A comparative life cycle assessment of tyre recycling using pyrolysis compared to conventional end-of-life pathways

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1 Abstract

End-of-life tyres (ELTs) are difficult to recycle due to their complex composition. Although there are several possible pathways to manage ELTs, in Germany roughly 50 % of the ELTs are still incinerated. One emerging technology that promises recycling of ELTs is the thermochemical processing through pyrolysis. This technology enables to recover carbon black and pyrolysis oil that can be reused for new tyres. Therefore, this study presents a comprehensive life cycle assessment to compare the environmental impacts from pyrolysis of ELTs against current dominant alternative end-of-life treatment pathways. The investigated alternative end-of-life pathways are (i) incineration in a cement plant, (ii) incineration in a dedicated incineration plant, and (iii) production of infills for artificial turfs. The results show that the recycling of ELTs for material recovery has lower environmental impacts in the categories global warming, particulate matter and resource use compared to end-of-life pathways that target energy recovery.

Keywords: Keywords: End-of-life Tyres (ELTs), Life cycle assessment (LCA), Pyrolysis, Infills, Incineration, Carbon black

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2 Introduction

End-of-Life tyres (ELTs) is a global issue and presents a significant environmental challenge. Although used tyres can be retreated and reused to a certain extent, this however is not feasible indefinitely, and sooner or later used tyres must reach their End-of-Life (EoL). Various synthetic and natural rubbers used in tyres, include polybutadiene, styrene-butadiene rubber (SBR), and (halo)butyl rubber, and can vary depending on the tyre type (Abbas-Abadi et al., 2022). Besides rubber, carbon black and silica are key components that contribute to mechanical integrity and performance of the tyres. Finally, textiles and metal add to the make-up of a tyre, wherein the commonly used textiles are rayon, nylon and polyester while the commonly used metal wires and belts are carbon steel and steel alloys (Evans and Evans, 2006). The difficulty in processing ELTs for recycling rises from this complex composition. ELTs can broadly be classified into three main categories – passenger vehicles, commercial trucks and off-the-road vehicles such as mining, farming, aircraft (Bulai, 2022). Depending on the type of tyre and technical requirements the composition of new tyres varies (Dong et al., 2021). Figure 1 shows a typical composition of the European car and truck tyres (Grammelis et al., 2021).

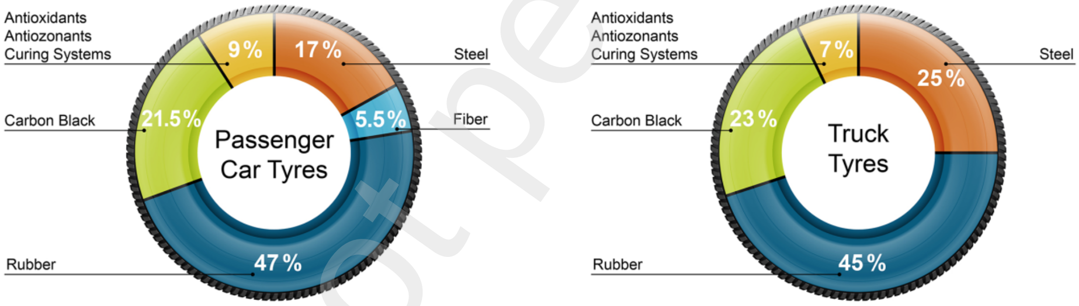

Figure 1: A typical car and truck tyre composition

With the amount of waste tyres rising each year and currently standing at 1.5 billion worldwide (Xiao et al., 2022), there is now a significant amount stockpiled or disposed of in landfills (Bulai, 2022). It is estimated that approximately 4 billion ELTs are currently in stockpiles and landfills worldwide (WBCSD, 2008), and this will rise to 5 billion by 2030 (Pacheco-Torgal et al., 2012). In Europe alone, ELTs stood at 3.26 million tons in 2019 (ETRMA, 2019). Most of these used tyres are either retreated, recycled, incinerated for energy recovery, or treated using pyrolysis for energy and material recovery (Valentini and Pegoretti, 2022, Xiao et al., 2022). According to the European Tyre & Rubber Manufacturers Association (ETRMA) in Europe 95 % of used tyres were collected and processed for material and energy recovery in 2019. Of which, recycling accounted for approximately 52%, while the remaining 48% were incinerated for energy recovery.

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Since ELTs are not listed under the Basel Convention, which oversees hazardous wastes and their disposal, they are traded freely and mostly end up in developing countries. Besides being a potent fire hazard, the toxic leachates from tyres discarded in landfills are found to contaminate waterbodies and pose a risk to both human health and the environment (Downard et al., 2015, Singh et al., 2015). Furthermore, being a stable component, rubbers in tyres take hundreds of years to degrade in landfills. Stockpiling of tyres have also been attributed to be a source for insects, pest and rodents that incubate and carry fatal diseases (Fiksel et al., 2011, Rubio et al., 2011). Finally, controlled incineration of ELTs is also not a sustainable option considering the sheer volume of ELTs worldwide. Through incineration the energy content in ELTs can be recovered, however, to avoid harmful atmospheric pollutants such as dioxins, NO_x, SO_x, dibenzofurans etc., incineration needs to occur at high temperatures and requires expensive downstream units to meet air quality specifications set by local and regional bodies (Valentini and Pegoretti, 2022). Furthermore, the amount of energy recovered from the incineration of ELTs amounts only to a fraction of the energy required in the production of a new tyre. A recent study shows that the energy recovered and the CO_2 emissions from the incineration of one tonne ELTs is 32 GJ/tonne and 2270 kg CO_2-eq./tonne respectively. In comparison, the energy required to produce one tonne new tyre is around 209 GJ/tonne (Valentini and Pegoretti, 2022).

Besides the recovery and reuse of metals, only a negligible proportion of other materials (e.g., recovered carbon black from Pyrum) are recycled on a commercial scale for new tyre production today. One key factor that effects the recyclability is the presence of impurities in reclaimed rubber that affects the overall performance of the recovered materials (Xiao et al., 2022). In addition, the complex crosslinked structure of vulcanized rubbers and the presence of heavy metals complicates the mechanical recycling process (Mohajerani et al., 2020). While there are some well-established technologies, such as cryogenic grinding to produce cleaner pulverized rubber without surface oxidation (Fazli and Rodrigue, 2020), and devulcanisation followed by revulcanisation to create the highest quality recycled materials (Valentini and Pegoretti, 2022); the majority of current ELT treatment methods still fall under a cascading use (mainly downcycling) than a closed-loop recycling. Most common cascading use applications for ELTs today include asphalt mix with crumb rubber in road construction, railway sleepers, and infills for artificial turfs. Other applications for recycled rubber granules include moulded rubber products such as wheels for caddies, dustbins, wheelbarrows and lawnmowers, urban furniture and sign posts (ETRMA, 2019). Recycled rubber granules and powder are also used to produce floorings for playgrounds, athletic tracks, shock absorbing mats, paving blocks or tiles for patios, swimming pool surrounds and roofing materials.

It should be noted that, besides material recovery, one emerging route is the thermochemical processing of ELTs through pyrolysis that produces a broad spectrum of intermediates, i.e., pyrolysis oil, pyrolysis gas and pyrolysis char that can be used in several downstream applications (Chen et al., 2022). The main objective of this research is to conduct a comprehensive life cycle assessment of one such patented technology developed by Pyrum Innovations® AG (Schulz, 2010) and compare the

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environmental impacts of pyrolysis of ELTs against current dominant alternative EoL treatment pathways. The alternative EoL pathways investigated are (i) incineration in a cement plant, (ii) incineration in a dedicated incineration plant, and (iii) production of infills for artificial turfs.

2 Methodology for the Life Cycle Assessment

The life cycle assessment (LCA) methodology applied in this study was structured and conducted according to the ISO 14040:2006 and ISO 14044:2006 standards (ISO, 2006a, ISO, 2006b). The LCA was modelled using the commercial software LCA for Experts (GaBi) version 10.7 and follows an attributional approach.

2.1 Goal and scope

The main objective of this study is to assess and analyse the environmental impacts generated from various EoL handling and processing pathways of scrap tyres. The investigated scenarios are presented in Table , wherein Scenario 1 evaluates the pyrolysis-based recycling technology. Scenario 2 explores the energetic treatment of ELTs in a cement plant. Scenario 3 analyses the use of ELTs in a dedicated incineration plant. Finally, Scenario 4 deals with the treatment of ELTs to produce infills to be used in artificial turfs.

Table 1: Investigated EoL scenarios for ELTs

Scenarios	Process	Final product from EoL treatment
1	Pyrolysis	Metal scrap, carbon black, pyrolysis oil, energy
2	Energy recovery in cement plant	Metal scrap, energy
3	Energy recovery in a dedicated incineration plant	Metal scrap, energy
4	Production of infill	Metal scrap, infill, energy

The investigated pyrolysis technology patented by Pyrum Innovations® AG (Schulz, 2010) is a multi-stage, energy self-sufficient, and continuously operating system. It is designed for the fractionated recovery of valuable substances and energy from pourable, cross-linked organic compounds of high molecular weights, specifically focusing on granules derived from end-of-life tires (ELTs). During operation, granules from ELTs gravimetrically pass through a vertical, multi-stage pyrolysis reactor from the top to the bottom, wherein said granulates are pyrolyzed at temperatures between 500 and 700ºC. A subsequent two staged fractionated condensation of the pyrolysis vapours allows the recovery of oil and gas compounds, and the use of pyrolysis gas in a combined heat and power plant (CHP) provides the electric power required for heating the pyrolysis unit.

Although the pyrolysis-based recycling technology is already commercially realized by Pyrum, it must however be noted that the other investigated EoL treatment options for ELTs have been established

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for decades. In addition, a further goal of this paper was to identify hotspots of the pyrolysis-based recycling technology for future optimisation. Although there are other pathways to treat ELTs such as the use of shredded tyres in steelworks equipped with electric arc furnaces to substitute anthracite and scrap metal, or to produce rubber modified asphalt, these alternative applications only play a minor role in Germany. In addition, truck tyres, in particular are retreaded to prolong their lifetime. This gives the retreaded tyres a new cycle of life. However, after second or third use (depending on the quality of the carcass), the tyre eventually needs to be disposed. Consequently, retreading is not regarded as a viable EoL option in the long run.

2.1.1 *Functional unit*

The functional unit (FU) for this study is defined as "*the treatment of 1 tonne ELTs*". The selection of this functional unit allows for comparison of different EoL treatment scenarios.

2.1.2 *Geographical and technological scope*

The geographical scope of the present study is Germany since the main technology under investigation currently is commercialised in Germany.

2.1.3 *System boundaries*

The LCA follows an attributional approach focusing on the EoL stage. It considers the environmental impacts derived from (1) the collection and transport of the ELTs to a treatment plant, (2) pre-treatment operations, (3) respective EoL treatment, and (4) substitutes for EoL products (see Figure 2).

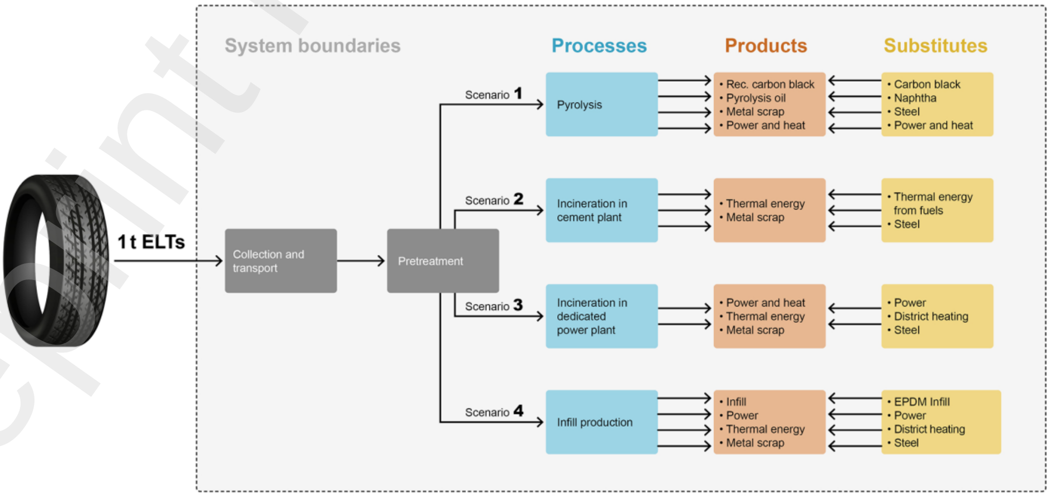

Figure 2: System boundary of the investigated scenarios

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2.1.4 Data collection and data quality

Primary data was collected for the pyrolysis process whereas data for the alternative EoL scenarios was taken from literature. The background data used for conducting the LCA models was based on the 2023.1 of Managed LCA Content database (Sphera Solutions GmbH, 2023) and for few cases where no representative data was found in the Sphera database, suitable datasets from ecoinvent database (ecoinvent v. 3.9) were used.

2.1.5 Multifunctionality

The issue of multifunctionality occurs when a system produces multiple product outputs or uses inputs that originate from another product life cycle. In this study, ELTs are the main input for all EoL treatment scenarios. As a waste, tt was assumed that the ELTs do not have any environmental burdens since all burdens are allocated to the tyre production itself. In the case of recycling or if ELTs are thermally treated, the system was expanded, and credits were given for replacing materials or energy. This approach is also referred to as "avoided burdens approach" and is often used in LCA studies of waste management systems (Heijungs and Guinée, 2007). The choice of this approach allows for a comparison between different EoL treatment options and therefore fits to the purpose of this study. The weak point of this avoided burdens approach, however, is that the number of "what-if" assumptions regarding the choice of substitution process can be so large that LCAs on the same topic lead to quite diverging results (Heijungs and Guinée, 2007). To address this weak point, sensitivity analysis will be conducted to assess the extent to which credits predominantly influence the LCA results.

2.1.6 Sensitivities

The key product of the pyrolysis technology under investigation is a carbon black substitute. Although the quality is not identical to that of virgin material, the product can substitute carbon black types N550 or N660 to a certain degree. Remaining quality challenges are the ash content and carbonaceous residues. In addition, carbon black can be produced through various technologies including the furnace black process, gas black process, channel black process, lamp black process, thermal black process and acetylene black process (European Commission, 2007). However, globally, approximately 95 % of the carbon black was produced in a furnace black reactor (European Commission, 2007), which is based on an incomplete feed combustion. The reason behind the extensive use of the furnace black process is that it allows producing nearly all carbon black grades required by the rubber industry. The furnace black process uses petrochemical oils, coal, tar oils and natural gas as feedstocks to produce carbon black. Depending on the feedstock, operation conditions, yields, and product specifications the environmental footprint varies. This study addresses the associated uncertainty by considering minimal and maximal footprint values.

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In the case of using ELTs as refused derived fuels in cement plants, the crucial question arises regarding the specific fuels that would be substituted in the cement plant. To address this issue, in addition to the German situation (base case), the European situation was investigated additionally.

Finally, it is important to note that the composition of ELTs (rubber composition, rubber content, iron content etc.) can impact the LCA results. This aspect will be discussed qualitatively.

2.1.7 Impact assessment methodology

The life cycle impact assessment (LCIA) methods chosen and applied in this study is based on the recommendations published by the International Reference Life Cycle Data System (ILCD) and the Product Environmental Footprint (PEF) method developed by the European Commission (EC) (European Commission, 2013, Zampori and Pant R, 2019). The selected impact categories are acidification (A), global warming (GW_{total}), freshwater, marine and terrestrial eutrophication (EU_F, EU_M, EU_T), ionising radiation (IR), land use (LU), ozone depletion (OD), particulate matter (PM), photochemical ozone formation (POF), resource use for energy carriers (RU_E), resource use for minerals and metals (RU_M), and finally water use (WU). The chosen impact category methods and their respective units are provided in the supplementary section (see Table S1). Human toxicity and ecotoxicity were not considered due to high uncertainties in both the life cycle impact assessment (LCIA) methods and data gaps in the life cycle inventory data.

3 Systems under study and life cycle inventories

3.1 Treatment of ELTs by pyrolysis

Figure 3 depicts the system boundary for the treatment of ELTs by pyrolysis. The process starts with the collection and transport of ELTs to the pyrolysis plant.

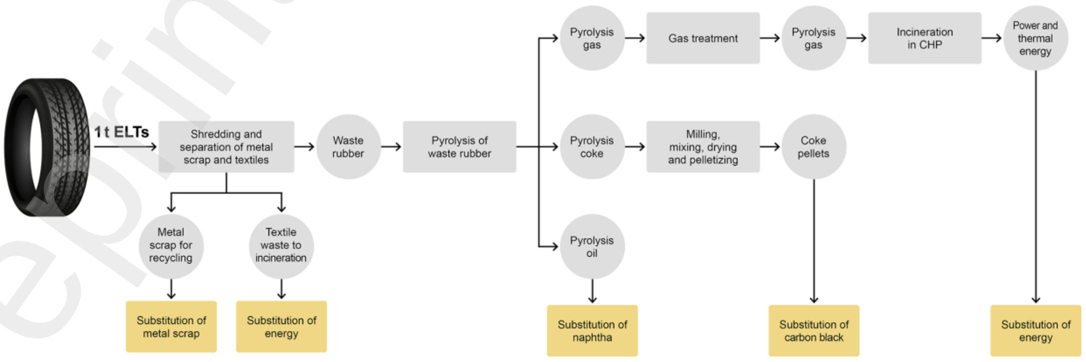

Figure 3: Treatment of ELTs by pyrolysis

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190 After storage, the ELTs are shredded, and the metals and textiles are separated from the rubber fraction. The metal scrap is sent for recycling, whereas the textiles are sold and are used as refused derived fuel in cement plants without any further pre-treatment. The heating value of the textile waste is 32 MJ/kg, and its carbon content is 76.9 %.

Table 2: Assumptions and data used for collection and treatment of 1 t ELTs

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
Amount of treated ELTs	1000	kg	FU (assumed)	
Average distance to transport ELTs to pyrolysis plant	50	km	assumed	GLO: Truck-trailer, Euro 6, 34 - 40t gross weight / 27t payload
Diesel demand for forklifts	5.48	kg	amount of fuel calculated based on annual consumption	DE: Diesel mix at filling station Sphera, Emissions based on (Fuc et al., 2016)
Electricity demand for shredding	211	kWh	measured	DE: Electricity grid mix (2020) Sphera
Water demand for washing	485	kg	DE: Tap water from surface water Sphera	DE: Tap water from surface water Sphera
OUTPUT FLOWS				
Rubber fraction for pyrolysis	636	kg	measured	
Metal scrap	269	kg	measured	DE: Recycling potential steel profile (D) Sphera Average distance to steel recycling is 26 km based on primary data
Textile waste	95	kg	measured	LCA model for combustion in cement plant and substitution of German fuel mix. Average distance to textile incineration is 700 km based on primary data
Wastewater	485	kg	measured	DE: Municipal wastewater treatment (mix) Sphera

195 The shredded rubber is fed to the pyrolysis plant where the products pyrolysis gas, pyrolysis coke, and pyrolysis oil are generated. The fluid phases are separated from each other by condensation steps. Apart from the target products, process water and pyrolysis sludge are produced as side streams and are incinerated. The heating value of the process water is 0.21 MJ/kg whereas the heating value of the pyrolysis sludge is 1.7 MJ/kg. The corresponding carbon contents are 0.03 and 0.43 kg C/kg.

200 **Table 3: Assumptions and data used for treatment of 1 t ELTs by pyrolysis**

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
Amount of rubber from	636	kg		

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Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
ELTs				
Electricity consumption for pyrolysis as well as cooling demand and own consumption of CHP unit	549	kWh	measured	Own dataset for production of electricity in a CHP
Water demand	358	kg	measured	DE: Tap water from surface water Sphera
Lubricants	0.007	kg	measured	DE: Lubricants at refinery Sphera
Oil consumption compressor	0.114	kg	measured	DE: Heavy fuel oil at refinery (1.0 wt.% S) Sphera
OUTPUT FLOWS				
Pyrolysis gas	135	kg	measured	
Pyrolysis coke	313	kg	measured	
Pyrolysis oil	166	kg	measured	
Process water	16	kg	measured	
Pyrolysis sludge	6	kg	measured	
Water evaporated	358	kg	calculated	

The pyrolysis gas is further treated using a fixed bed filter and iron-based sorbent and then incinerated in a combined heat and power plant (CHP) together with a small share of natural gas. The heating value of the pyrolysis gas is 46 MJ/kg, and its carbon content is 72 %. It was assumed that 73 % of the carbon is fossil-based and 27 % is biobased (natural rubber). The later was estimated based on an average natural rubber carbon content in new tyres of 17 % (Dong et al., 2021). The electrical efficiency of the CHP was 36 % and the thermal efficiency was 50 %. The emissions to air are presented in the SM table S2.

As sorbent, 9.45 kg iron-based pellets were used per FU. As a conservative assumption, the recycling of the sorbent was excluded. A major proportion of the produced electricity was used for heating the pyrolysis plant and only a small amount was used for other operations, while the rest was fed into the electricity grid. The thermal energy recovered from the off gas of the CHP (ca. 84 kW) was used for drying the agglomerated char in a fluidized bed dryer to achieve a residual moisture content of less than one mass percent. In addition, thermal energy from the cooling water of the CHP (low temperature, ca. 84 kW) was used for heating of the common rooms. The pyrolysis coke was milled using an impact classifier mill and agglomerated. The agglomerates ejected as the final product have a particle size of 1 – 2 mm. The exhaust air from the milling and agglomeration unit is purified with a filter system consisting of a PE needle felt.

Table 4: Assumptions and data used for processing the pyrolysis coke from treatment of 1 t ELTs

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
Pyrolysis coke	313	kg	Measured	
Electricity consumption	304	kWh	Calculated	DE: Electricity grid mix (2020) Sphera

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Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
Water demand	209	kg	Calculated	DE: Tap water from surface water Sphera
Thermal energy from CHP	211	MJ	Calculated	
Thermal energy from natural gas	629	MJ	Calculated	DE: Thermal energy from natural gas Sphera
OUTPUT FLOWS				
Carbon black agglomerates	313	kg	Calculated	

The pyrolysis oil is transported and used as a substitute for naphtha in an oil refinery. The substitution factor is based on the heating value of the pyrolysis oil and that of naphtha. In principle, the oil is converted to various chemical products including those used as additives in new tyres. A screenshot of the LCA model is provided in the supplementary materials in figure S3.

3.2 Treatment of ELTs in cement plants

To use ELTs as refused derived fuel in cement kilns they need to be grinded first. In addition, a part of the steel wire is separated from the ELTs and is sold as a by-product. Primary data on the pre-treatment of ELTs is sparse, however, Corti and Lombardi (2004) published LCI data for grinding based on Italian plants, considering yearly average values.

Table 5: Assumptions and data used for grinding of 1 t ELTs based on (Corti and Lombardi, 2004)

Mass/energy flow or process	Amount	Unit	LCI data set
Amount of ELTs	1000	kg	
Electricity consumption for grinding	47.2	kWh	DE: Electricity grid mix (2020) Sphera
Water demand	150	kg	DE: Tap water from groundwater Sphera
Steel	0.23	kg	EU: Steel cold rolled coil worldsteel
Oil consumption	0.011	kg	DE: Heavy fuel oil at refinery (2.5wt.% S) Sphera
OUTPUT FLOWS			
Ground tyres	966	kg	
Iron scrap	34	kg	DE: Recycling potential steel profile (D) Sphera

The ground tyres still containing iron scrap, textiles and rubber are further transported to a cement plant. According to Corti and Lombardi (2004) the transport distance varies between 35 and 100 km. In this study 50 km was assumed. The ground tyres are finally incinerated in co-combustion in cement plants. Greenhouse gas emissions were calculated based on a carbon content of 65 % in ELTs, and the assumption that 73 % of the carbon is fossil-based and 27 % is biobased (natural rubber). Emissions other than CO_2 were estimated based on data reported by European cement kilns (Schorcht et al., 2013). For the calculations, an average fuel demand of 3300 MJ/t clinker and a calorific value of 28 MJ/kg ground tyres (TabVDZ, 2020) was assumed.

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In the cement plant, the ground tyres replace the German fuel mix of cement plants which currently consists of 32.5 % primary fuels and 67.5 % of secondary fuels. The average heating value of the fuel mix is 23.5 MJ/kg whereas the CO_2 emission factor is 1.78 kg CO_2/kg fuel mix. Detailed information on the composition of the fuel mix, the heating values of the fuels, the carbon content and the biogenic share of carbon is given in the supplementary materials in table S3a. In Europe, the share of secondary fuels is lower compared to the German situation. According to Cembureau (2022) currently about 48 % of the fuel need in the EU is supplied by secondary fuels. To investigate the influence of a fuel mix with a higher share of fossil fuels, a conservative estimation for the European fuel mix for cement plants is considered. The fuel composition was taken from Merlin and Vogt (2020) and is also provided in the supplementary materials in table S3b.

For the reference system, it was assumed that all primary fuels are transported by truck over 150 km, whereas secondary fuels are transported 100 km. All waste used to generate the secondary fuels was assumed to be burden-free. In the case of used tyres, the same pre-treatment was assumed as described in table 4. For all other solid secondary fuels, for pre-treatment an electricity requirement of 320 kWh per ton of fuel was assumed.

Apart from the fact that the ground tyre replaces other fuels in the cement kiln, iron ore is also replaced, which is used as an additive in cement production (Ingemarsdotter et al., 2021). As substitution factor, a one-to-one replacement of iron scrap with iron ore was considered. Assuming an average metal content of 147 kg/t ELTs as well as the extraction of 34 kg iron scrap in the grinding step, 113 kg iron ore per FU are replaced by the metal content in the ground tyre.

Finally, assuming that the carbon is 100 % oxidized to CO_2, the incineration of 1 t ELTs produces 1681 kg CO_2. In addition, the calorific value of the ELTs is estimated based on 28 MJ/kg ground tyre. Assuming an efficiency of 100 %, the total energy generated per FU is 27 048 MJ. A screenshot of the LCA model is provided in the supplementary materials in figure S4.

3.3 Treatment of ELTs in dedicated power plants

Like the treatment of ELTs in cement plants, ELTs need to be collected and grinded to be further used as a fuel in a dedicated power plant. Primary data for ELT collection and grinding was taken from Malijonyte et al. (2016) and is summarized in table 5.

Table 5: Assumptions and data used for grinding of 1 t ELTs based on (Malijonyte et al., 2016)

Mass/energy flow or process	Amount	Unit	LCI data set
Amount of ELTs	1000	kg	
Transport ELTs from collection point to shredding facility	370	km	GLO: Truck, Euro 6 A-C, more than 32t gross weight / 24.7t payload capacity Sphera <e-ep>
Electricity consumption for grinding and metal separation	207	kWh	DE: Electricity grid mix (2020) Sphera

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Mass/energy flow or process	Amount	Unit	LCI data set
Lubricants for shredding	0.00022	kg	DE: Lubricants at refinery Sphera
OUTPUT FLOWS			
Amount of ground tyres	818.5	kg	Lower heating value 34.9 MJ/kg; Carbon content: 79 % (Malijonyte et al., 2016)
Amount of iron scrap	181.5	kg	DE: Recycling potential steel profile (D) Sphera

It was assumed that the ground tyres are transported 244 km from the shredding facility to the incineration plant (Malijonyte et al., 2016). For the incineration process, foreground data was taken from (Bianco et al., 2021) and partly adopted for German conditions. The efficiency of the dedicated power plant was assumed to be 15% electric and 37% thermal energy. These efficiencies reflect the average situation of German refused derived fuel (RDF) power plants in 2016 (Flamme et al., 2018). Relevant input and output flows used for incineration of 1 t ELTs are summarized in table 6. Emissions to air other than carbon dioxide were taken from (Bianco et al., 2021). A screenshot of the LCA model is provided in the supplementary materials in figure S5.

Table 6: Assumptions and data used for incineration of 1 t ELTs in dedicated power plants

Mass/energy flow or process	Amount	Unit	LCI data set
Amount of ground tyres	1000	kg	Lower heating value 34.9 MJ/kg; Carbon content: 79 % (Malijonyte et al., 2016)
Ammonia	3	kg	DE: Ammonia liquid (NH3) with CO2 recovery, by-product carbon dioxide (economic allocation) Sphera
Thermal energy	103	MJ	DE: Thermal energy from natural gas Sphera
Activated carbon	1	kg	DE: Activated carbon Sphera
Tap water	1770	kg	DE: Tap water from surface water Sphera
OUTPUT FLOWS			
Electricity	1456	kWh	DE: Electricity grid mix (2020) Sphera
Heat	12928	MJ	DE: District heating Sphera
Fly ash and scrubber sludge	40	kg	Europe without Switzerland: market for fly ash and scrubber sludge ecoinvent 3.9.1
CO_2 (fossil)	2120	kg	Calculated based on a carbon share of 79 % and a fossil share of 73 % in ground tyres
CO_2 (biogenic)	784	kg	Calculated based on a carbon share of 79 % and a biogenic share of 27 % in ground tyres

3.4 Treatment of ELTs for material use

Unlike the scenarios discussed earlier, ELTs can also be used for material recycling. Market share for end-of-life tyre applications is generally likely to differ based on variables like geography, industry trends, and the accessibility of substitute materials. Some key material applications for ELTs include civil engineering, noise barriers, beds for rail and tram and the production of tyre-derived products such as rubber mulch, shock pads, and infill materials (Fiksel et al., 2011, Magnusson and Mácsik, 2017, Patricio et al., 2021).

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Of these applications, infills for artificial turfs have increased exponentially in the past decade (Patricio et al., 2021). ELTs are reduced to smaller pieces or crumbs, that can be used as performance infill in third generation artificial turfs. Depending on the type of tyre being recycled, the tyre fragments' size and form can differ. Granulation, grinding, and cryogenic processing are a few techniques employed. Additional processing stages like screening, magnetic separation, and washing can enhance the final infill material's quality. The secondary fractions from infill production are steel and fabrics. The separated steel can be recycled, while the fabric fraction can be incinerated to recover energy (Fiksel et al., 2011, Magnusson and Mácsik, 2017).

The produced infills from the mechanical recycling of ELTs can be used to replace polymer-based infills from virgin materials such ethylene propylene diene monomer (EPDM) or thermoplastic elastomers (TPE) that are usually made from styrene ethene butene styrene copolymer (SEBS) (Merlin and Vogt, 2020). Relevant input and output flows used for production of infills from 1 t ELT are summarized in table 7.

Table 7: Assumptions and data used for production of infills from 1 t ELT based on (Johansson, 2018, Merlin and Vogt, 2020)

Mass/energy flow or process	Amount	Unit	LCI data set
Amount of ELT	1000	kg	
Electricity	276	kWh	DE: Electricity grid mix (2020) Sphera
Heat	69.6	MJ	DE: Thermal energy from natural gas Sphera
Tap water	45	kg	DE: Tap water from surface water Sphera
Diesel	4.18	kg	DE: Diesel mix at filling station Sphera
OUTPUT FLOWS			
Infill	725	kg	1 kg SBR from used tyres replaces 1.48 kg EPDM infills (Johansson, 2018)
Steel	147	kg	DE: Recycling potential steel profile (D) Sphera
Fabrics	124	kg	DE: Polyamide (PA) 6.6 in waste incineration plant Sphera
Inert	4	kg	EU-28: Inert matter (Unspecific construction waste) on landfill Sphera

Fabrics were assumed to be incinerated in a municipal solid waste incineration (MSWI) plant, and the scrap steel recycled, while the inert material was assumed to be sent to landfill.

The produced infills from ELTs is assumed to replace EPDM-based infills by volume (see table 8). Based on their respective density, 1 kg infills from ELTs replaces 1.48 kg EPDM-based infills (Johansson, 2018). A screenshot of the LCA model is provided in the supplementary materials in figure S6.

Table 8: Assumptions and data used for production of 1t EPDM infills based on (Johansson, 2018, Merlin and Vogt, 2020)

Mass/energy flow or process	Amount	Unit	LCI data set
EPDM rubber	220	kg	DE: Ethylene Propylene Diene Elastomer (EPDM) Sphera
Limestone	680	kg	DE: Limestone flour (50µm) Sphera

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Mass/energy flow or process	Amount	Unit	LCI data set
Mineral oil	80	kg	RNA: White mineral oil, at plant USLCI/Sphera
Phthalates	20	kg	DE: Di-isononyl phthalate (DINP) Sphera
Electricity	827	MJ	DE: Electricity grid mix (2020) Sphera
Diesel	5.77	kg	DE: Diesel mix at filling station Sphera
OUTPUT FLOWS			
EPDM Infill	1000	kg	1 kg EPDM infills from used tyres replaces 0.676 kg SBR (Johansson, 2018)

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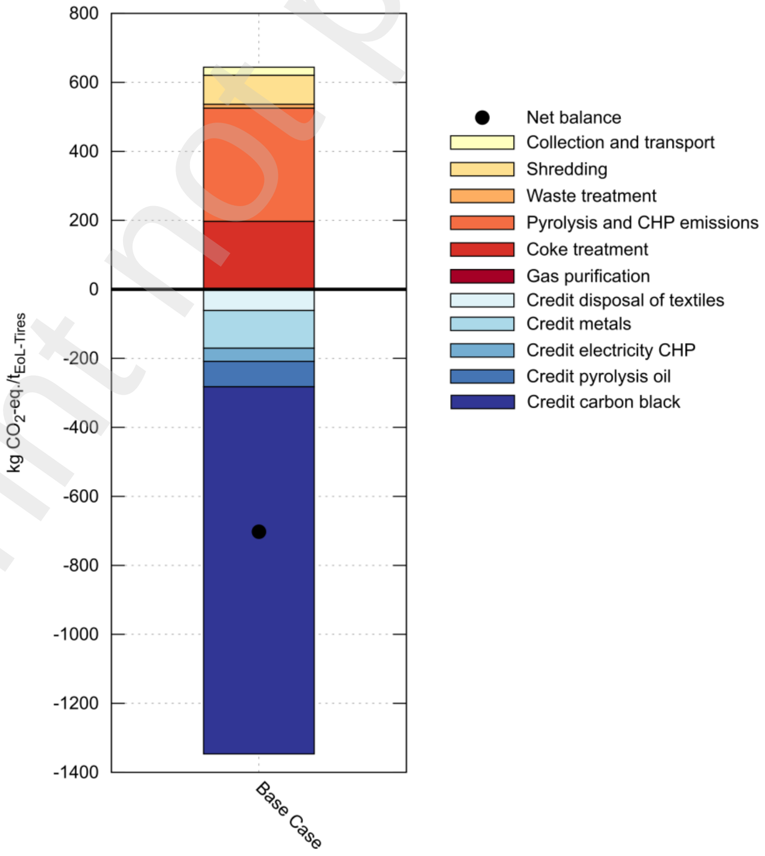
4.1 Environmental impacts of treatment of ELTs by pyrolysis

In total 644 kg CO_2-eq. are emitted from the treatment of 1 t ELTs. At the same time the system generates various co-products namely carbon black substitute, pyrolysis oil, metals, and a textile containing fraction which yields a combined credit of 1347 kg CO_2-eq./FU. In particular, the carbon black substitute gives a credit of 1064 and thus dominates the overall balance. In total, Scenario 1 315 generates greenhouse gas (GHG) savings of 703 kg CO_2-eq./FU (see Figure 4).

Figure 4: Treatment of ELTs by pyrolysis



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The incineration of the pyrolysis gas together with a small share of natural gas contributes the most to the global warming impact (GWI), amounting to 51 %. In addition, the treatment of the pyrolysis coke to produce carbon black substitute contributes to 30 % of the GWI. Shredding and metal separation contributes to 13 % of the GWI, collection and transport to 4 % and the treatment of sludge and wastewater to 2 % of the total GWI.

Savings are achieved in most of the investigated impact categories as shown in figure 5. Exceptions are freshwater eutrophication, ionising radiation, land use, ozone depletion and resource use minerals and metals. Ozone depletion is mainly driven by the background data used for iron-based pellets which are applied in gas treatment. As background data the ecoinvent 3.9.1 dataset "RoW: iron pellet production" was used due to unavailability of representative data for iron-based sorbent. However, the dataset used refers to data collected in 1999 which might explain the high impacts on ozone depletion. The other net-positive impacts mainly result from the additional electricity demand for shredding and further treatment of the pyrolysis coke. However, since nuclear power is no longer produced in Germany, the ionising radiation impacts are not time representative.

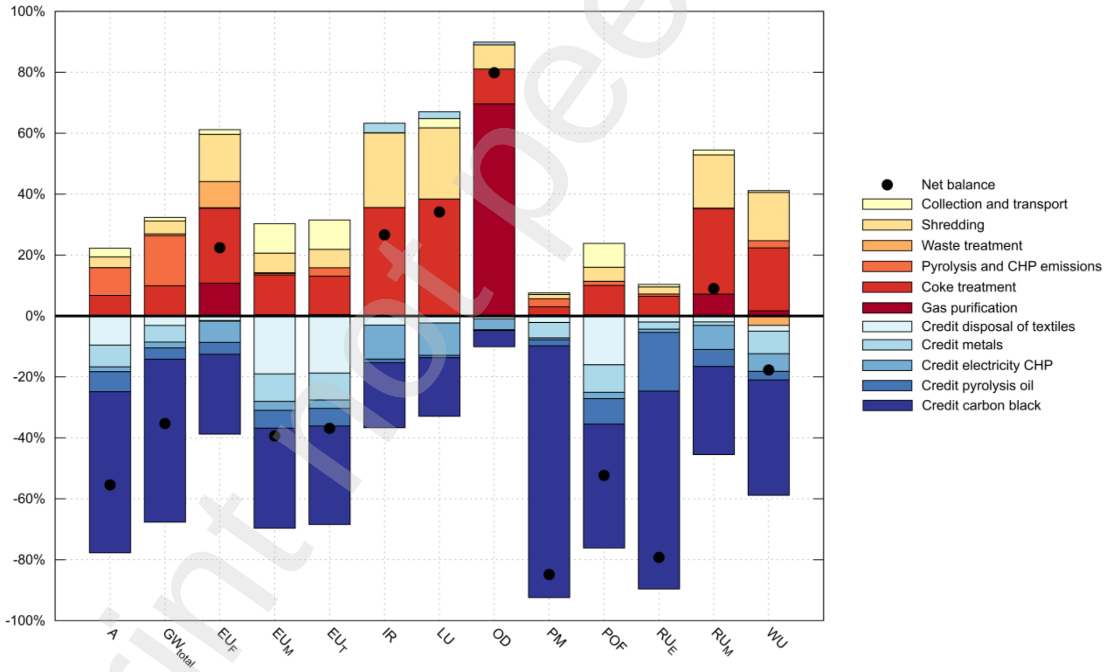

Figure 5: Contribution analysis for treatment of ELTs by pyrolysis

A: Acidification; GW,total: Climate Change – total; EUF: Eutrophication, freshwater; EUM: Eutrophication, marine; EUT: Eutrophication, terrestrial; IR: Ionising radiation; LU: Land Use; OD: Ozone depletion; PM: Particulate matter; POF: Photochemical ozone formation; RUE: Resource use, fossils; RUM: Resource use, mineral and metals; WU: Water use

4.2 Environmental impacts of treatment of ELTs in cement plants

In total, the impact on global warming from the treatment of 1 t ELTs in cement plants is minus 309 kg CO_2-eq./t ELTs. The provision of the ELTs emits 33 kg CO_2-eq./t ELTs. The incineration of ELTs contribute to 1681 kg CO_2-eq./t ELTs, whereas 2023 kg CO_2-eq./t ELTs can be avoided by replacing

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the fuel mix of German cement plants. In addition, 10 kg CO_2-eq./t ELTs are avoided by substituting iron in cement plants. Net savings are achieved in all investigated impact categories (see table S5a in SM). Significant savings are achieved in the categories freshwater eutrophication, ionising radiation, land use, ozone depletion, resource use (fossils, mineral and metals) and water use. The savings in freshwater eutrophication and land use are mainly affected by the avoided electricity demand for fuel preparation of secondary fuels for cement plants as well by the avoided biodiesel demand for transportation of fuels. Savings in ionising radiation, resource use (fossils, mineral and metals), and water use are mainly driven by the avoided provision of fossil fuels used in cement plants.

4.3 Environmental impacts of treatment of ELTs in dedicated power plants

The incineration of 1 t ELTS in dedicated power plants including energy recovery results in 679 kg CO_2-eq. The main GHG emissions occur during incineration (1825 kg CO_2-eq./t ELTs), while the main GHG savings result from the generation of electricity (-474 kg CO2-eq.) and thermal energy (-726 kg CO_2-eq.). Transport, tyre shredding, and the recovery of metals play only a minor role in all investigated impact categories. In many impact categories, the environmental impact is higher than the savings from energy recovery. A summary of all environmental impacts is provided in the SM in table S6.

4.4 Environmental impacts of treatment of ELTs for infill production

The production of infills from the treatment of 1 t ELTs for use in artificial turfs for sports applications was found to be 302 kg CO_2-eq/FU. The emissions occur during the production of infills and the incineration of fabrics. The system also produces steel as a co-product which yields a credit of 60 kg CO_2-eq/FU. The produced infills can substitute traditional infill materials such as EPDM or TPE. The credits from the substitution of EPDM-cased infills is 1076 kg CO_2-eq. As a result, scenario 4 generates GHG savings of 829 kg CO_2-eq./FU. If the produced infills from ELTs are used to substitute TPE-based infills, the overall GHG savings are even higher at 1468 kg CO_2-eq. Savings are also realized in all other investigated impact categories except of water use. A summary of all environmental impacts is provided in the SM in table S7.

4.5 Comparison of environmental impacts of EOL options for ELTs

Scenarios 1 and 4 investigate material recycling of ELTs while scenarios 2 and 3 look at two different options to treat ELTs energetically. The material recycling shows advantages regarding the impact categories climate change and conservation of fossil resources. In addition, comparatively fewer particulate matter emissions are released. In contrast, the pyrolysis process shows comparatively high impacts in the categories freshwater eutrophication, ionising radiation, and land use, since comparatively

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more power is needed for the treatment of ELTs. In addition, the thermal EoL processes for ELTs benefit from the fact that they substitute the German electricity mix and fuel mix of cement plants and thus receive credits in the impact categories acidification, eutrophication, ionising radiation, land use (bioenergy) and particulate matter.

When comparing incineration of ELTs in cement plants (scenario 2) with incineration of ELTs in dedicated power plants (scenario 3), scenario 2 performs better in most impact categories due to a higher efficiency of energy recovery and the substitution of fuels with high environmental impacts. However, regarding ionising radiation, land use (bioenergy) and water use scenario 3 performs better since not only thermal energy is replaced but also the German electricity grid mix.

Figure 6 illustrates the emissions from each EoL scenario, their savings realised by their substitution potential, as well the net values (net values are also summarized in table S7 in the SM). It can be clearly seen that credits often contribute most to the overall results, in particular in scenario 2 and 3 leading to net savings. However, it must be noted that the greater the share of renewables in the energy mix, the lower the credits for energy recovery. The same applies to the case of greener fuels in cement plants.

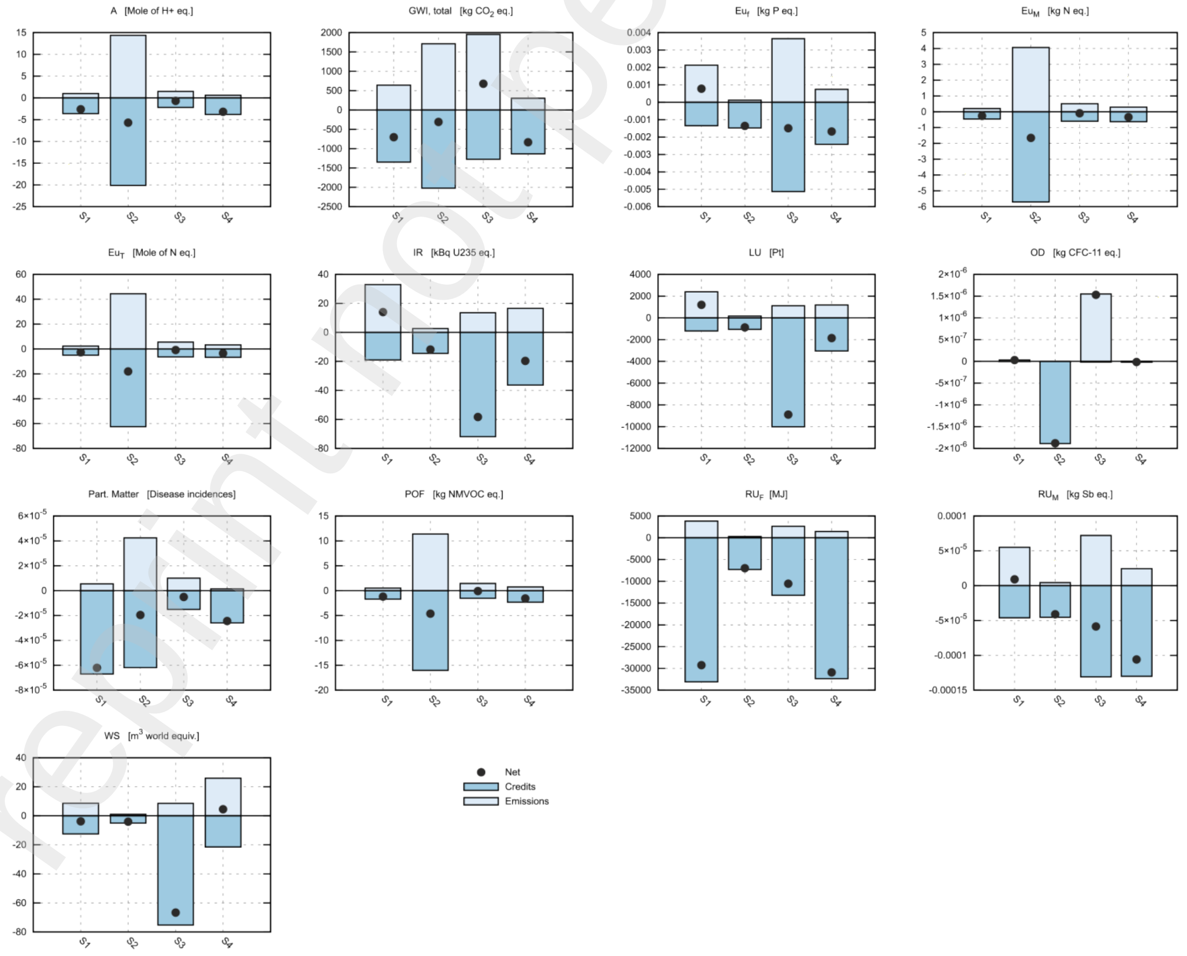

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Figure 6: All investigated LCIA results for scenarios 1-4 (A: Acidification; GW, total: Climate Change – total; EUF: Eutrophication, freshwater; EUM: Eutrophication, marine; EUT: Eutrophication, terrestrial; IR: Ionising radiation; LU: Land Use; OD: Ozone depletion; PM: Particulate matter; POF: Photochemical ozone formation; RUE: Resource use, fossils; RUM: Resource use, mineral and metals; WU: Water use)

To support the interpretation, the LCIA results were normalised and weighted. The applied normalisation factors (NF) and weighting factors (WF) are recommended for PEF 3 and exclude toxicity-related impact categories (European Commission et al., 2018). NF and WF are provided in the SM in table S8. The normalised and weighted results clearly show that scenarios one and four result in the highest environmental benefits, whereas scenario 2 shows smaller benefits and scenario 3 leads to net environmental impacts. Most relevant impact categories are climate change and fossil resource use. Minor contributions come from acidification, particulate matter, and photochemical ozone formation. All other impact categories hardly contribute to the overall normalised and weighted results, as presented in figure S7 in the SM.

4.3 Sensitivity analysis

4.3.1 Influence of the choice of carbon black substitute

As stated above, the environmental footprint for the pyrolysis scenario is strongly influenced by the credits given for the carbon black substitute. Key indicators influencing the footprint of carbon black are the yield of the process (efficiency of extracting carbon from the feedstock), valorisation of the by-product tail gas, as well as the method to account for the added value of the recovered energy generated from the incineration of the tail gas (Chikri and Wetzels, 2020). Depending on the quality of carbon black, the yield of carbon black varies between 10% and 65%. In general, the yield of rubber carbon black (reinforcing and semi-reinforcing carbon black) is between 40 and 60 %, while the yield of pigment black is lower, ranging between 10 and 30 % (van Veen & Leendertse, 2002).

In addition, the footprint of the product is influenced by the primary feedstock fuels, typically carbochemical or petrochemical oils such as steam cracker oils, catalytic cracker oils, or aromatic concentrates. The secondary feedstock, often natural gas, also influences to the overall carbon footprint of the product.

Carbon Footprints of carbon black are summarized in Table 9 and range between 3.44 (Sphera dataset economic allocation, base case) and 2.39 kg CO_2-eq./kg carbon black (Sphera dataset with system expansion). Comparably low carbon footprint values (2.55 kg CO_2-eq./kg) are reported by Orion Engineered Carbons GmbH (Orion Engineered Carbons GmbH, 2018), who apply system expansion as well in order to account for the added value generated from thermal energy and electricity. Cabot B. V. estimates a carbon footprint of 3.1 kg CO_2-eq. per kg carbon black (Chikri and Wetzels, 2020) when applying energetic allocation.

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Table 9: Carbon Footprint of Carbon Black according to different sources

Carbon Footprint [kg CO2-eq./kg]	Reference
2.39 (worst case)	Sphera-dataset: DE: Carbon black (furnace black; general purpose) - thermal energy credit Sphera
3.44 (base case)	Sphera-dataset: DE: Carbon black (furnace black; general purpose) (economic allocation) Sphera
3.3	Cabot B. V. average plant, 2016, energetic allocation (Chikri and Wetzels, 2020)
3.1	Cabot B. V. estimated for rubber black, 2016, , energetic allocation (Chikri and Wetzels, 2020)
3.2	(Kuile, 2016) based on data taken from BREF report (European Commission, 2007)
2.55	Orion Engineered Carbons GmbH, Sustainability report 2018, credits for avoided burdens (Orion Engineered Carbons GmbH, 2018)

Apart from these values, the ecoinvent 3.9.1 dataset: “carbon black production” (cut-off database) provides a carbon footprint of 2.3 kg CO_2-eq. per kg carbon black. The data is based on a mixture of literature references and expert judgments. The corresponding life cycle inventory, in particular the direct CO_2 emissions, do not seem to be plausible and therefore the dataset is not considered due to low data quality. Consequently, the Sphera-dataset “DE: Carbon black (furnace black; general purpose) - thermal energy credit Sphera” is used for sensitivity analysis. When using this dataset, the GHG savings for Scenario 1 are reduced from 702 kg CO_2-eq./FU to 366 kg CO_2-eq./FU. A similar tendency can be observed in other investigated impacts categories, but differences are often smaller compared to global warming and range between 1 and 83 %. LCIA results and difference are presented in table S8 in the SM.

4.4.2 Sensitivity on the substitution of fuels in cement plants

When considering the average fuel mix of European cement plants instead of the German fuel mix (see tables S3a and S3b in the SM), the highest changes occur in global warming. GHG savings increase from 309 kg CO_2-eq./FU to 660 kg CO_2-eq./FU. Higher environmental benefits are also realized in other impact categories but less strongly. The savings range from 1 % (water use) to 64 % (resource use, fossils). All LCIA results and differences to the base case are shown in table S9 in the SM.

4.4.2 Sensitivity on tyre composition

In case of the pyrolysis technology (Scenario 1), mainly truck tyres and discarded truck tyres from retreading are used as raw material. Therefore, the share of fibres is lower (truck tyres do not use textiles) and the metal content is higher. In addition, truck tyres contain a higher proportion of natural rubber in the total rubber than passenger car tyres. In case of discarded truck tyres from retreading the share of rubber content is lower. Although there is no available primary data for treating car tyres, it is obvious

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that switching to car tyres the carbon black yield would increase due to a higher rubber content of approximately 10 %. Since the credit for carbon black dominates the LCIA results, most of the investigated impact categories would show lower impacts or rather higher savings. Although it was not possible to consider the same composition of ELTs in each scenario, the influence of varying compositions is expected to be small.

5 How circular are the investigated treatment options?

For the design of circular products, the so-called R-strategies serve as a framework. Several definitions exist; of which the most commonly quoted is the 10-R strategies (Potting et al., 2017). The R-strategies are ranked according to their increasing levels of circularity. The strategies reduce (R2), reuse (R3), repair (R4), refurbish (R5), remanufacture (R6), repurpose (R7), and recycle (R8) contribute the most to circularity while refuse (R0), rethink (R1) and reduce (R2) try to avoid unnecessary products and energy recovery (R9) is the final means, if the quality of the material is too low for other options (Minguez et al., 2021). In this study we compared different EoL solutions for scrap tyres which fall under different R strategies: Scenario 1 considers recycling (R8), Scenario 2 and Scenario 3 involves energy recovery (R9) and finally Scenario 4 refers to remanufacture (R6).

The general idea behind the R-strategy framework is to optimize circularity by reducing dependency on primary raw materials. However, the remanufacture or repurpose often leads to a cascading use. This means that recycled materials cannot be reutilised within the same product and value chain. This is also the case in Scenario 4. The recycled rubber can no longer be used for new tyres but is used in downcycling applications such as infills, which must be disposed of after about 8 to 10 years. In contrast, the investigated pyrolysis process captures a high fraction of the carbon contained in the tyres in the form of recovered carbon black and pyrolysis oil (80%-85% depending on feedstock), which are general purpose raw materials. These materials can be used very flexible, and many different new products can be produced, including new tyres. Therefore, a real circularity is possible.

So far, Scenario 1 uses the produced pyrolysis-gas for electricity generation to supply the unit with electricity, causing CO_2 emissions in the process. This is due to unsolved technical burdens of capturing the pyrolysis gases and economic feasibility. In the future, it might be possible to capture the pyrolysis gas and reintroduce it to the chemical industry achieving a nearly complete capturing of the carbon content. In this case the pyrolysis reactor should be heated with green electricity so that no CO_2 is released to the atmosphere.

The recovered carbon black is the valuable product which gives high credits when replacing virgin carbon black in the tyre industry. However, the pyrolysis produces more pyrolysis char than the amount of carbon black in the tyres, i.e., a complete closed loop in the tyre industry is not possible. In addition,

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the properties of the recovered carbon black differ from virgin carbon black limiting straight-away substitution.

6 Summary of results and conclusion

This paper applied the LCA methodology to investigate the environmental impacts generated from four EoL handling and processing pathways for ELTs. The functional unit of the study was the treatment of 1 tonne ELTs. The results show that the recycling of ELTs (Scenarios 1 & 4) have lower environmental impacts regarding global warming, particulate matter and resource use compared to energetic treatment (Scenarios 2 & 3). When comparing the thermal treatment of ELTs in cement plants or in dedicated power plants, the thermal treatment in cement plants shows higher savings in most impact categories due to higher efficiencies. The recovery of carbon black via pyrolysis shows similar benefits as the production of infill. However, in general it is also possible to recycle the used infill via pyrolysis at its EoL. Such a second or third life was not considered by this LCA study. Moreover, the material use of ELTs could be legally restricted in the future. The European Chemicals Agency ECHA currently works on a proposal to ban rubber infills for sports pitches (ECHA, 2019). Just in April 2023 the European REACH Committee voted for the proposals to restrict microplastics introduced intentionally to various products. Therefore, the investigated thermochemical technology could play a key role in treating ELTs.

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