Technical Substitutability of Recycled Materials in Life Cycle Assessment: A Comprehensive Review and Framework for Quantification.

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ABSTRACT

In evaluating environmental sustainability with methodologies like life cycle assessment (LCA), recycling is usually credited for avoiding impacts from virgin material production. Consequently, the LCA results are influenced by the manner in which the substitutability of virgin by recycled materials is estimated. This study reviews how the scientific community assesses the technical substitutability of recycled materials in LCA. Accordingly, 49 peer-reviewed papers were in-depth analysed, considering aspects such as materials studied, type of substitution, recycled material (rMaterial) application, and life cycle stages (LCSs) where substitution was evaluated. The results show that 49% of the papers investigated material substitutability through technical and economic aspects. 51% of the articles did not consider the final application of the rMaterial. Plastics were the most studied material, and mass was the most used property to quantify technical substitutability. Certain materials were more analysed in specific LCSs (e.g., metals in the natural resource extraction stage). As 51% of the papers developed a new approach for substitutability calculation, this shows that substitutability is still a concept in development. It was noticed in 33% of the papers that substitutability values were taken from

external sources, and in some cases were used without considering whether they were representative for a specific case. Aspects such as harmonization, transparency, and consideration of the application of recycled materials, therefore, require more attention in substitutability evaluation. Based on the results, a step-wise framework to measure technical substitutability at different LCSs was developed to guide researchers in including substitutability in LCA studies.

KEYWORDS

Substitutability, substitution, displacement, recycled materials, secondary materials, material quality, LCA.

1. Introduction

Life cycle assessment (LCA) is a frequently used methodology applied to analyse the potential environmental impacts attributed to materials, products and processes, and its results are used as guidance for decision-makers in both the public and the private sectors (European Commission, 2003). Recycling activities are recognized as a strategy to reduce the use of virgin materials by substituting them with secondary materials (European Environment Agency, 2019). These activities are typically credited for avoiding the environmental impacts associated with the supply of virgin materials (Huang et al., 2013; van der Harst et al., 2016). Consequently, the assumptions relating to the virgin material substitution (substitutability) can dominate the overall environmental balance of an LCA study (Jeswani et al., 2021; Lazarevic et al., 2010; Vadenbo et al., 2017). Jeswani et al. (2021) performed a sensitivity analysis varying the quality of plastic recyclates from mechanical recycling and compared the climate change impact results of those recyclates to the ones from chemical recycling via pyrolysis. Their results showed that if mechanical recycling produces a recyclate with a quality similar to its virgin alternative, its climate change impact would be 21% lower than that of pyrolysis;

otherwise, its impact would be 8% higher than that of pyrolysis. Despite the importance of considering the material quality in substitutability, several LCA studies assume a full substitution (1:1) of primary materials for recycled ones, without mentioning a clear motivation (Geyer et al., 2016; Rigamonti et al., 2020; Vadenbo et al., 2017).

Some guidelines have been provided to address the substitutability of recycled materials in LCA. According to the European Commission (2010), substitution can be applied in LCA to solve multifunctionality in waste treatment processes (European Commission, 2010). In essence, it is necessary to quantify the difference in functionality between the recycled material resulting from waste treatment and its corresponding substituted virgin counterpart. This quantification can be done in two ways, firstly, by considering the mass of the recycled material that can substitute a determined mass of virgin material for a given application; and secondly, in case of undefined applications by including the quality aspect/applicability of the secondary resource, for example by the market-price ratio of secondary over virgin material (European Commission, 2010). Hossain et al. (2017), Neo et al. (2021), and Rigamonti et al. (2014) are examples of studies that employed the mass ratio method for calculating substitutability, in line with the European Commission's recommendation (2010). Conversely, Bovea et al. (2010), Di Maria & Micale (2014), and Giugliano et al. (2011) considered the disparity in market prices to estimate substitutability.

Besides the guidelines from the European Commission (2010), different approaches have been taken to estimate substitutability. Authors such as Rigamonti et al., 2020 considered the difference in technical properties between the recycled and the virgin material, incorporating in some cases weighting factors to include the importance of the application of the recycled material to the substitutability calculation (Demets et al., 2021; Golkaram et al., 2022). Other methodologies considered a combination of technical and market data, sometimes adding the importance of the application through weighting factors (Bala et al., 2015; Huysveld et al., 2022). Finally, Vadenbo et al. (2017) have established a framework for the estimation of the substitution potential related to secondary resources (γ), which was defined as the product of four different factors (Equation 1). They are: (1) the physical amount of secondary resources inside a waste stream for treatment (U^{rec}), (2) the share that is expected to be recovered and used, the resource recovery or recycling efficiency (η^{rec}), (3) the degree of functional equivalence between the secondary resource and the competing product for a specific end use or application ($\alpha^{rec:disp}$), and (4) the expected change in consumption levels of the replaced product system, the market response (π^{dis}).

$$\gamma = U^{\text{rec}} * \eta^{rec} * \alpha^{rec:disp} * \pi^{dis} \qquad \text{Equation 1}$$

In this framework, the authors distinguish 'substitution potential' (γ) from 'technical substitutability' ($\alpha^{rec:disp}$), the latter being part of the calculation of the former and the subject of this article. In other words, technical substitutability of recycled materials can be understood as the extent to which the recycled materials (rMaterials) can replace virgin materials (vMaterials) in specific applications, considering functionality (Tonini et al., 2022). This reveals that the concept of substitutability is linked to the quality of recycled materials, because the latter directly affects the former, and that considering the specific application of the rMaterials is a key requirement for its estimation (Dahlbo et al., 2018; Demets et al., 2021; Golkaram et al., 2022; Huysveld et al., 2022; Pehlken et al., 2014; Tonini et al., 2022). Whether a certain technical property should be high or low depends on the application, for example, plastic packaging film requires a low stiffness, while a plastic pipe requires a high one (Demets et al., 2021).

The various approaches to estimating substitutability highlight that there is still no harmonized practice in the this field. At the time of writing this introduction, there are a few articles that performed a critical reviewed related to the technical substitution approaches taken by the scientific community. Sazdovski et al. (2021) performed a systematic review of 51 articles on beverage packaging to evaluate the extent to which the new circular economy (CE) paradigm has been integrated into the LCA methodology. Only five of the reviewed articles considered a substitutability value. Schrijvers et al. (2016) reviewed end of life (EoL) allocation procedures for recycling in LCA and recommended the use of substitutability in the EoL through quality correction factors. Van der Harst et al. (2016) reviewed methods for including recycling in LCAs, such as substitution (quantified by e.g., quality correction factors), allocation based on the number of recycling loops, the recycled-content method, and the equal-share method. The authors recommended, when applicable, incorporating quality reduction of recycled materials rather than disregarding it in LCAs. Tonini et al. (2022) discussed available studies on recycling quality from which, according to the authors, substitutability is part. They summarized and linked the existing approaches and suggested a way to make the quality concept useful to support circular economy policies and monitoring. Tonini et al. (2022) discussed substitutability in a general, yet concise, way, and explained relevant articles. However, they did not provide an in-depth analysis of how the scientific community approaches substitutability, as their focus was primarily on quality.

Lazarevic et al. (2010) conducted a review of ten LCA case studies to determine whether a consensus existed regarding preferred EoL treatment options for post-consumer plastic in Europe. However, the analysed papers were published more than 12 years ago and the motivation for the selected substitutability ratio is not discussed (Lazarevic et al., 2010). Viau et al. (2020) investigated the modelling of substitution in LCAs of municipal solid waste management, including 51 case studies published between 2003 and 2017. These studies focused not only on recycled materials but also on energy, ashes, fuels and compost. The authors evaluated the extent to which the studies followed the different contributing factors identified by Vadenbo et al. (2017) (physical resource potential; resource recovery/recycling efficiency; substitutability; market response) and if there was a justification for the substitutability value. Viau et al. (2020) found that 65% of the analysed LCA studies did not provide justification for the substitutability values used. Furthermore, among the studies that did offer justification, the substitutability values did not represent physically realistic substitutions. This means that the substitution was not based on the physical characteristics of recycled materials or on the effects on their functionality compared with primary materials. Rigamonti et al. (2020) proposed a harmonized approach for calculating the substitutability of secondary materials in LCA and they also provided a brief overview of the state of the art regarding technical substitutability presenting a list of sixteen technical substitutability values. However, the authors did not extensively review the way in which material substitutability was approached in the literature.

Concluding, there exists a variety of material substitutability calculation approaches. There is a need to understand what material substitutability factors measure specifically and on which information or data they are based. The aim of this paper is to provide a comprehensive understanding of the existing material substitutability factors and their calculation methods in order to assist the scientific community in the application of substitutability factors in LCA case studies and their further development. This work relies on an extensive literature review of material substitutability, mainly from a technical perspective, focusing on studies published between 2010 and May 2022, in which the substitutability calculation method is clearly reported. Our work includes an in-depth analysis of 49 papers addressing aspects such as the quantitative base for substitutability calculation, the applicability in terms of material types and recycling type (open-loop versus (semi-)closed), the calculation complexity, the representativeness for a specific application, etc. Finally, this paper also presents a framework with recommendations on technical parameters that can be considered to measure substitutability at different life cycle stages (LCSs). It can function as a guide towards LCA practitioners on how to include substitutability in their studies, considering the available information.

2. Methodology

2.1. Scope

This review focuses on studies with the following characteristics; firstly, the articles have applied substitutability to material recovery via recycling, thus energy recovery (electricity, heat and fuels) and material recovery via processes carried out by biological organisms (e.g., composting, anaerobic digestion, etc.) were excluded. Secondly, only studies evaluating the physical quality loss of the materials after recycling were considered. Studies that focused on the dynamics of supply and demand in market substitution were not included as these approaches are not based on physical causality (Geyer et al., 2016). Besides articles that address physical quality loss in a direct way through e.g., technical properties, this work also covers substitutability quantified based on two indirect approaches, including (i) the price ratio of the substitutable materials, which can reflect material quality and, hence, is used as a proxy of material quality drop, recommended by the European Commission, (2010), and (ii) the market share/percentage of recycled material that is accepted in certain market segments, depending on its quality and/or legal aspects (Golkaram et al., 2022). Articles that considered full substitutability (1:1 substitution) and, hence, considered closed-loop recycling were excluded, regardless of whether or not motivation or justification was included. This is because a few of the papers found estimated full substitutability with a valid motivation (e.g., Beigbeder et al., 2013), while several studies assume this scenario without clear justification (e.g., Ghose et al., 2017; Pires et al., 2011; Sevigné-Itoiz et al., 2015). Hence, to decrease the possibility of papers without motivation being considered in the analysis, all the ones in which 1:1 substitution was applied, were left out.

2.2. Literature review searching process

The literature search process is illustrated in Figure 1, including keywords used, articles obtained after each step, and filters related to the date of publication, type of publication and language applied.

The search was carried out using a combination of three strategies, electronic database search in Web of Science and Scopus, academic networks, as part of the personal knowledge strategy, and the snowballing process defined as an approach for systematic literature studies (Greenhalgh & Peacock, 2005; Wohlin, 2014). Applying the academic network's strategy, a relevant paper that was under revision but not yet approved in an academic journal and three other relevant articles cited in it were included. This paper was published in August 2022 (Huysveld et al., 2022). In the snowballing process, the reference list of a related paper and/or the citations on it, are used to identify additional relevant papers; it can be complemented with a systematic way of looking at where papers are referenced and where papers are cited (Greenhalgh & Peacock, 2005; Wohlin, 2014). This combination allowed for obtaining applicable papers for material recycling substitutability. Using the electronic database search, several non-pertinent papers were also obtained. For example, the term substitution is also used in the context of finding raw materials that can replace critical raw materials (Pavel et al., 2016). Additionally, with some of the keywords, papers studying the effect of the inclusion of recycled materials on the performance of construction materials were obtained. However, in these, substitutability was not calculated, as an established percentage of recycled material was applied, and later this was varied to evaluate the performance of the final material (Juan-Valdés et al., 2018, 2021; Kočí et al., 2021).

Figure 1 also shows that 49 out of the 71 papers retrieved during the screening and searching underwent an in-depth analysis. These 49 papers comply with the following characteristics: (i) the substitutability was considered in a quantitative way, hence it was not only discussed or recommended, and (ii) there was transparency about the origin of the data considered for the substitutability calculation. A complete list of the 71 papers, containing title, authors, year of publication, the term(s) used to refer to substitutability, and a summary of the goal of the paper, among other information can be found in the Supporting material (SM) (Tables S1 and S2). The reasons why 22 papers were excluded from the in-depth analysis are presented in the SM (Table S2). The aspects considered during the in-depth analysis of the 49 papers are presented in Section 2.3.

2.3. Aspects considered during the in-depth analysis and definitions

The aspects evaluated during the in-depth analysis are presented in Table 1, followed by the definitions.

Materials studied: the recycled material (rMaterial) and substituted material (sMaterial) that were compared in the substitutability calculation. The rMaterial can either replace the same or a different sMaterial (e.g., recycled polyethylene (rPE) from plastic packaging replaces virgin polyethylene (vPE) in packaging, or rPE replaces virgin wood (vWood) in a street bench).

Substitutability type (S type): defined based on the relationship under which rMaterials and sMaterials were compared (quantitative base for comparison). It can be:

1. Technical (TS): the substitutability calculation considered material technical properties as a proxy for quality and/or the number of times that the material is or can be recycled (recycling cycles).
2. Economic (ES): the substitutability calculation considered the difference in prices between the rMaterial and the sMaterial as a proxy for quality and/or the percentages of rMaterial that is accepted in certain market segments depending on its quality and/or legal aspects.
3. Technical and economic (TS+ES): the substitutability calculation considered technical and economic information either combined in one calculated value or not.

Quantitative base for comparison: relationship under which the secondary material was compared to the virgin one. TS can be based on the number of recycling cycles or the difference in technical properties. The latter can be of three types, i.e. physical, physical-mechanical, and/or properties related to the processability of the material (i.e., physical-processability). ES can be based on the price ratio or the market shares difference.

Recycled material (rMaterial) application considered: evaluates whether the specific application of the rMaterial was considered or not during the substitutability calculation. When the application of the rMaterial was not specified, it was assumed that this was not considered.

Origin of data: evaluates where the data used for the substitutability calculation comes from. It can be:

1. Expert judgment (EJ): data based on communication with experts from recycling companies, consortiums, institutional entities or research institutes.
2. Modelled data (ModD): based on information from mathematical models (e.g., material flow analysis (MFA)). Simple mathematical estimations as averages were not considered as modelled data.
3. Measured data (MeasD): primary data obtained from laboratory, pilot and/or working plant experiments.
4. Economic data: based on the market shares (percentages) of recycled or virgin material used in a certain market segment or sector, or data on the price of recycled and virgin materials.

Methodological innovation: evaluates what was done with substitutability in the article:

1. New approach: Authors developed a new method to calculate substitutability, which may build upon existing approaches. For example, Eriksen et al. (2019) considered market shares in which the rMaterial with a specific quality level has potential to substitute vMaterial. That approach is analogous to substitutability proposed by Vadenbo et al. (2017), who divided the functionality of the rMaterial with the one of the sMaterial.
2. Calculated: one or more substitutability values were determined using and referring to an existing method in the literature.
3. Used: one or more substitutability values were retrieved from literature and used in an LCA case study.

Calculation complexity:

1. Simple ratio: substitutability calculation considering the ratio of a single variable. This ratio can be either part of a broader calculation (see 3. Part of a broader calculation) or not.
2. Elaborated: calculation involved a mathematical operation between multiple variables. The only objective of the calculation was the determination of the substitutability (e.g., different properties were multiplied with weighing factors that represent the relative importance of each property).
3. Part of a broader calculation: the substitutability calculation was part of another calculation of which the main objective was not the determination of the substitutability e.g, inside an EoL formula, a circular performance indicator or a circular economy benefit formula (Hermansson et al., 2022; Huysman et al., 2017; Huysveld et al., 2019).

Life cycle stage (**LCS)** considered for the substitutability determination: evaluates where in the life cycle, the rMaterial substitutes the sMaterial. This is also known as the point of substitution (Schrijvers et al., 2021).

Note that the LCS was identified by the properties used in the substitutability calculation. These LCSs are based on the work from Dewulf et al. (2015) and can be:

1. Natural resource extraction (NRE): compares the concentration and/or the recovery coefficient of the substance or material of interest in the natural resource to its concentration and or recovery coefficient in the waste stream.
2. Raw material production (RMP): considers the easiness of transforming (purifying) virgin (compared to recycled) resources into raw materials for the manufacturing sector.
3. Manufacturing (MF): considers properties measuring the easiness of processing the rMaterial into components and/or into products, compared to the virgin one. Note that when the substitutability is only based on prices or market shares, the substitutability was considered to be evaluated at the manufacturing stage. Indeed, when the rMaterial is available on the market, it will be purchased instead of the virgin one, to be manufactured into components and/or products.
4. Use: considers (based on technical properties) how functional the rMaterial in certain applications is compared to the virgin one. This encompasses both intermediate products that need further processing (e.g., pellets) into an application, and final products and/or components ready for application. Note that the categorization of articles into distinct LCSs is guided by the properties used in substitutability calculations. Therefore, if the substitutability analysis focuses on functional properties, articles will be placed in the use LCS, even if the recycling process yields intermediate products.
5. EoL: considers the number of recycling cycles, meaning the number of times, after the EoL, that a rMaterial can be recycled again compared to the virgin one.

Recycling type: the type of recycling considered in the substitutability determination:

1. Closed-loop: the rMaterial was used in the same application as the original application from which it was recovered without losing quality (Civancik-Uslu et al., 2019; Huysman et al., 2015; Pires et al., 2011; van der Harst et al., 2016). This is out of the scope of the analysis in this article, because the substitutability will be 1:1.
2. Semi-closed: the rMaterial was used in the same application as the original application from which it was recovered but with a reduction in quality. Consequently, in some cases, virgin material was added (Huysman et al., 2017). In this case, the rMaterial replaces the same sMaterial.
3. Open-loop: the rMaterial was used in a different application than the original application in the previous life cycle. This new application can have a lower or higher value than the original one (Civancik-Uslu et al., 2019; Ragaert et al., 2017). There are two types of open-loop recycling. In the first one, the rMaterial replaces the same material but in a different application (e.g., rPE from plastic packaging was used to replace vPE in toys). In the second one, the rMaterial replaces a different sMaterial in a different application (e.g., rPE from plastic packaging replaces wood in a street bench). When the future application of the rMaterial was not specified, it was also considered in our analysis as open-loop recycling based on the ILCD handbook (European Commission, 2010).

3. Results and discussion

For the 49 papers that underwent an in-depth analysis, the most outstanding results for the evaluated aspects defined in Table 1 are discussed hereafter (Sections 3.1 to 3.6). Relationships between the aspects were studied to figure out relevant connections. The analysis is presented by the number of articles. Note that a single article can be counted for multiple categories within the defined aspects. For example, Rigamonti et al. (2010) studied the substitutability of plastics, paper and wood, hence, this paper appears three times in the type of material studied. Consequently, the quantity of all the materials studied can be higher than that of the papers analysed. Also, Section 3.4 provides an example of the analysis performed on all articles by evaluating the aspects outlined in Table 1, using the recent work by Golkaram et al. (2022). Finally, the findings from the literature review are translated into a framework for LCA practitioners considering recommendations on technical parameters that can be used to measure substitutability at different LCSs (Section 3.6).

3.1. Comparison of methods

Due to the importance of the consideration of the final application of the rMaterial in the substitutability calculation, this was analysed in relation to the substitutability type, the recycling type, and the methodological innovation (Figure 2).

Figure 2a shows that the substitutability type covered in almost half of the papers (24 out of 49 analysed papers or 49%) was based on both technical and economic properties (TS+ES), followed by technical substitutability (TS) in 19 papers (39%). Only six papers (12%) considered exclusively economic substitutability (ES).

Figure 2a also shows that more than half of the papers (25 out of 49 analysed papers or 51%) did not consider the application of the rMaterial in the substitutability calculation. Technical substitutability was the only substitutability type in which more than half of the papers (12 out of 19 papers or 63%) considered the future application of the rMaterial. On the contrary, in none of the papers applying only economic substitutability the application of the rMaterial was considered. Due to the importance of considering the rMaterial application, the representativeness of economic substitutability can be questioned.

In Figure 2b it is observed that most of the papers (65%) evaluating substitutability considered only open-loop recycling (32 out of 49 analysed papers), while nine papers (18%) focused on both open and semi-closed loop recycling, and seven papers (14%) only on semi-closed loop recycling[1]. This could be explained by the fact that open-loop recycling is still more common than semi-closed loop recycling (European Commission JRC, 2010). 21 out of the 32 papers (66%) evaluating open-loop recycling did not consider the final application of the rMaterial in the calculation of the substitutability. On the contrary, in five out of seven papers (71%) studying semi-closed loop recycling, the final application of the rMaterial was considered. This might be because, in semi-closed loop recycling, the rMaterial goes back to the original application in the previous life cycle, hence, its future application is clear (Huysman et al., 2017). On the contrary, in case of open-loop recycling, the future use of the rMaterial can be known or not (Ragaert et al., 2017).

Figure 2c shows that the majority of the papers (25 out of 49 analysed papers or 51%) developed a new approach for calculating substitutability, which may build upon existing approaches. This reveals that substitutability is still a concept in development that lacks standardization for its calculation. In 16 papers (33%), the substitutability values were only retrieved from other studies. In the latter case, it is necessary to meticulously choose the substitutability value in order to ensure that it is applicable to the specific case, hence representative. However, it was found that seven papers published between 2010 and 2020 (Andreasi Bassi et al., 2017; Bovea et al., 2010; Di Maria & Micale, 2014; Ferrara & De Feo, 2020; Giugliano et al., 2011; Hossain et al., 2017; Tunesi et al., 2016) used the same substitutability value for plastics (0.81:1), developed by Rigamonti et al. (2009, 2010), based on prices from 2008 in Italy. Knowing that prices are constantly fluctuating, for example, according to Eurostat (2021), from 2012 to 2020, the average price of plastic waste

[1] Vadenbo et al. (2017) which is one of the most well-known articles developing a new approach to calculate substitutability, is part of the 49 papers analysed in detail. However, it presents a case of energy substitutability, and none on material recycling substitutability. Hence, it was not classified in the aspects "recycling type", "material type" and "quantitative base for comparison (properties)".

in Europe varied from 334 to 247 EUR/ton plastic, it is likely that the same substitutability value is not representative in all those studies.

Figure 2c also depicts that most of the papers (18 out of 25 analysed papers or 72%) that developed a new calculation approach for substitutability considered the final application of the rMaterial. In contrast, in most of the papers only using a substitutability value from another literature source (14 out of 16 analysed papers or 87%), the final application of the rMaterial was not considered. Finally, in exactly 50% of the papers in which the substitutability was calculated and used, the future application of the rMaterial was considered.

Bearing in mind the importance of the future application of the rMaterial in the calculation of substitutability, it might not be reliable and hence, not advisable to apply the substitutability value defined from one work to another one. If needed, it should only be applied when the rMaterial application and the conditions considered for the calculation of the substitutability value are the same in both cases.

3.2. Comparison of studied materials

The most studied material in the 49 analysed articles was plastic (39) (Table 2 in Section 3.2.1), followed by paper/cardboard (13), metal (6), wood (3), and others (6), including tires, glass, carbon fibres and materials from construction and demolition waste. This result shows the awareness of the scientific community about quality degradation of plastics and paper/cardboard after recycling, two material groups in which this discussion is indeed very pertinent. Consequently, the study of plastic has shown more substantial advancements in terms of models for estimating substitutability. These models incorporate multiple technical properties, market data, and the inclusion of weighting factors. Note that a single paper could be counted for multiple materials studied (first paragraph of Section 3). The evaluation of materials in the reviewed articles considered two aspects: their quantitative basis for comparison (Section 3.2.1) and the LCS at which substitutability is assessed (Section 3.2.2).

3.2.1. Relationship between the studied materials and the quantitative base for comparison

Table 2 presents, in terms of the number of papers, the properties considered in the technical substitutability calculation for the different materials. Bearing in mind that a single paper can study different properties to calculate substitutability, the number of times that all the properties were studied was higher than the total number of analysed papers. For example, Demets et al. 2021 (a single paper) calculated technical substitutability considering five properties; ease of flow, i.e., melt-flow index (MFI), elastic modulus (E), yield strength (σ_y), impact strength (a), and strain at break (ε_b).

In 39 out of the 49 articles analysed, plastic substitutability was studied. 22 of the 39 papers evaluating substitutability for plastics considered technical properties (physical, physical-mechanical and physical-processability) for the estimation. Four papers considered solely physical-mechanical properties, 15 considered physical properties and three considered both physical-mechanical and physical properties. Most of the articles evaluating technical properties in the substitutability calculation for plastics focused on physical properties (mainly mass), followed by physical-mechanical properties (mainly elastic modulus and tensile strength). The latter two physical-mechanical properties look at the ability of a material to bend and the ability to withstand an applied load without failure, respectively (Demets et al., 2021; Rufe, 2013). The fact that mass was most frequently considered is probably due to the simplicity of quantifying it, which represents an advantage of considering this property in substitutability calculations. On the contrary, determination of physical-mechanical properties is less straightforward and requires a laboratory test in which a load is applied to evaluate how the material reacts (Rufe, 2013). Moreover, the widespread use of mass in the substitutability calculation can be explained by the well-known concept of recycled content referring to the percentage of the total mass of a product that comes from rMaterials (Eriksen et al., 2020; Horodytska et al., 2020; Neo et al., 2021). However, mass alone does not accurately reflect the quality and functionality of recycled materials (rMaterials). During the recycling process, factors such as contamination control and processing techniques can lead to rMaterials becoming unsuitable for high-end applications, consequently diminishing their effectiveness as a substitute for virgin materials (Grant et al., 2020; Tonini et al., 2022).

Additionally, 17 other papers considering substitutability of plastics did not study technical properties at all. These based exclusively on price (12), market shares (2) or on the number of recycling cycles (1). The remaining two considered "quality characteristics" such as colour and odour to determine the market position of rPlastics, but these were not used as a basis for determining substitutability. While price and market share data can serve as proxy estimates for substitutability in cases of undefined applications, it is susceptible to market volatility, thus introducing uncertainty (European Commission, 2010; Golkaram et al., 2022). Regarding the utilization of the number of recycling cycles, despite its consideration of quality losses linked to recycling, practically estimating the number of recycling cycles is unfeasible due to challenges in monitoring recycled plastic. This requires the application of experiments to obtain the needed data for estimating the recycling cycle count.

Among the 13 articles studying paper, only one considered physical-mechanical properties for estimating substitutability, while 12 relied on the number of times paper can be recycled. Concerning the six articles examining metals, one article based its comparison on mass concentration of the target component (grade content) and recovery efficiency, another on recovery efficiency and market price, one solely on mass concentration of the target component, and three articles focused on economic parameters. The choice of concentration and recovery efficiency for metals is driven by feasibility considerations. Recycling metal waste becomes viable when certain thresholds are met, such as a minimum concentration (e.g., 24% for manganese) and a coefficient of recovery for the target component (Jandieri, 2022).

In the case of the three articles focused on wood, physical-mechanical properties (specifically, elastic modulus and longitudinal bending strength) were considered for estimating substitutability. These properties are important for wood as they are associated with maintaining the structural integrity of wood-based structures and supporting loads or resisting bending forces, respectively. In conclusion, the properties considered during the substitutability calculation, for all materials, should align with the specific requirements of their defined applications (Demets et al., 2021; Golkaram et al., 2022).

Table 2 also indicates that among the 49 articles analyzed, six examined the substitutability of other materials. These materials encompass tires, construction and demolition waste (C&DW), glass, and carbon fibers. For the first three materials, the evaluation of substitutability relied on mass, with the additional consideration of

C&DW's composition (i.e., soil content). In the case of glass, the analysis factored in thermal conductivity, while for carbon fibres, substitutability was based on the physical-mechanical attribute of tensile strength.

Additionally, in six out of the 49 papers (12%), the rMaterial substituted a different sMaterial. For example, rPE from household waste replaced cast iron and hardwood in a street bench, or rFoam from glass cullet substituted vInsulation material[2] (Haupt et al., 2018; Huysman et al., 2015, 2017). Hence, in all of these six papers, the recycling type studied was open-loop, and the calculation of substitutability was based on mass. This can be explained by the fact that material properties are specific to material types, however, mass is a property applicable for all material types, then the comparison is feasible. In general, two approaches to quantify substitutability considering mass were identified. In the first one, the mass of rMaterial needed to replace the sMaterial is directly used as a substitutability value. This is typically the case when the rMaterial replaces a different sMaterial e.g., rPE is used to produce a street bench normally made of 63 kg cast iron. If the bench is made of rPE, 95.5 kg of rMaterial is required, hence 1 kg rPE substitutes 0.65 kg cast iron (Huysman et al., 2017). In the second one, a technical property is first used to estimate how much of the mass of rMaterial is required to replace sMaterial. For instance, van Eygen et al. (2018) considered the thermal conductivity and density of recycled and virgin EPS to calculate the amount of both materials needed to provide a thermal insulation of one m^2K/W, resulting in 2.43 kg virgin/kg recycled. Hence, when identifying the quantitative base for comparison, this paper was classified under others and not under mass. The second approach provides an advantage by allowing the estimation of the rMaterial mass that can be included in the product, without its manufacturing. Thus, substitutability can be included in LCA of products that are not yet produced due to time or budget constraints. In 27 of the 32 papers studying open-loop recycling and in eight

[2] Mix of: EPS: Expanded polystyrene, XPS: extruded polystyrene, PUR: Polyurethane, mineral wool and glass wool.

of the nine papers considering open and semi-close-loop recycling, the rMaterial substituted the same sMaterial. In those cases, not only mass was used as quantitative base for comparison.

3.2.2. Relationship between the studied materials and the LCS considered

Figure 3 presents, in terms of the number of papers, the materials studied according to the LCS that was considered for their substitutability calculation.

It was found that the only materials for which natural resource extraction was considered in the substitutability calculation were metals. As described in Section 3.2.1, the comparison was made between ore grade (i.e., metal concentration in the ore) and metal content in the waste stream (Hossain et al., 2017; Jandieri, 2022) or the ease of extracting metal from the ore versus from scrap (Jandieri, 2022). However, as per the substitution potential framework established by Vadenbo et al. (2017) (Equation 1), the concentration of the metal and ease of extraction are factors more closely related to the physical amount of secondary resources within a waste stream for treatment (U^{rec}) and the expected recovery and utilization rate (resource recovery or recycling efficiency, η^{rec}). Thus, even though the authors referred to it as the substitution ratio and ore substitution index, their studies may not be (directly) measuring substitutability or substitution potential.

The results also pointed out that none of the articles considered the raw material production stage for the substitutability calculation. This shows that the transformability (i.e., easiness of transforming) of the virgin (compared to recycled) resources into raw materials for the manufacturing sector was not studied for substitutability calculation. An example where this LCS could be relevant is in case of chemical recycling of plastic waste; the substitutability calculation could be performed comparing the easiness of transforming the recycled oil or recycled naphtha fraction to that of the virgin fossil-based oil or naphtha.

Out of the 49 articles evaluated, 32[3] articles assessed substitutability in the manufacturing stage. Note that the articles using a substitutability type based on market shares and/or prices were all considered to evaluate substitutability in the manufacturing LCS (Section 2.3). Only two of the 32 articles considered technical properties related to material processability. These articles, authored by Demets et al. (2021) and Golkaram et al. (2022), introduced new methods for determining technical substitutability. Both articles studied plastics, and looked into the melt-flow index (MFI) as a processability property, which is defined as the mass of polymer extruded under a fixed load over a specified time through a die of specified length and diameter at a fixed temperature. The study from Demets et al. (2021) emphasized that the chosen processing technique and its associated processing property, such as MFI, should be considered carefully as it affects the intended application. For example, the MFI for film applications is typically between 0.25 and 4 g/10 min, whereas for injection moulding of rigid products it is between 0.8 and 20 g/10 min (Demets et al., 2021).

32[3] of the 49 articles evaluated the substitutability at the use stage, considering properties such as mass, tensile strength, E modulus, etc. Note that within these 32 articles, the recycling process could yield intermediate products (e.g. pellets) for further processing, as demonstrated by Andreasi et al. (2017), or final products (or its components) ready for use as shown by Rigamonti et al. (2020). Importantly, all articles within the use LCS

[3] Vadenbo et al. (2017), which is one of the most well-known articles developing a new approach to calculate substitutability, is part of the 49 papers analysed in detail. However, it presents a case of energy substitutability, and none on material recycling substitutability. Hence, it was not classified in the aspects "recycling type", "material type" and "quantitative base for comparison (properties)". However, their approach looks at the degree of functional equivalence between the secondary resource and the competing product for a specific end use or application and to the expected change in consumption levels (market shares). Hence, it looks at the manufacturing and use LCSs.

consistently factored in functionality-related properties. Among the 32 articles in the use stage, six performed the substitutability calculation by comparing the rMaterial to a (different) sMaterial, based on mass (Section 3.2.1). Note that the assessment of substitutability at the use stage does not necessarily entail the consideration of the future application of the rMaterial. For example, Hermansson et al. (2022) evaluated the substitutability of rCarbon fibres against virgin ones by examining the difference in tensile strength, a property associated with the use LCS. Nonetheless, the study did not specify the future application of the rCarbon fibres, indicating that the application of the rMaterial was not a part of the assessment.

Figure 3 depicts that most of the articles that looked at the number of recycling cycles (i.e., EoL stage) for the substitutability calculation focused on paper/cardboard. This might be explained by the fact that the number of times that paper can be recycled is well-known, in contrary to other materials such as plastics (Rigamonti et al., 2009, 2010). Rigamonti et al. (2009, 2010), based on the ISO/TR 14049, applied this approach considering that paper can be recycled up to five times based on expert judgment, and thus, the environmental impacts of the production of virgin paper (vPaper) can be allocated among six use phases. They calculated the environmental impact of one kg of recycled paper (rPaper) pulp by adding 1/6 (= 0.167) of the environmental impacts of one kg of vPaper pulp to the environmental impacts of the recycling process. Based on this, they assumed that one kg of recycled pulp (rPulp) and 0.167 kg of virgin pulp (vPulp) can replace one kg vPulp, or one kg of rPulp can replace 0.833 (1-0.167) kg of vPulp. According to the authors, this value reflects the difference in physical-mechanical properties and colour between the virgin and the recycled pulp and thus the quality loss caused by recycling.

Finally, it was noticed that the substitutability of plastics, the most studied material (Section 3.2), was investigated in all of the LCSs, except natural resource extraction and raw material production. This indicates that there is room for further investigation.

3.3. Comparison of calculations and data

The most frequently used origin of data was expert judgment, which was applied in 34 out of the 49 analysed papers. This was utilized in 20 of the 24 papers that considered both technical substitutability (TS) and economic substitutability (ES), and in 14 of the 19 papers that only considered TS. The preference for expert opinion can be attributed to several reasons: its perceived reliability as firsthand information and its accessibility through expert reports, eliminating the need for experiments to obtain measured data. However, it is important to note that while expert judgment is often considered reliable, the desired material properties can vary based on conditions and specific applications. Therefore, it is recommended to combine expert opinion with measured data to ensure a comprehensive analysis and understanding of material or product characteristics (e.g., Demets et al., 2021; Golkaram et al., 2022; Huysveld et al., 2022).

The second most commonly used source of data was measured data, which was utilized in 19 of the 49 articles. It was applied in nine of the 24 articles that considered both TS and ES, and in 10 of the 19 papers that only considered TS. Market prices and market shares data were used in 18 and three of the 48 analysed articles, respectively. In the articles that only applied ES, the majority (5 out of 6) relied solely on prices, while in the articles that considered both TS and ES, price data was used in 13 out of the 24, and market shares data was used in 11 of the 24 articles. Finally, the least utilized source of data was modelled data, which was used in three of the 49 analysed articles. Regarding the calculation complexity, the majority of the papers (32) calculated substitutability as a simple ratio, followed by a more elaborated calculation (27) and, lastly, as part of a broader calculation (10). Note that the results are given by the number of papers, and that one paper can include multiple substitutability values and therefore multiple calculation complexity and origins of data (first paragraph of Section 3). More information on these aspects for all analysed papers can be found in the SM (Table S1).

3.4. An example of the in-depth analysis applied to a method to quantify substitutability

Golkaram et al. (2022) presented a model to estimate the quality of rPlastics, incorporating degradation, degree of mixing and contamination. The aspects considered were material properties (physical, physical-mechanical, and sensory), percentage of impurities, permissible value per property, ideal value for the property and relative importance for the property depending on the application (i.e., weighing factors). The model gives a quality value between zero and one which was used as the substitution factor in LCA cases. Analysing the article from Golkaram et al. (2022), it was found that the application of the rMaterial was considered for the calculation. Regarding the methodological innovation, a new approach to calculate technical substitutability, using physical, physical-mechanical and sensory properties, was developed. Furthermore, this approach was also applied (calculated) and used in LCA case studies. Considering the properties used (melt flow rate, tensile strength, impact strength, E modulus, etc.), the substitutability was evaluated in the manufacturing and use LCSs. The calculation complexity was elaborated since it is the product of different properties applying weighing factors. The origin of the data was expert judgment and measured data. The recycling type was open-loop since the waste comes from fridges and will be recycled to be used in toys, cheese packaging and food packaging (these are not the same application; however, they are all high-end applications). In the discussion section, the authors claimed that their approach is more accurate than conventional substitution methods used in the context of LCA and compared the model developed with others available in the literature, including economic value (e.g., Rigamonti et al. (2010)), market shares (e.g., Eriksen et al. (2019)) and single property indicators (e.g., Rigamonti et al. (2020)). It was stated that the first two methods represent quality yet suffers from volatility, the second basically categorizes quality in low, medium and high values which remain constant, while the last is often not comprehensive and not application based. Nevertheless, the single property approach developed by Rigamonti et al. (2020) is based on the future application of the rMaterial. Additionally, the importance of basing the calculation of substitutability on experimental data (measured data) was mentioned.

3.5. Further discussion

The use of different terms in the context of substitutability (e.g., displacement, quality factor, replacement coefficient, substitution) shows a lack of standardization in its terminology and concept. Consequently, articles addressing substitutability but referring to it with terms different from the keywords used during the literature search in the present study (Figure 1) may have been left out of the analysis. For example, Kusenberg et al. (2022) evaluated the use of pyrolysis oil from plastic waste recycling as a steam cracking feedstock compared to fossil-based (naphtha) feedstocks. The authors found that blending with fossil naphtha is necessary to meet steam cracking specifications for contaminants (e.g., nitrogen, sulfur and oxygen compounds) and thus a "dilution ratio" was calculated. Despite potentially exploring a new approach to calculate substitutability in the raw material production LCS, the article does not use the terms "substitutability" or "substitution", causing it to be missed in the literature search.

Furthermore, the concept of substitution is defined differently by various authors. Some define it broadly (*sensu latu)*, taking multiple factors into account, such as the framework to calculate the substitution potential established by Vadenbo et al. (2017) (Equation 1). Others define it more strictly (*sensu stricto)*, considering only one or two factors from previously mentioned framework. This is seen in the works of Jandieri et al. (2022) and Andreasi et al. (2017) in which different aspects are considered for the calculation but the term "substitution" is used in both of them (Section 3.2.2). Jandieri et al. (2022) calculated the "ore substitution index" by multiplying the coefficient of recovery from secondary raw materials (recovery efficiencies) by the content of the material of interest in the waste stream (Table 3). Andreasi et al. (2017) estimated the "substitution ratios" by multiplying the recovery efficiencies by the market ratios. This lack of harmonization in the definition of substitution can result in under/overestimation of substitution when LCA practitioners use values from literature without caution.

In considering a more circular economy, where rMaterials are used for production, it may become important to account for the possibility of comparing the rMaterials with replaced (other) rMaterials in the substitutability calculation (Bala et al., 2015). Bala et al. (2015) proposed a new method to calculate environmental credits associated with material recycling using their "Inside Impact Avoided Formula". This method considers the physical-mechanical properties of virgin and recycled materials, the proportion of recycled and virgin material in the market mix, and the environmental impacts of both recycling and virgin production. Hence, it assumes that the rMaterial will replace not only vMaterial in the market but a mix of recycled and virgin material. According to the authors, this approach aligns better with the attributional approach in LCA compared to the assumption of a 1:1 substitutability (Bala et al., 2015). Notably, this approach has been adopted by Bala et al. (2020) and Civancik-Uslu et al. (2019).

When calculating substitutability, some authors (e.g., Demets et al., 2021; Huysveld et al., 2022) allow a value for substitutability between zero and one, meaning that even if the rMaterial could replace more vMaterial or if its quality would be higher (upcycling), the maximum substitutability value equals one. Contrary, other authors (e.g., van Eygen et al., 2018 and Rigamonti et al., 2020) provided final substitution factors higher than one. It may be of importance to consider when a limit of one is valid and when not.

The results of the in-depth analysis could be affected by the assumptions adopted in the present study. For example, all papers focusing only on economic substitutability were classified as considering the manufacturing stage for substitutability determination, increasing the number of papers evaluating the substitutability in this LCS. Additionally, when the application of the rMaterial was unknown or unclear, the application of the rMaterial was classified as "not considered" and it was assumed that open-loop recycling was the focus. Furthermore, due to the time window of the literature search (up to May 2022), some very recent relevant papers, such as the study by Schulte et al. (2023), were not included in the analysis. Schulte et al. (2023) introduced the concept of Conservation Potential, which incorporates both quantity and quality conservation, aiming to evaluate the substitutability of rMaterials based on functional requirements and weighted technical properties relevant to specific applications. This innovative approach holds promise for LCA experts in enhancing the assessment of substitutability for rMaterials (Schulte et al., 2023).

3.6. Recommendations and framework for evaluation of substitutability in LCA

Based on the findings from the literature review, recommendations for the evaluation of substitutability in LCA are presented in this section. Firstly, it is suggested to consider the decrease in material quality after recycling by calculating its substitutability value (based on primary data). When possible, this assessment should be as comprehensive as possible, taking into account physical, physical-mechanical, physical-processability, and sensory properties, as presented in the work by Golkaram et al. (2022).

When there is primary technical data available, it is possible to calculate the technical substitutability. The specific technical parameters used for the calculation can vary depending on the LCS that is focused on for the evaluation, as illustrated in Figure 4. Note that the LCS at which substitutability can be evaluated in the case study depends on the research question(s) being addressed, which is also related to scope, i.e. the system boundaries, to be defined in the LCA study. It is essential for the practitioner to have a clear understanding of the key characteristics of the secondary materials intended to obtain from the recycling process and the specific material(s) it aims to replace, depending on the targeted application. Depending on the point of substitution, the focus may be on replacing extracted natural resources, treated or refined natural resources (primary raw materials), materials to be manufactured into products, or even finished products ready for use. However, as it is the case within LCA, altering the research question(s) and thereby modifying the system boundaries will affect the LCS at which substitutability is evaluated. Moreover, the assessment can yield different results depending on which LCS the substitutability is evaluated. For instance, considering the "use LCS" for a chemically recycled plastic would likely show a high substitutability (often assumed as 1:1), as the chemically recycled plastic shares the same characteristics and, therefore, functionality with virgin plastic. However, assessing substitutability at the "RMP LCS" could result in lower substitutability coefficients. This is due to the potential lower quality of th" obtain"d recycled pyrolysis oil, which might contain higher contaminants and lower paraffin content compared to virgin fossil naphtha. Consequently, additional purification steps and/or dilution with fossil naphtha are needed to match the quality of virgin naphtha and enable its processing into polymers. To ensure a consistent substitutability value aligned with LCA, the appropriate LCS for evaluating substitutability should be selected based on the LCA boundaries.

At the natural resource extraction stage (NRE), the comparison between the rMaterial and the sMaterial can be based on the concentration of the material of interest in the waste and in the natural resource (Δ concentration), or on the ease of extracting the material of interest from the waste compared to the natural resource. This is presented in the works by Jandieri et al. (2022) and Hossain et al. (2017).

At the raw material production stage (RMP), substitutability can be assessed based on the ease of transforming recycled (compared to virgin) resources into raw materials for manufacturing (Δ transformability). For instance, the substitutability in the context of chemical recycling of plastics could be calculated by comparing the easiness of transforming the rOil or rNaphtha fraction into new plastics to the one from virgin fossil-based oil or naphtha. Factors such as impurities and the tolerance of steam crackers with respect to their intake can affect this transformability. This could be done considering the blending of rOil with fossil vNaphtha to reach the specifications to be further process into plastics as approached by Kusenberg et al. (2022). Note however that in the work by Kusenberg et al. (2022) the terms "substitutability" or "substitution" are not used.

When substitutability is evaluated at the product manufacturing stage (MF), processability properties for specific manufacturing processes can be used (Δ processability). This approach was only observed so far for plastics, based on properties such as melt viscosity, melt-flow index, and intrinsic viscosity (Demets et al., 2021; Golkaram et al., 2022).

At the use LCS, substitutability can be assessed based on properties related to the functionality of products for specific applications (Δ functionality), such as mass (directly or indirectly used (Section 3.5)), tensile strength, and E modulus. If the rMaterial is compared to a (different) sMaterial at the use LCS, the mass of substituted material replaced by the vMaterial can be the primary factor in assessing substitutability (Δ functionality). This approach is observed in several articles, including Demets et al. (2021), Huysveld et al. (2022) and Rigamonti et al. (2020).

When substitutability is evaluated at the EoL stage, the number of recycling cycles that rMaterials can withstand can be used to estimate the substitutability (Δ recycling cycles). This was applied by Rigamonti et al. (2009, 2010).

Table 3 provides a framework for evaluating technical substitutability at different LCSs. It includes three steps: (1) Identify the LCS at which substitutability can be evaluated in the case study by looking at the research questions to be answered, (2) Look for available approaches to quantify substitutability in the selected LCS from step 1, and (3) Gather the necessary data to apply the approach(es) identified in step 2. Table 3 also provides some examples of past studies utilizing the approach along with the materials considered.

Note that substitutability estimates in certain LCSs can be combined with those in other LCSs. For example, Demets et al. (2021) took as final substitutability value the limiting factor between the processability recycling quality factor (MF LCS) and the mechanical recycling quality factor (Use LCS). Golkaram et al. (2022) multiplied processability properties (melt flow rate) with technical and sensory properties (tensile strength, modulus, colour, odour etc.) related to the application of the rMaterial to calculate the substitution ratio.

Table 3 also shows that depending on the LCSs in which the comparison between the rMaterial/resource and the substituted one is done, the future application of the rMaterial should be taken into account. This can be done, for example, by adding weighing factors, giving high values to the properties that are most important for the performance of the material in a specific application (Demets et al., 2021; Huysveld et al., 2022). However, when the rMaterial application is unknown, it is possible to apply economic substitutability based on market prices and/or shares, nevertheless, due to its potentially low representativeness, a sensitivity analysis is recommended.

The technical data can be combined with economic data (if 32vailablee) to calculate both the technical and economic substitutability. This has been done by Civancik-Uslu et al. (2019), who multiplied the composition of the rMaterial by the prices of each component material. Another approach was presented by Huysveld et al. (2022), in which the overall substitutability was calculated as the multiplication of technical substitutability (based on the ratios of multiple physical-mechanical properties for specific applications) and market substitutability (based on the potential share of the total market size of the sMaterial that can be replaced by the rMaterial considering legislative constraints). Finally, if primary data is unavailable, substitutability values can be taken from literature, while ensuring that the intended use and conditions match those of the case study, yet a sensitivity analysis is suggested.

4. Conclusions, recommendations and perspectives

Reviewing the literature on (technical) substitutability of recycled materials, this article revealed that most of the analysed papers (49%) investigated the material substitutability through a combination of technical aspects and economic aspects (prices or market shares). As for the consideration of the recycled material application, 51% of the papers did not include this in their substitutability estimation. It was also found that the consideration of the recycled material application was closely linked to the type of substitutability being evaluated. Technical substitutability was more likely to contemplate the recycled material application compared to economic substitutability.

Concerning methodological innovation, 51% of the papers developed new ways to calculate substitutability, while 33% of the papers took the substitutability value from an external source. This shows that the substitutability concept and its calculation is still in development. Considering the materials studied, plastics were the most analysed material, and their technical substitutability was mainly evaluated through mass-related physical properties.

Regarding the material life cycle stage at which substitutability was evaluated, it was found that most of the papers considered the manufacturing and the use life cycles stages. A pattern was also identified; certain materials were more analysed in some life cycles stages, e.g., at the natural resource extraction stage only metals were studied, and at the end of life stage, paper/cardboard were the most studied materials. Plastics were evaluated in all the life cycle stages except at the natural resources extraction and raw material production stages.

The concept of substitutability lacks harmonization in the scientific literature, with some authors estimating it by considering multiple factors, while others focus on only one or two factors. This variation in conceptualization might lead to some inconsistencies in research findings and presents a challenge to achieve a unified understanding.

Based on the findings, a step-wise framework to asses technical substitutability at different life cycle stages was developed to guide researchers in including substitutability in LCA studies. This considers the identification of the life cycles stages at which substitutability can be evaluated by looking at the research questions to be answered, the identification of available approaches to quantify substitutability for the respective life cycle stage, and the required data to apply the approach(es) identified. The use of the framework can serve as a guidance to increase transparency and harmonization in the estimation of substitutability for LCA studies and thus improving their the reliability and comparability.

Further recommendations were also proposed. First, it is advised to estimate technical substitutability considering not only physical-mechanical but also physical-processability and sensory properties. Second, more attention needs to be given to the consideration of the recycled material application. Third, if substitutability values are taken from an external source, these need to be carefully selected to be representative, by verifying that aspects such as the material evaluated, the application of the recycled material and the conditions considered for the calculation of the substitutability value, are the same as the ones in the study in which it is going to be used. Nevertheless, performing a sensitivity analysis is recommended when using substitutability values that are not based on primary data. Finally, due to the absence of considering the recycled material application, when applying economic-based substitutability, a sensitivity analysis should be carried out to ensure the representativeness of the results.

Based on this work, it can be concluded that the concept of substitutability is as a crucial aspect within LCA studies involving secondary resources due to its potential large influence on the results. However, it becomes evident that further advancements and standardization are necessary to incorporate substitutability effectively into LCA, as various methods currently exist without a universally agreed-upon approach. Further research can be conducted on the substitutability of material in LCS preceding the use of the secondary material, i.e.at the natural resource extraction and the raw material production). This would be particularly useful within the context of chemical recycling of plastics for which substitution methods could be developed for intermediate products such as oil or naphtha.

5. 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.

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FIGURES AND TABLES

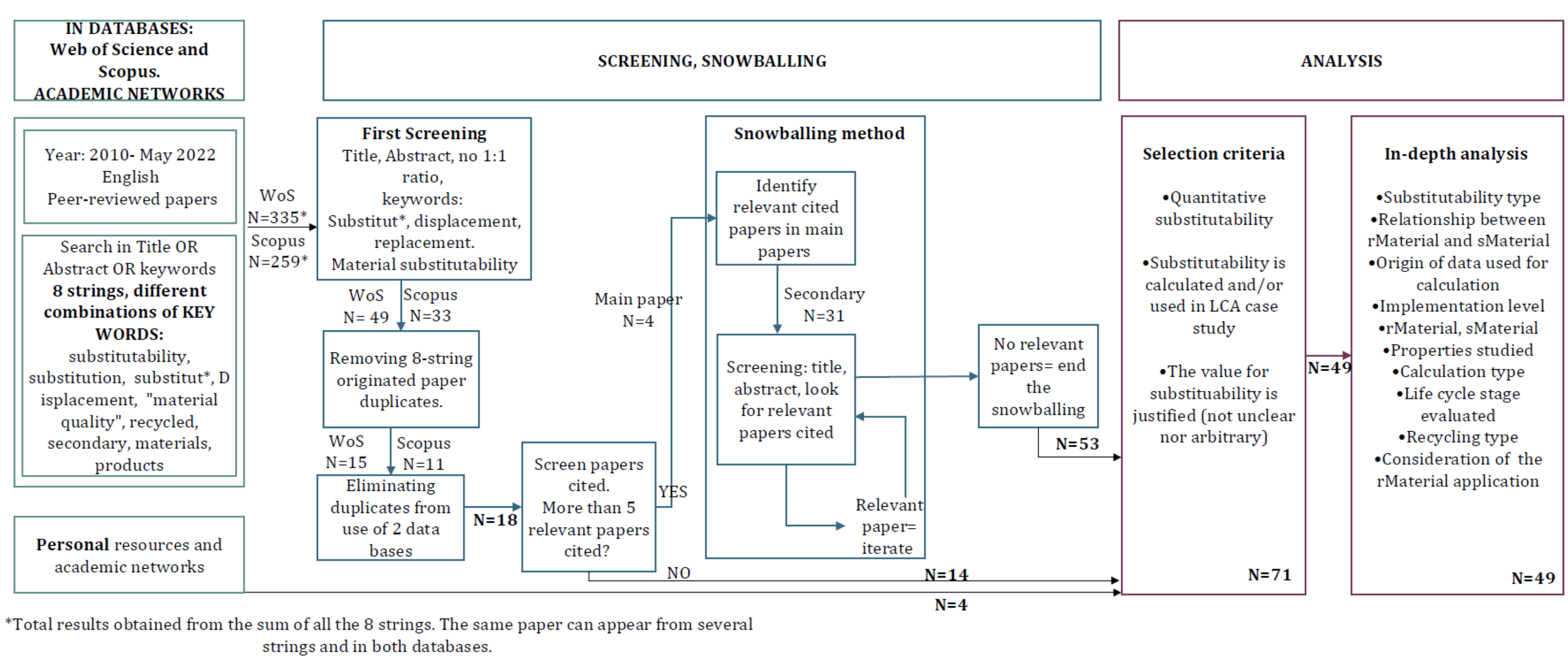

Figure 1. Overview of the literature search and selection process

Table 1: Summary of aspects considered in the in-depth analysis. Between brackets is the number of categories evaluated within the aspects.

ASPECTS	CATEGORIES
Materials studied (2)	1. Recycled material, 2. Substituted material
Substitutability type (3)	1. Technical, 2. Economic, 3. Technical and economic
Quantitative base for comparison (2)	1. Technical: properties (including physical, physical-mechanical and physical-processability)*, number of recycling cycles, 2. Market: Price ratio, market shares**
Recycled material application considered (2)	1. Yes, 2. No
Origin of data (4)	1. Expert judgment, 2. Measured data, 3. Modelled data, 4. Market data: price ratio, market shares
Methodological innovation (3)	1. New approach, 2. Calculated, 3. Used
Calculation complexity (3)	1. Simple ratio, 2. Elaborated, 3. Part of a broader calculation
Life cycle stage (5)	1. Natural resource extraction, 2. Raw material production, 3. Manufacturing, 4. Use, 5. EoL
Recycling type (2)	1. Semi-Closed loop: substituted material substituted by the same recycled material in the same application, 2. Open-loop: substituted material substituted by different recycled material or substituted material substituted by the same recycled material in a different application.

*Applicable only when technical properties are considered. **Applicable only when market substitutability is considered

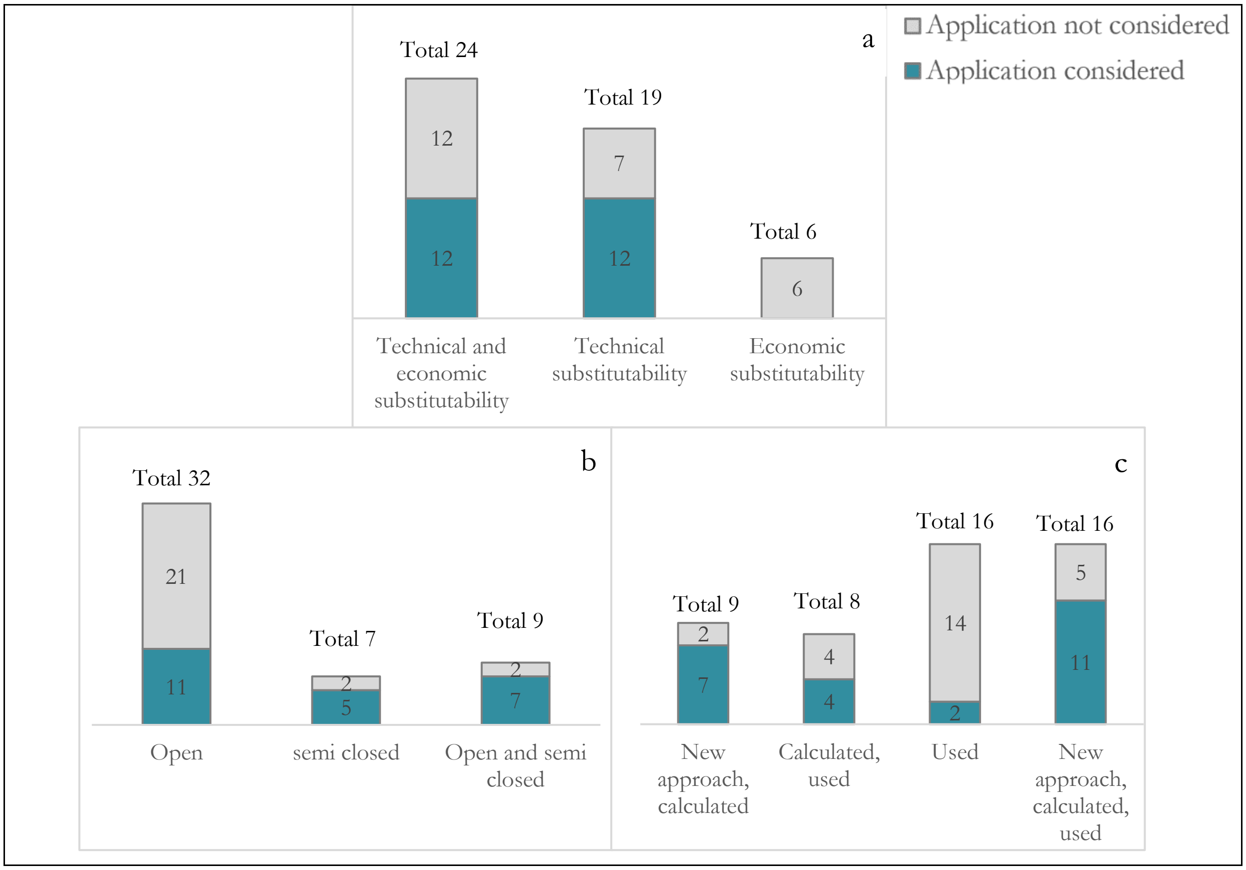

Figure 2: Relationship between consideration of the final application of the recycled material (rMaterial), the substitutability type (a), the recycling type (b) and methodological innovation (c) The presented values are the number of papers.

*Table 2: Properties considered in the technical substitutability calculation for the different materials. The values presented between brackets are the number of articles. A single article can consider different properties, hence the number of properties can be higher than the total number of articles. *Others (6): density, thickness, molecular weight, interfacial tension, thermal conductivity, colour, haze, gloss, coefficient of friction, water vapor transmission and odour. N/A: The articles based their substitutability calculation only on market prices/shares and/or in the number of recycling cycles.*

MATERIAL	PROPERTIES TYPE (QUANTITATIVE BASE FOR COMPARISON)	PROPERTIES
Plastics (39)	Physical	Mass (15)
		Others (6)*
	Physical- mechanical	Tensile strength (4)
		Elastic modulus (4)
		Impact strength (3)
		Strain at break (3)
		Flexural modulus (1)
		Yield strength(1)
		Tear strength (1)
	Physical- processability	Ease of flow (2)
	N/A (17)	
Paper/cardboard (13)	Physical- mechanical	Tensile strength (1)
	N/A (12)	
Metals (6)	Physical	Mass concentration (2)
	Others	Recovery efficiency (2)
	N/A (3)	
Wood (3)	Physical	Mass (1)
	Physical- mechanical	Elastic modulus (3)
		Longitudinal bending strength (2)
Others: tires, C&DW, glass, carbon fibers (6)	Physical	Mass (5)
		Thermal conductivity (1)
	Others	Composition (2)
	Physical- mechanical	Tensile strength (1)

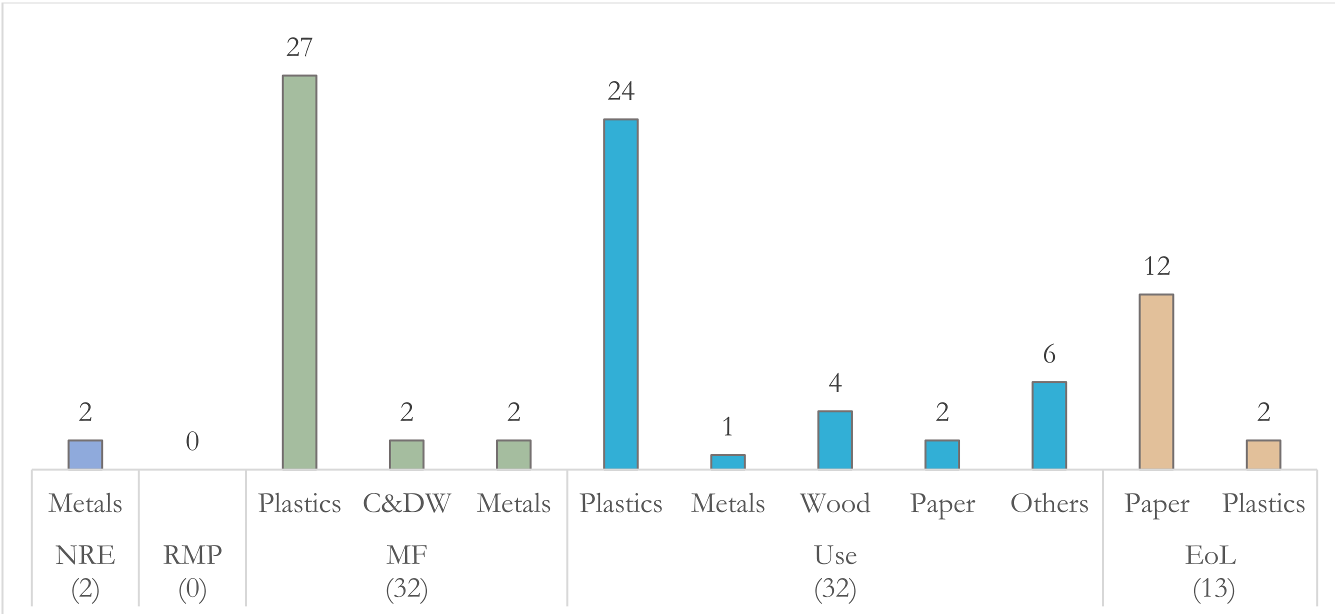

Figure 3: Point of substitution in life cycle and materials studied. The presented values are the number of papers. The values above the bars do not add up to the values below the graph because a single paper can study multiple materials. NRE: natural resource extraction; RMP: raw material production; MF: Manufacturing, EoL: End of life.

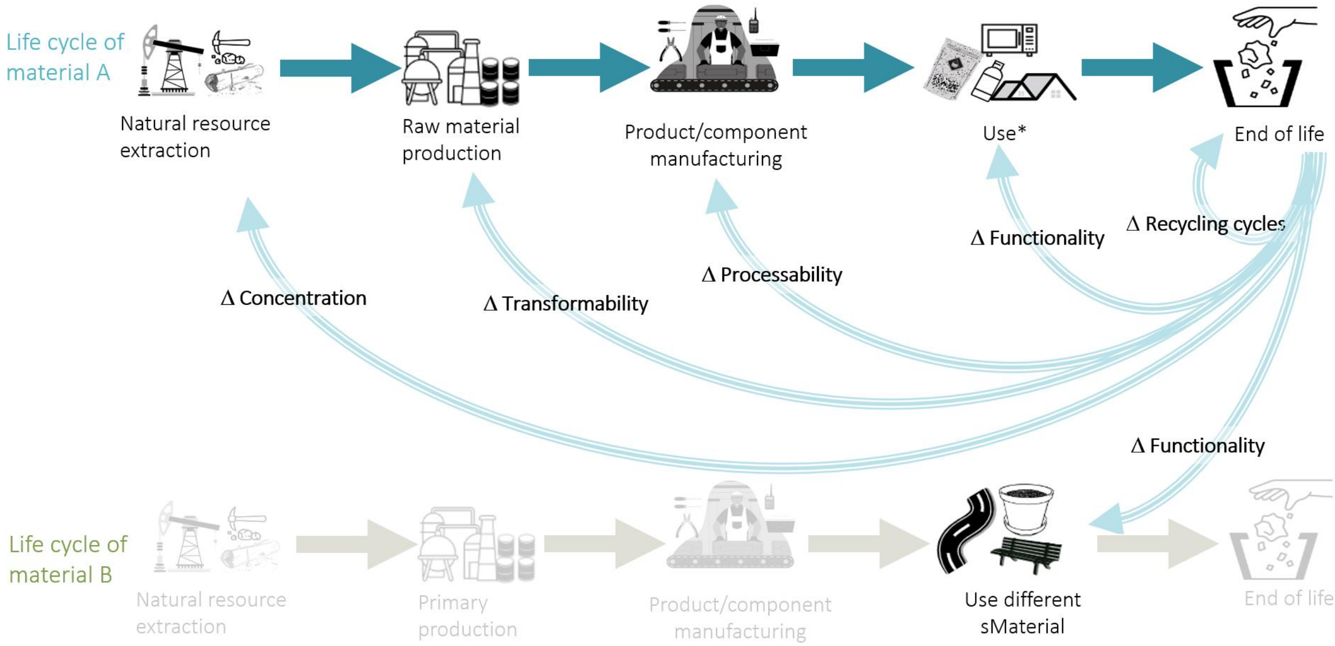

*Figure 4. Technical substitutability can be evaluated at different LCSs. The light blue arrows represent where in the life cycle the rMaterial is compared to the sMaterial (i.e., where the substitutability calculation takes places). *Use: Includes articles in which the recycling process results in a component or final product ready for use by the consumer or an intermediate product for further processing.*

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Table 3. Framework to guide LCA practitioners on how to include technical substitutability in their studies based on the LCS where substitutability is evaluated.

LCS	**Step 1: Research question to be answered**	**Step 2: Example equation**	**Step 3: Data requirements**	**Exemplary studies**	**Materials**
NRE	To what extent is it possible to replace natural **resources** (e.g., metal ore) with secondary resources (e.g., metal scrap)?	$I_{os} = \frac{X_{i_{SR}} \times R_{i_{SR}}}{X_{i_{NR}} \times R_{i_{NR}}}$ I_{os}: ore substitution index X_i: content of target component (in secondary (SR) or natural resource (NR)) (%) R_i: recovery coefficient of target component (in secondary or natural resource) *Adapted from Jandieri et al. (2022)*	• Recovery coefficient of the target component from the secondary and natural resource •Concentration of target component in secondary and in natural resource	Hossain et al., 2017. Jandieri et al., 2022.	Metals
RMP	How easy is it to transform the secondary **resource** (e.g., rNaphtha) (compared to the virgin one) into raw materials for manufacturing?	$DF = \frac{V_{SR}}{V_{NR}}$ DF: Minimum dilution factor V_{SR}: Value for certain parameter (s) (in secondary (SR) or natural resource (NR)) *Adapted from Kusenberg et al. (2022)**	•Characterization (composition) of secondary resource •Requirements (desired composition) for raw material production	Kusenberg et al., 2022. *	Plastics*
MF	How easy is it to process the r**Material** (e.g., rPE) (compared to the sMaterial (e.g., PE) into specific applications?	$RQ^{proc} = f(F_i^{vir}, F_i^{rec})$ RQ^{proc}: processability recycling quality factor *f*: scoring function (between 0 and 1) F: flow property of virgin and recycled materials *Taken from Demets et al. (2021)*	•Application of the rMaterial •Range of desired value of processability property for a certain manufacturing method (the vMaterial falls in that range), as wells as the value for the rMaterial	Demets et al., 2021. Golkaram et al., 2022.	Plastics Metals

Use	To what extent is it possible to replace s**Materials** (e.g., PE) with rMaterials (e.g., rPE) considering their functionality (physical, physical-mechanical, sensory properties) in certain applications?	$$RQ^{mech} = \sum_{i=1}^{n} w_i \cdot f\left(P_i^{vir}, P_i^{rec}\right)$$ RQ^{mech}: mechanical recycling quality factor w_i: weighing factor *f*: scoring function (between 0 and 1) P: property of recycled and virgin material *Taken from Demets et al. (2021)*	•Application of the rMaterial • Range of desired value of properties for certain application (the vMaterial falls in that range) and the value of that property for the rMaterial, or technical properties of the rMaterial and sMaterial. •Importance of properties for the application: weighing factors based on expert judgment	Demets et al., 2021. Huysveld et al., 2022. Rigamonti et al., 2020.	Plastics Wood Metals Paper
EoL	How many recycling cycles can a material withstand without a significant drop in quality for a certain application?	$$TS = 1 - \frac{1}{n_{rc} + 1}$$ n_{rc}: number of recycling cycles *Based on Rigamonti et al. (2010)*	•The number of recycling cycles	Rigamonti et al., 2010	Paper Plastics

*Although no previous studies were found that explicitly address the concept of "substitutability" of recycled materials by virgin ones during the RMP LCS phase, Kusenberg et al. (2022) appear to be exploring a new method to determine substitutability at this stage.