LIFE CYCLE ASSESSMENT OF A FIBRE-REINFORCED POLYMER MADE OF GLASS FIBRE AND PHENOLIC RESIN WITH BROMINATED FLAME RETARDANT

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Abstract

A life cycle assessment (LCA) for fibre-reinforced polymers consisting on glass fibre and phenolic resin with a brominated flame retardant additive is presented. Due to its toxicity and difficulties in recycling, new environmental friendly materials are being designed. It is intended that this LCA, which is based on new data life cycle inventory (LCI) serves as a benchmark.LCA results obtained with these new LCI are compared with results from LCI of commercial database. The environmental impacts associated with energy consumption and air emissions were assessed, as well as other environmental impacts resulting from the extraction and processing of materials and fibre-reinforced polymers manufacturing. End-of-life scenarios for recycling, incineration and landfilling,including the environmental impacts of brominated flame retardants,were also compared for this material.

Keywords:*life cycle assessment; fibre-reinforced polymer; glass fibre; phenolic resin; brominated flame retardant*

Resumen

Se presenta el análisis del ciclo de vida (ACV) para un polímero de resina fenólica reforzado con fibras de vidrio y con retardante de llama bromados como aditivo. Debido a su toxicidad y dificultades en el reciclaje, se están buscando nuevos materiales alternativos. Se pretende que este ACV, que está basado en datos de nuevos inventarios del ciclo de vida (ICV) sirva como base de comparación. Los resultados del ACV obtenidos con estos nuevos ICV se comparan con los resultados de ICV de bases de datos comerciales. Se han evaluado los impactos ambientales asociados al consumo de energía y emisiones al aire, así como otros impactos resultantes de la extracción y procesado de los materiales y fabricación de los polímeros.Además, se han comparado tres escenarios de fin de vida: reciclaje, incineración y disposición en vertedero, incluyendo en todos ellos los impactos ambientales de los retardantes de llama bromados.

Palabras clave: *análisis del ciclo de vida; polímero reforzado con fibras; fibra de vidrio; resina fenólica; retardante de llama bromado*

1. Introduction

Fibre-reinforced polymer (FRP) materials are commonly used in the aerospace, automotive, marine, and construction industries. FRP is a composite material made of a polymer matrix reinforced with fibres. The fibres are usually glass, carbon, basalt or aramid, although other fibres such as paper, wood or asbestos can also be used. The polymer is usually an epoxy, vinylester or polyester thermosetting plastic, or phenol formaldehyde resins. The FRP considered in this research comprise glass fibre, phenolic resin and Deca-BDE as flame-retardant additive. These are noxious materials and this composite is not easily recyclable.

Deca-BDE is a brominated flame retardant used in polymers and composites for applications that demand high fire resistance. Deca-BDE belongs to the group of polybrominated diphenyl ethers (PBDEs). PBDEs are persistent, bioaccumulative, and toxic to both humans and the environment. Human exposure to PBDEs may occur through occupations that manufacture flame retardants or products that contain flame retardants, as well as in end-of-life operations.

The European Community and the United States government have expressed concern about the use of PBDEs as flame retardants. The EU Directive restricting the use of hazardous substances in electrical and electronic equipment (RoHS Directive or Directive 2002/95/EC) prohibits the use of Deca-BDE since July 2006. In December 2012, the European Chemicals Agency announced the inclusion of the flame retardant Deca-BDE in the Candidate List of Substances of Very High Concern (SVHCs) for Authorization under REACH regulation. The authorization procedure aims to assure that the risks from SVHCs are properly controlled and that these substances are progressively replaced by suitable alternatives while ensuring the good functioning of the EU internal market. In June 2013, Norway announced that it submitted a proposal to consider the listing of Deca-BDE under the Stockholm Convention as a persistent organic pollutant (POP). The Stockholm Convention aims for the elimination of POPs and requires that they are disposed of in such a way that the POP content is 'destroyed or irreversibly transformed so that they do not exhibit the characteristics of POPs'. It also permits to dispose of POPs in an environmentally sound manner 'when destruction or irreversible transformation does not represent the environmentally preferable option' or 'when the POP content is low'. If the proposal of Norway is finally adopted, the inclusion of Deca-BDE in the Candidate List of SVHCs will cease to be valid and the use of Deca-BDE will be definitively prohibited as previously happened with other PBDEs listed as POPs in the Stockholm Convention (e.g., Penta-DBE and Octa-BDE).

Furthermore, the question of how to dispose of end-of-life thermoset composite parts is growing in importance. Traditional disposal routes such as landfill and incineration are becoming increasingly restricted, and composites companies and their customers are looking for more sustainable solutions.

A life cycle assessment (LCA) has been carried out for FRP made of glass fibre and phenolic resin with a brominated flame retardant additive to be used for future comparative assessments with other environmental friendly substitutes. Besides gaining knowledge about the environmental impact of FRP by the results of LCA, one additional aim of the study was to obtain new life cycle inventories, which environmental results are compared to LCI of commercial databases.

The LCA was further extended by the consideration of flame retardants at the end-of-life calculations. As a result of LCA, environmental impacts by life-cycle stages and by FRP components furthermore the end-of-life impacts by incineration, mechanical recycling and landfilling is discussed in the paper.

2. Materials and methods

The LCA methodology was used in this study to calculate the environmental impacts of a FRP made of glass fibre and phenolic resin and a brominated flame retardant additive. LCA was applied according to the guidelines provided by the International Organization for Standardization (ISO, 2006a, 2006b). The LCA software application SimaPro® was used to tackle the development of the study more effectively. The ReCiPe method (Goedkoop et al., 2009) was then used to assess the environmental impacts according to two sets of impact categories: midpoint categories and endpoint categories. Midpoint impact categories are climate change, fossil depletion, and another sixteen impact categories. Endpoint impact categories are damage to human health, damage to ecosystem diversity, and damage to resource availability. The Cumulative Energy Demand method was also used to assess the total energy.

2.1 Goal and scope definition

The present study aimed to calculate the environmental impacts of a FRP made of glass fibre and phenolic resin with flame retardant. One kilogram of FRP is assumed here as the functional unit, which is a reference unit to which the results of the LCA are related. These results will serve as a baseline for comparison with novel FRP materials made from renewable polymers and natural fibre reinforcements, which will be assessed in further studies.

The scope of this study includes all processes from raw material extraction until delivery of the FRP at plant (cradle-to-gate analysis). The end-of-life of the FRP is also considered in the study to compare the environmental impacts of different treatments that can be applied to FRP products when these become waste. The life cycle of the FRP being studied can thus be divided into these major stages or sub-systems: (1) materials, (2) manufacturing, and (3) end-of-life.

2.2 Inventory analysis

2.2.1 Materials

The FRP being studied is composed of glass fibres and phenolic resin modified with a halogenated flame-retardant additive (decabromodiphenyl ether or Deca-BDE). Table 1 shows the composition of the finished FRP and the total amount of each material required for manufacturing one kilogram of FRP, including rejects and waste.

Table 1: Materials required for manufacturing 1 kg of FRP

Material	Composition of FRP (% by weight)	Total amount, including waste (kg)
Phenolic resin	34.4	0.406
Glass fibre	60.0	0.708
Deca-DBE	5.6	0.066

Phenolic resins are synthetic polymers obtained by reaction of phenol with formaldehyde. The phenolic resin used in the FRP studied is composed of 95% phenol and 5 % formaldehyde. The LCI of formaldehyde was taken from the Ecoinvent® database (Althaus et al., 2007) and includes data for all processes from raw material extraction until delivery of formaldehyde at plant. The LCI of phenol was based onthe Eco-profiles of the European Plastics Industry (Boustead, 2005) and it also includes data for all processes from raw material extraction until delivery of phenol at plant. Additional data for the production of phenolic resin were also taken from the Ecoinvent® database (Althaus et al., 2007), including transport of materials to the manufacturing plant, infrastructure of the plant, water consumption, process energy demand, and emissions to air and water from production.

Glass fibres are prepared from a mixture of the so-called E-glass in the form of continuous strands with a size coating and a binder. A LCI of glass fibres was available from the Ecoinvent® database (Kellenberger et al., 2007), which includes data for all processes from raw material extraction until delivery of glass fibres at plant. An alternative LCI of glass fibres was developed here based on the reference document on best available techniques for the manufacture of glass (Joint Research Centre, 2013), which includes a specific section for glass fibre products. This inventory includes the resources, raw materials and chemicals used for production (silica sand, limestone, dolomite, aluminium oxide, and so forth); process energy demand (light fuel oil and natural gas used for melting of glass, and electricity used for forming glass strands); emissions to air and water from production; and wastes occurring during the production process. Table 2 shows the LCI of one kilogram of glass fibres manufactured with best available techniques.

Table 2: LCI of 1 kg of glass fibres manufactured with best available techniques.

Input/output	Amount	Unit	Input/output	Amount	Unit
MATERIALS					
Silica sand	0.565	kg	Aluminium oxide	0.135	kg
Limestone	0.205	kg	Titanium dioxide	0.0075	kg
Dolomite	0.025	kg	Potassium oxide	0.005	kg
Colemanite	0.025	kg	Sodium hydroxide	0.005	kg
Borax	0.025	kg	Fluorspar	0.005	kg
ELECTRICITY AND FUELS					
Electricity	4.1	MJ	Natural gas	6.2	MJ
Light fuel oil	6.2	MJ			
EMISSIONS TO AIR					
Carbon dioxide	0.1	kg	Selenium	0.00000445	kg
Particulates	0.000245	kg	Chromium VI	0.0000045	kg
Fluoride	0.00009	kg	Antimony	0.000014	kg
Chloride	0.00005	kg	Lead	0.000014	kg
VOC	0.0003	kg	Chromium III	0.000014	kg
Arsenic	0.0000045	kg	Copper	0.000014	kg
Cobalt	0.00000445	kg	Manganese	0.000014	kg
Nickel	0.00000391	kg	Vanadium	0.00001292	kg
Cadmium	0.00000448	kg	Tin	0.000014	kg
EMISSIONS TO WATER					
COD	0.0004385	kg	Copper	0.00000195	kg
Suspended solids	0.000195	kg	Crcompounds	0.00000195	kg
Fluoride	0.000039	kg	Cdcompounds	0.000000325	kg
Hydrocarbons	0.0000975	kg	Ni compounds	0.00000325	kg
Lead compounds	0.00000113	kg	Ammonia	0.000065	kg
Antimony	0.00000325	kg	Boron	0.000013	kg
Arsenic	0.00000195	kg	Sulfate	0.0065	kg
Barium	0.0000195	kg	Phenol	0.0000065	kg
Zn compounds	0.00000325	kg	Tin	0.00000325	kg
WASTE TO TREATMENT					
Inorganic waste	0.155	kg	Inert waste	0.007	kg

Decabromodiphenyl ether (Deca-BDE) is a brominated flame retardant used in polymers and composites for applications that demand high fire resistance. Deca-BDE belongs to the group of polybrominateddiphenyl ethers (PBDEs). The LCI of Deca-BDE was taken from the Ecoinvent® database (Sutter, 2007) and includes the raw materials and chemicals used for production, transport of materials to the chemical plant, infrastructure of the plant, water consumption, process energy demand, and emissions to air and water from production of diphenyl ether compounds.

2.2.2 Manufacturing

The different materials composing the FRP are processed and mixed at the manufacturing plant to produce a sheet moulding compound. Sheet moulding compound (SMC) is an integrated ready-to-mould composition of glass fibres, polymer resin, fillers and additives. This compound is made by chopping continuous fibres onto the surface of a resin paste that is conveyed on a thin plastic carrier film. This mixture of fibres and resin is further covered by another layer of resin on a second plastic carrier film. Compaction rollers knead the fibres into the resin for uniform fibre distribution and wetting. The compound sandwiched between the carrier films is gathered into rolls and stored until it matures. The carrier films are then removed and the material is cut into charges of predetermined weight and shape. The charges are placed on the bottom of two mould halves in a compression press. Heat and pressure act on the charge until it is fully cured, and then it is removed from the mould as the finished FRP product. Figure 1 shows the manufacturing process for FRP products.

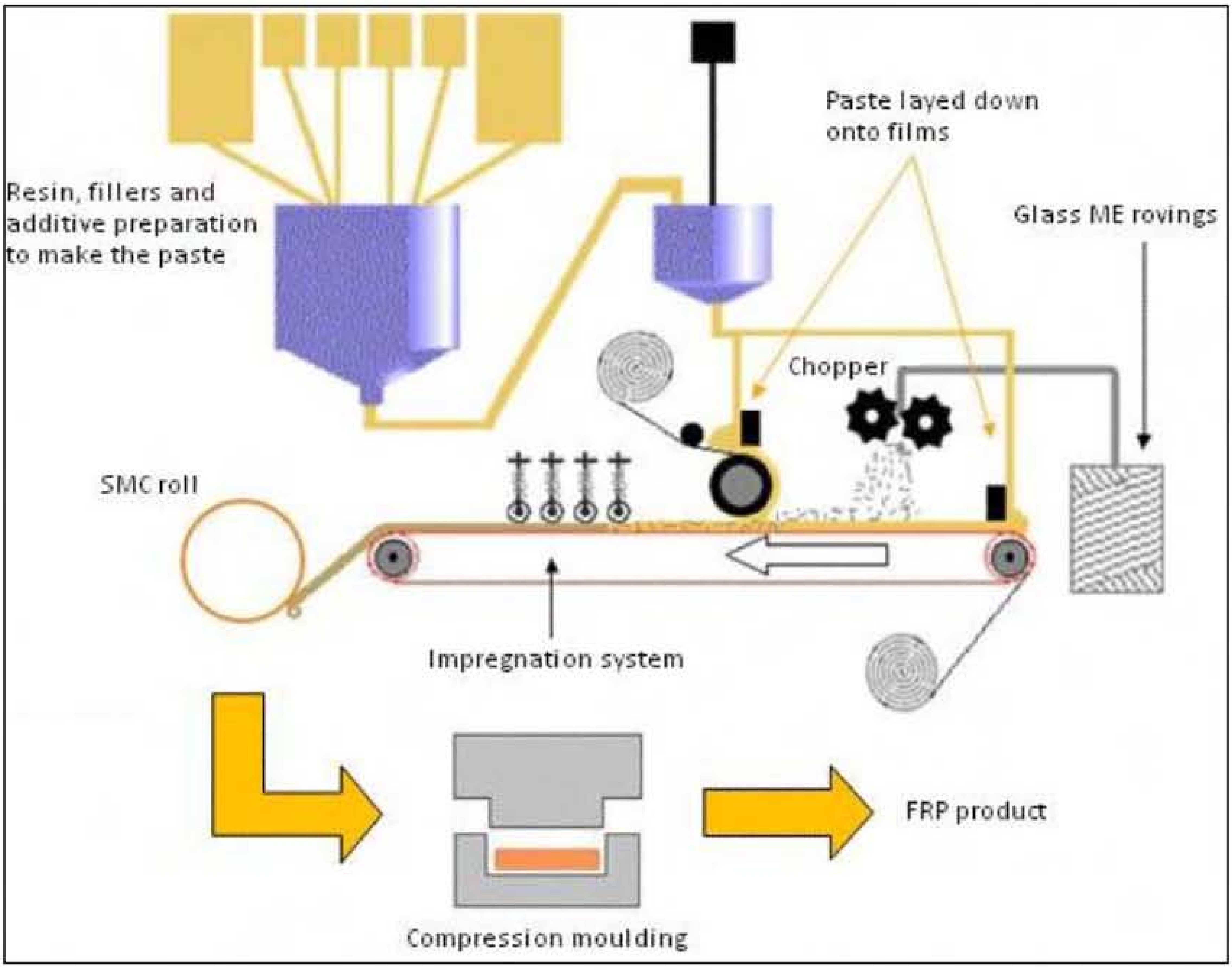

Figure 1: Manufacturing process for FRP products (Aimplas, 2010).

Data on auxiliaries, process energy demand, and wastes occurring during the manufacturing process were based on field data from an international Spanish company. Table 3 shows the LCI of the production of one kilogram of FRP product.

Table 3: LCI of the manufacturing of 1 kg of FRP product.

Input/output	Amount	Unit
ELECTRICITY		
Electricity, medium voltage	4.55E+01	MJ
AUXILIARIES		
Hydraulic oil	5.00E-03	kg
Water	5.64E-01	kg
EMISSIONS TO AIR		
Particulates	1.52E-01	g
WASTE TO TREATMENT		
Polymer waste	1.79E-01	kg
Hydraulic oil	2.41E-03	kg
Wastewater	2.85E-06	m^3

2.2.3 End-of-life

The last stage in the life cycle of the FRP is the end-of-life. When FRP products reach this stage, the alternatives considered in this paper are: disposed of in a landfill, recycled or incinerated (with the possibility of energy recovery). The environmental burdens associated with this stage can be quite different depending on the end-of-life strategy that is chosen.

2.2.3.1 Mechanical recycling

Mechanical recycling process starts with the size reduction of the composite scrap by low speed cutting or crushing (to 50–100 mm). The size is then reduced down to 10 mm to 50 µm through a hammer mill or other high speed millings for fine grinding. Mechanical treatment of FRP produce short milled fibres used as filler reinforcement materials.

The LCI of mechanical recycling of thermosetting-based panels was based on a study by Hedlund-Åström (2005), who measured the fuel and electricity consumption for the different processes involved (compression, cutting, shredding, and grinding). It was assumed that the production of 1 kg of filler (limestone) from raw materials is avoided as a result of recycling 1 kg of residual polymer composite.

2.2.3.2 Incineration

The model of Doka (2009) was used to estimate the LCI of waste disposal in a municipal solid waste incinerator (MSWI). The incineration of waste leads to direct air and water emissions as well as land use burdens. Indirect burdens are also originated from the consumption of auxiliaries, energy and infrastructure materials. The solid wastes remaining after incineration are landfilled: slag (bottom ash) is landfilled in a slag compartment, while fly ash and scrubber sludge are solidified with cement and disposed of in a residual material landfill.

Brominated flame retardant requires special attention during incineration. Research from Wang et al. (2010) was used to quantify PBDEs, polybrominateddibenzo-p-dioxins and dibenzofurans (PBDD/Fs), and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs).

In this paper, based on the findings of Wang et al. (2010), it was assumed that 2.45E-06% of Deca-BDE is undestroyed in the incineration process. The amount of undestroyed Deca-BDE was allocated between the three routes: bottom ashes, 91.2%; fly ashes, 1.9%; and the stack flue gases, 6.9%. Furthermore, PBDD/Fs in the input materials were found not destroyed in the combustion. Moreover, a ratio of 3.05E-04 was assumed between Deca-BDE and PBDD/Fs emissions. The abundant 1,2,3,4,6,7,8-HpBDF and OBDF in the bottom ashes may be the side compounds in the commercial Deca-BDE mixtures (Hanari et al.,2006). The disposal of bottom ashes of MSWIs in sanitary landfills should contribute PBDEs into the environment, and not be ignored from the developing PBDE inventory. Furthermore, the high PBDE content in bottom ashes may also contribute the PBDE exposures of the workers who handle and operate with these substances. The burdens associated with the incineration of one kilogram of Deca-BDE were thus inventoried as shown in Table 4.

Table 4 Burdens from the incineration of 1 kg of Deca-BDE.

Input/output	Amount
	Stack flue gases (emissions to air)
Deca-BDE (kg)	1.70E-09
PBDD/Fs (kg)	5.18E-13
	Bottom ashes (waste to sanitary landfill)
Deca-BDE (kg)	2.23E-08
PBDD/Fs (kg)	6.82E-12
	Fly ashes (waste to hazardous waste landfill)
Deca-BDE (kg)	4.70E-10
PBDD/Fs (kg)	1.44E-13

2.2.3.3 Landfill

The model of Doka (2009) was used to estimate the LCI of waste disposal in a municipal solid waste landfill (MSWL). The incineration of waste leads to direct air and water emissions as well as land use burdens. Indirect burdens are also originated from the consumption of auxiliaries, energy and infrastructure materials.

Brominated flame retardant requires also special attention during landfilling.The expected leachate from deca-BDE flame retardant will be 3, 81E-20 kg (Danon-Schaffer MN, 2006).

3. Results

The environmental impacts of this material were assessed both at the midpoint level and the endpoint level. Sensitivity of LCA to different inventory databases were analysed and LCA results by life cycle stages, material components and end-of-life scenarios were discussed.

3.1 Comparison of results of different inventory data

LCA from inventory data of phenolic resin from Eco-profiles of the European Plastics Industry (Boustead, 2005) have been compared with LCA from inventory data from Ecoinvent® database (Althaus et al., 2007). The results from Eco-profiles of the European Plastics Industry are the most reliable due to the updated database. The phenolic resin from Eco-profiles obtain better results in issues as human toxicity, photochemical oxidation, ionizing radiation, fresh water eutrophication, marine eutrophication, fresh water eco-toxicity, marine eco-toxicity, agricultural land occupation, water depletion, land depletion and fossil depletion. Values in which phenolic resins is worse are in climate change, ozone depletion, particular matter, terrestrial acidification, terrestrial eco-toxicity and natural land transformation (Figure 2).

Impacts of phenolic resin calculated from Eco-profiles data are less harmful regards in points as fossil depletion due to the optimization of energy consumption along the time. In other aspects the impacts of phenolic resin from Eco-profiles are worse and more restrictive than impacts of resin from Ecoinvent due to that the resin from Eco-profiles has been conducted with a more comprehensive study.

LCA from the inventory data of glass fibres from reference document on best available techniques for the manufacture of glass (Joint Research Centre, 2013)have been compared with LCA from the inventory data from Ecoinvent® database (Kellenberger et al., 2007) and Idemat®. The results from BAT document are the most reliable due to that come from a most representative industrial survey. The impacts of glass fibre calculated based on the BAT document database is higher in some points as in Terrestrial and Freshwater Ecotoxicity with respect to glass fibre of Ecoinvent due to that the glass fibre from BAT document has in consideration more heavy metals and mainly has in consideration the fluorides (Figure 3). Respect to the rest of the aspects the glass fibre from BAT document is less harmful because of the optimization of energy consumption along the time for points as climate change, fossil depletion, etc.

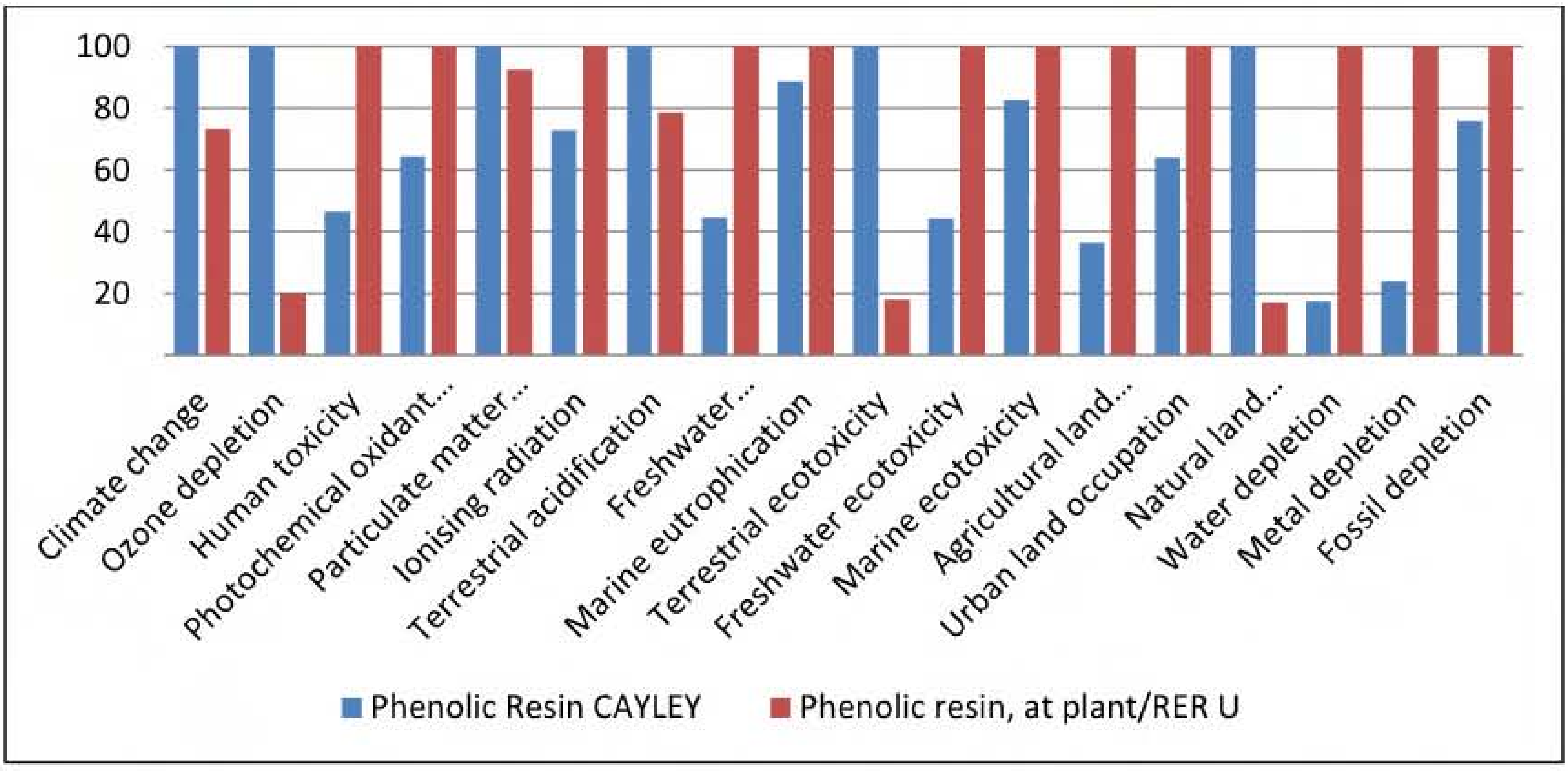

Figure 2: Comparison 1kg of phenolic resin from `Ecoinvent` and 'Eco-profiles', Method: Recipe Midpoint (H) V1.06/ Europe ReCiPe H/ Characterization.

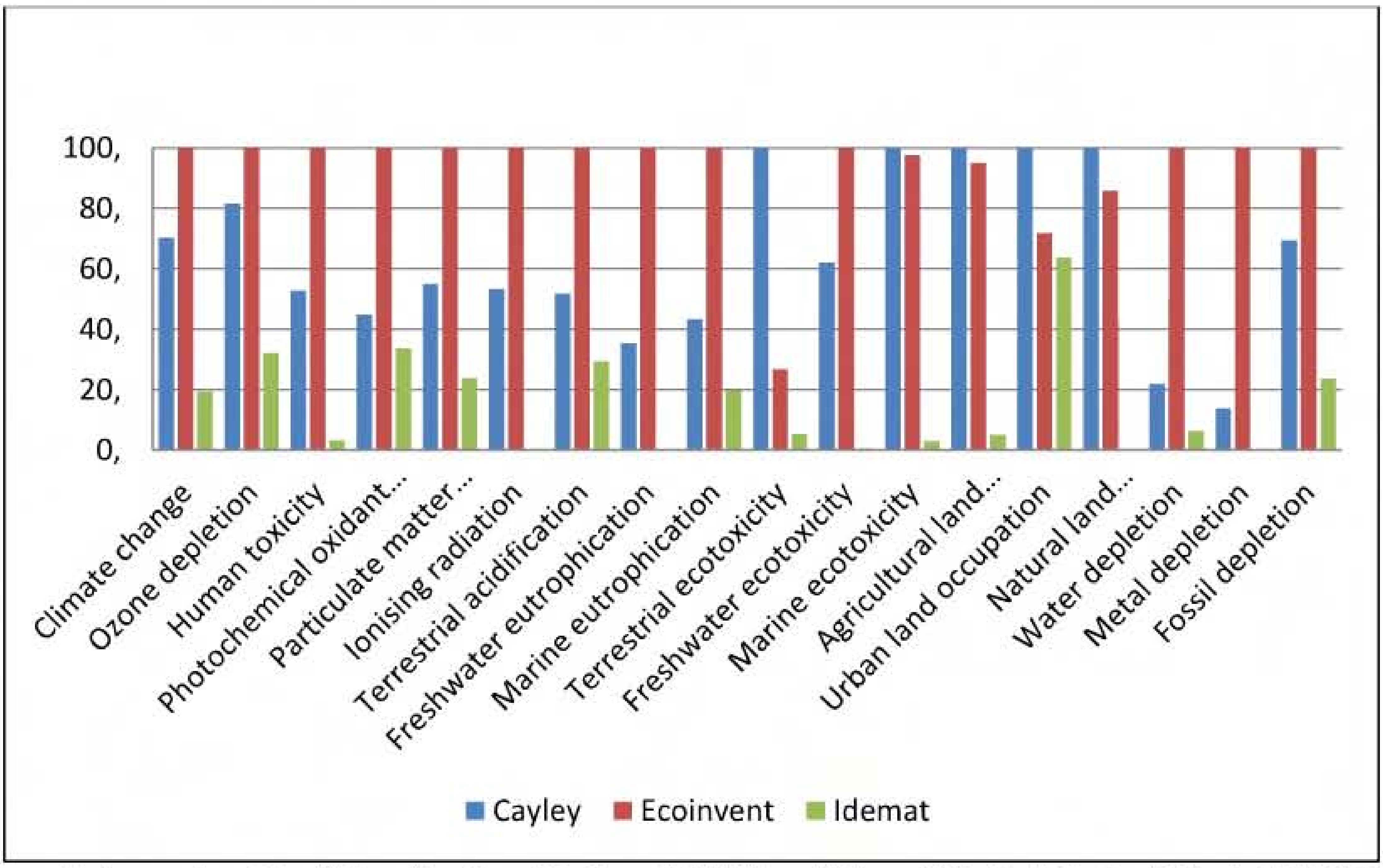

Figure 3: Comparison 1kg of glass fiber from `Ecoinvent`, 'BAT', and `Idemat` Method: Recipe Midpoint (H) V1.06/ Europe ReCiPe H/ Characterization.

3.2 Environmental impacts by life cycle stages

The environmental impacts of each life cycle stage were assessed at the endpoint level according to three endpoint impact categories: damage to human health (HH), damage to ecosystem diversity (ED), and damage to resource availability (RA). The endpoint impacts by life cycle stage are shown in Figure 4 to Figure 9.

FRP manufacturing stages became the stage of the life cycle with the greatest impacts in HH. The materials stage caused damage to ED and RA with a contribution to the overall damages of 47.2 % and 52.5 %, respectively. The FRP manufacturing caused damage to ED and RA with a contribution to the overall damages of 52.1 % and 47.5 %, respectively. The end-of-life stage, in this case for the end-of-life option landfilling was chosen, caused the lower impact in every end point category.

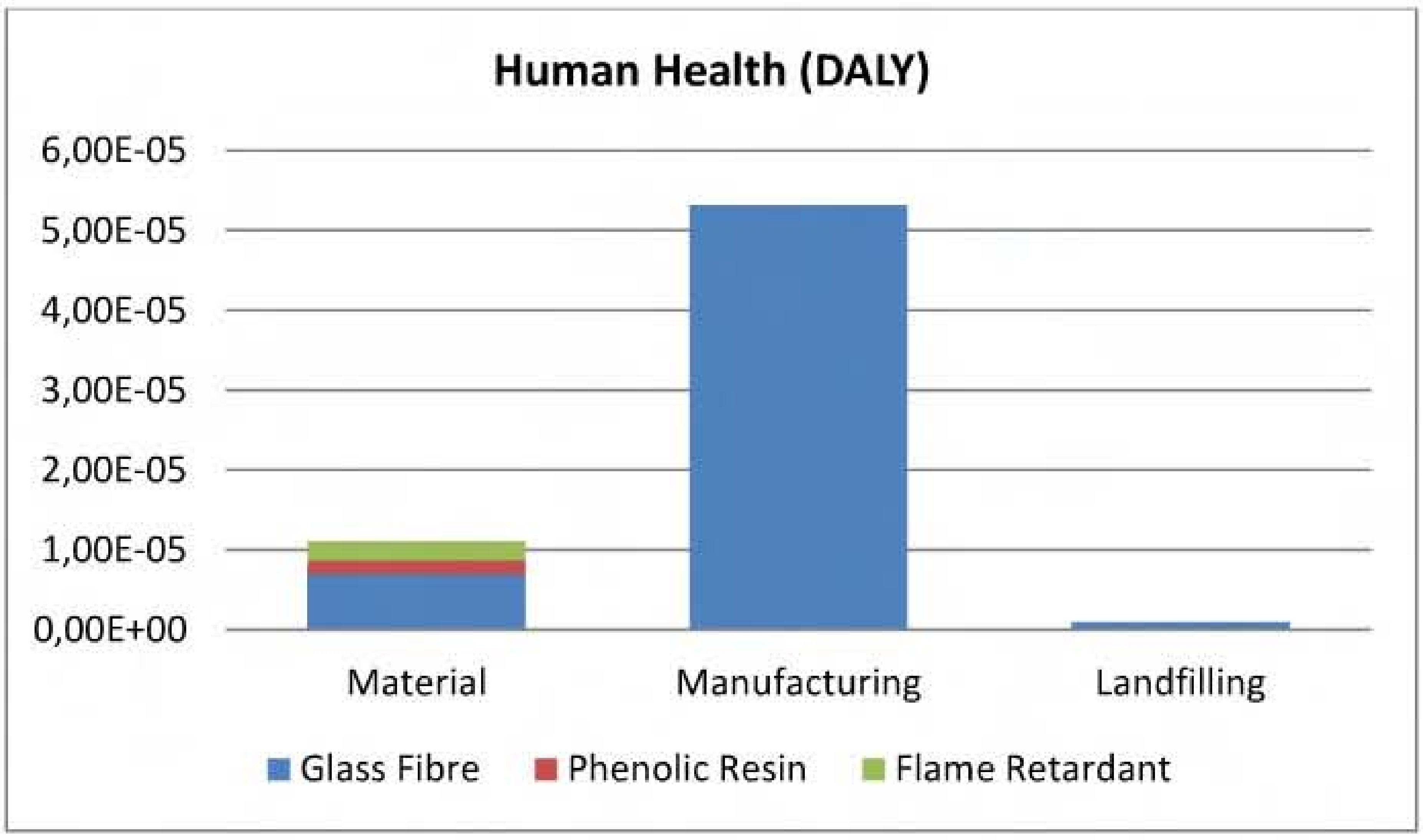

Figure 4: Damages to HH per Kg of skin made of glass fibre and phenolic resin

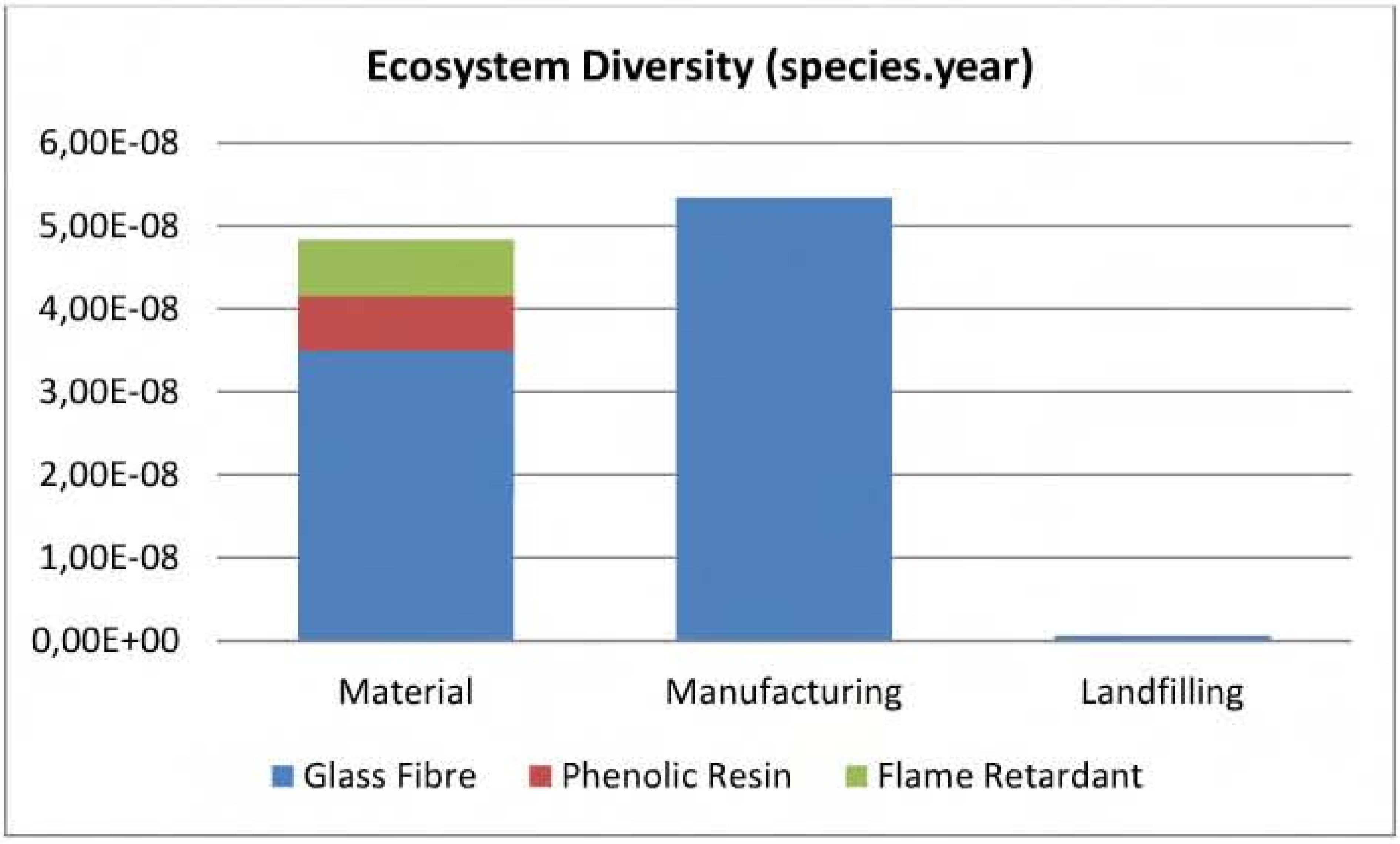

Figure 5: Damages to ED per Kg of skin made of glass fibre and phenolic resin

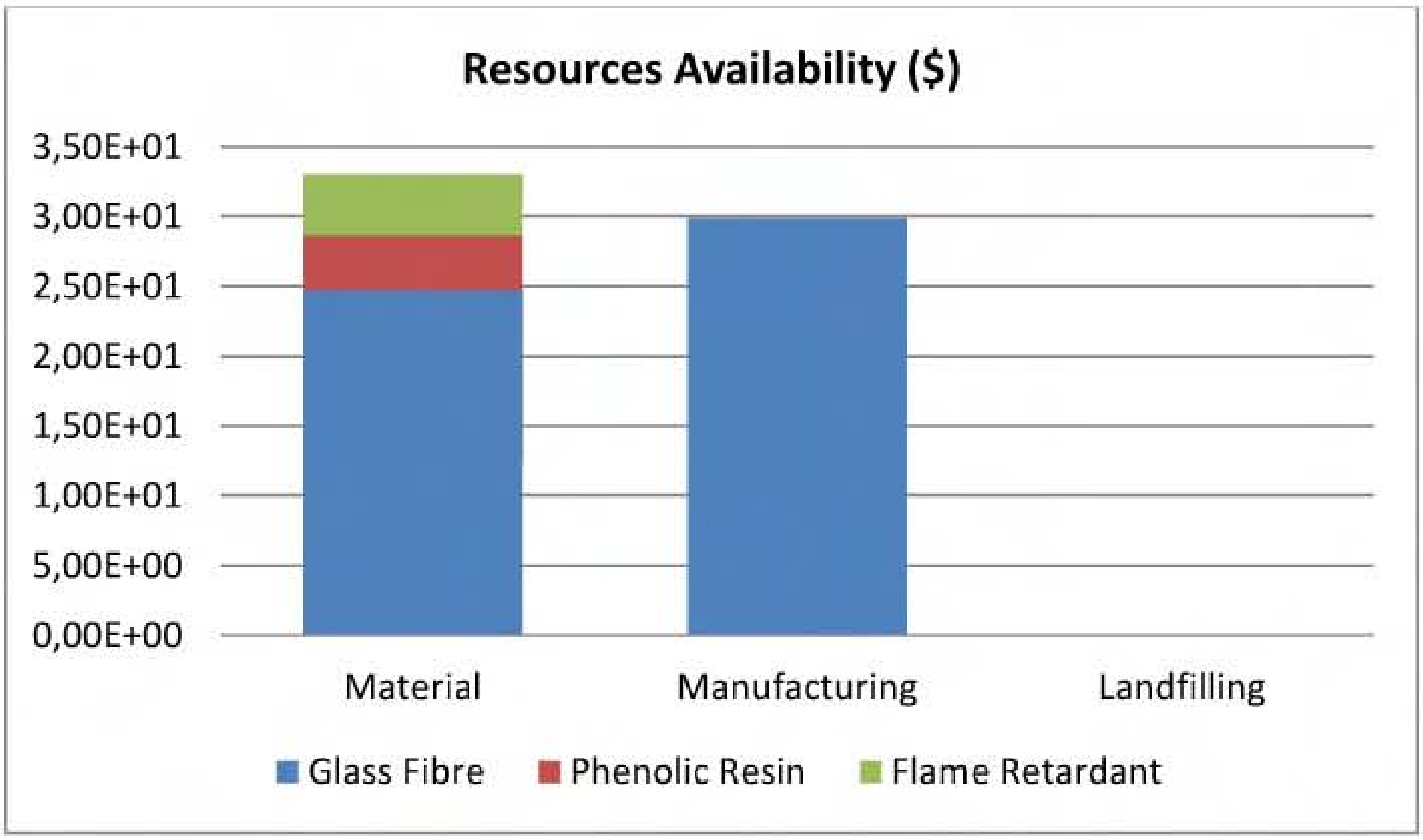

Figure 6: Damages to RA per Kg of skin made of glass fibre and phenolic resin

3.3 Environmental impacts of materials

The environmental impacts of the materials stage were assessed in more detail to determine the impacts of each of the main components of the FRP. To this end, the environmental impacts of the phenolic resin, glass fibre and flame retardant (decaBDE) were assessed at the midpoint level according to nineteen midpoint impact categories. The impact on climate change (CC) and cumulative energy demand (CED) for the above components are shown in Figure 7 and Figure 9.

It can be observed that the phenolic resin is the component with the greatest impacts, with a contribution to the overall impacts of the materials stage of 52.84% for CC and 52.53% for CED. Although glass fibre is the dominant component of FRP (60% by weight is glass fibre), the higher energy demand of phenolic resin leads to significant air pollutant emissions from the life cycle of the electricity used, therefor it has the most impacts for CC and CED out of the 3 investigated components.

Relatively important are the impacts of the brominated flame retardant in the two impact categories shown in Figure 7 and Figure 9 and much more in ozone depletion Figure 8.

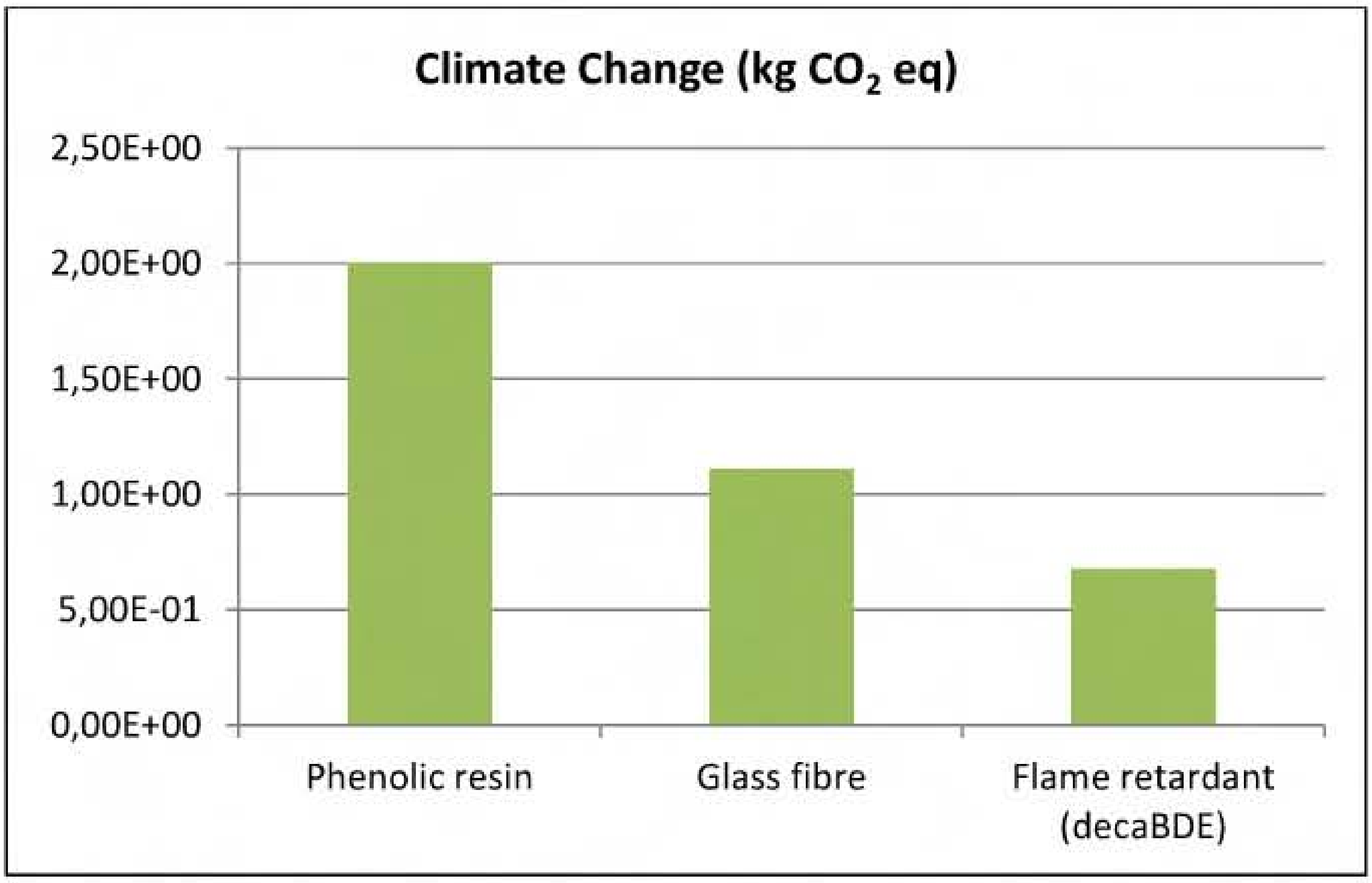

Figure 7: Impact on CC of the materials stage per kg of fibre-reinforced polymer by components

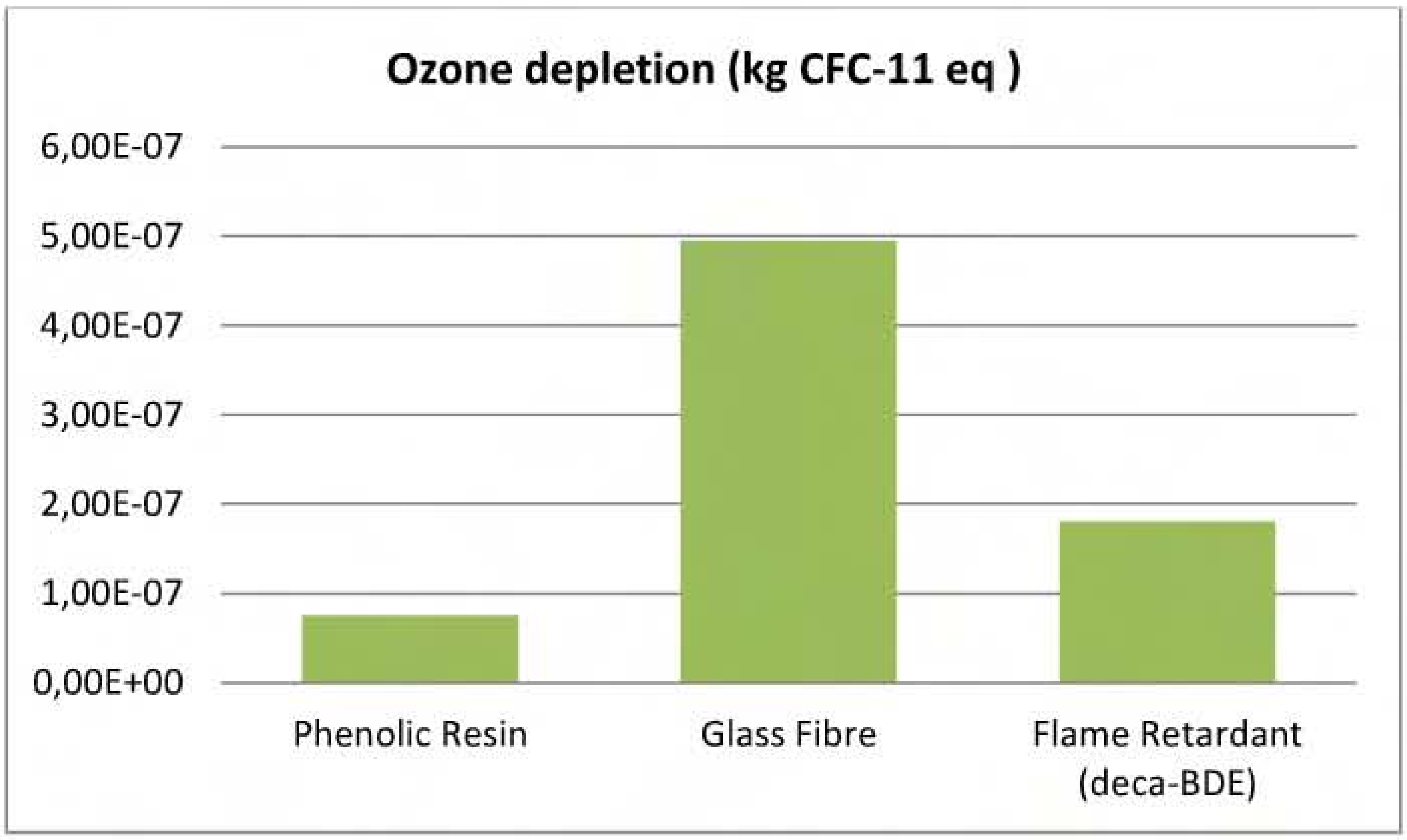

Figure 8: Impact on OD of the materials stage per kgof fibre-reinforced polymer by components

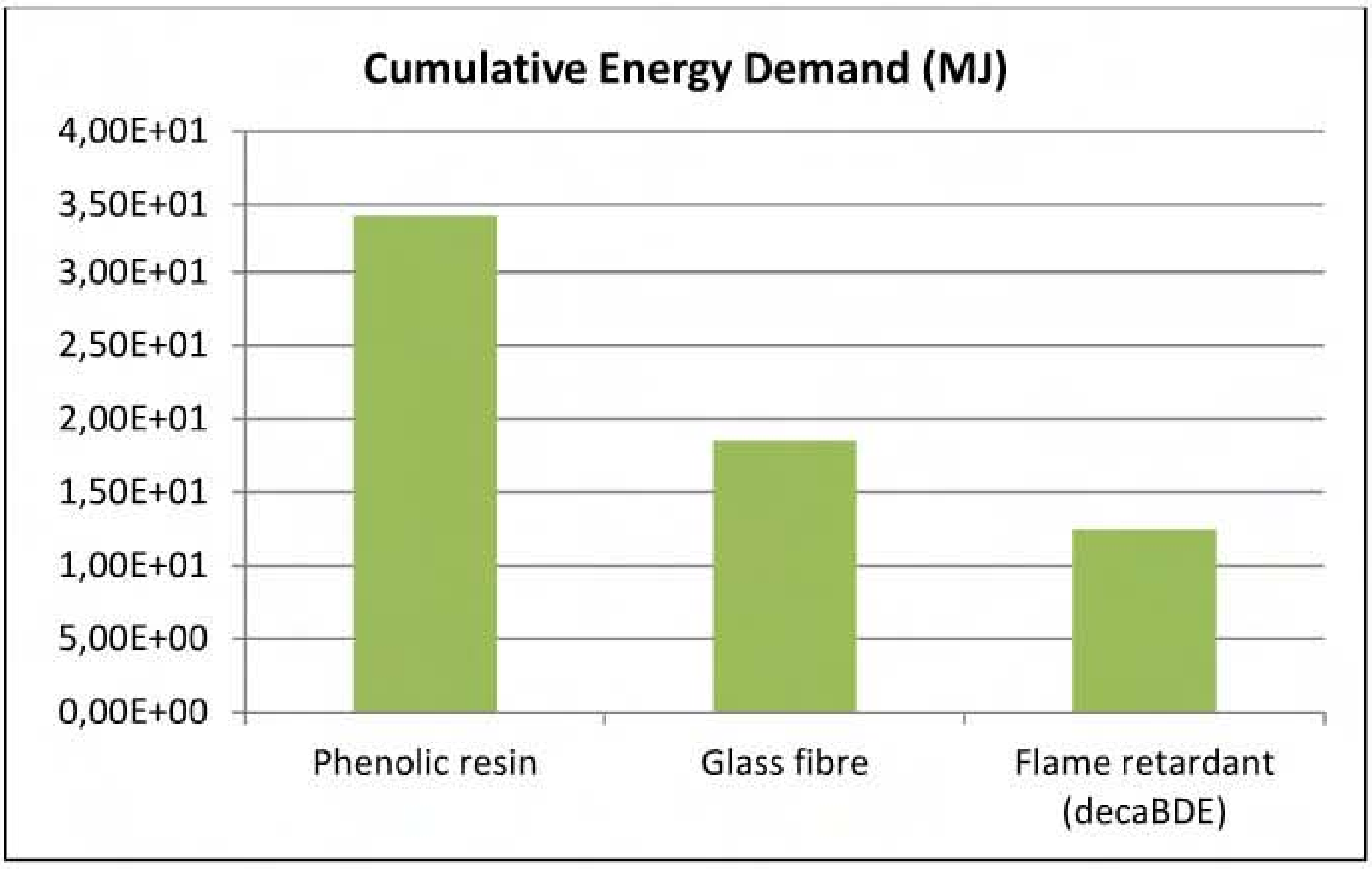

Figure 9: CED of the materials stage per kg of fibre-reinforced polymer by components

3.4 Environmental impacts of end-of-life

In Section 3.1, it was assumed that panels are deposited on landfills at the end of their useful life. However, other end-of-life strategies are commonly used for products made of fibre-reinforced polymer composites. Three end-of-life scenarios were assessed in this study: mechanical recycling, incineration and landfilling. The impacts on CC and CED for all the three scenarios are shown in Figure 10 and Figure 11.

outputs associated with each stage of the life cycle.

In order to use the best and most up-to-date inventory database, different inventory databases were compared and environmental impacts of these different databases were studied. For the phenolic resin database from the Eco-profiles of the European Plastic Industry proved to be more reliable compared to Ecoinvent® database due to its updates. For glass fibres Ecoinvent® database were compared to inventory data of glass fibres from reference document on best available techniques for the manufacture of glass. The results from BAT document were considered more reliable in this study due to that come from the last actualization of Best Available Techniques. However it is important to note that life cycle inventories use mean values which cause uncertainty in LCA. In reality there might be difference between the inventory values and the practical values.

FRP manufacturing stages become the stages of the life cycle with the greatest impacts in damage to human health. The materials stage caused damage to ED and RA with a contribution to the overall damages of 47.2 % and 52.5 %, respectively. The FRP manufacturing caused damage to ED and RA with a contribution to the overall damages of 52.1 % and 47.5 %, respectively.The end-of-life stage, however, caused the lower impact in every end point category.

Considering the impacts of the materials of FRP, phenolic resin part of the fibre-reinforced polymer is the component with the greatest impacts, with a contribution to the overall impacts of the materials stage of 52.84% for climate change and 52.53% for cumulative energy demand.

Investigation of end-of-life impacts could show the best waste management for a given material by giving its environmental impacts quantitatively. Recycling, incineration and landfilling were compared as end-of-life scenarios in this study. As only a slight part of FRP can be recycled from which filler can be produced, recycling shows positive environmental impacts with a significant energy demand (highest of the three scenarios).The incineration was also considered here as an alternative end-of-life scenario for panel waste. A portion of the thermal energy contained in waste can be recovered in the incineration, but it is not enough to offset the energy demand and the impacts due to air pollutant emissions from incineration. The third scenario was landfilling which has no energy demand but its continuity in the future is uncertain if decaBDE is finally included in POP list.

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