable feedstock, and to derive renewable energy from biomass and biorefinery residual streams and other sources such as wind and solar. The authors recommend that analyses also incorporate a prospective approach in dealing with such topics.
- Given the diverging approaches and findings of these biorefinery LCA studies, future research should seek to understand the possibilities of developing or implementing Product Environmental Footprint Category Rules (PEFCRs) for each biorefinery product. While it may be difficult to harmonize approaches due to the wide range of biorefinery feedstock, process and product routes, it may be possible to establish a degree of harmonization in relation to how different feedstocks are treated (e.g., proper categorization of feedstocks), and to agree a minimum level of harmonization in relation to certain aspects of biorefinery value chains (e.g., how biogenic carbon is treated), although this may be difficult to achieve at product level.

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CRediT authorship contribution statement

James Gaffey: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Maurice N. Collins:** Supervision, Writing – review & editing. **David Styles:** Supervision, Writing – review & editing.

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

Data will be made available on request.

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Appendix A. Supplementary data

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