An Evaluation of Life Cycle Assessment and its Application to the Closed-Loop Recycling of Carbon Fibre Reinforced Polymers

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Abstract

Life cycle assessment (LCA) is a valuable tool for establishing the environmental burdens of a composite material over its lifetime. It is therefore of importance to the composites industry as a material selection tool when determining the applicability of recycled composites in the component design phase. This review paper evaluates the LCA framework and its ability to accurately determine the benefits of closed-loop composite recycling, with the aim of aiding future material selection for recycled CFRP. LCA is a powerful tool for CFRP assessment when used in combination with an economic and technical component as covered by the integrated Life Cycle Engineering approach. The broad range of values available in LCA databases may prove an issue for cross comparison between studies and provide disparate results leading to impractical conclusions. The use phase offers the greatest potential for CFRP emissions savings in the transport sector; the advent of closed-loop recycling for CFRP may provide the multiple use phases required to breakeven on the significant energy burden of production or possibly provide net environmental savings gains over traditional materials.

Keywords: Life cycle assessment; Polymer-matrix composites (PMCs); Recycling.

1. Introduction

LCA uses an internationally standardised methodological framework for analysing the environmental impacts associated with the life cycle phases of products, processes or activities over their entire life, typically from cradle-to-grave. For a product, this is accomplished through: *1) the summation of the relevant inputs and outputs of a collection* *of processes, 2) the evaluation of potential impacts of this list, and 3) the final interpretation of results in the context of the goal and scope, as defined at the beginning of the assessment* [1].

LCA is a valuable decision-making tool, useful for generating insight into environmental 'pinch-points', savings opportunities, and process trade-offs. These benefits, along with a shift in public opinion towards environmental protection, have led to LCA becoming a recognised industrial tool for the evaluation and selection of novel materials and processes. Its presence is gradually increasing in the construction [2–4], aerospace [5,6], wind [7,8] and automotive [9,10] sectors. Current requirements for the automotive industry are to reduce vehicle emissions in use [11], and increase recycling at the end of life (EOL) [12]; with the aim of meeting recycling targets (in EU at least 85%) [13]. Assessment criteria from LCA can provide the valuable process modelling and quantitative analysis required to make informed improvements to meet these goals. In contrast, the composites industry also uses the LCA approach to highlight the benefits of lightweight as alternatives to conventional materials [14]. Typically, lightweighting materials such as aluminium, magnesium, carbon fibre reinforced polymer composites (CFRP), and glass fibre reinforced composites (GFRP) are compared with conventional steels, over a product's lifetime [10,15,16].

CFRP can provide weight savings of up to 65 % when compared to steel automotive parts [15] and up to 20% when replacing aluminium in aviation [17]. However, the suitability of CFRP for a given application is not solely driven by weight, there are many other considerations such as part complexity, production volume, manufacturing lead time, environmental impact, and costs [18]. The production and manufacturing burden (PMB) of CFRP *i.e.* the total environmental impact, cumulative energy demand (CED), and financial cost of production, rules them out as an alternative material for many industries as they do not meet industrial legislative or commercial requirements [19]. One method of reducing this burden is through recycling, as the energy required for recycling is typically far less than that of primary production [10]. However, these benefits have so far not been widely quantified or, in most cases, developed into a practically feasible technology. The application of LCA to evaluating the benefits of closed-loop recycling is still in its infancy, particularly for CFRP structures, and a suitably comprehensive framework, capable of capturing the impacts over multiple lifetimes, is not currently available. This study reviews the disadvantages associated with current LCA practices and aims to provide guidance for the evaluation of emerging closed-loop recycled CFRP.

From these findings, the available strategies for defining and evaluating the benefits of a closed-loop recycling process, or a closed-loop recyclable material, are analysed. This is achieved through various EOL allocation approaches and the application of multiple use-phases. A closed-loop process is defined as one which requires no additional material to propagate, once the initiator material has been added [20]. Closed-loop processes for composites manufacturing and recycling are in accordance with the Circular Economy paradigm presented by the Ellen MacArthur foundation [21] and encouraged by the Composite Leadership Forum in the UK Composite Strategy (2016) [22] for new composite materials.

2. Life cycle assessment methodology

The LCA framework used is as stipulated by the International Organisation of Standardisation (ISO) standards (ISO 14040 2006; ISO 14044 2006), an LCA consists of four categories: *1) goal and scope definition, 2) life cycle inventory analysis, 3) impact assessment, and 4) interpretation of results* [23,24].

The goal and scope outline the system to be studied, describe the environmental impact categories, and identify any limitations or assumptions made during the assessment. It is important to first establish the decision that will be informed by the result of the assessment, for material selection this is a comparison of alternative systems for a reference unit, *i.e.* a *functional unit*, this can be a specific amount of material [25], or a specific component, *e.g.* an aircraft undercarriage stay beam [26].

The life cycle inventory (LCI) comprises the identification, and summation, of all relevant unit process flows associated with the product system. Product systems for CFRP encompass all the interconnected *unit processes* of a products life cycle, from raw material extraction to EOL processing. The LCI (Fig. 1) is composed of the key phases of the functional unit life cycle as limited by the system boundaries. For most product-focussed LCA this spans the cradle-to-grave life cycle of a product, which in turn comprises the following phases; a) raw material production, b) manufacturing, c) use and d) EOL [27].

Fig. 1. Schematic of a typical LCI.

For an automotive CFRP component, this would include every unit process from acrylonitrile treatment, through fibre sizing, matrix polymerisation, resin transfer moulding of the panel, panel lifetime in vehicle, disassembly, and EOL. Each of these unit processes is joined by its input and output flows where each flow could be materials, resources or emissions. One of the most common inputs of a unit process is the CED (units MJ/kg), *i.e.* required energy for all processing operations.

The databases available for LCA include Ecoinvent [28], GaBi [29], European reference Life Cycle Database (ELCD) [30] and, specifically for composites, the European Composites Industry Association (EuCIA) Eco Impact calculator database [31]. In addition to these, direct data from experimentation or industry have been used in previous studies, despite being in limited supply [6,32]. In general, one of the main drawbacks of LCA databases, is that the data are typically acquired from the product manufacturers themselves and are rarely audited. Moreover, there is also the limitation of misrepresentation as databases are seldom updated from their initial inception providing outdated and potentially unreliable data points; most of the CF data available is over 10 years old, concerningly outdated given the advances in CF production efficiency in that time [31]. EuCIA developed an Eco Impact calculator tool, c.2016, for evaluating the environmental impact of composite products without the user requiring specialised knowledge on LCAs, this is especially valuable to the small composite manufacturing companies that are prevalent in the industry [33].

It is useful to create limitations in the amount of unit processes considered in the analysis *i.e.* create a *system boundary*, however this is a trade-off between the accuracy of the evaluation and the time required to complete it [34]. Ignoring all common operations between two alternatives for a functional unit is a typical example of this. The necessity of setting a system boundary will undoubtedly lead to the omission of external effects that could result in a significant underestimated result [32]. Therefore, LCA is very much a user specific evaluation based upon subjective interpretation of the importance of the many facets of the assessment and its conclusions. This can make comparison of LCAs on the same topic complex and regularly impractical.

The suitability of an LCI process is determined by its contribution to the *impact categories* selected in the life cycle impact assessment (LCIA). The LCIA applies a variety of impact categories to the LCI which best contrast their quantified impacts in the context of the assessment scope [27]. There is a significant range of impact categories to choose from, which typically cover: Global Warming Potential (GWP), *i.e.* Greenhouse Gas (GHG) emissions, Fossil Fuel Depletion, and Ozone Depletion, *etc.* [23,24]. However, GHG (unit $kgCO_{2eq}/kg$) is the most widely reported metric used for environmental impact across industry and academia.

The selection of the most appropriate inputs/outputs of the LCI is an important factor when determining the validity of the LCA, especially when considering the system boundaries. For example, the number of carcinogens emitted during production may be insignificant in comparison to the product mass, however the other indirect environmental impacts may be considerable, and therefore of consideration. This is especially pertinent when most of the impact from a process is in a category that lies outside the system boundaries or, conversely, the most impactful process, within a given boundary, has not been included in the LCI. Despite the real advantages of LCA there are drawbacks associated with its key stages that can result in widely varying results. As a LCA is user constructed, there are elements which require personal judgement, *i.e.* the system boundary breadth and the detail employed in the mapping of production phases [10]. There is also the issue of *allocation*, this describes the need to ensure the assignment of an impact to only one process in the system; some impacts appear in other supply chains within a shared system boundary and therefore should not be accounted for twice [35].

It is important to combine economic as well as environmental assessments, especially when considering the comparison of composite materials with conventional materials as the production of CFRP is an expensive process [36]. Life Cycle Costing (LCC) is an integrated LCA approach with the additional metric of economic cost. LCC considers all of the relevant projected costs associated with a product in its life cycle, which can be split into the same four key stages as an LCA [37]. It's use has been driven primarily by cost sensitivity, especially during the research & development and design stages [37–39]. Witik *et al.* performed an integrated LCA/LCC evaluation of a glass fibre sheet moulding compound (GF SMC), glass matt thermoplastic (GMT), reaction injection moulded carbon fibre (RIMCF), injection moulded glass fibre, and magnesium, to replace a conventional steel vehicle bulkhead [9]. The study showed that GF SMC provided the lowest environmental impact and economic cost for the entire cradle-to-grave life cycle, despite offering the lowest weight savings [9]; an important additional metric given the potential for cost and weight sensitivity in composite design.

When considering material selection from a wide range of material types, *e.g.* composite, metal, and wood, it is also important to include a technical dimension to the evaluation as the resulting component will have a design envelope with mechanical property limits and varying manufacturing requirements. The Life Cycle Engineering (LCE) framework has seen recent development due its ability to capture environmental, economic and technical aspects of the material selection process [40,41]. Materials are scored based on the three aspects, and can be compared by either the *CLUBE* or *ternary* mapping methods depending on the importance placed on the technical performance [40]. Integrated LCE methods, (LCA + LCC + LCE) are also useful for highlighting areas where cost is unnecessarily high, or areas in which it may be possible to reclaim some of the financial expenditure to help reduce the overall economic burden of a material. For example, it is possible in some recycling procedures to recuperate some of the costs by down-cycling recyclate or energy recovery, even if the production of high performance recycled materials should be favoured [5,10]. In some cases, the financial savings and environmental benefits achieved over the life cycle of a functional unit may be offset by the costs, and environmental impact, accrued during material manufacture or use phases. The fully comprehensive account of economic and environmental impacts framed within the engineering requirement, should therefore be the most desirable for the holistic analysis of the complex impacts of composite materials.

3. Composite material life cycle inventory (LCI) phases

3.1 Material production

The initial phase of the material life cycle depicts the production of constituent fibre and matrix. For fibre and matrix manufacture, the conversion of raw material into a usable format can vary significantly in CED, due to the variation in CED of the unit processes involved. The production CED, GHG emissions and cost of common reinforcements and matrices for FRPs obtained in the literature survey are collated in Table 1. Both the literature and LCA databases report significantly broad range of values for environmental impact, cost, and CED, for the same constituents, although the magnitude of the differences varies in each case. The literature covered in this review use an equally broad range of values for the same constituents which makes comparative assessment problematic. The reasoning that led to the selection of these values is beyond the scope of this review. However, its aim is to highlight that the selection of the best representative value, and the subsequent comparison with alternative studies, should be approached with caution.

Individual countries usually rely on different energy source ratios, *i.e.* fossil fuels, natural gas, renewables and nuclear, for electrical power. For example, a significant portion of global CF production comes from Japan which has high average GHG emissions (484 gCO_{2e}/kWh) associated with energy demand contribution from grid electricity. Alternatively, Sweden has lower emissions per MJ of electricity produced as its electricity production is predominantly provided by renewable sources [42]. This has a significant effect on the accuracy of the final composite impacts and, in the authors opinion, is the most substantial cause of value discrepancy and required dutiful consideration at the early stages of a composite LCA.

Resource use, *i.e.* energy, water, and capital, vary depending on the infrastructure, technology, and methodology used; this range is broader in more common, widely available materials as the variety of production technologies increases. Resource depletion for raw material production can also vary as a result of economies of scale between companies of small and large scale production [32]. Multinational corporations typically have processes with optimised consumptions, *i.e.* iron and steel suppliers, which result in substantial energy savings over smaller competitors. This can make for a complex comparison of relatively nascent CFRP production techniques with metal production established over decades of process optimisation.

The PMB of CF is significantly greater than that of conventional materials; it is widely considered as the main deterrent to the adoption of CFRP, although this is predominantly cost based [14]. Reduction of PMB has been investigated using renewable fibre precursors, more efficient production techniques, and renewable energy sources [43]. Natural fibres (NF) and bio-derived synthetic fibres have been investigated extensively as potential replacements for CF; LCA can be a useful tool in determining if they are indeed a useful alternative [26,44]. Joshi *et al.* stated that NF production is of lower environmental impact across all categories [45]. Wötzel *et al* also reported a 45 % decrease in energy requirements for NF production over CF. However, this is in combination with an increase in water emissions due to the fertilizer use during cultivation [46]. The major drawback with NFs as CF alternatives is the substantially inferior specific mechanical performance. The fibre content required to get a competitive mechanical performance with GFRP is high [47], which makes CF equivalence unrealistic. This is another example of where an LCE approach has merit, the environmental benefits of using NF may be negated by the increased content required for an NFRP to compete, although attaining performance equivalence is doubtful.

Table 1. Table summarising EI, GHG and costs of common CFRP constituents found in the literature.

Material	CED	GHG	Cost
	MJ/kg	kg CO2eq/kg	£/kg
Steel	13-56 [10]		
	25.0-44.6 [48]	2.26-2.49 [48]	0.48-0.58 [49]
Recycled Steel	9.0-52 [10]		
Aluminium	197-298 [50]		
Fibre			
Virgin Carbon	171 [51]		
	183-286 [16,52]	-	-
	353 [53]	-	-
	478 [16,52,54]		
	198-595 [55]	-	-
	771 [56]		
	286-704 [57]	24.4-31.0 [49]	
	-	-	21.2-47.0 [58,59]
Glass	13.0-54.3 [32,60]	-	-
	45.6 [38]	2.50 [38]	
	48.3 [45]	2.04 [45]	-
			1.59-3.54 [49,58]
Aramid	222-245 [49]	16.4-18.2 [49]	21.2 [9]
Flax	6.50-11.6 [59]	0.45 [59]	1.57-3.14 [49]
Matrix			
Thermoset			
Epoxy	140-144 [45,61]		3.00-15.0 [62]
	76-80 [16,38]		
	76-137 [57]	4.7-8.10 [57]	
Polyester	63-78 [16,63]	2.8-3.10 [16,63]	1.00-2.00 [62]
Thermoplastic			
ABS	95 [45,64]	3.10	1.22
PVC	53-80 [65–67]	2.20 [32]	1.36 [32]
Polypropylene	22.4-112 [16,45,61]	1.85-2.60 [16,45,61]	1.23 [49,64]
Nylon	139-145 [61,68]	6.50-8.33 [61,68]	1.66-2.55 [49,64]
PC	80-112 [32,69]	6.00-7.50 [32,69]	1.82 [49,64]
LDPE	65-92 [32,70]	1.80 [67]	1.22 [66]

3.2 Manufacture

A variety of manufacturing procedures exist for CFRP manufacture therefore selection is driven by the design requirements of the application. The process CED and typical production volumes for the most common CFRP manufacturing processes can be found in Table 2; these data are typically quoted for the manufacturing process only and therefore do not include those required to produce constituent materials.

Generally, the largest proportion of manufacturing energy is spent in the application of the heat and pressure needed for matrix curing and/or fibre impregnation. Parameters such as manufacturing rate and component complexity are not considered in the calculations, yet they have consequences in processes down-stream that can lead to significant increases in environmental impact. For example, pultrusion is considered a low-energy process however it is limited to non-complex parts with simple cross-sections [32]. LCE may provide a solution to this issue as a part complexity and manufacturability parameter can be factored into the technical dimension of the assessment.

Table 2. Table summarising CED and production volumes of CFRP manufacturing processes. For production volumes Low < 5k ppa, Medium = 5k – 15k ppa and High = 15k – 100k.

Process	Process CED*	Production volume[1]
	MJ/kg	Parts per annum
Autoclave	21.9 [32]	Low
Spray up	14.9 [16]	
RTM (CF)	12.8 [16]	Medium
RTM (GF)	11.6 [71]	Medium
LRI/VARI	10.2 [16]	Medium
Cold press	11.8 [16]	High
Preform matched die	10.1 [16]	
SMC	3.5-3.8 [16]	High
Thermoplastic moulding	-	High
ATL	-	
ilament winding	2.70 [16]	Medium
Pultrusion	3.10 [16]	High
Injection moulding	19-29.9 [32]	High
Prepreg (CF UD)	40 [16]	Low
Comp. mould.	7.2-15.9 [14]	

[*] Process CED is equivalent to onsite energy, [†] Costs are approximated ranges, real values are highly material specific. RTM = Resin transfer moulding. LRI = liquid reactive injection. VARI = vacuum assisted reactive injection. ATP = Automated tape laying. Comp. mould. = Compression moulding

Production volumes and manufacturing times can also alter the suitability of a material for a given application. Simões *et al* reported a win-win scenario for the replacement of a single, stainless steel storage tank with a GFRP counterpart [39]. The LCA/LCC evaluation showed the composite part to have a lower environmental impact and part cost over the life cycle. However, when considering increased production volumes, the extensively optimised production infrastructure for stainless steel enables the production of far greater quantities in less time than the current manufacturing processes for composite materials can permit [15]. If the system boundary in the study undertaken by Simões *et al* was expanded to include a larger production volume, the impacts of the LCA and LCC may not favour production using GFRP.

3.3 Use

The use phase refers to the period of a component life cycle in which it is functioning in its predetermined application. For any application the associated CED, environmental impacts and costs of the use phase can be split into those incurred through general usage and those from maintenance activities. For example, the major metrics for evaluating the CED and emissions of vehicles are lifetime travel distance and fuel economy. Any maintenance/repair contributions are insignificant compared to fuel consumption [10]. This phase dominates the life cycle energy consumption of vehicles; contributing anywhere between 60 - 84 % of the total life cycle energy consumed, primarily due to impacts of vehicle weight, *i.e.* fuel economy [16,72]. Comparatively, the manufacturing phase contributes only 4 - 7 % of the total lifetime energy demand of a passenger vehicle produced in mild steel with the currently available technologies [73]. 75 % of fuel consumption is directly dependent on vehicle weight, mostly due to reduced powertrain demands, *e.g.* rolling resistance and acceleration and a 6 – 8 % increase in fuel economy can be realised with every 10 % weight reduction [74]. Therefore, the lightweighting benefits of CFRP are greater realised in the transport sector where use phases are the longest; the PMB may potentially be offset by the fuel consumption savings made during the use phase, providing it is of suitable length.

For components in passenger vehicles the use phase covers the lifetime of the vehicle, typically 10-20 years covering a range of distances, anywhere between 57,000 - 350,000 km, depending on geography and culture [10,73]. The United States Environmental Protection Agency (US EPA) and Canadian Standards Association (CSA) stipulated a 'useful lifetime distance' value of 240,000 km and 250,000 km for the average American and Canadian vehicle, respectively [73,75]. However, European studies have used an average of 150,000 km [5,9,53]; the selection of average lifetime distance is therefore important to clarify in breakeven comparisons.

The total net change in the use phase energy demand, ΔE_{Use_i}, of a lightweighting material, i, over a conventional baseline material, b, can be determined following 3.1 [75,76].

$$\Delta E_{Use_i} = C_i \times E_{FP} \times R_{WTW} \times \rho_F \qquad 3.1$$

where, E_{FP} is the energy required to produce 1 kg of petrol (MJ/kg), R_{WTW} is the conversion factor used to determine the energy demand of petrol from well-to-wheel (WTW). ρ_F is the density of petrol (kg/L) and C_i is the total mass-induced fuel savings which was calculated according to 3.2 [75,76].

$$C_i = (m_i - m_b) \times F_{CP} \times LTDD_v \qquad 3.2$$

where, m_i and m_b are the lightweight and baseline masses respectively. F_{CP} is the mass-induced fuel consumption change potential value, *i.e.* fuel (L) required to move 100 kg by 100km. $LTDD_v$ is the chosen lifetime driving distance (km). As $m_i < m_b$, negative fuel consumptions are returned from Equation 3.1 which represents a fuel saving compared to the baseline material with a fuel saving value of zero.

A useful tool in use phase comparisons is the determination of the breakeven distance which describes the minimum distance required for the energy savings accrued through use to offset the energy demand amassed during production. Recent literature shows that the breakeven point for virgin CFRP (vCFRP) automotive *versus* steel can be between 132,000-250,000 km, however this is entirely dependent on a number of factors including vehicle location, vehicle type, energy demand of fuel production, CFRP production CED and the primary energy source used during processing [9,65,77,78]. The mass-induced fuel consumption change potential value had a significant effect on the breakeven distances, for a 50 % substitution of steel to CFRP a change of 0.5 to 0.15 L/(100 km, 100kg) resulted in a 70 % increase in breakeven distance to 250,000 km [78]. Finally, Witik *et al.* reported similar findings when comparing rCFRP and vCFRP against a vGFRP component; the rCFRP component, reclaimed from pyrolysis, was able to break even at 41,000 km, compared to vCFRP which did not break even in the vehicle lifetime (200,000 km) [43].

Durability of a material can dramatically reduce its operational lifetime; therefore, durability is an important consideration when performing material selection. Some constituents can provide peak performance for longer durations than others, resulting in materials with shorter use-phases and higher maintenance/replacement costs for the same functional unit, this is especially true for NF. In some studies NF composites have exhibited lower cost, weight and environmental impact for functionally equivalent cases [79–81]. However, Corbiere-Nicollier *et al.* indicated that a NF composite pallet only remained environmentally favourable to a GFRP counterpart provided that a minimum lifetime of 3 years could be achieved [26]. Moreover, if the lifetime was shorter than 5 years, the GFRP equivalent would perform better due to necessity to use multiple NF pellets to meet the product function (ability to transport 1000 km per annum for 5 years) [26]. This is important as there is insufficient data for the fatigue and longevity of NF composites therefore there are no guarantees that the NF composite can provide the purported environmental benefits. Mechanical performance inequalities can also drastically alter the environmental impacts and costs of composites in an LCA. Dufluo *et al.* conducted a comparative study between flax fibre (FF) and GF reinforced polypropylene. The inferior mechanical performance of the FF composite limited its potential as a replacement material, despite FF offering lower global warming potential [82]. Equal strength equivalent material required a higher FF volume fraction which reduced the environmental savings to below what was required for it to be a feasible alternative. [82]

3.4 End of Life (EOL)

The final stage of an LCA evaluates how a material is processed when it reaches the end of the use phase. For steel and aluminium, this involves the melt reprocessing of EOL scrap which varies in yield but is commonly around 95 % [15,75]. The predominant EOL treatments for CFRP are landfill, incineration, and, more recently, recycling processes. These are typically mechanical grinding or fibre reclamation techniques. Currently, the two main routes for higher value constituent reclamation are *via* thermal processes (pyrolysis [83], micro-wave pyrolysis [84], fluidised bed pyrolysis [85]) and chemical process (solvolysis [59], acid digestion [86], super-critical fluid solvolysis [87]); these vary in production from commercial to lab scale operation. Reclaimed fibres are then remanufactured into new feedstock material through various potential approaches as described by Pimenta & Pinho [19] and Oliveux *et al* [36]. The availability of commercial scale remanufacturing techniques is currently limited, resulting in a negligible market for rCF. The vast majority of LCA applied to CFRP recycling are only capturing fibre reclamation and do not incorporate a realistic remanufacturing step.

Landfill has been the most common disposal route for CFRP material as it is traditionally the most economical and can easily handle large waste quantities [88]. Incineration has been an alternative to landfilling in many LCA studies as there is the potential for energy recovery. Simões *et al.* showed that the LCA performance of a GFRP storage tank could be improved by selecting incineration with energy recovery as its EOL treatment over landfilling [39]. This can reduce the overall CED, despite the higher process emissions compared with landfill, as the energy recovered is deducted from the total CED [9,43,50,53]. However, the significance of the impact is determined by the scope of study, *i.e.* cost or emissions driven application, therefore the benefits are subjective. Witik *et al.* reported that the CO_2 emissions avoided by the reuse of recovered energy were not sufficient to offset those produced during incineration, making landfilling the most appropriate from an emissions perspective [43]. The problem with landfill and incineration is that most or even all the embodied energy of the CFRP is lost. Recycling is an EOL alternative that has the potential to provide recyclate of value typically through down-cycling into separate product systems; this could be as filler, or as a low-performance reinforcement. For FRPs, in general, mechanical grinding is an alternative to landfilling as it has a lower energy demand and can provide some recyclate value as a filler material, however this is a fraction of the original virgin material cost and is rarely economically viable [89,90].

Pyrolysis can produce recycled CF (rCF) as a high value recyclate, at a commercial scale (over 1,000 tonnes/year) [91] and is reported as having a much lower energy demand than virgin CF (vCF) production [9,15,65,92]. Pyrolysis is viewed as a superior alternative to landfill and incineration because rCF can be reused, reducing the total life cycle CED despite pyrolysis having far greater process CED and emissions. The environmental impacts of landfilling, incineration and reclamation processes have previously been contrasted in the literature and the conclusions of each vary depending on energy source [25,43,90]. A summary of the CED, emissions and costs of state-of-the-art fibre reclamation processes for CFRP can be found in Table 3.

Table 3. Energy intensity, environmental impact and recyclate value estimates for CFRP recycling technologies.

Process	Process CED	GHG
	MJ/kg	kg CO2eq/kg
Landfilling	0.11-0.4 [93,94]	0.09-4.61 [34,43]
Incineration	32-34 * [43,53]	2.17-3.05 [94]
Incineration (energy recovery)	(-)31.7 to (-)34 † [93]	2.01-3.4 [93,94]
Mechanical grinding	0.14-51 [95,96]	-36 [97]
FB Pyrolysis	7.7-30 [65]	5.4-11 [43,97]
Microwave Pyrolysis	-	-
Pyrolysis	2.8-30 [43,93]	-
High voltage fragmentation	4 [90]	-
Solvolysis	15-64 [98,99]	-
Steel recycling	11.7-19.2 [93]	0.5-1.2 [93]
Aluminium	2.4-5.0 [93]	0.3-0.6 [93]

* Based on CFRP epoxy component bond energies, † Lit value based on CFRP epoxy calorific content with unknown recovery efficiency. FB = Fluidised bed.

3.4.1 EOL recycling allocation

In LCA the EOL stage CED, emissions, and costs are typically a summation of the EOL operations required to dispose of, or, to convert the component into a reusable material; such as disassembly, size reduction, transport, recycling and landfill. The reuse of material results in a reduction in the amount of production energy required, how this reduction is accounted for, or *allocated*, can produce substantial variation in results. Recycling credits can be assigned based upon *recycled content* or through EOL approaches, in which there are subtle difference categorised as *Cut-Off*, *Closed-loop* and *Substitution* [100]. There are subtle differences in the EOL subcategories; Nicholson *et al.* compared the impact of each EOL method on the validity of two materials in a comparative LCA. It concluded that the differences in EOL approach are insignificant until the PMB of a material is far greater than an alternative [101]. The recycled content approach assigns credits for any secondary material in the total virgin production energy value [100]. For example, the reuse of manufacturing waste in the same process will be accounted for by a reduction in the total virgin production energy equal to that of the reused material. It lends itself well to the traditional linear production processes in that it doesn't account for material that may be used in other product streams, *i.e.* down-cycling.

For closed-loop product systems, where there are no changes in the inherent properties of the recycled material, such as steel and aluminium production, the EOL approach is best suited. The EOL approach assumes that there is a material *pool* available where any unrecycled material is compensated by primary material; recycling therefore offsets primary production by the given recycling factor for the material. The recycling factor is determined by balancing the scrap output with scrap input to avoid double counting. The total production CED for a metal is therefore the primary production CED multiplied by the recycling factor, generally 0.16 for steel and 0.72 for aluminium, which accounts for the proportion sourced from recycled scrap [14,75,76]. This results in a reduced production stage CED and an EOL stage contribution from the recycling process CED. Closed-loop recycling of metals has been practiced for decades so the LCA framework to evaluate the EOL allocation is well defined. Closed-loop CFRP recycling research is in its infancy, therefore detailed LCA frameworks are limited. The standard Closed-Loop approach is not applicable as the recycled material is not directly reusable and has reduced mechanical performance. LCA evaluations that include CFRP approach this in different ways depending on the goal and scope of the study.

If the study compares the environmental impacts of an EOL operation, vCFRP has an EOL stage contribution for landfilling, incineration or pyrolysis and solvolysis [43,78,94,102–104]. The energy recovery credits from incineration are counted as a negative burden and cause a reduction in life cycle CED. The CED of pyrolysis and solvolysis methods result in an increased life cycle CED however the GWP benefits, and economic cost reductions due to recyclate resale, can provide overall life cycle improvements. If the study is comparing the total life cycle environmental profile of a rCFRP over vCFRP, and other lightweighting materials, this is approached in a similar manner to the EOL approach. In the literature, it is usually presented in two distinct ways:

1. The rCFRP, made from reclaimed rCF, is treated as a separate material option and compared with alternatives. The production CED does not include the raw material energy required for vCF but accounts for the pyrolysis CED in the EOL stage [65,70,76].
2. The vCFRP is evaluated over multiple life cycles, where the EOL stage of the first life cycle accounts for the CED required for recycling. The production stage of the second life cycle includes a reduction for the CF sourced from the previous recycling operation [16,105]. This is closer to the Cut-Off subcategory of the EOL approach found in the literature [106].

For CFRP, the recycled content approach may provide scenarios that better reflect the current recycling market for rCFRP which has no large-scale market of the rCF produced. An EOL approach will be the best suited for future scenarios where an appropriate recycling infrastructure is in place for large-scale production and recycling of CFRP [73]. Suzuki and Takahashi followed the Cut-Off EOL approach and suggested a 3R (reduce, recycle, and reuse) system for LCA evaluation of recycled CFRP [16]. The 3R system enabled three use phases for an automotive component which provided the lifetime savings required to make CFRP a superior lightweighting alternative to steel. However, this study only provides a model for a 3R product system and does not offer any real industrially applicable technologies for how the CFRTS/TP material will be reclaimed and remanufactured. The central problem that the industry faces for rCF remanufacture is that the current rCFs are produced in a filamentised, randomly oriented, and low-density-packing form [19]. As well as being fragmented into shorter lengths due to waste size reduction [107]. Without significant alignment, high fibre volume fractions are difficult to achieve and remanufacture typically results in rCFRP with significant mechanical performance reductions [108].

Until recently LCA has not accounted for the economic influences or technical feasibility of the real world recycling market which play a vital role in passenger vehicle production [109]. Without a high value application for the rCF recyclate the purported financial and environmental savings cannot be achieved [19]. A recent LCA by Meng *et al.* evaluated the environmental impacts of rCFRP material and compared them with steel and aluminium using the recycled content approach. The rCFRP materials were made by a range of remanufacturing techniques, notably compression moulding aligned rCF fibre mats and virgin epoxy matrix. The highly aligned rCFRP component (60 % Vf_F) offered the lowest life cycle energy demand and GHG of all materials, with a 94 % reduction compared with steel alone. It stated that vCFRP becomes favourable to steel at a lifetime driving distance > 250,000 km, but that rCFRP can offset production CED at distances of < 50,000 km [65].

4. Interpretation and discussion

The current PMB of virgin CFRP, as widely reported in LCA databases and the literature, remains significantly high despite the reduction efforts of recent years. There are a variety of different data sources available when selecting the production process for a CFRP product system and care should be taken when choosing the most appropriate representation. In addition to the environmental burdens the financial cost of fibre production is in constant flux. The reduction in CF cost is a heavily researched area that is slowly making improvements; however, the cost of CFRP remains far from competitive. From an LCA perspective the recuperation of energy can help to reduce total energy demand and environmental impact, however this may only be a fraction of the PMB and does not reclaim any of the value of the constituent materials. As environmental impact is rarely as influential as cost for a material selection metric in the design phase, the reclamation of high value fibre and matrix is equally as important as reclaiming energy for many industries. The variation in source data for both CED values and others used in calculations, *i.e.* lifetime travel distance, fuel-consumption change potential, can result in significant variance in the final life cycle values. Most studies have applied a sensitivity analysis on the LCA results to account for the substantial variation in input data, reporting the variation in the final results obtained [9,15,43,65,70,109,110].

Fig. 2. Breakeven envelope plot showing sensitivity analysis of life cycle energy demand. The envelope represents the potential impact values based on the range of values available in databases and the literature.

The variations in the literature PMB, EOL and total lifetime distance values can cause substantially different results in LCA. The significance of these are explored in the Fig. 2 sensitivity analysis. It shows the range of potential life cycle CED values for a component from using the range of input data available in the literature. The functional unit is a 10 kg CFRP component made from woven epoxy prepregs (60 % Vf_F). It offers a 60 % weight saving over a steel baseline for a vehicle lifetime, *i.e.* PMB through Use to EOL. The weight saving was calculated using the mass ratio expression in Equation 4.1 [111]:

$$R_{mass} = (\rho_C/\rho_S) \times (E_S/E_C)^{1/\lambda} \quad 4.1$$

where, ρ is the density, E is the stiffness and the subscripts C and S refer to CFRP and steel, respectively. λ is a structural index which can be used to predict weight savings from material substitution when the part stiffness is assumed constant [111]. It expresses the response of the design to a change in thickness afforded using lightweighting materials and is effectively a measure of thickness-dependent non-linearities in stress distribution in a thin-walled structure [111]. λ can be anywhere between 1 and 3 however most automotive components are between 1 and 2 therefore an average value of 1.5 was selected. A CFRP stiffness and density of 69 GPa and 1.5 g/cm^3 and a steel stiffness and density of 210 GPa and 7.85 g/cm^3 were applied to produce a mass ratio of 0.4 and thus a 60 % weight saving.

The envelope is constructed using the maximum, minimum, and average PMB and EOL values found in the literature, these are displayed in Table 1, Table 2, and Table 3. PMB and EOL values include a reduction for the replaced steel burden. Maximum, average and minimum values for steel production were used with a reduction factor of 0.16 applied following the Closed-Loop EOL allocation approach assuming virgin steel had an 84 % recycled steel content. Maximum values are combined for the upper envelope boundary and the minimum values are combined for the lower boundary to create the life cycle CED envelope. The CSA lifetime driving distance of 250,000 km was selected however the US EPA (240,000 km) and average literature value (150,000 km) were provided for sensitivity analysis. Equations

3.1 and 3.2 were used to determine the mass-induced fuel savings, where, E_{FP}, ρ_F and F_{CP} were 43.5 MJ/kg, 0.74 kg/L and 0.16 L/(100km, 100kg) respectively. R_{WTW} can vary depending on geography therefore a maximum (1.47 MJ/MJ), minimum (1.05 MJ/MJ) and average (1.36 MJ/MJ) value was applied to the relative boundary combination using the values obtained from Sweden, Europe, and other international sources [112], respectively.

EOL allocation for CFRP followed the recycled content approach and assumes that the fibre weight fraction (0.67) are reclaimed by pyrolysis and the epoxy weight fraction (0.33) is treated as landfilled as this is where the resultant char would likely be disposed of; there is no application of energy recovery as this is not common practice, although some pyrolytic processes have utilised it in the literature [113]. As the rCF is downcycled into other product streams no recycling credits are applied in the CFRP PMB. The maximum EOL burden is used in combination with the maximum PMB for the envelope, however in reality the EOL burden magnitude is independent from that chosen for PMB. The plot highlights the large range of possibilities for lifetime energy demand of a vehicle based on the significant variation in literature values. The maximum and average PMB do not breakeven within any lifetime distance. The minimum PMB results in a breakeven distance within the US EPA and CSA vehicle lifetime distances providing net energy savings after 180,000 km. Net fuel savings are also ensured after application of the minimum EOL burden which does not intercept again.

Both scenarios are likely widely misrepresentative of a real CFRP lifetime energy summation. The central dashed line represents the average burden determined using the literature range and may be considered a more accurate depiction of the burdens observed in a real CFRP part. The use phase savings are almost enough to breakeven on the PMB and, when combined with the EOL burden, a second use phase is required to breakeven. No matter the data source or LCI employed the energy demand of CFRP production and manufacture is significant (165-595 MJ/kg). The use phase offers the greatest potential for CFRP emissions savings therefore providing multiple use phases will enable enhanced reductions that could breakeven or possibly provide net environmental savings gains over vCFRP.

It is increasingly apparent that the best-case scenario for rCFRP is that it can be remanufactured into high-value applications to provide additional use phase savings. The tandem breakeven plot in Fig. 3 illustrates how an additional use phase can provide net energy savings in the life cycle of a CFRP material.

Fig. 3. Tandem breakeven plot showing the lifecycle energy demand of a CFRP material with two use phases i.e. virgin and recycled. Use phase fuel savings are compared against a steel baseline. The second use phase assumes rCFRP performance equivalency with virgin CFRP to represent potential gains.

The plot is based upon the same functional unit and use phase calculations as Fig. 2 however only the average PMB values are used. The CFRP part has two use phases, virgin and recycled, after the second use phase the component is disposed of *via* landfill. The initial PMB CED includes primary production and manufacture with a deduction for the steel PMB. The EOL burden applied is the same used in Fig. 2. The second PMB CED does not include a CF contribution, following the Closed-Loop allocation approach; only the CED for the weight fraction of epoxy and the part manufacturing CED is applied.

The PMB is not offset within any of the lifetime driving distances in the first use phase. The part breakeven distance is within the second use phase, at 115,000 km even after paying for the pyrolysis of the fibres, landfill of the epoxy, steel production and CFRP remanufacture. To simplify the construction and interpretation of the graph, the recycled material is assumed to have the same mechanical performances, and therefore the component has the same function, as the virgin predecessor.

The tandem plot is a useful method for displaying the potential benefit of CFRP closed-loop recycling from an LCA perspective. Despite the use of averaged PMB, EOL, lifetime distances and other constants the conclusions drawn from the plot are valid for a realistic component. However, the suggested benefits are only attainable with the application of a recycling process that can reclaim and remanufacture the virgin constituents into a recycled component of equivalent performance, this has already been proved possible on a lab scale [114,115].

5. Conclusions

CFRP has become a popular material for many industries based principally on its potential for weight savings over conventional materials. Despite the prospective benefits there are considerable financial cost and environmental impact burdens associated with CFRP production that impede its utilisation in some applications. LCA is a useful tool for comparative studies of lightweighting materials, the following conclusions are apparent form the literature:

- LCA can provide valuable information for material selection, design and optimisation by generating insight into environmental ‘pinch-points’, savings opportunities and trade-offs.
- It is important to develop a standardised LCA framework for material selection for automotive materials so that LCA’s conducted in different institutions are not isolated in their conclusions but can be combined easily for a broader understanding of the true applicability of CFRP.
- LCA should not be considered as an isolated assessment for material selection involving CFRP as there a variety of additional design aspects that, without consideration, will provide a widely inaccurate evaluation and impractical conclusion. The increased capital costs, part complexities, and moulding

capability of composites manufacture, compared to conventional metals, are aspects that must be considered, reinforcing the importance of the LCE holistic approach.

- The variance in PMB has shown to have a dramatic effect on the breakeven point for CFRP material which will be a deciding factor in its adoption.
- The use phase of most vehicles has a substantial emissions profile and a longevity enough for CFRP to provide emissions reduction through weight savings, however in several applications this is not enough to offset the impact of production. The vehicle lifetime duration is also susceptible to significant variance, depending on the geographical region taken into consideration in the literature, which makes comparison difficult.
- Recycling and reuse technologies have the potential to provide valuable recyclate at industrial scales. However, without appropriate remanufacturing technologies able to deliver high performance recycled material, the maximum emissions reductions, and financial savings, from multiple use phases, may not be attained. It is apparent that emerging research effort should concentrate on recycling technologies that can reclaim high quality constituents and remanufacture them into high performance materials; ideally with the goal of providing virgin CFRP performance equivalency.

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All data required for reproducibility are provided within the paper.

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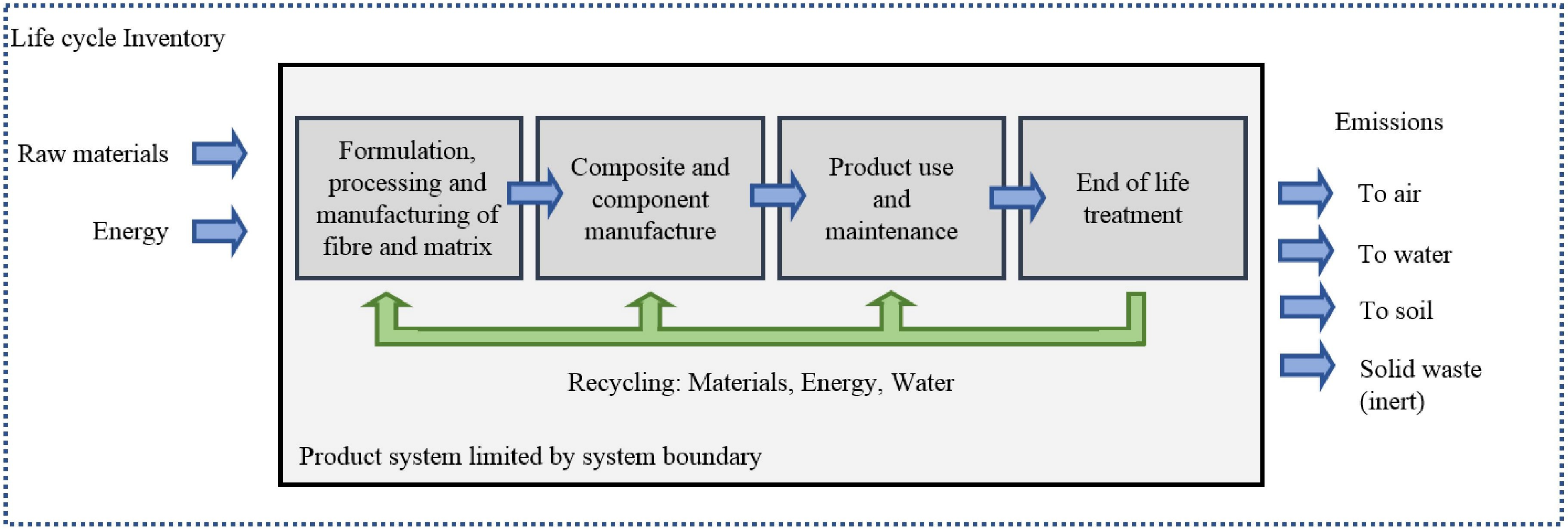
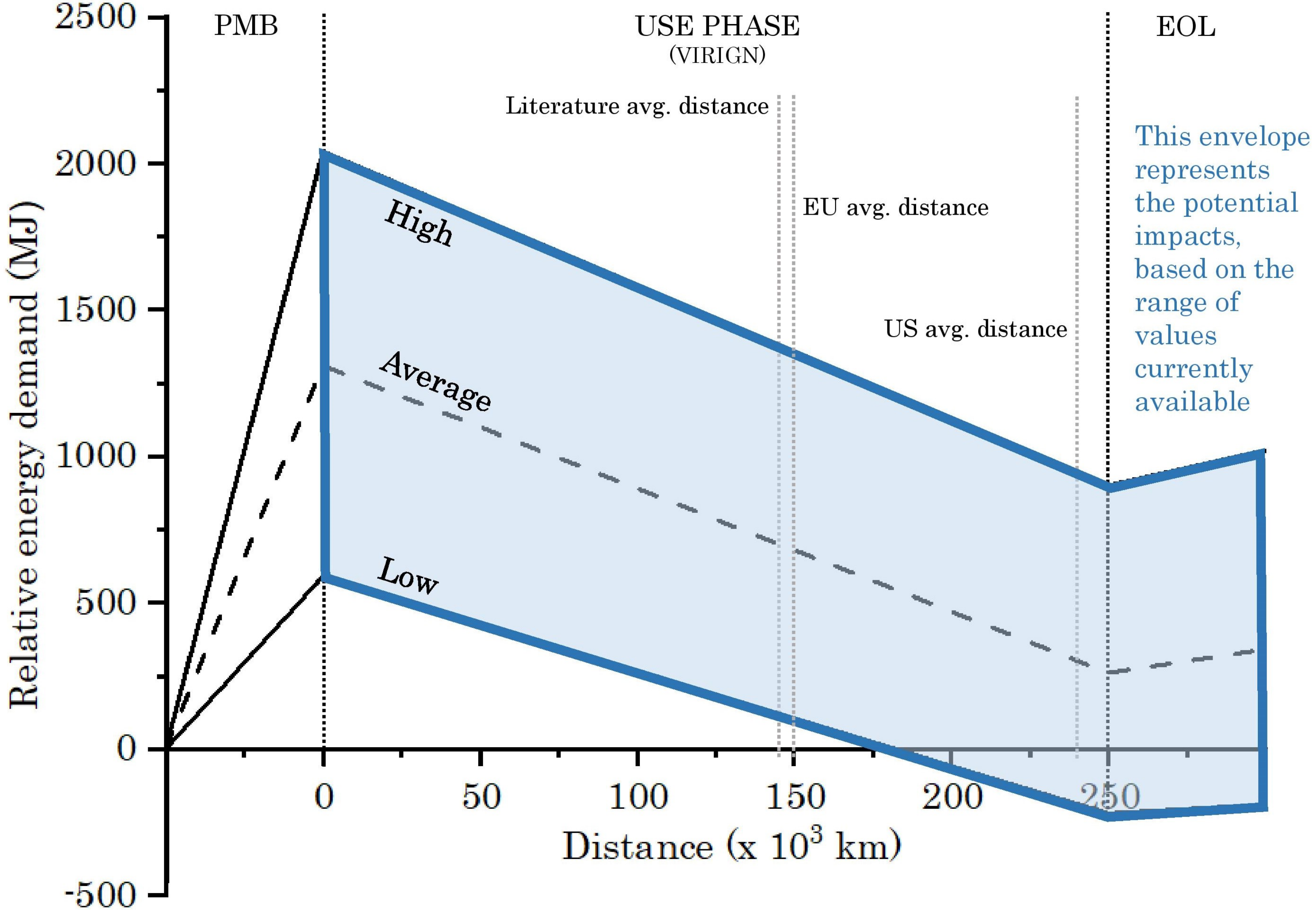
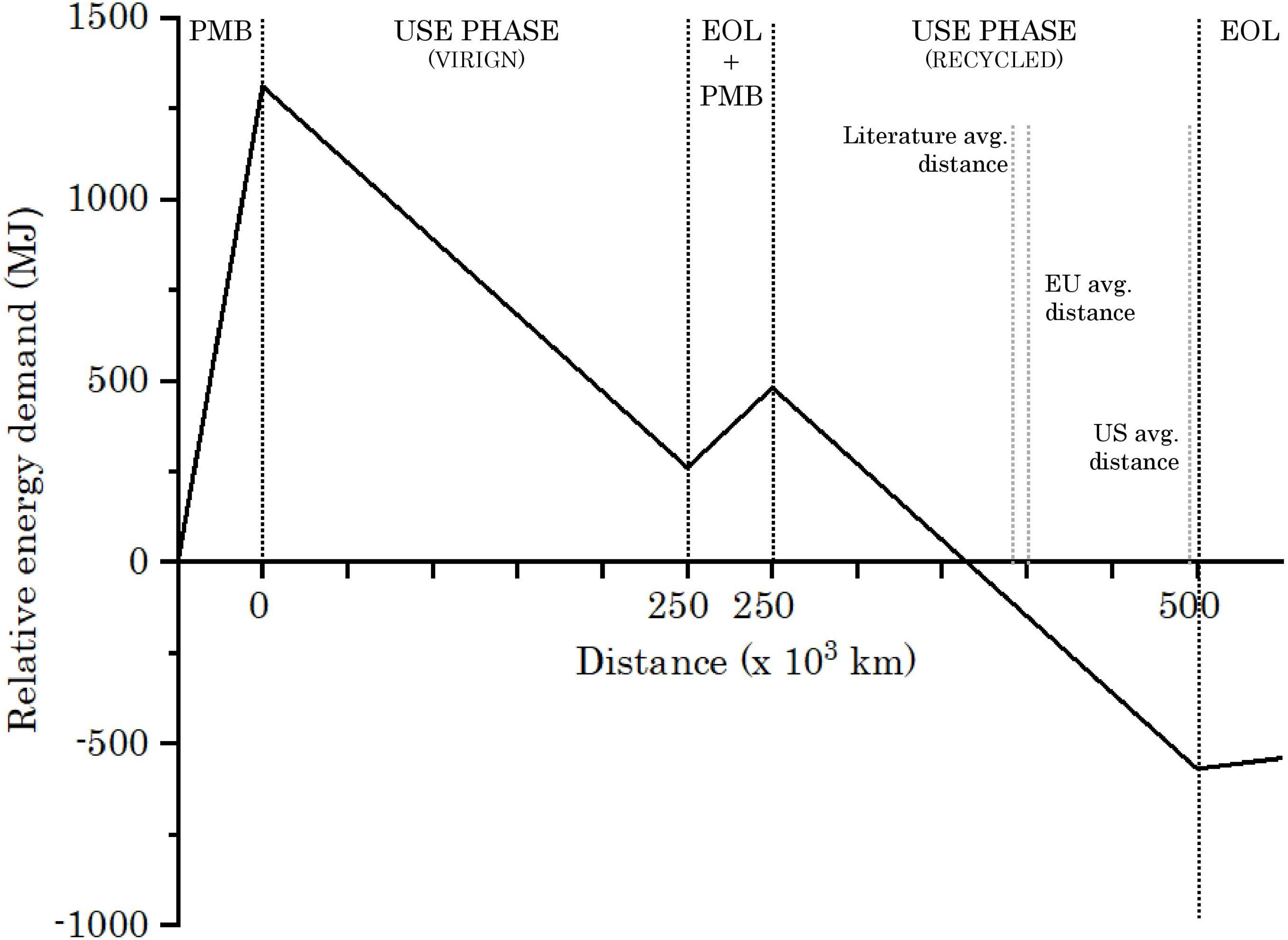

Life cycle Inventory
Raw materials
Energy
Formulation, processing and manufacturing of fibre and matrix
Composite and component manufacture
Product use and maintenance
End of life treatment
Recycling: Materials, Energy, Water
Product system limited by system boundary
Emissions
To air
To water
To soil
Solid waste (inert)


Relative energy demand (MJ)
2500
2000
1500
1000
500
0
-500
PMB
USE PHASE
(VIRIGN)
EOL
Literature avg. distance
EU avg. distance
US avg. distance
High
Average
Low
This envelope represents the potential impacts, based on the range of values currently available
0
50
100
150
200
250
Distance (x 10^3 km)



PMB
USE PHASE
(VIRIGN)
EOL
+
PMB
USE PHASE
(RECYCLED)
EOL
Literature avg. distance
EU avg. distance
US avg. distance
1500
1000
500
0
-500
-1000
Relative energy demand (MJ)
0
250
250
500
Distance (x 10^3 km)