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LCI METHODOLOGY AND DATABASES

Allocation in recycling of composites - the case of life cycle assessment of products from carbon fiber composites

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Abstract

Purpose Composites consist of at least two merged materials. Separation of these components for recycling is typically an energy-intensive process with potentially significant impacts on the components' quality. The purpose of this article is to suggest how allocation for recycling of products manufactured from composites can be handled in life cycle assessment to accommodate for the recycling process and associated quality degradations of the different composite components, as well as to describe the challenges involved.

Method Three prominent recycling allocation approaches were selected from the literature: the cut-off approach, the end-of-life recycling approach with quality-adjusted substitution, and the circular footprint formula. The allocation approaches were adapted to accommodate for allocation of impacts by conceptualizing the composite material recycling as a separation process with subsequent recycling of the recovered components, allowing for separate modeling of the quality changes in each individual component. The adapted allocation approaches were then applied in a case study assessing the cradle-to-grave climate impact and energy use of a fictitious product made from a composite material that in the end of life is recycled through grinding, pyrolysis, or by means of supercritical water treatment. Finally, the experiences and results from applying the allocation approaches were analyzed with regard to what incentives they provide and what challenges they come with.

Results and discussion Using the approach of modeling the composite as at least two separate materials rather than one helped to clarify the incentives provided by each allocation approach. When the product is produced using primary materials, the cut-off approach gives no incentive to recycle, and the end-of-life recycling approach and the circular footprint formula give incentives to recycle and recover materials of high quality. Each of the allocation approaches come with inherent challenges, especially when knowledge is limited regarding future systems as in prospective studies. This challenge is most evident for the circular footprint formula, for example, with regard to the supply and demand balance.

Conclusions We recommend modeling the composite materials in products as separate, individual materials. This proved useful for capturing changes in quality, trade-offs between recovering high quality materials and the environmental impact of the recycling system, and the incentives the different approaches provide. The cut-off and end-of-life approaches can both be used in prospective studies, whereas the circular footprint formula should be avoided as a third approach when no market for secondary material is established.

Keywords Life cycle assessment · Allocation · Recycling · Composites · Carbon fibers · Climate impact · Energy use

1 Introduction

Recycling of products reduces the need for waste disposal. When the recovered material substitutes primary material in products, it also reduces the need for primary material production. Increased recycling reduces the environmental impacts when the impacts of recycling are less than the impacts of waste disposal and primary material production

that can be avoided. To avoid double-counting of this net environmental benefit in life cycle assessment (LCA), it can be assigned to either the product life cycle that generates material for recycling or to the product life cycle where the recovered material is used. Alternatively, the net environmental benefits of recycling can be divided between the two product life cycles. The allocation problem at recycling can be defined as the task to decide how the impacts and benefits of recycling should be divided between these two life cycles (Ekvall et al. 2020).

Many different approaches to allocation in recycling are available (see, for example, Ekvall and Tillman 1997; Allacker et al. 2017; Ekvall et al. 2020). Previous research has shown that the choice of allocation approach will inevitably influence the final results of the assessment (Allacker et al. 2017). Furthermore, it has been claimed that different allocation approaches reflect different subjective values (Ekvall and Tillman 1997). We believe that the practitioner's choice of allocation approach would therefore be aided by clear descriptions of what values or views are inherent to different allocation approaches.

Composites are materials that consist of at least two materials that have been merged into a new one. The properties of the composite are more advantageous than those of the individual materials; for example, the composite may exhibit a combination of high strength and low weight and is then desired in products where this combination is advantageous. The share of contained recycled material can vary between components in the composite. After use, the composite can sometimes be recycled as a single material, if it is reused or reshaped. However, the secondary material can often only substitute material in a product application with lower or different material requirements, or, if components of the composite can be separated, recycled as separate materials. These materials may meet very different fates depending on how much of their quality can be maintained during recycling. In some cases, only one component can be recycled, while in other cases both components can even be merged again into a new composite of similar quality as the primary one, something that is likely to become more common as recycling technologies develop. Most approaches to allocation model the recycling of one material at a time. This makes modeling the recycling of a composite a challenge, since the components of the composite may have different recycling rates and quality degradation. In fact, a composite might not even be conceptualized as consisting of different materials.

When different materials of a composite are being separated in a recycling process, impacts related to this separation process need to be partitioned not only between the separated material components, but also between the product sent to recycling and the products where the materials are used after recycling. As a parallel, complex products, such as cars, are dismantled before recycling and the dismantling impact should ideally be shared in the same way as was just described for recycling of composite materials. However, the environmental impact of such dismantling is typically small (Schmidt et al. 2004), which means that the approach for modeling dismantling has little influence on the LCA results. Many separation processes for composites, in contrast, are energy-intensive and cannot be disregarded in the assessment without significantly influencing its results.

A common challenge in LCA concerning end-of-life treatment is that the recycling generally happens in the future. In LCA of long-lived products, such as buildings, the recycling may not be taking place until several decades into the future. Furthermore, for many composite materials, recycling technology is not yet widely available. This adds the general challenges of prospective modeling in LCA, as described by, for example, Arvidsson et al. (2018).

This article discusses and explores how some prominent allocation approaches can be applied in LCA that involves composite recycling and what incentives the different approaches provide. We model the composite by considering the individual material components it consists of, to accommodate for allocation of impacts related to the separation process, as well as the potentially different recycling rates and functional and quality changes of the individual components. We adapt the selected allocation approaches to make this possible and apply them to a fictitious case of a product made from carbon fiber composite that is recycled. This is done to test (and for the purpose of this article, illustrate) how these recycling allocation approaches can be applied to composite recycling, how they influence the LCA results, and what incentives they provide for recycling. We also discuss the main challenges and opportunities of the allocation approaches, especially when applied in prospective contexts.

2 Theory and adaption of allocation approaches

An approach to allocation when there is recycling in a product life cycle specifies how the production and disposal of the material in question should be modeled. It accounts for the recycled share of the material (R_1) and/or the recycling rate of the material when it leaves the product life cycle (R_2). The boundaries between the processes, in particular what activities that count as a disposal process or a recycling process, can vary depending on the allocation approach used.

In an LCA, the chosen allocation approach should be applied to all environmentally significant materials in the product investigated if they are recycled into the product or recycled output from the product life cycle. To make the LCA complete, the impacts of manufacturing, distribution, and use of the product should be added, if significant for the LCA results. An exception is for allocation at the point of substitution, where

the impacts of manufacturing, distribution, and use are all accounted for in the allocation (Jolliet et al. 2015).

2.1 Selection of allocation approaches

Our study includes the cut-off approach and the end-of-life recycling approach, because they are both frequently used in LCA (Frischknecht 2010; Nordelöf et al. 2019). Frischknecht (2010) claims that these approaches complement each other: the cut-off approach reflects a strong interpretation of sustainability because uncertain benefits of future recycling are not included in the calculations, whereas the end-of-life approach reflects a weaker interpretation of sustainability as it accounts for the expected benefits of future recycling.

In addition to these two approaches, the Circular Footprint Formula (CFF) (European Commission 2018) was selected because it is part of the Product Environmental Footprint (PEF) framework, which we expect to be used by many LCA practitioners in the future. The PEF framework has been introduced by the European Commission in order to (among other things) increase the harmonization of environmental assessments of different products, services, and organizations (Manfredi et al. 2012).

A brief overview of the three selected approaches is found in Table 1; they are described in more detail in Sects. 2.1.1–2.1.3. Variables used in the three approaches are found in Table 2 and those common for the three approaches are illustrated in Fig. 1. Different guidelines suggest that different allocation approaches are to be used in different situations; these suggestions and how our case study applies the three allocation approaches are discussed in Sect. 4.3.

2.1.1 Cut-off approach

The cut-off approach (sometimes referred to as the recycled content approach or the 100:0 approach) is specified in, for example, PAS 2050:2011 (BSI 2011) and in the Greenhouse Gas Protocol (WBCSD & WRI 2011). This approach accounts for no impacts or avoided impacts beyond the boundary of the product life cycle. This means that flows related to raw material extraction, processing, etc. are attributed to the product where the primary material is used, while the impacts of waste disposal are assigned to the product where the material is ultimately lost. The impacts of recycling are typically assigned to the product where the recycled material is used, but researchers and guidelines disagree on to what life cycle that collection and sorting of recyclable waste belong (Nordelöf et al. 2019). In this study, we assume that the life cycle of the product ends after waste collection, which means that processes after the collection and onwards are allocated to the life cycle of the product where the recycled material is used.

With this interpretation of the cut-off approach, the part of an LCA that is associated with the production and disposal of a material in the product (E_M) is calculated using Eq. 1.

$$E_M = (1 - R_1) * E_P + R_1 * E_{R,in} + (1 - R_2) * E_D \qquad (1)$$

2.1.2 End-of-life recycling approach

The end-of-life recycling approach (also known as closed-loop approximation or the 0:100 approach) is specified in, for example, the PAS 2050:2011 (BSI 2011) and the

Table 1 Recycling allocation approaches included in this study. The allocation approaches are described in more detail in Sects. 2.1.1–2.1.3

Allocation approach	Description	Equation	Reference
Cut-off approach	Impacts of primary material production are allocated to the product where the primary material is used. Impacts of the recycling process are allocated to the product where the recycled material is used. Impacts of treatment of waste not recycled are allocated to the product generating the waste	Eq. 1	For example, PAS 2050:2011 (British Standards Institute (BSI, 2011)), Greenhouse Gas Protocol (World Business Council for Sustainable Development and World Resource Institute (WBCSD &WRI) 2011)
End-of-life recycling approach	Impacts of primary material production are allocated to the product, regardless of the product's recycled content. Impacts of recycling and a credit for avoided primary material production are assigned to products generating material for recycling. Impacts of treatment of waste not recycled are allocated to the product generating the waste	Eq. 2	For example, PAS 2050:2011 (BSI 2011), Greenhouse Gas Protocol (WBCSD & WRI 2011)
Circular Footprint Formula	Impacts of recycling and a credit for avoided primary material production are partitioned between products generating material for recycling and products where recycled material is used. This allocation reflects quality losses and the supply and demand balance on the market for the recycled material. Impacts of treatment of waste not recycled are allocated to the product generating the waste	Eq. 3	European Commission (2018)

Fig. 1 A general outline of a product life cycle that includes recycling, also showing what parts that variables in Table 2 (common for all three allocation approaches) relate to. Note that E_P and E_D are a part of the allocation issue, but $E_{Manu.}$ and E_U are not
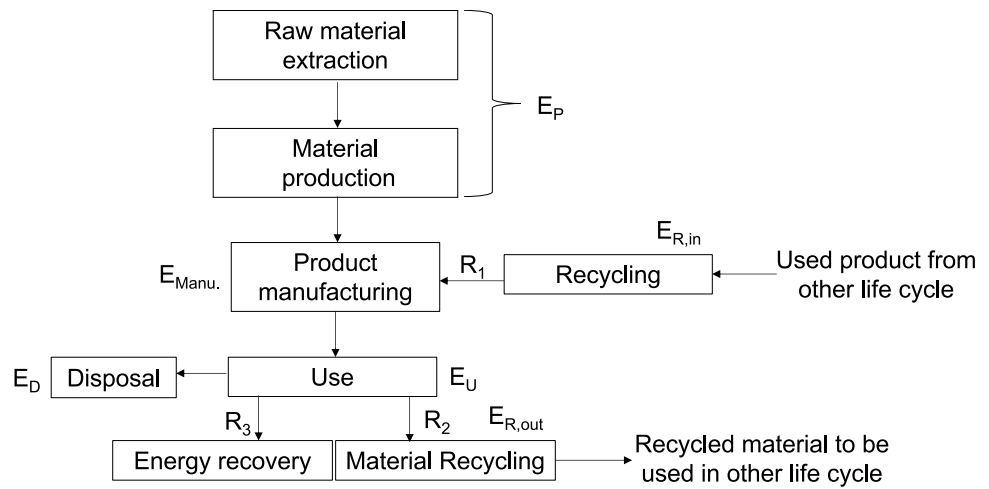
Greenhouse Gas Protocol (WBCSD & WRI 2011). This approach allocates the recycling impact in full to the life cycle of the product generating the recycled material (Allacker et al. 2017). There are many versions of the end-of-life recycling approach; in this study, we chose to use the version with a quality correction factor as described by Allacker et al. (2017) as shown in Eq. 2:

$$E_M = E_P + R_2 * (E_{R,out} - E_P^* * \frac{Q_{S,out}}{Q_P}) + (1 - R_2) * E_D$$

$$(2)$$

The use of quality correction factors to reflect these changes during recycling is also supported by for example

Annex D in ISO 14044:2006/Amd 2:2020 (International Organization for Standardization 2020). Other versions, such as the one in PAS 2050:2011 (BSI 2011), do not include avoided primary production (E_P^*), nor any quality correction factor. Instead, the avoided primary production is accounted for by subtracting the amount of recycled material from the quantity of material used in the product (E_P).

2.1.3 Circular footprint formula

The CFF is an approach to modeling recycling, energy recovery, and waste disposal stipulated in the Product Environmental Footprint Category Rules Guidance (PEFCR

Table 2 The variables used in the allocation approaches

Variable	Explanation
E_M	Environmental impact associated with the production and waste management of a material in the product investigated
E_P	Impact related to the primary material production, which includes both raw material extraction and the production of material
E_R	Impact related to recycling; $E_{R,in}$ for recycling of the material in the beginning of the product life cycle and $E_{R,out}$ for recycling of the material at the end of the product life cycle
E_D	Impact related to the disposal of the material
R_1	Share of recycled content in the material used in the product life cycle
R_2	Share of material from the product life cycle that is recycled into the next system
E_P^*	Impact connected to primary material assumed to be substituted by recycled material
Q_P	Quality of the primary material
Q_S	Quality of the secondary (recycled) material; $Q_{S,\,in}$ for the quality of secondary material coming in and $Q_{S,\,out}$ for secondary material going out
Only used in CFF	
A	Allocation factor for burdens and credits between supplier and user of recycled material
B	Allocation factor for energy recovery processes
E_{ER}	Impacts related to energy recovery processes
R_3	Share of the material being used for energy recovery at end of life
$X_{ER,heat}$	Efficiency of the energy recovery process, for heat
$X_{ER,\,elec}$	Efficiency of the energy recovery process, for electricity
$E_{SE,\,heat}$	Impact related to the substituted energy source, for heat
$E_{SE,\,elec}$	Impact related to the substituted energy source, for electricity
LHV	Lower heating value (MJ) for material used for energy recovery

Guidance) provided by the European Commission (2018). The CFF, and the PEF methodology in general, was developed through a comprehensive consensus process, involving researchers, industry, and authorities, with an aim to increase reproducibility and comparability of LCA results of goods and services. The CFF is expressed by Eq. 3.

$$
\begin{aligned}
E_M = \left(1 - R_1\right) * E_P + R_1 * \left(A * E_{R,in} + (1 - A) * E_P * \frac{Q_{S,in}}{Q_p} \right) \\
+ (1 - A) * R_2 * \left(E_{R,out} - E_P^* * \frac{Q_{S,out}}{Q_P} \right) + (1 - B) * R_3 \\
* \left(E_{ER} - LHV * X_{ER,heat} * E_{SE,heat} - LHV * X_{ER,elec} * E_{SE,elec} \right) \\
+ \left(1 - R_2 - R_3\right) * E_D
\end{aligned}
$$

$$(3)$$

In the CFF, both impacts and credits (referred to as burdens and benefits in the PEFCR Guidance (European Commission 2018)) from using and providing recycled materials are considered. These impacts are distributed between the product life cycles using an allocation factor A. The PEFCR Guidance presents default values of A, which are based on the market supply and demand balance of the recyclable materials, in the range of 0.2 to 0.8 for many common materials (see Annex C in the PEFCR Guidance (European Commission 2021)). A low A factor reflects a low supply but high demand of recyclable material and vice versa. When no A factor can be found in the list of default values, 0.5 should be used.

In essence, CFF can be seen as a compromise between the cut-off approach and the version of the end-of-life recycling approach we are using in this study. In cases when the secondary material has the same quality as primary material, A = 0.5 makes the CFF a 50:50 approach to modeling of recycling.

2.2 Redefining the composite as components

As composites in fact consist of several material components that have been merged into a new material, a recycling process will often involve a separation (or deconstruction) process with potentially multiple useful materials as outputs. This means that the recycling process introduces an allocation problem of its own: allocation between the different material components (i.e., "co-products" of the recycling process). The same is true for any deconstruction process in the life cycle of complex products (e.g., the dismantling of a car) as well as for any raw material processing that generates multiple products (e.g., in oil refineries or biorefineries). If the recycling process can have a significant contribution to the total impacts, it is important how this allocation is done. For composites, successfully recycling the material components typically requires considerable efforts, e.g., a large energy input, and how allocation is done may therefore have a significant impact on the LCA results.

Dealing with the allocation problem for the separation process may require that the components of the composite are modeled as individual materials (e.g., a polymer matrix and carbon fibers in the case of carbon fiber composites) rather than a single composite material, as functional and quality changes may differ between the components. When this is needed, the considered recycling allocation approaches must be adapted to allow for consideration of multiple materials with different fate. Equations for adapted recycling allocation approaches were therefore developed and these are described in Sect. 2.3. The impacts of the recycling process are allocated to each material i using the allocation factor α_i, which can be based on, for example, the mass or energy content, or economic value of the materials. For more discussion on approaches for co-product allocation, see Hermansson et al. (2020).

2.3 Adapted allocation approaches

Equations 4–6 show the adapted allocation approaches, where each composite component is handled separately. Note that the equations only describe the impact that is allocated to a considered composite material that is recycled and that can be part of a product life cycle. Other materials and various other activities, like the product use, might have to be added in an LCA. The variables that have been added or changed compared to what is presented in Table 2 are provided and explained in Table 3. Note that the composite recycling is now conceptualized as a separation or deconstruction process.

Equation 4 shows the adapted cut-off approach.

$$
E_M = \sum_{i=1}^{n} \underbrace{\left(1 - R_{1,i}\right) * E_{P,i}}_{} +
$$

Impact related to primary material production

$$
\sum_{i=1}^{n} R_{1,i} * \underbrace{\left(\underbrace{E_{Separation,in,i} * \alpha_{i,in} + E_{R,in,i}}_{E_{R,in}} \right)}_{} +
$$

Impact related to recycling process for recycled material input

$$
\sum_{i=1}^{n} \underbrace{\left(1 - R_{2,i}\right) * E_{D,i}}_{}
$$

Impact related to disposal of material not entering a second life cycle

$$(4)$$

Table 3 Variables changed in or specific to the adapted allocation approaches

Variable	Explanation
$E_{Separation,in,i}$	Impact related to the separation of components in a composite used in a prior product life cycle, for material i—allocated between components and between product life cycles.[*]
$E_{R,in,i}$	Impact of treatment of separated composite component material i at the beginning of the product life cycle—not allocated between components, but between product life cycles
$\alpha_{i,in}$	Allocation factor for distributing impact related to separation between components in a composite for material i used in a prior life cycle
$E_{Separation,out,i}$	Impact related to the separation of components in a composite after use, for material i—allocated between components and between product life cycles
$E_{R,out,i}$	Impact of treatment of separated composite component material i at the end of the product life cycle—not allocated between components, but between product life cycles
$\alpha_{i,out}$	Allocation factor for distributing impact related to separation between the components in a composite for material i, $\sum_{i=1}^{n} \alpha_i = 1$, where n is the number of materials from the separation process

[*]Note that if the incoming material comes from a product life cycle without a separation process, $E_{Separation,in,i}$ is 0

Equation 5 shows the adapted end-of-life recycling approach.

$$E_M = \sum_{i=1}^{n} \underbrace{E_{P,i}}_{} +$$

Impact related to primary material production

$$\underbrace{\sum_{i=1}^{n} R_{2,i} * (\underbrace{E_{Separation,out,i} * \alpha_{i,out} + E_{R,out,i}}_{E_{R,out}} - \underbrace{E_{P,i}^{*} * \frac{Q_{S,out,i}}{Q_{P,i}}}_{\substack{\text{Credit for providing recycled} \\ \text{material}}}) +}_{\text{Impact related to recycling process and credit for providing recovered material}}$$

(5)

$$\sum_{i=1}^{n} \underbrace{(1 - R_{2,i} *) * E_{D,i}}_{}$$

Impact related to disposal of material not entering a second life cycle

Equation 6 shows the adapted version of the CFF.

$$E_M = \sum_{i=1}^{n} \underbrace{(1 - R_{1,i}) * E_{P,i}}_{\text{Impact related to primary material production}} +$$

$$\sum_{i=1}^{n} R_{1,i} * (A_i * \underbrace{(E_{Separation,in,i} * \alpha_{i,in} + E_{R,in,i})}_{E_{R,in}} + (1 - A_i) * \underbrace{E_{P,i} * \frac{Q_{S,in,i}}{Q_{P,i}}}_{\text{Credit from recycled material input}}) +$$

$$\underbrace{}_{\text{Burdens and benefits related to the use of recycled material}}$$

$$\sum_{i=1}^{n} (1 - A_i) * R_{2,i} * (\underbrace{(E_{Separation,out,i} * \alpha_{i,out} + E_{R,out,i})}_{E_{R,out}} - \underbrace{E_{P,i}^* * \frac{Q_{S,out,i}}{Q_{P,i}}}_{\text{Credit from recycled material output}}) +$$

$$\underbrace{}_{\text{Burdens and benefits of providing recycled material for the next life cycle}}$$

(6)

$$\sum_{i=1}^{n} (1 - B) * R_{3,i} * (\underbrace{E_{ER,i}}_{\substack{\text{Impact related to energy} \\ \text{recovery}}} - \underbrace{LHV_i * X_{ER,heat} * E_{SE,heat} - LHV_i * X_{ER,elec} * E_{SE,elec}}_{\text{Credit from energy recovery}}) +$$

$$\underbrace{}_{\text{Burdens and benefits related to energy recovery}}$$

$$\sum_{i=1}^{n} \underbrace{(1 - R_{2,i} - R_{3,i}) * E_{D,i}}_{\substack{\text{Impact related to the share of the material not being recycled} \\ \text{or sent to energy recovery}}}$$

3 Case study description

The recycling of a fictitious product manufactured from carbon fiber composite (also known as carbon fiber reinforced polymer; CFRP) was chosen as a case study for application of the adapted recycling allocation approaches. CFRPs consist of carbon fibers and a polymer matrix. They are appreciated for their low weight and high strength and are often considered interesting for replacing, for example, heavier materials in vehicles. However, due to the highly energy intensive carbon fiber production process, the use of these composites to replace other materials does not automatically lead to a reduction in environmental impacts (Hermansson et al. 2019). One possibility for addressing this is to recycle the composite to reuse at least the carbon fibers in new applications.

Three different recycling technologies (see Sect. 3.1) were chosen to explore how the recycling allocation approaches can be applied and how the approaches rank the recycling options and why. The different recycling technologies were

compared to a case with no recycling where the product is sent to landfill. The total life cycle environmental impact of the product can be calculated by adding the environmental impacts of the material production and end-of-life treatment calculated in Eqs. 4–6 to those of the product manufacturing ($E_{Manu.}$) and of the use phase (E_U) as seen in Eq. 7.

$$Total\ life\ cycle\ impact = E_{Mas\ in\ Eq.\ 4-6} + E_{Manu.} + E_U \quad (7)$$

Note that Eq. 7 in this form is a simplified approach to calculating the life cycle environmental impact and is valid for a product produced using only the composite and that does not need any distribution or similar. If the product is produced using additional materials as well, any other impacts related to the production of these, as well as any impacts associated with distribution, need to be added as well.

3.1 System description

The case study is used here for illustration of the influence of applying different allocation approaches. Therefore, a fictitious product made from only primary composite material was chosen and one such product weighs 1 kg. A passive product type is considered to allow for the simplification that the use phase can be neglected. Several other simplifications were also made as described below. The functional unit was the service provided by one piece of fictitious product φ used for σ years.

It was assumed that the product was manufactured, used, and discarded in Germany. Any transports and collection of waste were excluded from the study as these were deemed to be negligible in relation to the impacts of the other processes.

Product φ is assumed to be manufactured by means of injection molding. The product is assumed to consist of 30% carbon fibers and 70% polyamide (PA). Any impact for cutting or arranging the fibers before adding the polymer matrix has been excluded because of lack of specific data and assumed low influence on the results. The product is considered to be produced only from primary materials, which reflects the current situation for CFRP. We also assume that there are no material losses in the product manufacturing, which is a simplification.

For simplicity, the use-phase is also assumed to have negligible impact (i.e., $E_U = 0$) and, hence, can be excluded from this particular case study. This adequately represents the use of the composite in a passive product, such as a man-hole cover, but not in an active product such as a vehicle.

All data were, if not stated otherwise, taken from the Ecoinvent APOS 3.3 database (Wernet et al. 2016). All LCA modeling was done using the software OpenLCA v.1.10.

The polyacrylonitrile (PAN) precursor fiber production data was based on a dataset by Fazio and Pennington (2005) in the European reference life cycle database (ELCD), which was modified to fit the Ecoinvent nomenclature. Carbon fiber production inventory data were taken from Romaniw (2013). Data for PA production was approximated by a nylon 6–6 production process.

Three different recycling methods were considered: grinding (Case A), pyrolysis (Case B), and supercritical water treatment (Case C). Data for the recycling processes were taken from Dong et al. (2018) who base their data on the work by Hedlund-Åström (2005), Witik et al. (2013), and Knight (2013). It was assumed that when recycling is not done, the discarded product is sent to landfill. The modeled systems are shown in Fig. 2. Details on the
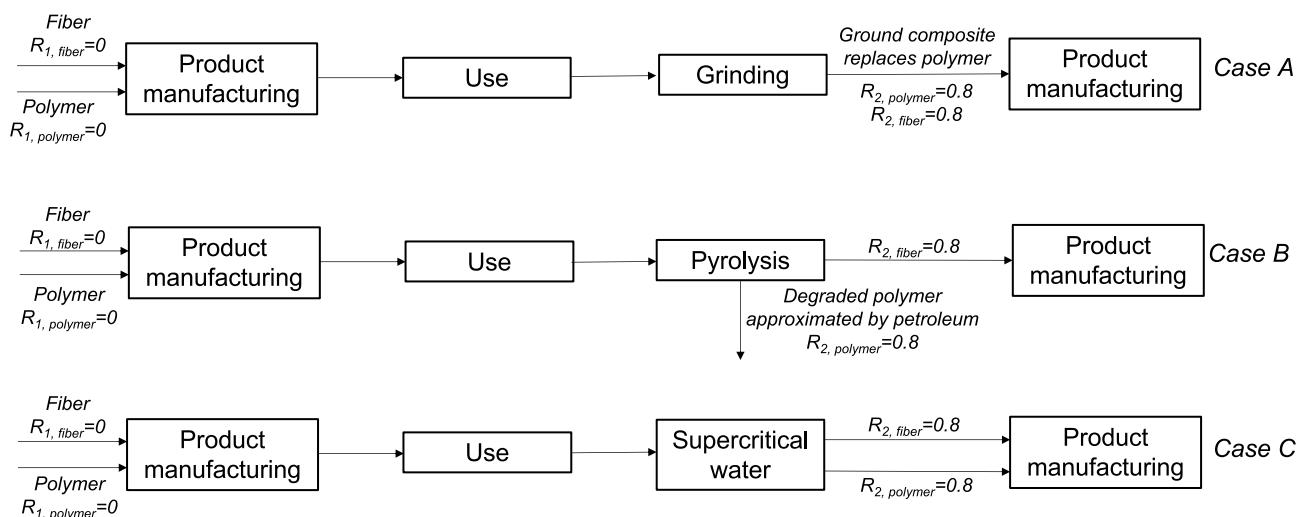

Fig. 2 A schematic illustration of the three recycling Cases; A, B, and C, considered in the assessment. The considered recycling rates (R_1 and R_2) are provided for each component and recycling method.

Note that the assessment only considers a product made from primary materials, and that the use phase was considered to have negligible impacts in the case study and was therefor excluded

modification of datasets and on system modeling can be found in the supplementary material.

Grinding of a composite can in some cases separate the polymer matrix and the fiber (Dong et al. 2018). In this study, however, for illustration purposes, the composite is ground in Case A and the resulting powder of mixed material is assumed to replace only polymer in a secondary composite material application.

In pyrolysis (Case B), high temperatures are used to decompose the polymer matrix and to liberate the carbon fibers. The energy carrier is assumed to be electricity (Dong et al. 2018). Clean fibers are recovered, and thermal energy and/or secondary fuels or chemicals can be produced (Yang et al. 2012). For example, Cunliffe et al. (2003) write that the polymer matrix can be turned into oil and gas products. In this study, the decomposition of the polymer is assumed to generate a chemical comparable to petroleum, and therefore substituting the production of petroleum. Note that we assume that 100% of the polymer is decomposed into an oil; in reality, this value is likely lower.

The process used in Case C is supercritical water treatment. The energy carriers are assumed to be electricity and heat generated from natural gas (Dong et al. 2018). The inventory data used are for the recycling of a composite with epoxy matrix, but in our case study, the composite contains a thermoplastic polymer; this was not corrected for, but is not expected to have influenced the results in important ways. It is assumed that the recovered fibers can substitute fibers with a quality very close to primary fibers, and that the recovered polymer substitutes a similar polymer produced from fossil resources with negligible quality degradation and losses in the recycling process.

The impacts of the recycling process were allocated between the composite material components based on mass for comparability and data availability reasons. As a simplification, it is assumed that none of the recovered materials in the three cases needs any (intensive) processing before entering the second product life cycle. This is more likely to be true for the fibers, but the polymer matrix would need some processing before the next application as the polymer has likely been degraded or contaminated to a certain extent depending on the recycling method used. However, as this need will also vary depending on the secondary application of recycled materials, which in our case study is not specified, it was excluded from further assessment.

3.2 Impact assessment methods

This study considered the environmental impact categories of energy use and climate impact. The selection was based primarily on data availability, but it was also deemed sufficient for the purpose of the study as it serves to illustrate how the choice of allocation approach influences the results. In addition to this, as climate impact often correlates well with other emission-based impact categories, it can be seen as a proxy for other impacts (Janssen et al. 2016). The impact assessment methods used were the cumulative energy demand and GWP100 in CML2001 as provided by Ecoinvent 3.3 (Wernet et al. 2016).

3.3 Case study-specific values for the variables

3.3.1 Recycling rates for incoming and outgoing materials

As shown in Fig. 2, R_1 is set to 0 and R_2 is set to 0.8 for both the fiber and the polymer in all three cases.

3.3.2 Factor A

For most polymers, the default value is $A = 0.5$, which corresponds to an equilibrium between demand and supply; this is also the value applied to the polymer in all three cases in our study. There are no A factors available for carbon fibers in the list of default values, so according to the PEFCR Guidance, this value should be set to 0.5. This would, however, not necessarily reflect the true supply and demand balance for recycled carbon fibers, especially as large-scale carbon fiber recycling is not available today and future market developments are unknown. Therefore, this study explores the options of applying either $A = 0.2$ or $A = 0.8$ for the fibers in Cases B and C. In Case A, the recycled fiber is assumed to substitute polymer, thus adopting the A value for polymer (i.e., 0.5).

3.3.3 Quality of primary and secondary materials

Both the end-of-life recycling approach and the CFF account for the difference in quality between primary and recycled material. Annex C in the PEFCR Guidance provides default quality ratios (Q_S/Q_P) for several common materials. For example, the quality ratio for most plastics is set to 0.9. We use this default value as a quality correction factor both for the CFF and for the end-of-life recycling approach in Cases A and C. As the polymer in Case B is assumed to substitute production of petroleum, which is considered a new product rather than a degraded form of polymer, no quality correction factor for the degradation of the polymer is needed.

Determining the quality ratio of the fibers is not as straightforward as for the polymer since there are no default values available in the PEFCR Guidance. The PEFCR Guidance states that quantification of the quality ratio should be based on either (1) the price of secondary materials compared to primary materials, or (2) physical factors when they are more relevant. The production cost, and as a consequence price, of

carbon fibers is generally determined by their quality (Fang et al. 2017), but as there is currently still no large-scale production and market for recycled carbon fibers, costs are highly uncertain. In this study, we instead base the quality ratio, and thus the quality correction factor, on the loss of tensile strength. We assume a tensile strength degradation of 18% for Case B which corresponds to the degradation in tensile strength for recycled fibers using the fluidized bed method (Pickering et al. 2015), and 2% for Case C which is the highest retention in the tensile strength of the carbon fibers as reported by Zhang et al. (2020) (who in turn base this value on results from Henry et al. 2016). There is no need to assess the quality of recycled fibers in Case A since they are not separately recovered; they are assumed to replace polymer in the next application. As the fibers are assumed to be indistinguishable from the polymer after grinding, we used the same quality correction factor for these as for the polymer.

3.3.4 R_3 and energy recovery

In this study, the share of the product that is not recycled is assumed to go to landfill, meaning that R_3 is set to 0 for both the polymer and the fiber in all three cases, and there is no need to determine a factor B, $X_{ER, heat}$, or $X_{ER, elec}$.

4 Results and discussion

4.1 A composite should be modeled as multiple materials

The changes in the modeling of the composite into several parallel material flows were proven useful in the modeling of recycling of a composite. The conceptual split of the composite into different material flows may seem obvious once it is established, but a composite is not necessarily thought of as several material components that will have different fates in the end-of-life process, as well as in subsequent product life cycles. If the composite had been modeled as a single material at recycling, the differing properties, quality degradations, and supply and demand of the different components would not have been captured. We argue that this approach for assessments that include recycling of composites is useful and likely applicable also to many other types of composites, allocation methods, and recycling cases, than the ones used here, but we have not tested this.

4.2 The influence of allocation approach on the LCA results

The adapted allocation approaches, described in Eqs. 4–6, were applied to three different cases of

recycling of a product manufactured from CFRP. The resulting climate impacts and energy uses are seen in Figs. 3 and 4. Note that the results are based on information found in literature and databases and are generated for the purpose of illustration of the influence of the allocation approaches on the cradle-to-grave environmental impact only. The modeling was not done with any specific real product in mind.

The cut-off results are identical for all recycling options. This means that the cut-off approach gives no incentive to improve recycling processes. In fact, it hardly gives an incentive to send the product to recycling at all. The only difference between no recycling and the cut-off approach is that the disposal part in the cut-off approach is slightly smaller as parts of the product's materials are recycled into a second life cycle. The impacts related to the disposal process are small, and this difference is hardly visible in Figs. 3 and 4. The cut-off approach does, however, give incentives to use recycled materials in product manufacturing (see Eqs. 1 and 4). Adding recycled input as a parameter in this case study would have made the modeling of different scenarios complicated and difficult to interpret and we deemed this to be beyond the scope of this study. However, a simplified calculation shows that recycling components as in Case C and using them in a secondary product application would result in both the climate impact and energy use of the secondary product being reduced with around 50% compared to using only primary materials (i.e., half of what is presented in Figs. 3 and 4). This approach thus strongly rewards using recycled materials in composite-based product manufacturing. The use of recycled materials in product manufacturing should be considered in future research on recycling allocation; this is particularly important in prospective studies and for composites that already today include recycled materials.

The end-of-life recycling approach results in lower climate impact and energy use for Case C compared to Cases A and B. This is because of the relatively large credits associated with the avoided production of fibers and polymers due to the high quality of the recovered components. In Case A, the carbon fibers do not substitute other carbon fibers but instead a polymer and the product is therefore awarded the relatively smaller credit corresponding to replacing polymer. In Case B, the polymer is degraded and assumed to replace petroleum production, which gives a smaller credit than when the recycled polymer is assumed to replace polymers. Note that the larger reduction awarded due to substitution in Case C is partly counteracted by the relatively high impacts of the supercritical water recycling process. Otherwise, the climate and energy advantage of Case C would have been even greater. This illustrates how some allocation methods reward recovering high-quality materials from the product, and thus highlights the importance of capturing this in the
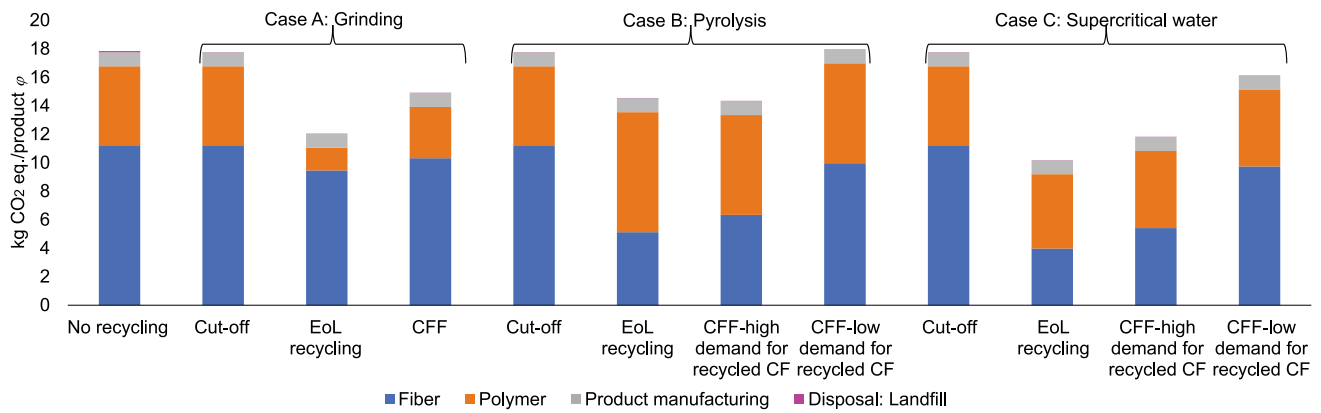
Fig. 3 The climate impact of the fictitious product φ in the three different recycling cases, using different allocation approaches

allocation procedure. It also shows that the end-of-life recycling approach gives not only an incentive to recycle the product, but also to prioritize the recovery of that material for which the primary material production has the highest environmental impact. What the method does not do is to give any incentive to use recycled materials in production of composites (see Eqs. 2 and 5). The strong influence of what is substituted and of the considered quality degradations in this method highlights the importance of relevant input data. Accurate data are particularly difficult to obtain when modeling future recycling systems, in this case, the pyrolysis and supercritical water treatment processes. In such cases, key parameters could be varied in a sensitivity analysis.

When using the CFF, the climate impact and energy use results strongly depend on the supply and demand balance of the market (i.e., allocation factor A). For example, in Case A (grinding), the fibers are not reused; instead, the powder containing both polymer and fiber is reused and replaces only polymer matrix in a new composite. This means that the output of recycled polymer material is higher than the input, as it is diluted with milled fibers. As a consequence, the climate

impact and energy use results are lower for Case A compared to Cases B and C when there is a low demand for recycled carbon fibers. This result is intuitive as it suggests that when the demand for recycled fibers is low, it is better to replace the polymer than the fiber. With a high demand for recycled fibers, though, the CFF results indicate that fibers should be recycled separately, particularly if the recycling process does not degrade the fibers significantly (for Case C). Hence, the CFF gives incentives not only to provide high-quality recycled materials to the next product life cycle (just like the end-of-life recycling approach) but also to recycle and recover materials for which there is a demand on the market. This is true even if the recycling process itself has a high environmental impact. Note that the degree of complexity of the CFF makes it particularly challenging to apply in studies of future recycling systems, as it considers both recycling rate, the market supply and demand balance, what is substituted and quality changes of each component in recycling. As society changes and technology develops, all these variables are expected to change. A way to manage this in future-oriented studies could be to look into best- and worst-case scenarios.
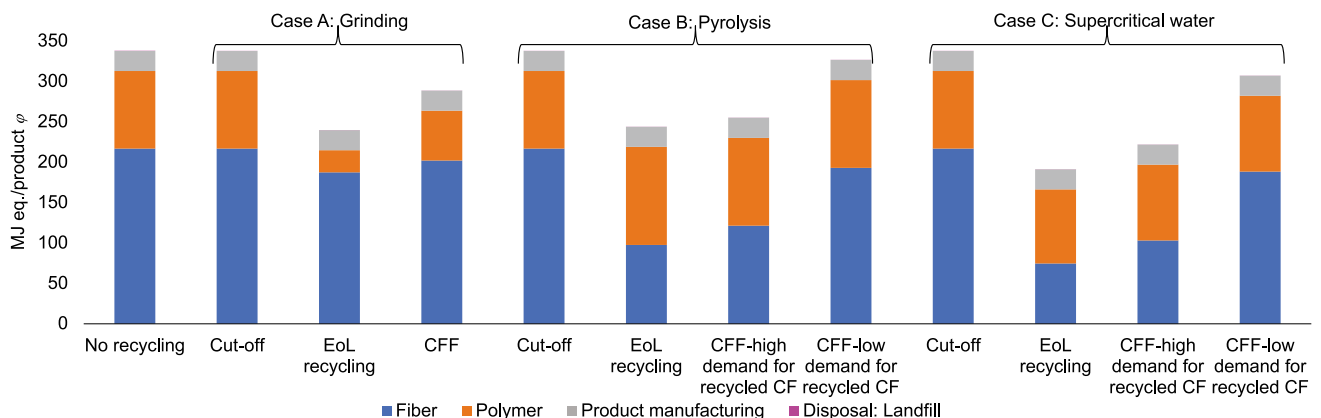

Fig. 4 The cumulative energy demand of the fictitious product φ in the three different recycling cases, using different allocation approaches

4.3 On choosing allocation approach

What is clear from the results in this study is that the different allocation approaches provide different incentives for recycling. Differences in incentives provided by different recycling allocation approaches have been discussed before in the literature on a more generic level, for example, by Frischknecht (2010) who relates the cut-off and end-of-life recycling approaches to different views on sustainability (see Sect. 2.1). Frischknecht (2010) writes that it is not possible to claim that one approach is more correct than another, but that both approaches can be rational depending on one's value system.

It is not possible to provide general guidance on which allocation method is more relevant as this depends on the context and purpose of each LCA study. There are, however, some recommendations in the literature: BSI (2011) stipulates in PAS 2050:2011 that the cut-off approach is to be used if the recycled material does not keep the same inherent properties as the primary material, and the end-of-life recycling approach is to be used if the recycled material does keep the same inherent properties as the primary material. Likewise, the Greenhouse Gas Protocol by WBCSD & WRI (2011) stipulates that the end-of-life recycling approach is to be used in processes where the recycled material can be used to replace primary materials with the same inherent properties, and the cut-off approach is to be used in open-loop recycling systems which have recycled material inputs and outputs. In some cases, both the end-of-life recycling approach and the cut-off approach could be relevant. In such a situation, the Greenhouse Gas Protocol (WBCSD & WRI 2011) suggests that the cut-off approach is more appropriate to use when the product contains recycled input but the product is not recycled, in cases where the markets for recycled materials are saturated, when the use phase of the product is long or uncertain, and when the content of recycled materials in the product is influenced by the companies' activities alone. The end-of-life recycling approach, on the other hand, is to be used when the recycled material content of the product is unknown, when the market for the recycled material is not saturated, and when the use phase of the product is short or well known.

When attempting to apply the guidelines by the BSI (2011) and WBCSD & WRI (2011), described above, to our three recycling cases, it can be argued that the changes in quality of both fibers and polymer mean that the cut-off approach is more suitable for Cases A and B, whereas the end-of-life recycling approach is more suitable for Case C. However, it could be argued that today's market is not saturated for recycled carbon fibers, and the end-of-life recycling approach could be seen as the most appropriate approach for Case B as well. This highlights again the importance of the choice of allocation approach in future-oriented studies,

and ideally, both the cut-off and the end-of-life recycling approaches could be used to illustrate the potential effect of changes in both market saturation and technology development, where, for example, quality degradation could decrease with continued technology development over time.

Section 4.2 highlights the importance of the allocation factor A in the CFF, which allows for accounting for relevant market mechanisms in the modeling of recycling. This inclusion allows for a more accurate modeling of recycling in cases when the market for the secondary material is sufficiently established for its bottlenecks to be identified. When there is not yet an established market, the PEFCR Guidance stipulates A = 0.5 (European Commission 2018). This makes the CFF a compromise between the cut-off and the end-of-life approaches. As an alternative, different values for A can be applied to illustrate the uncertainty, as in our case study, where different assumptions on the future demand for recycled carbon fibers were included (see Sect. 3.3.2). Note, though, that the PEFCR Guidance allows for 0.2 < A < 0.8 only, which does not cover the full range of possible situations. This full range is better illustrated by the two extreme approaches: cut-off and end-of-life recycling (see Figs. 3 and 4).

5 Conclusions

In LCA, we recommend that the material components in composites are modeled as separate and parallel flows to make it possible to account for the different quality changes in recycling. We also recommend that the impacts of the recycling process that separates the material components are allocated between these flows.

It was found that the inherent incentive structures of different allocation approaches clearly influence assessment results when recycling of products is involved. In essence, we saw that the cut-off approach provided no incentives to recycle the composite, whereas both the end-of-life recycling approach and the CFF give incentives to provide recycled materials of high quality at the end-of-life. We recommend that the LCA practitioner consciously selects and clearly states and motivates the choice of allocation approach and discusses how the resulting environmental impacts are influenced by the choice.

The case study showed that when a recycling allocation approach puts a large importance on the quality of recovered materials, the recovery of high-quality materials could decrease the total environmental impacts of a product manufactured from primary composites, even when the recycling method itself has a relatively high impact.

Each of the allocation approaches considered in this study come with their own challenges when applied in prospective studies. We recommend to use both the cut-off approach and the end-of-life recycling approach to capture

the extremes. When doing so, variables such as quality degradation and products being substituted should be carefully modeled using a prospective perspective and, ideally, varied in a sensitivity analysis.

When the cut-off and end-of-life approaches are both used in a prospective LCA of products where markets for secondary materials have not yet been established, we advise against using the CFF as a third method. To be accurate, the CFF depends on information on the supply and demand on the market for secondary materials—information that is unavailable if the market is not yet established. Without such information, the CFF is merely a compromise between the cut-off and end-of-life recycling approaches and, hence, adds little new information to the study.

Declarations

Competing interests The authors declare no competing interests.

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