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Environmental assessment of rubber recycling through an innovative thermo-mechanical devulcanization process using a co-rotating twin-screw extruder

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Abstract

Under the umbrella of the transition toward a circular economy, the evaluation of the environmental sustainability of processes aiming to the recycling of materials is essential. Rubbers, like ethylene-propylene diene monomer (EPDM), represent a class of materials that have become an environmental, social and economic issue, so the evaluation of the end-of-life management of these materials is of primary relevance. This study investigates the environmental burdens associated with a thermo-mechanical devulcanization process for recycling of EPDM wastes by co-rotating twin-screw extruders. The Life Cycle Assessment methodology was used to carry out the analysis and eight environmental impact categories have been evaluated: Climate change, Ozone depletion, Photochemical ozone formation, Acidification, Eutrophication (freshwater), Ecotoxicity (freshwater), Resource use (fossils) and Resource use (minerals and metals). In the first step, the analysis focused on the identification and quantification of main environmental hotspots of the process. Then, the study was extended by including two comparative analyses, to better understand the magnitude of the environmental burdens generated by the devulcanization process. The results of the hotspots analysis showed that the main contribution to the environmental impacts of the devulcanization process is due to energy consumption. However, the comparison underlines how the devulcanization process for recycling of EPDM waste allows to reduce the environmental burden associated with the life cycle of rubber products.

Keywords

Life Cycle Assessment; Environmental impact; Thermo-mechanical devulcanization; Extrusion; Recycling; Rubber waste

Number of words in the manuscript: 6486

1. Introduction

The current models on which the development of modern society is based led to an unsustainable consumption of resources, with associated important environmental and social impacts (Hanumante et al., 2019). This approach involves an ever-increasing production of wastes, and its management remains a challenge (Fan et al., 2021).

In 2020 the European Commission, by adopting a Circular Economy Action Plan, has defined the path to follow to achieve a more sustainable model for economic development (European Commission, 2020). The objective of the EU action plan, based on the circular economy paradigm, is to exploit the full potential of resources and materials, keeping them in the economy for as long as possible. At the same time, this objective towards a greater sustainability is pursued by minimizing the production of waste (European Commission, 2015).

Elastomers, and more specifically rubbers, although indispensable for today's life, represent a class of materials with important environmental, social and economic impacts. These issues are not only associated with the production phase, which requires high intensity of labor, energy and raw materials, but also with respect to the final disposal of wastes (Dunuwila et al., 2018; Garcia et al., 2015; Fazli and Rodrigue, 2020).

1.1 Rubber End-of-Life

Nowadays, thanks to the development of new processes, there are several different options that can be identified for the end-of-life management of these materials. One of the most common solution is to use the rubbers as fuel in the cement industries and blast furnace or, more generally, to burn this waste with associated energy recovery (Isayev, 2013; Ramarad et al., 2015). Another solution is to recycle this material through a mechanical process, by grinding the waste to convert it into powder or granulates, that can be used for different applications: bituminous mixtures, concrete, reinforcing fillers in polymers, rubber pads (Farina et al., 2017; Isayev, 2013; Li et al., 2010; Si et al., 2018; Sienkiewicz et al., 2017; Zanetti et al., 2015). If rubber materials, in the last part of their life cycle, are not directed towards a recycling process, the remaining destination is landfill disposal, with the consequent possible development of diseases and ecological contaminations (Molino et al., 2018). However, to fulfil the objectives defined by a circular economic model and therefore try to keep the quality of materials unchanged for as long as possible, it is necessary to rely on other processes, which allow a recovery of the material while preserving its physicochemical properties.

1.2 Devulcanization processes

A possible solution to recycle rubbers is the devulcanization processes (Seghar et al., 2019). Devulcanization is a process that allows the breaking of the crosslinked bonds between polymeric chains, obtaining a material that can be mixed, processed, and vulcanized again (Isayev, 2013). The process can be carried out using physical methods, i.e. ultrasound, microwave, mechanical stress (Aoudia et al., 2017; Mangili et al., 2015; Seghar et al., 2015), thermo-mechanical methods (Meysami et al., 2017; Seghar et al., 2019) or chemical methods (Jana et al., 2006; Joseph et al., 2015; Kojima et al., 2005). Previous works suggested that the thermo-mechanical devulcanization is one of the most suitable methods to be implemented at an industrial level. In fact, it allows to obtain a high degree of devulcanization and is carried out using a simple extrusion process, which is a very widespread technology in the world of rubber and polymeric materials (Fukumori et al., 2006; Seghar et al., 2019; Yazdani et al., 2011).

1.3 Life Cycle Assessments

To our knowledge, objective evaluations of possible environmental effects associated with the use of a thermo-mechanical devulcanization process for the recycling of elastomeric materials are not reported in the literature. In fact, only limited studies have examined the environmental burdens of the devulcanization process and references are always made to chemical methods preceded by mechanical grinding treatments. Li et al. (2010) compared, from an environmental point of view, different technologies for the end-of-life treatment of tires. In this study, the environmental impacts of a devulcanization process (dynamic devulcanization), carried out using chemical agents, were also assessed. The analysis showed that the best environmental profile for end-of-life treatment can be identified in the use of pyrolysis technology, followed by dynamic devulcanization and environmental grinding. The main impacts for all treatments are mainly due to energy consumption, which is very high for the devulcanization process. However, the authors believe that dynamic devulcanization treatment is the preferable approach in China; in fact, by optimizing this process, it is possible to recover materials and reduce the dependence on the extraction of new raw resources. In another study, Li et al. (2014) investigated, through the Life Cycle Assessment (LCA) methodology, the environmental profile of ground rubber produced from scrap tires. The recycling phase of scrap tires was also modelled, using a chemicals-assisted devulcanization process. The results showed that the main impacts (more than 66% of the total) of the devulcanization are due to the high energy consumption. However, a more in-depth investigation of the purely thermo-mechanical process for the devulcanization of rubber, and therefore of its recycling, is lacking.

1.4 Aims of the study

The aim of this study is to investigate the environmental impacts of an integrated system able to carry out a thermo-mechanical devulcanization process of rubber at industrial scale. Therefore, the analysed process, without requiring devulcanizing agents, allows the recycling of rubber-based components, extending the useful life of the material. More in detail, the investigated recycled materials are gaskets in ethylene-propylene diene monomer (EPDM) (Restrepo-Zapata et al., 2013), sulfur vulcanized, deriving from the automotive sector. Although different thermo-mechanical devulcanization processes by co-rotating twin-screw extruders (cTSE) have been proposed (Meysami et al., 2017; Diaz et al., 2018; Pirityi and Pölöskei, 2021), their practical implementation has not been completely satisfactory till now. The process here considered, i.e. the thermo-mechanical devulcanization process by cTSE, patented by Maris SpA, allows the devulcanization without the use of devulcanizing additives, supercritical CO_2 or solvents, obtaining a product re-usable in a virgin blend without altering its characteristics. Moreover, most sulfur/peroxide-crosslinked rubbers can be recycled in this way. The methodology used to perform the analysis of environmental impacts is the LCA and data to model the system refer to a real plant (Maris EVOREC RUBBER line, with a 58mm cTSE) that carries out the devulcanization of EPDM waste. First, the attention was focused on a hotspot analysis of the devulcanization process, in order to identify the steps in the process responsible for the greatest environmental burden. Subsequently, the actual consequences of the recycling of EPDM were investigated; a comparative analysis between the use of a virgin EPDM compound and a devulcanized EPDM compound was carried out. This comparison is necessary to obtain information on the actual environmental implications of the recycling process. In fact, it is of primary importance to carry out comparative evaluations of the processes and technologies considered enabling for sustainable development, with respect to existing technologies and approaches. In addition, at the present time, there is no effective analysis of the environmental implications of recycling these materials in a closed cycle compared to the use of virgin rubbers.

2. Materials and methods

LCA methodology, through the collection and quantification of resources, energy consumption, emissions, and waste, allows to quantify and interpret the environmental burdens associated to the entire life cycle of a product or a process (Baumann and Tillman, 2004). The approach to LCA analysis has been carried out following the UNI EN ISO 14040-44 standard and the guidelines of the International Reference Life Cycle Data System (ILCD) (European Commission - Joint Research Centre - Institute for Environment and Sustainability, 2010; ISO, 2006a, 2006b). According to the requirements of the ISO 14040 and 14044 standards, four steps are required to conduct an LCA: goal and scope definition, inventory analysis, life cycle impact assessment and interpretation of the results. The whole work was carried out using the SimaPro 9.2 software and the Ecoinvent v.3.7.1 database.

2.1 Goal and scope definition

The definition of the objectives and the field of application is the first step of an LCA analysis, and it defines the purpose of the study, the system boundaries, and the functional unit for all flows (Curran M.A., 2017). The analysis examined the production of the integrated system that allows the devulcanization process of EPDM. As anticipated, the goal is to identify, within the studied system, units and processes that involve high environmental impacts (defined as hotspots). To carry out a more accurate analysis, the study has also examined the use phase, in order to divide the environmental burdens generated by each process unit over the entire life cycle of the system. The identified functional unit (FU), to which all inventory flows refer, corresponds to the production of 120 kg of devulcanized EPDM in 1 hour. Such amount has been chosen in order to represent the real situation, in fact it is the real quantity of EPDM treated in 1 hour by the cTSE. To facilitate data collection and process modelling, the integrated system, capable of carrying out the devulcanization of the EPDM, has been divided into unit processes. Fig. 1 reports a graphic representation of the devulcanization line.

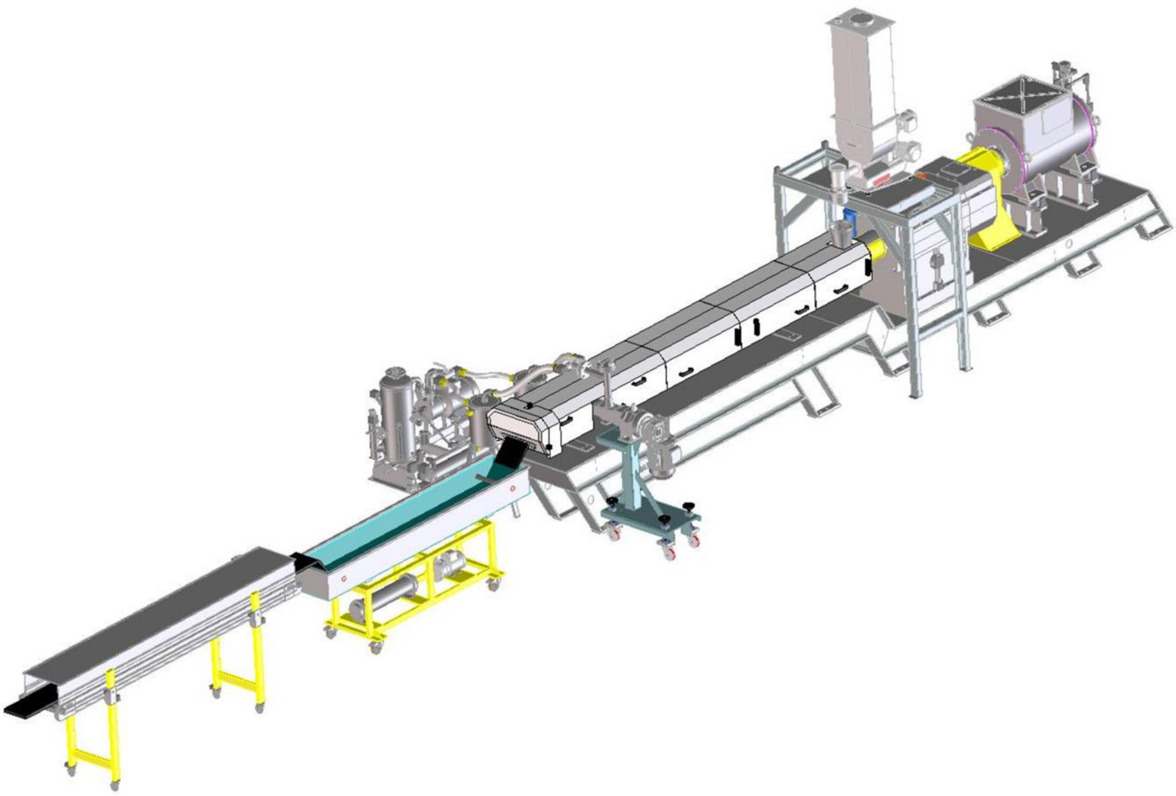

Fig. 1 Graphic representation of the devulcanization line (Maris S.P.A. property).

Figure 2 shows the scheme of the units that make up the system and the dashed box reports the system boundary for the environmental analysis. The analysis takes into consideration both the materials used for the production of all the physical units of the system and the materials and energies spent during the use phase.

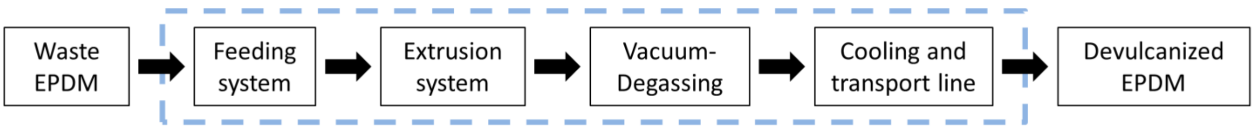

Fig. 2 Scheme of the units that allow the thermo-mechanical EPDM devulcanization process; the dashed box indicates the system boundary for the LCA.

The devulcanization process, shown in Fig. 1, allows to break C-S and S-S bonds, which are generated during the vulcanization step of EPDM. In fact, selective breaking of the crosslinking points, limiting the C-C bonds breaking, can occur thanks to the application of adequate temperatures and shear stresses during the devulcanization process (Fukumori et al., 2003; Jana and Das, 2005; Sabzekar et al., 2015).
The analyzed plant basically consists of an extruder, which allows to obtain 120 kg of devulcanized rubber per hour. The first part of the system consists of a gravimetric feeding system that allows to feed free-flowing rubber waste into the extruder in a constant and continuous way. Then, there is the central part of the system where the co-rotating twin-screw extruder is located, followed by elements that allow vacuum extraction and degassing. As the rubber passes through the extruder, it is subjected to specific temperatures and shear forces that allow the breaking of the bonds created during vulcanization. Finally, to decrease the temperature of the material leaving the extruder, there is a cooling tank, followed by a transport line that collects the devulcanized EPDM.

2.2 Inventory analysis

The inventory analysis is the second phase of the LCA, with the aim of identifying and quantifying the raw materials, resources, energy consumption, outputs and emissions related to the functional unit considered (Rebitzer et al., 2004). Collecting all this information allowed to build a model that represents the real system investigated. The data to model the system were obtained from direct measurements (primary data) and from the literature; as describe by Sonnemann et al. (2010), an attributional approach was used to compile the inventory. Table 1 shows details of the inventory of the various components contributing to the production and use phase of the integrated system. The system use phase involves the use of consumables (Table 1) and electricity (Table 2). The “life time” column shows the estimated life of each input, at the end of this period the material is considered therefore to be replaced. In this regard, it should be noted that, apart from consumables and components subject to wear, for which the duration of the life cycle is exactly known, for all other inputs the reported values have been estimated based on the knowledge of professionals in the sector and of the possible market trend. The system is assumed to operate 8 hours/day for 253 days/year.

Process phase	Input	Amount	Ecoinvent dataset	Lifetime
Feeding system	Feeding unit**	10 kg	Steel. low-alloyed	10 years
	Steel (feeding system)	63 kg	Reinforcing steel	20 years
	Steel (feeding system)	27 kg	Steel. chromium steel	20 years
Extrusion system	Extruder motor	1150 kg	Electric motor. vehicle	20 years
	Steel (extrusion system)	817 kg	Steel. unalloyed	20 years
	Steel (extrusion system)	2180 kg	Cast iron	20 years
	Steel (extrusion system)	2453 kg	Reinforcing steel	20 years
	Lubrication pump**	25 kg	Pump	10 years
Vacuum-degassing system	Vacuum system – pump**	40 kg	Pump	10 years
	Side degassing unit**	24 kg	Pump	10 years
	Steel (vacuum system)	80 kg	Reinforcing steel	20 years
	Steel (vacuum system)	80 kg	Steel. chromium steel	20 years
	Steel (Degassing system)	123 kg	Reinforcing steel	20 years
	Steel (Degassing system)	53 kg	Steel. low-alloyed	20 years
Cooling and transport line	Pump (cooling tank)**	20 kg	Pump	10 years
	Conveyor belt**	20 kg	Conveyor belt	20 years
	Air knife**	30 kg	Steel. low-alloyed	10 years
	Steel (cooling tank)	114 kg	Reinforcing steel	20 years
	Steel (cooling tank)	266 kg	Steel. chromium steel	20 years
	Steel (conveyor belt)	350 kg	Reinforcing steel	20 years
Consumables	Steel (bushings)	100 kg	Steel. chromium steel	8000 hours
	Steel (screws)	120 kg	Steel. chromium steel	4000 hours
	Lubricating oil (extrusion system)	120 kg	Lubricating oil	4000 hours
	Lubricating oil (degassing system)	3 kg	Lubricating oil	4000 hours
	Anti-stick	0.012 kg/FU	Zinc Stearate*	/
	Activated carbon**	0.06 kg/FU	Activated carbon	/
Process phase	**Output**	**Amount**	**Ecoinvent dataset**	**Lifetime**
Waste treatment	Condensed degassing liquid	0.6 kg/FU	Treatment of spent solvent mixture	/

Table 1 Inventory of the materials required to build and use the entire integrated system; *modelled by authors; ** data with the greater uncertainties for modelling (object of study in the sensitivity analysis).

Since it is rather complex to trace all materials that make up the entire system, some assumptions have been fixed to simplify the modelling of the process. To model the extruder motor, the dataset, present on the Ecoinvent database, relating to an electric motor (Electric motor, vehicle {GLO} | market for | Cut-off, U), suitably scaled according to mass, was used as input. The dataset relating to a pump (Pump, 40W {GLO} | market for | Cut-off, U) was used to represent: the lubrication pump, the vacuum system - pump, the side degassing unit, and the pump for the material recirculation in the cooling tank. Also in this case, the input has been scaled based on the mass of each component. In the process plant, each component corresponding to a single step not outlined before (feeding system, extrusion system, vacuum-degassing system, cooling, and transport line) is mainly constituted on steel. So, the inventory was completed by reporting the specific quantities, broken down by the type of steel. The feeding unit and the air knife were modelled by introducing a quantity of steel equal to the original weight of this system component. Among consumables, an anti-stick agent is used in the cooling tank; this material, on a commercial level, can assume different compositions and in this specific case it consists of zinc stearate. This substance was modelled using stearic acid and zinc as input, in stoichiometric quantities according to the chemical composition (for 1 kg of substance: 0.897 kg of stearic acid and 0.103 kg of zinc).

During the modelling phase of the unit processes of the integrated system, it was assumed that, on average, the system operates 8 hours a day, for 253 days a year.

Table 2 shows the energy consumed by the plant in 1 hour of operation during the devulcanization process by the various components of the integrated system. The reported energy consumptions are average values, measured during the devulcanization of different EPDMs. All information reported in Table 2 are primary data, i.e. directly measured during the plant operation. It was assumed that the devulcanization process takes place in Italy, therefore the energy consumption was modelled using electricity data obtained from the national grid (Ecoinvent dataset: Electricity, medium voltage {IT} | market for | Cut-off, U).

Process phase	Input electricity	Amount / kWh
Feeding system	feeding unit	0.5
Extrusion system	extruder motor	58
	extruder heaters	28
	lubrication pump	1.3
	extruder thermostatic unit	3.5
Vacuum-degassing system	vacuum system	2.5
	side degassing unit	0.9
Cooling and transport line	pump cooling tank	1.1
	cooling tank exchanger	14
	air knife	0.5
	conveyor belt	1.0

Table 2 Inventory of the energy requirements per functional unit.

2.3 Impact assessment

In the third phase of LCA, the set of resources and emissions, which constitute the inputs and outputs of the inventory, are converted into a series of environmental impact categories. In this study, the environmental indicators, evaluated at midpoint level using the EF 3.0 method (Fazio et al., 2018), are: Climate change, Ozone depletion, Photochemical ozone formation, Acidification, Eutrophication (freshwater), Ecotoxicity (freshwater), Resource use (fossils), Resource use (minerals and metals).
To determine the relative contribution of the considered categories, the impacts were also assessed on a global level by a normalization. The used normalization factors of the Environmental Footprint (EF) 3.0 method are reported in Table 3 (Crenna et al., 2019). The normalized results allow to have an indication to what extent the analyzed product system contributes to the total environmental burdens, compared to the impacts generated in a year by a person.

Impact category	Unit per year	Global NF for EF per person
Climate change	kg CO_2 eq	$8.05*10^{+03}$
Ozone depletion	kg CFC-11 eq	$4.83*10^{-02}$
Photochemical ozone formation	kg NMVOC eq	$4.06*10^{+01}$
Acidification	mol H^+ eq	$5.55*10^{+01}$
Eutrophication (freshwater)	kg P eq	$1.61*10^{+00}$
Ecotoxicity (freshwater)	CTUe	$4.26*10^{+04}$
Resource use (fossils)	MJ	$6.50*10^{+04}$
Resource use (minerals and metals)	kg Sb eq	$6.37*10^{-02}$

Table 3 Environmental Footprint (EF) 3.0 normalisation factors (NF): global impact per year per person.

To facilitate the modelling, as reported in Table 1, data for feeding unit, lubrication pump, vacuum system - pump, side degassing unit, pump (cooling tank), conveyor belt, air knife and activated carbon have been defined by assumptions. The uncertainties associated with these inputs were evaluated by means of a sensitivity analysis, as reported below in Section 3.1: the individual impacts of the inputs have been increased and decreased by 50%, in order to assess the relative weight of their contributions on the total impact for each category.

2.4 Comparative analysis

In addition to the evaluation of the integrated system, that allows to carry out the devulcanization of the EPDM, the study was extended by including two comparative analyses. These comparisons were made to understand the magnitude of the environmental burdens generated by the devulcanization process modelled in a more realistic context. The goal is to highlight whether the here analyzed large-scale use of the thermo-mechanical devulcanization process can lead to an effective reduction of the environmental impacts when compared to a classic linear use (i.e. production, use, dispose) of EPDM materials. This reduction in impacts could allow a decrease in the use of virgin EPDM, with a consequent reduction of the quantity of rubbers disposed of in landfills.

2.4.1 First comparison

For the first case, to highlight the impacts generated by the production step, the analysis compared the production of 1 kg of virgin EPDM compound (v-EPDM) with 1 kg of EPDM obtained from the thermo-mechanical devulcanization process (r-EPDM). In this case, a "cut-off" approach was used, which involves the exclusion from the system boundaries of all the impacts attributable to the previous life of the material. As regards the r-EPDM, only the devulcanization process is considered in the modelling, as if the material had never previously generated any environmental impact. Thus, we applied a linear proportion of the previously obtained results. The functional unit for this comparison was the production of 1 kg of EPDM compound. The choice to focus the attention exclusively on the production of 1 kg of EPDM was aimed to simplify the system studied. The impacts to produce v-EPDM were estimated by considering the Ecoinvent dataset “Synthetic rubber {RER} | production | Alloc Rec, U”. This dataset considers the polymerization, extrusion, and vulcanization phases of EPDM elastomer production; therefore, since the objective is to evaluate the impact for a generic v-EPDM compound (pre-extrusion and vulcanization), contributions related to the extrusion and vulcanization phases have been neglected from the values of the Ecoinvent dataset. Consequently, only contributions from materials (base polymers and additives) and energy to obtain an EPDM elastomer sheet were considered (Kellenberger et al., 2000). The main objective of this first comparison is to highlight the different burdens on the different impact categories analyzed, generated by the production of v-EPDM and r-EPDM.

2.4.2 Second comparison

In the second case, a more realistic and structured system was examined: it was assumed to compare, through the “system expansion” approach (Shen et al., 2010), two different system models, each of which allows to obtain two rubber products. This comparison is necessary due to the different physicochemical properties of the devulcanized materials compared to the virgin ones. To take this difference into account, it was assumed to model the production of the second product (Product 2, in Fig. 3) using a combination of 30% r-EPDM with 70% v-EPDM; the product 2 obtained in this way shows the same physicochemical properties of a product generated using 100% of virgin material. If higher percentages of r-EPDM would be used, there would be a loss of mechanical properties and, consequently, the final application would be different. Fig. 3 shows the life cycle diagram of the two compared systems: for System 1, both the two products are manufactured using v-EPDM compounds, whereas in System 2, v-EPDM is used for Product 1 and both v-EPDM and r-EPDM are used for Product 2.

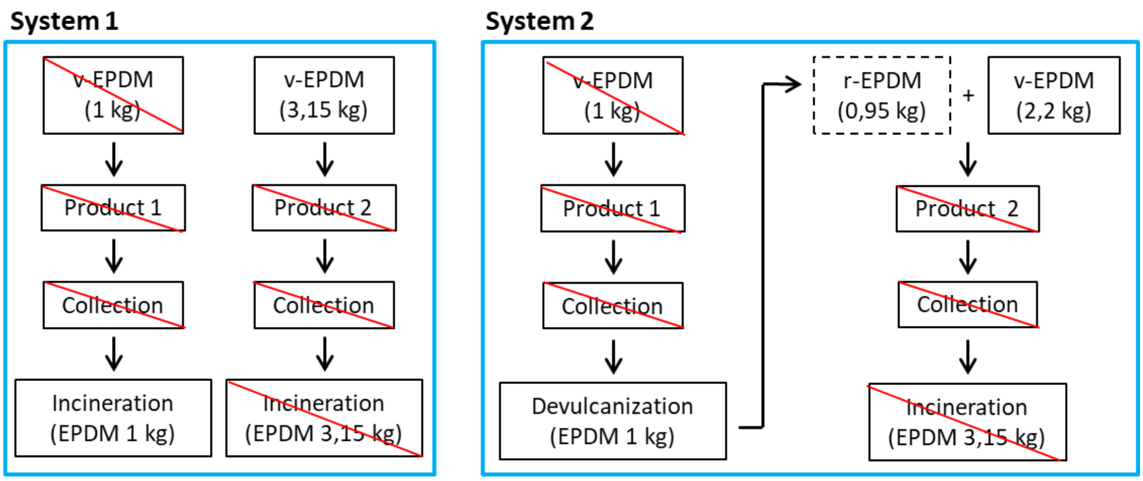

Fig. 3 Comparison, through the "system expansion" approach, of two systems to produce EPDM rubber products. EPDM amount for each product is indicated. The boxes crossed out in red correspond to the equivalent unit processes between the two systems.

It can be observed that in System 1 the end-of-life phase occurs by incineration (both for the first and for the second product), while in System 2 the first product undergoes a devulcanization process that allows its recycling. The quantities of raw material (EPDM) for product 2 have been assumed to use all the devulcanized EPDM, and the mixing ratio between v-EPDM and r-EPDM is 70:30. It should be noted that there is a 5% loss of the material during the devulcanization process and therefore 0.95 kg of recycled material is obtained following the devulcanization process of 1 kg of EPDM (as obtained from product 1). As shown in Fig. 3, the analysis of the two systems is greatly simplified if the equivalent processes are neglected (boxes crossed out in red). For System 1, only impacts associated with the incineration of 1 kg of rubber and the production of 3.15 kg of virgin EPDM compound need to be considered. Instead, for System 2, it is enough to consider the impacts for the devulcanization process, which allows to obtain 0.95 kg of r-EPDM, and the impacts to produce 2.2 kg of v-EPDM compound.

The incineration process has been modelled using the dataset, present on the Ecoinvent database, "Waste rubber, unspecified {Europe without Switzerland}| treatment of waste rubber, unspecified, municipal incineration | Cut-off, U"; the avoided impacts by the production of heat and electricity, obtained from the incineration process, were also taken into account. As mentioned previously, the impacts to produce v-EPDM compound were estimated by considering the modified dataset "Synthetic rubber {RER} | production | Alloc Rec, U".

The results shown in the second comparison were enriched by carrying out an uncertainty analysis of the data at the inventory level. The Pedigree matrix was applied to estimate the uncertainty caused by temporal, geographical and technological gaps (Frischknecht et al., 2007; Guo and Murphy, 2012). Based on the uncertainty of the inventory data, expressed as a probability distribution, the Monte Carlo function, created thanks to the SimaPro software, was performed with 1000 iterations at a significance level $\alpha=0.05$.

3. Results and discussion

Table 4 shows the environmental impacts for the devulcanization process of 120 kg of EPDM, using the here investigated thermo-mechanical technology. In detail, the table reports the results of analysed impact categories grouped per: system (physical components of the devulcanization system), consumables (inputs subject to wear and consumption), electricity and waste. To facilitate the understanding of the table, a heat map is introduced, for each impact category (each column), with a color gradient: the greatest impacts are indicated in red, instead the lowest in green.

Impact category		Climate change kg CO_2 eq	Ozone depletion kg CFC11 eq	Photochemical ozone formation kg NMVOC eq	Acidification mol H^+ eq	Eutrophication (freshwater) kg P eq	Ecotoxicity (freshwater) CTUe	Resource use (fossils) MJ	Resource use (minerals and metals) kg Sb eq
Total		52.63	$6.94*10^{-06}$	0.11	0.23	0.01	531.93	762.22	$1.62*10^{-04}$
System	Feeding system	0.01	$3.24*10^{-10}$	$2.54*10^{-05}$	$3.22*10^{-05}$	$2.50*10^{-06}$	0.18	0.07	$1.17*10^{-07}$
	Extrusion system	0.38	$1.96*10^{-08}$	$1.78*10^{-03}$	$2.08*10^{-03}$	$2.71*10^{-04}$	21.31	4.00	$4.03*10^{-05}$
	Vacuum-degassing system	0.03	$1.67*10^{-09}$	$1.44*10^{-04}$	$2.08*10^{-04}$	$2.61*10^{-05}$	2.33	0.34	$5.18*10^{-06}$
	Cooling and transport line	0.16	$9.32*10^{-09}$	$6.23*10^{-04}$	$7.50*10^{-04}$	$7.45*10^{-05}$	4.84	1.76	$3.42*10^{-06}$
Consumables	Bushings & screws	0.18	$8.67*10^{-09}$	$6.45*10^{-04}$	$1.15*10^{-03}$	$6.17*10^{-05}$	6.26	1.99	$6.65*10^{-06}$
	Lubricating oil	0.04	$2.34*10^{-08}$	$8.16*10^{-04}$	$2.70*10^{-04}$	$1.18*10^{-05}$	1.12	1.90	$6.21*10^{-07}$
	Anti-stick	0.01	$6.87*10^{-10}$	$4.13*10^{-05}$	$6.14*10^{-05}$	$6.43*10^{-06}$	0.88	0.13	$2.05*10^{-06}$
	Activated carbon	0.48	$1.30*10^{-08}$	$1.48*10^{-03}$	$3.60*10^{-03}$	$2.81*10^{-04}$	15.52	6.58	$2.45*10^{-07}$
Electricity	EPDM feeding	0.20	$2.76*10^{-08}$	$4.33*10^{-04}$	$8.84*10^{-04}$	$4.44*10^{-05}$	1.90	3.01	$4.15*10^{-07}$
	Extrusion system	40.97	$5.58*10^{-06}$	$8.74*10^{-02}$	$1.78*10^{-01}$	$8.95*10^{-03}$	383.04	607.28	$8.38*10^{-05}$
	Vacuum-degassing system	1.53	$2.09*10^{-07}$	$3.27*10^{-03}$	$6.68*10^{-03}$	$3.35*10^{-04}$	14.34	22.74	$3.14*10^{-06}$
	Cooling and transport line	7.47	$1.02*10^{-06}$	$1.59*10^{-02}$	$3.25*10^{-02}$	$1.63*10^{-03}$	69.82	110.69	$1.53*10^{-05}$
Waste	Spent solvent treatment	1.19	$3.99*10^{-08}$	$5.45*10^{-04}$	$7.14*10^{-04}$	$1.69*10^{-04}$	10.39	1.74	$7.28*10^{-07}$

Table 4 Environmental impacts for the EPDM devulcanization process.

The table illustrates that main impacts are associated to the electricity consumption. In detail, the energy required by the extrusion system, to heat (extruder heaters) and rotate (extruder motor) the screw extruder, causes the highest impacts for all the categories. In fact, as shown by the inventory analysis, the technology underlying the devulcanization process is quite simple and the contributions of the units for the construction of the integrated system are spread over a considerable time span. On the contrary, the energy demand to make the process running is particularly marked, thus determining the highest impacts. The cooling and transport system can be considered as the second main cause of impacts for almost all categories; these impacts are again due to the consumption of electricity. The Resources use (minerals and metals) category differs from this trend; in fact the second major cause of impact is due to the extrusion system (mainly made up of steel).

In general terms, it can be said that the consumption of electricity is the main responsible for the environmental impacts of the thermo-mechanical devulcanization process for the EPDM recycling.

Fig. 4 reports the LCA results of the whole system normalized with respect the impact generated in a year by a person.

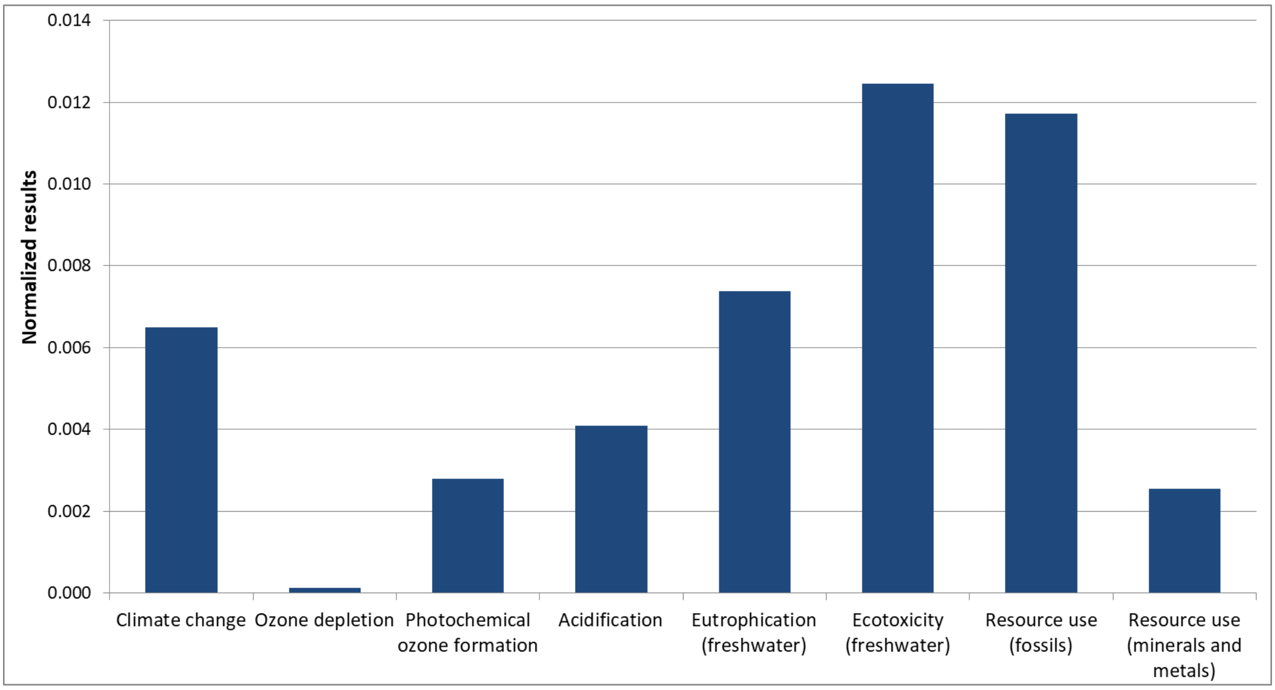

Fig. 4 Normalized results for the EPDM devulcanization process.

It can be observed that the category that shows the greatest relevance is that of Ecotoxicity (freshwater), followed by the impact on Resource use (fossils), Eutrophication (freshwater), Climate change, Acidification, Photochemical ozone formation, Resource use (minerals and metals), and Ozone depletion. The impact on the latter category seems negligible compared to all the others. The burdens of the Ecotoxicity (freshwater) category are mainly associated with the use of hard coal, that represents one of the energy sources for Italian electricity production (the input data used in the Ecoinvent database referred to the year 2017, when around 13% of Italian electricity production was made using hard coal). The use of fossil resources for energy production is obviously also highlighted by the impact of the category Resource use (fossils). It worth noting that the impact of the category Resource use (minerals and metals), partly caused using steel in the extrusion system, shows little relevance compared to the other impact categories.

3.1 Sensitivity analysis

Fig. 5 shows the percentage change of the burdens for each category, obtained by varying the impacts of individual inputs, modelled with the greatest uncertainties. The inputs in question are feeding unit, lubrication pump, vacuum system - pump, side degassing unit, pump (cooling tank), conveyor belt, air knife and activated carbon. As previously described, the individual impacts of these inputs have been increased and decreased by 50% with respect to the reference values (i.e with no variation) reported in Table 4, associated with the devulcanization process of 120 kg of EPDM.

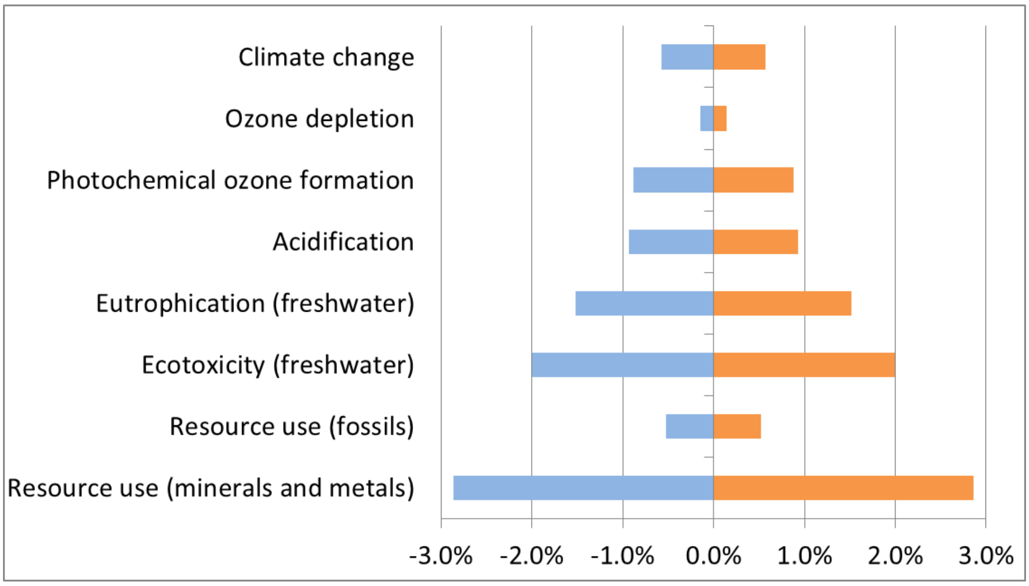

Fig. 5 Sensitivity analysis: percentage change in impacts for each category.

In Fig. 5 it can be seen how the percentage variation of the impacts for the individual inputs, modelled with high uncertainty, affects the final burdens very slightly. The major variations were obtained for the categories: Resource use (minerals and metals) (±2.9%), Ecotoxicity (freshwater) (±2.0%) and Eutrophication (freshwater) (±1.5%). All the other categories show changes in impact less than 1%. The lowest absolute variation was reported for the category Ozone depletion (±0.1%), which, as already shown in Fig. 4, shows very minimal impacts. In general, since the variations for all the categories are less than 3%, they can easily be considered negligible, confirming the validity of the original modelling.

3.2 Comparative analysis

3.2.1 LCA results - first comparison

To assess the relative environmental burdens of the recycled EPDM, obtained through the devulcanization process, the production of 1 kg of v-EPDM, before the extrusion and vulcanization, was compared with 1 kg of r-EPDM, as obtained from the devulcanization process. The results are reported in Table 5, which shows the environmental impacts calculated at the step of characterization.

Impact category	Unit	v-EPDM	r-EPDM
Climate change	kg CO_2 eq	$2.12*10^{+00}$	$4.39*10^{-01}$
Ozone depletion	kg CFC11 eq	$6.17*10^{-07}$	$5.79*10^{-08}$
Photochemical ozone formation	kg NMVOC eq	$1.04*10^{-02}$	$9.43*10^{-04}$
Acidification	mol H^+ eq	$1.14*10^{-02}$	$1.90*10^{-03}$
Eutrophication (freshwater)	kg P eq	$6.40*10^{-04}$	$9.89*10^{-05}$
Ecotoxicity (freshwater)	CTUe	$4.34*10^{+01}$	$4.43*10^{+00}$
Resource use (fossils)	MJ	$7.14*10^{+01}$	$6.35*10^{+00}$
Resource use (minerals and metals)	kg Sb eq	$4.91*10^{-05}$	$1.35*10^{-06}$

Table 5 Environmental impacts for 1 kg of virgin EPDM compound (v-EPDM) compared with 1 kg of devulcanized EPDM (r-EPDM).

Looking at the results reported in Table 5, it is possible to note that the devulcanization process allows to obtain EPDM with considerably lower impacts compared to the virgin one. In fact, the value for the r-EPDM is on average one order of magnitude smaller than those calculated for v-EPDM.

Fig. 6 shows the comparison between the normalized environmental impacts of 1 kg of v-EPDM and r-EPDM, with respect to the factors of the EF 3.0 method.

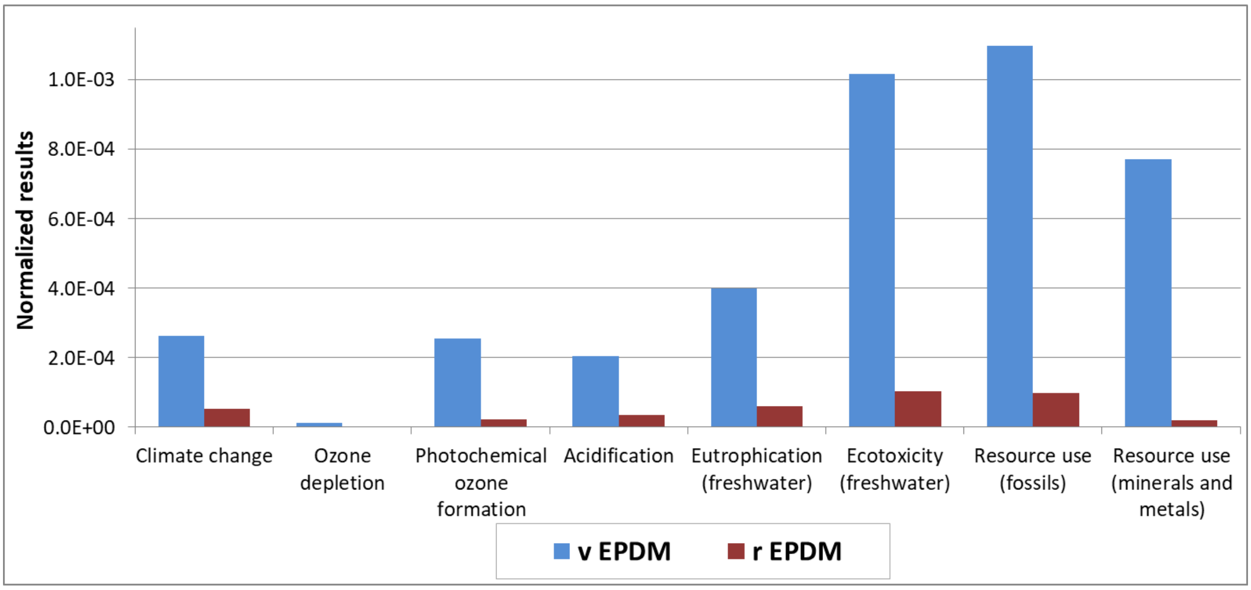

Fig. 6 Normalized environmental impacts for the comparison of 1 kg of v-EPDM (blue bars) and 1 kf of r-EPDM (red bars).

The normalized results were reported to show the relative contributions of the impacts among the considered categories, when switching from v-EPDM to r-EPDM. As regards the v-EPDM, the most relevant impact is related with the categories: Resource use (fossils), Ecotoxicity (freshwater) and Resource use (minerals and metals). The impacts on these categories are mainly caused using carbon black, respectively Ecotoxicity (freshwater) and Resource use (fossils) and of the catalyst to activate the polymerization reaction for the category Resource use (minerals and metals). Instead, as already seen in Fig. 4, the devulcanization process, that allows to produce r-EPDM, shows the major impact among the categories Ecotoxicity (freshwater) and Resource use (fossils). As already pointed out, these impacts are mainly related to the energy consumption, from the Italian national mix, during the phases of the devulcanization process.

3.2.2 LCA results - second comparison

The results, with the associated uncertainties, of a comparison based on the "system expansion" approach, as previously described, are reported in Fig. 7. To facilitate the comparison between the two systems and the impact categories considered, results have been normalized, according to the factors reported in Table 3. Results for System 1 consider the reduction of the impact due to the recovery of energy and heat, following the EPDM incineration process.

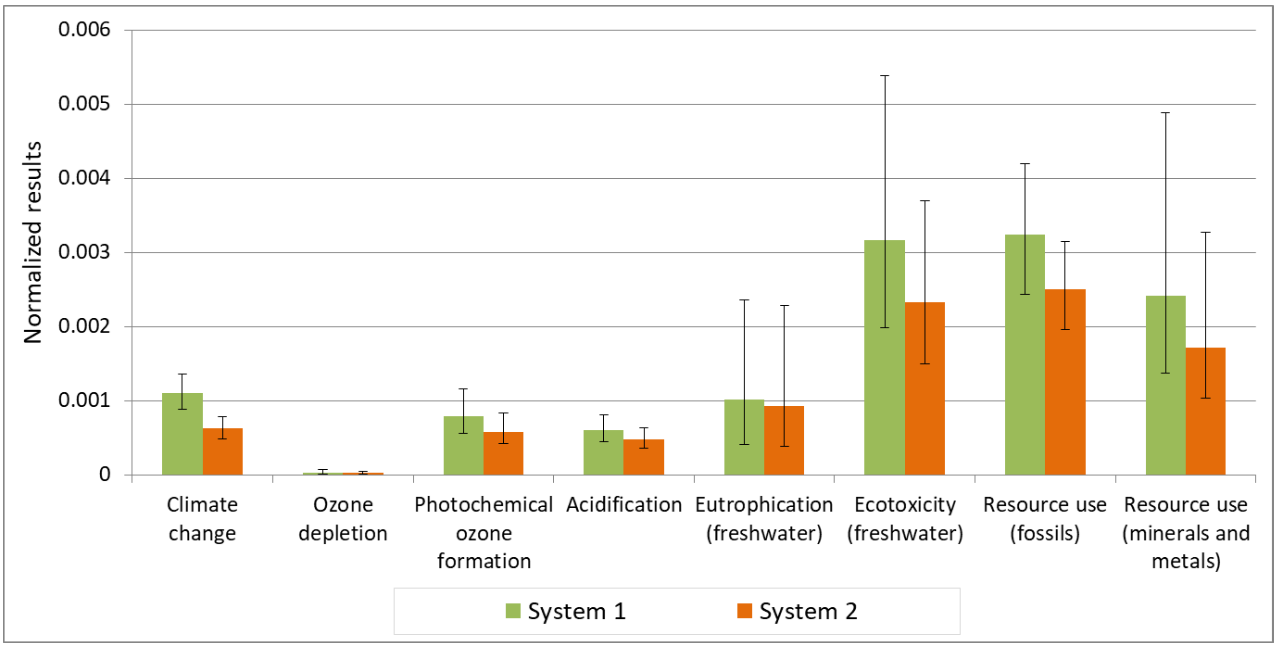

Fig. 7 Normalised results, with associated uncertainties, for the comparison between the two systems using the "system expansion" approach.

Fig. 7 shows that impacts generated in System 2, modelled assuming one recycling phase of the EPDM, are lower than impacts in System 1 (modelled using always virgin EPDM); this trend is common for all impact categories. The impact for the category Ozone depletion is difficult to be distinguished, because it is one order of magnitude lower than all other values (i.e. System 1: $3.51*10^{-05}$; System 2: $2.63*10^{-05}$).
For both compared systems, the main impacts are obtained for the categories: Resource use (fossils), Ecotoxicity (freshwater) and Resource use (mineral and metals). This result is mainly attributable to the use of virgin EPDM, which, as shown in Table 5, shows the greatest relative impacts for these categories. The most marked reduction in impact linked to the use of r-EPDM is obtained for the category Climate change (approximately 43% of impact reduction); this result is obtained because the EPDM incineration phase, despite allowing the recovery of energy and heat, generates CO_2 eq emissions higher than those avoided. On the other hand, the category Eutrophication (freshwater) shows only a 8% reduction of impact, moving from System 1 to System 2. The results obtained by modelling System 2 show an impact reduction between 20% and 29% for all the other categories (Ozone depletion 25%; Photochemical ozone formation 26%; Acidification 21%; Ecotoxicity (freshwater) 26%; Resource use (fossils) 23%; Resource use (minerals and metals) 29%).
Figure 7 also shows the uncertainty ranges for the normalized LCA profiles of the two systems analyzed. The error bars, calculated individually for each system, suggest how great uncertainty is introduced for different impact categories, in both systems. The most affected category is Eutrophication (freshwater), followed by Ecotoxicity (freshwater) and Resource use (mineral s and metals). Contrary all the other categories show low variance. The higher confidence associated with the results on Climate change, Ozone depletion, Acidification, Resource use (fossils) indicates that the environmental profiles of the systems analyzed represent reality with respect to these impact categories and also provides a better basis for comparisons between the two systems. To avoid misinterpretation relating to correlations, a Monte Carlo simulation was performed to estimate the uncertainties generated by comparisons between the two systems. The results, shown in Figure A.1 (see Appendix A) show the graphical distribution of the difference between System 1 (A) and System 2 (B). It is possible to observe that there is over 94% probability that the impacts generated in System 2 are lower than those associated with System 1, for almost all the categories considered. The only exception is represented by the category Eutrophication (freshwater); for this

category the analysis shows a lower probability, equal to 72%, that the environmental impact generated by System 2 is actually lower than that obtained by System 1. In this case the certainty with which to state which system represents the most environment friendly solution, is therefore slightly lower.

Both comparisons made in this study underline how recycled EPDM, obtained through a thermo-mechanical devulcanization process, allows to limit the environmental impacts compared to the production of virgin material. It should be noted that, although the second comparison already represents a more realistic situation, for an even more extensive and more reliable modeling it would be better to also evaluate the environmental burdens of the entire end-of-life phase of the EPDM waste: i.e. collection, sorting and transport towards the treatment plant. However, the contributions of these phases are expected to generate limited environmental impacts over the entire life cycle of the material. This reason, associated with the lack of information and / or specific technologies, to be used on a large scale, are the main motivation behind the choice to neglect these aspects in the analysis.
At the same time, in order to obtain a better and more comprehensive evaluation of all the EPDM recycling solutions, a comparison with other technologies applied for the treatment of EPDM waste materials should be addressed in future works.

4. Conclusions

This work investigated the environmental impacts associated with a thermo-mechanical devulcanization process at industrial level, which allows the recycling of EPDM waste from automotive gasket. Main conclusions can be summarized as follow:

- The analysis focused on the identification of the main environmental hotspots of the process and then the focus of the study was expanded by carrying out comparative analyses of devulcanized EPDM with the alternative represented by virgin EPDM compound. Eight impact categories have been evaluated: Climate change, Ozone depletion, Photochemical ozone formation, Acidification, Eutrophication (freshwater), Ecotoxicity (freshwater), Resource use (fossils) and Resource use (minerals and metals). The results were estimated both at the characterization phase and after normalization with respect to the average impact generated by a person in a year.
- The analysis of hotspots highlighted that, at the characterization level, the main contribution of the devulcanization process to the environmental impacts, for all categories, is due to energy consumption.
- The normalized results showed that the categories most affected by the devulcanization process are Ecotoxicity (freshwater) and Resource use (fossils). The burdens of the components of the process, modeled with high uncertainty, were tested by varying the relative impacts (range ±50%) and subsequently the contribution of this variation on the final impact was assessed. The analysis showed that energy consumption is responsible for most of the impacts and this is minimally influenced by the possible variations of the elements, with the greatest modeling uncertainty.
- The comparative analysis was initially conducted using a simple “cut-off” approach, comparing 1 kg of virgin EPDM compound with 1 kg of recycled EPDM. The results showed that the devulcanized product offers a constantly better environmental profile than the virgin alternative. Subsequently, the comparison was conducted by a "system expansion" approach, where two different systems were compared, considering the entire life cycle (from cradle to grave) of two products made of EPDM. This analysis made it possible to model a situation close to reality and, therefore, to obtain more reliable results. What emerged from this comparison is how the devulcanization process,

which allows the recycling of EPDM waste, reduces the environmental impact associated with the life cycle of the rubber products.

As a conclusion, the thermo-mechanical devulcanization of EPDM has proved to be a process that allows both to recycle waste and to limit the environmental burdens of this material. The results presented in this work have made it possible not only to evaluate a specific process, but in the perspective of a sustainable development and a transition to the circular economy, they can be used as a motivation to push companies and policy makers to incentivize the collection and recycling of EPDM based materials. Since the impacts of this process are mainly related to the type of energy source used, it can be assumed that they can be further reduced by relying on renewable sources and optimizing the system to avoid any possible energy dissipation.

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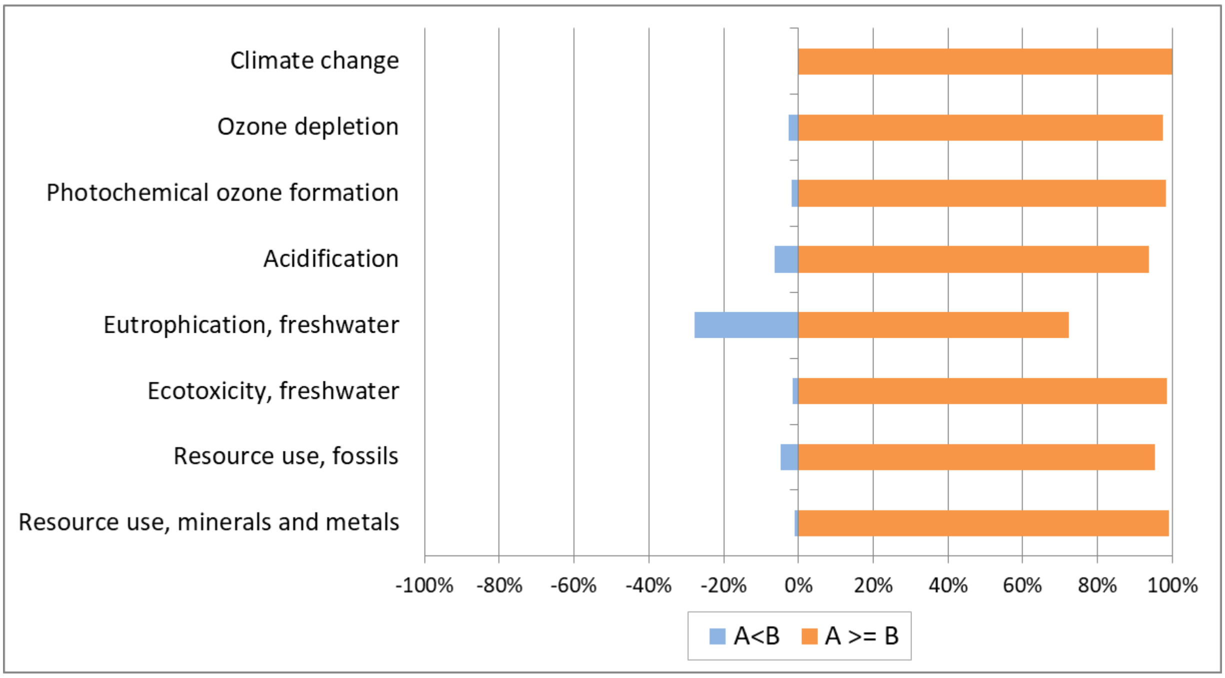
Appendix A

Figure A.1 Monte-Carlo simulation results of normalized impact assessment comparison between System 1 and System 2 (unit: A =System 1; B =System 2).



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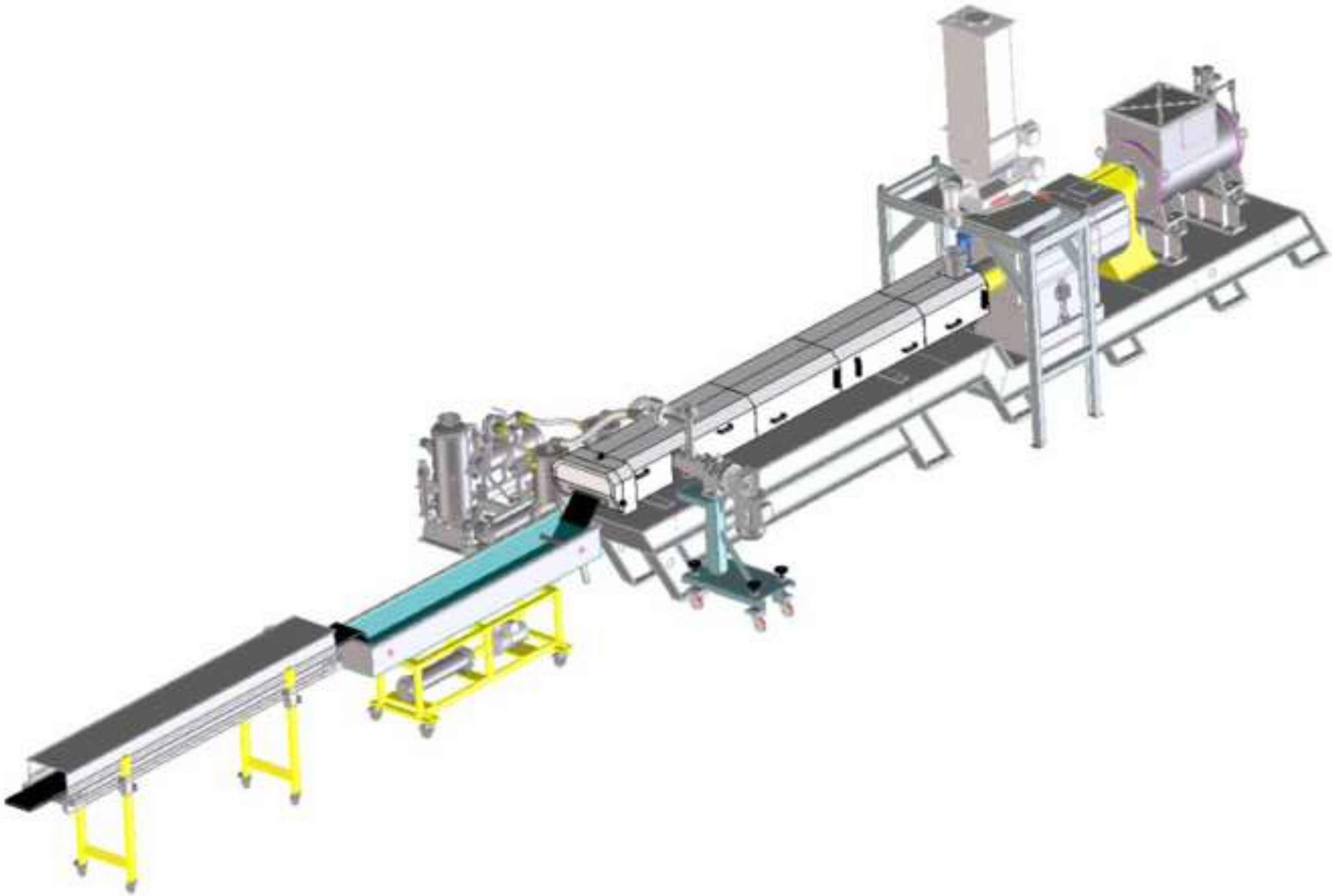

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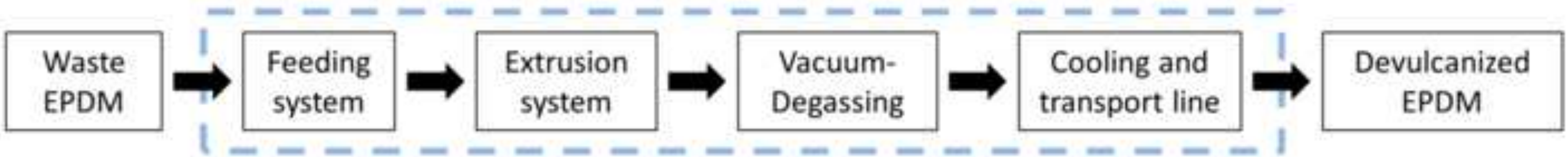

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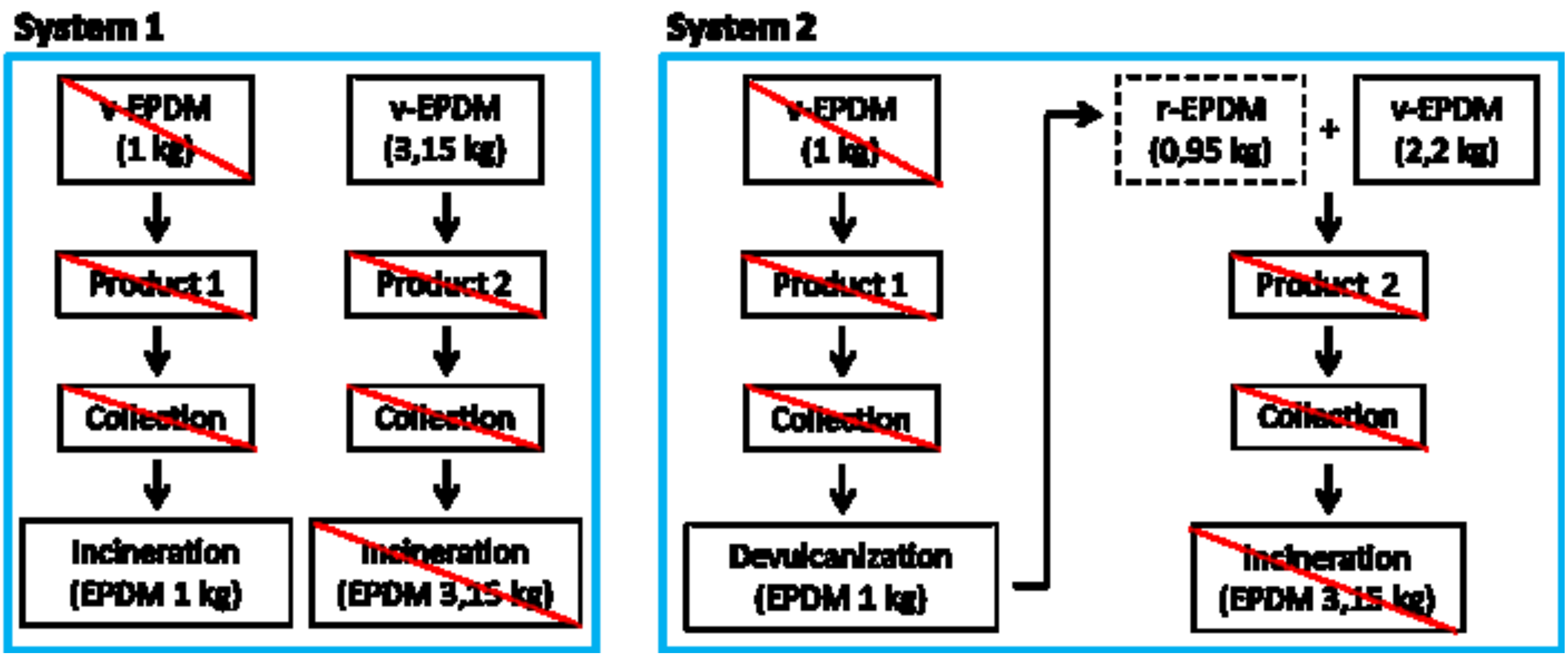

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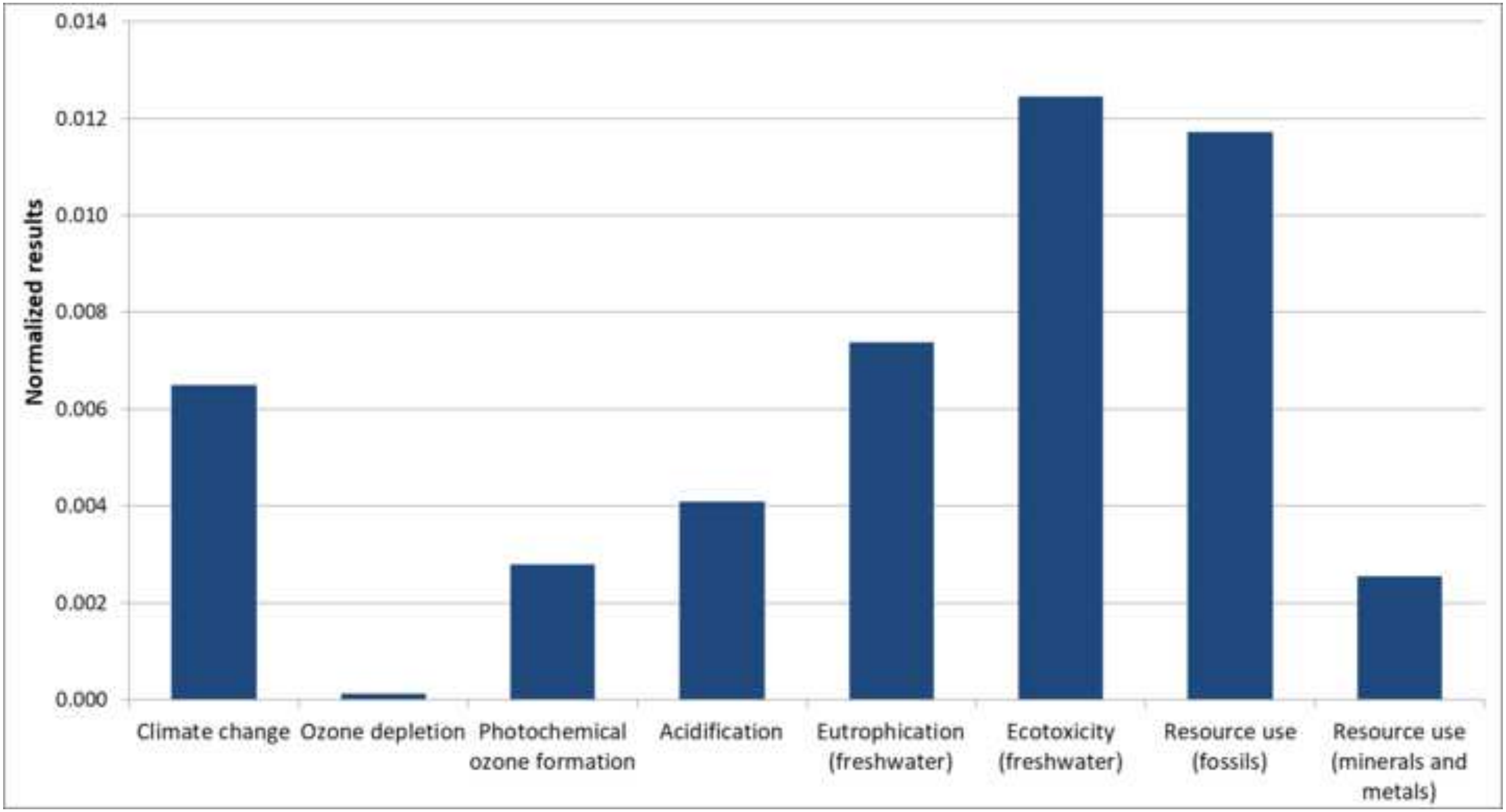

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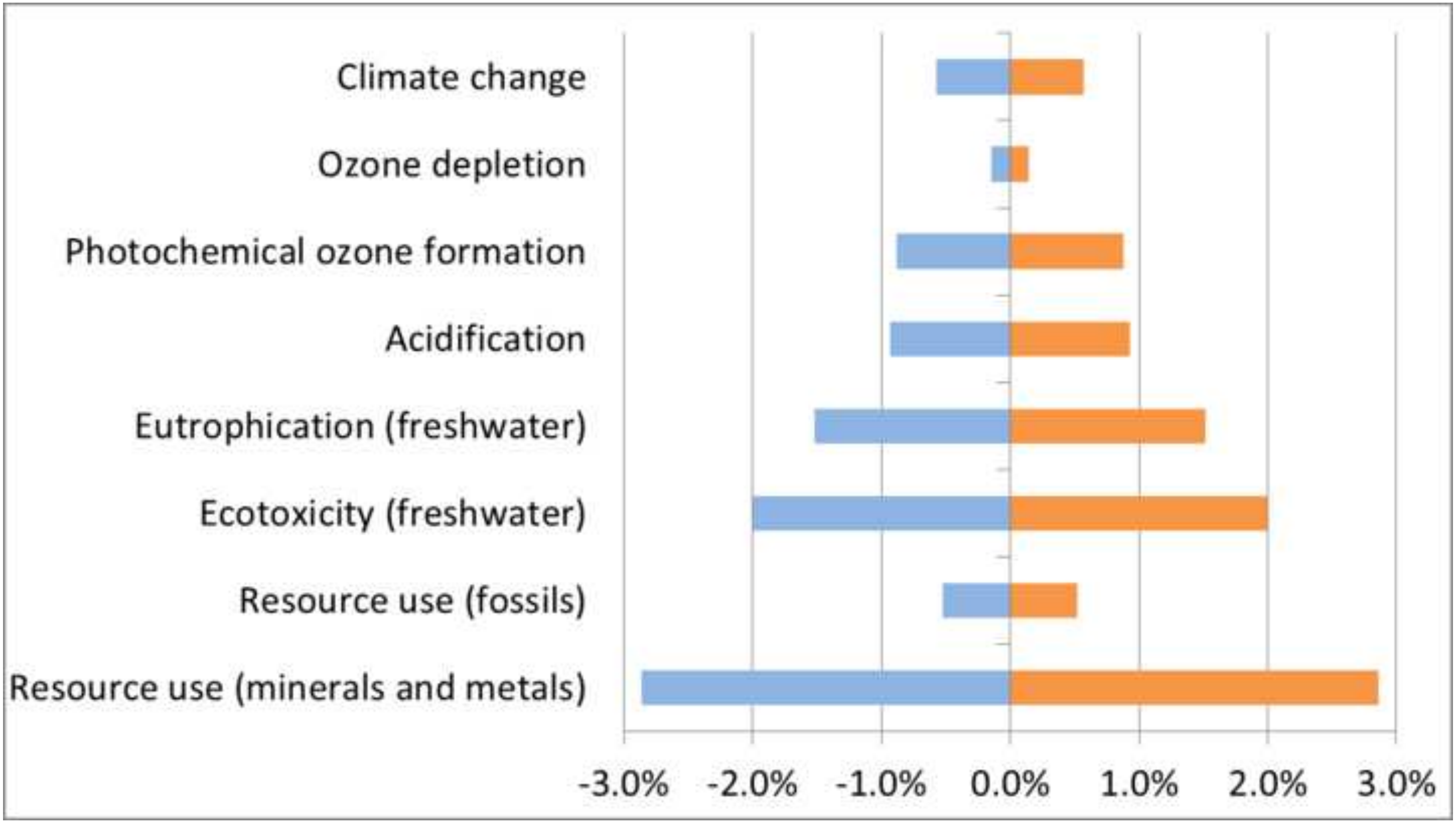

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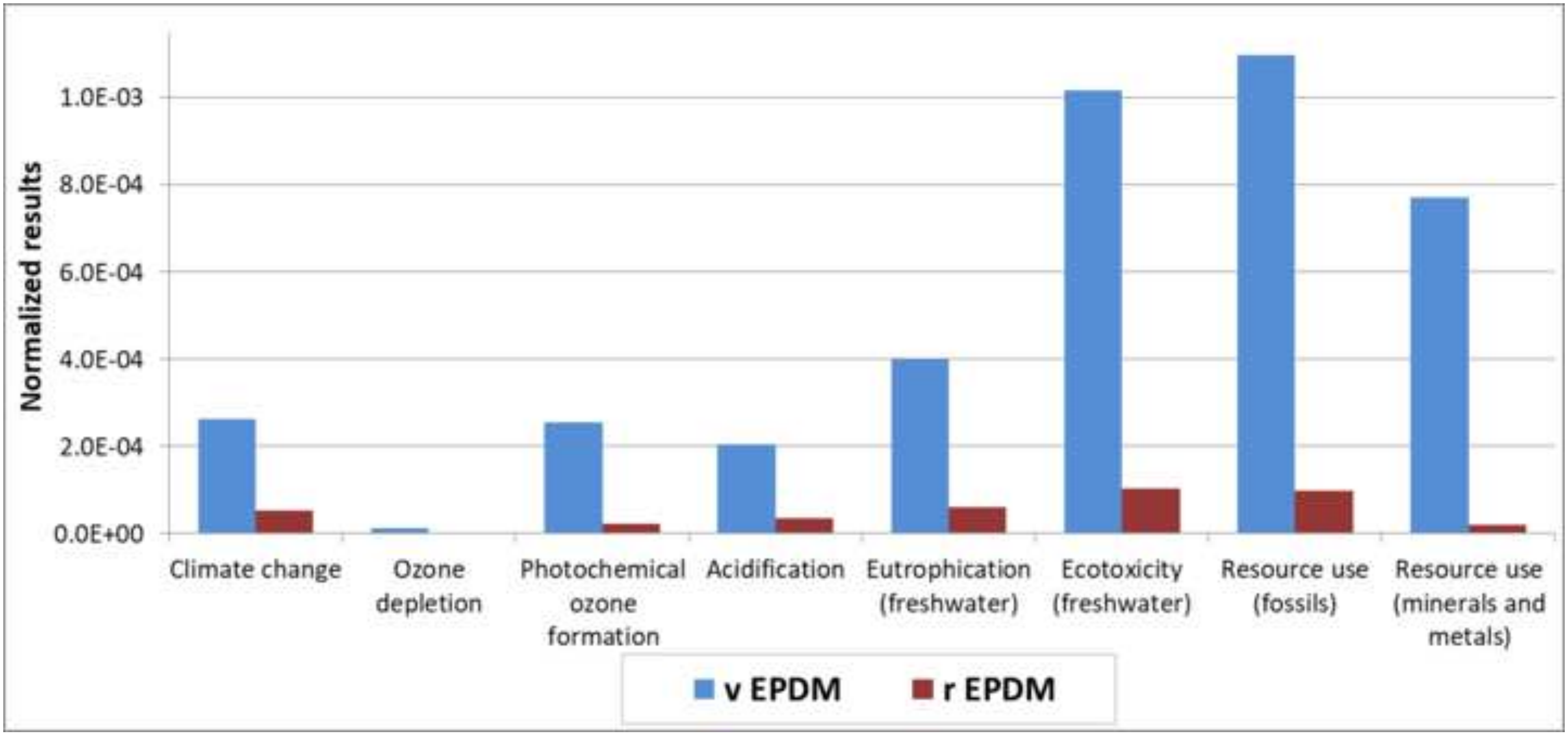

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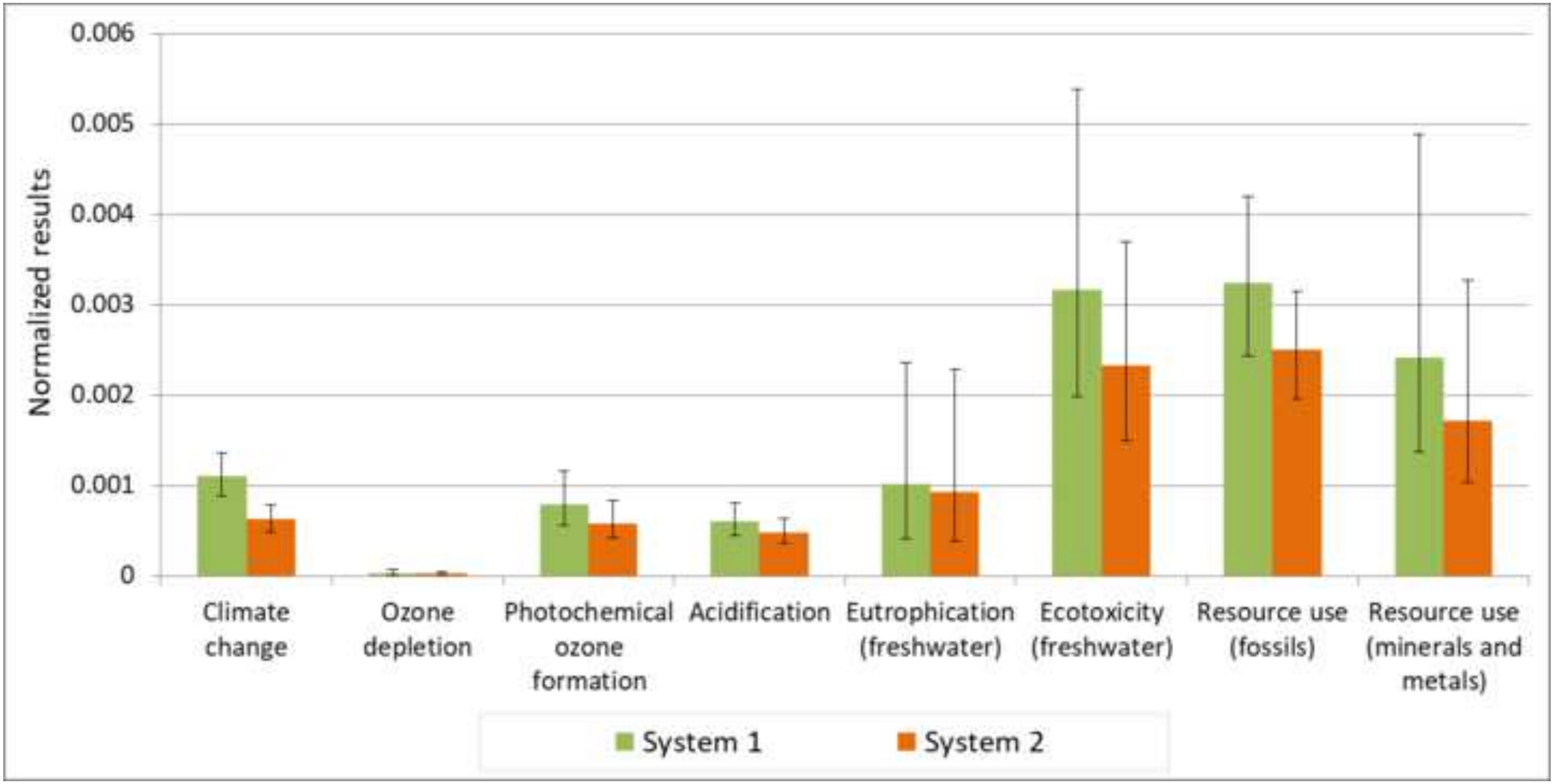

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