

Carbon footprint of different recovery options for the repulping reject from liquid packaging board waste treatment process

Khan Md.Musharof Hussain, Laitinen Vilma, Havukainen Jouni, Horttanainen Mika

This is a Publisher's version of a publication
published by Elsevier
in Waste Management

DOI: 10.1016/j.wasman.2021.10.003

Copyright of the original publication:

© 2021 The Authors. Published by Elsevier Ltd.

Please cite the publication as follows:

Khan, M., Laitinen, V., Havukainen, J., Horttanainen, M. (2021.) Carbon footprint of different recovery options for the repulping reject from liquid packaging board waste treatment process. Waste Management, vol. 136, pp. 93-103. DOI: 10.1016/j.wasman.2021.10.003

**This is a parallel published version of an original publication.
This version can differ from the original published article.**



Carbon footprint of different recovery options for the repulping reject from liquid packaging board waste treatment process

Md.Musharof Hussain Khan^{*}, Vilma Laitinen, Jouni Havukainen, Mika Horttanainen

Lappeenranta-Lahti University of Technology LUT, School of Energy System, Department of Sustainability Science, Yliopistonkatu 34, P.O. Box 20, 53851 Lappeenranta, Finland

ARTICLE INFO

Keywords:

Liquid packaging board waste
Repulping reject
Beverage food packaging
Mechanical recycling
Pyrolysis
Incineration

ABSTRACT

Liquid packaging board (LPB) is an integral part of storing and transporting liquid food. In addition to its significant advantages, LPB has been challenging the existing waste management sector since its introduction into the market. In most European countries, LPB waste is either incinerated or recycled in the recycling facilities where fibre is recycled, and the repulping reject is separated for incineration. Mechanical recycling and chemical recycling processes are other options for repulping reject treatment. This study used life cycle assessment (LCA) to compare the environmental impacts of three treatment processes, incineration, mechanical recycling and chemical recycling; each was considered with the functional unit of 1 tonne of repulping reject. Furthermore, two sub-scenarios based on the substituted heat produced by energy from the treatment processes were considered. In substituting biomass-based heat sources, chemical recycling generated the lowest greenhouse gas (GHG) emissions, about 560 kg CO₂ eq. tonne⁻¹ repulping rejects, followed by the mechanical recycling process (approximately 740 kg CO₂ eq. tonne⁻¹ repulping reject). The maximum amount of GHG was emitted from the incineration scenario, which was about 1900 kg CO₂ eq. tonne⁻¹ repulping rejects. By substituting natural gas-based heat sources, chemical recycling generated about 290 kg CO₂ eq. tonne⁻¹ repulping rejects. On the contrary, the mechanical recycling process generated about 430 kg CO₂ eq. tonne⁻¹ repulping rejects and incineration process generated 960 kg CO₂ eq. tonne⁻¹ repulping rejects. Uncertainty analysis showed that some assumptions significantly impact the results; however, the chemical recycling process had the lowest environmental impact in almost all uncertainty analysis.

1. Introduction

Waste generation is increasing due to population growth, economic growth and urbanisation (Kaza et al., 2018). Liquid packaging board (LPB) waste is one fraction of the waste that is generated. It has become an integral part of transporting and storing liquid food and beverages to keep the food fresh, safe, odour and taste-neutral and untainted over the products' entire lifetime and prevent food wastage (ACE, 2020). Also, its lightweight reduces fuel consumption during transport and decreases transportation costs (Storaenso, 2020). However, the thriving use of LPB is putting additional pressure on the waste management sector (Verghese et al., 2012). In 2014, global LPB production was 3.3 million tonnes; this was expected to increase to 3.6 million tonnes by the end of 2020 (Mohan, 2015). In the European Union (EU), most LPB waste is recycled, followed by recovery and landfilling (Verghese et al., 2012). However,

the recycling and recovery rate of LPB waste varies from country to country in the EU. According to the Alliance of Beverage Carton and the Environment (ACE), 51% of the LPB waste was recycled in 28 countries in the EU, where some countries such as Germany and Belgium had a 70% recycling rate (Robertson, 2021). LPB waste is challenging to recycle because its multilayer structure contains different materials. Aseptic LPB contains around 80% fibre, 15% plastic and 5% aluminium (Al) (Arvanitoyannis, 2005). Fibre is recycled by the re-pulping process, whereas plastic and Al are separated as a reject of the repulping process.

The EU has set a recycling target for packaging waste for 2030 and 2050. For example, by 2030, the target is 85% for paper and cardboard packaging recycling and 55% for plastic packaging recycling (Eurostat, 2021). In Finland, the recycling rate of plastic packaging was 31% in 2018, which is lower than the average recycling rate in the EU (40%) (Eurostat, 2021). In this case, recycling the repulping reject, especially

^{*} Corresponding author at: Lappeenranta-Lahti University of Technology LUT, School of Energy System, Department of Sustainability Science, Yliopistonkatu 34, P. O. Box 20, 53851 Lappeenranta, Finland.

E-mail address: musharof.khan@lut.fi (Md.Musharof H. Khan).

<https://doi.org/10.1016/j.wasman.2021.10.003>

Received 15 April 2021; Received in revised form 27 September 2021; Accepted 1 October 2021

0956-053X/© 2021 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

the plastic components, could increase the plastic recycling rate and help achieving the stipulated recycling targets.

To date, recycling of repulping reject is financially unfeasible (Lahme et al., 2020). Recycling repulping reject containing plastic and Al is not a straightforward process as cardboard. In most cases in Europe, these residues are used in cement kilns, where the plastic fraction is used as an alternative fuel, and Al is used as a bauxite substitute (Pretz and Pikhard, 2010). Plastic residues are also used in the waste incineration plant for recovering energy. However, LPB rejects can also be recycled by chemical and mechanical recycling processes. Chemical recycling utilises reprocessing technologies that affect the formulation of the polymeric material or polymers, turning them into other products, such as monomers, basic chemicals or something else with value, but this process excludes energy recovery (Plastics Recyclers Europe, 2018). The mechanical recovery options could include traditional mechanical recovery or recovery as a composite. Traditional mechanical recovery separates the plastic fractions from Al and the fibre residues. Thus, the plastic is recovered as a material, and the rest is recovered as energy. It should be noted that each of the recycling processes of plastic residues has environmental impacts. Therefore, finding a recycling option with a better environmental impact is difficult without conducting a life cycle analysis (LCA).

LCA is a useful method to quantify the environmental impact of a product or service throughout its life cycle (ISO, 2006). Numerous LCA studies have been conducted on the environmental impact of an LPB system. Most of the studies have concentrated on cradle-to-gate analysis of LPB (Vergheze et al., 2012; Von Falkenstein et al., 2010; Wellenreuther et al., 2012; Jelse et al., 2009). The environmental impact from the materials of LPB was presented in Harst-wintraecken (2015). By analysing several articles, three options for LPB waste management were found: (1) fibre production from cardboard (Ruttenborg, 2017; Bisinella et al., 2018; Vergheze et al., 2012), (2) energy production from incinerating repulping reject residues (Ruttenborg, 2017; Bisinella et al., 2018) and (3) landfilling of LPB (Vergheze et al., 2012). Although several studies have highlighted the end of life (EoL) of repulping rejects, there is a lack of knowledge about the environmental impact of other possible waste management options, such as chemical recycling and mechanical recycling. To the best of our knowledge, no study has compared the incineration of repulping rejects with mechanical and chemical recycling systems. By addressing the knowledge gap, this study aimed to answer the following questions:

- What is the environmental impact of the treatment of LPB repulping rejects that is separated from LPB treatment?
- Which treatment option has the lowest environmental impact, and why?

2. Materials and methods

LCA is a well-established method used to evaluate the environmental impacts of products and services throughout their life cycle (ISO, 2006). The LCA framework, methods and procedures were developed by the Society of Environmental Toxicology and Chemistry (SETAC) and the International Organisation for Standardisation (ISO), resulting in two international standards: ISO 14040 and ISO 14044. By following ISO 14040 and ISO 14044 standards, the present study defined its scope and goals and utilised life cycle inventory (LCI) analysis and life cycle impact assessment (LCIA) to obtain its results. This study also conducted an uncertainty analysis and drew its conclusions based on the outcomes of the study. This research study used the following software: GaBi professional, version 9.5, by Sphera as the LCA modelling tool and Microsoft Excel as the data analysis tool.

2.1. Goal and scope definition

This study compared the environmental impact from different waste management options for repulping reject residues of LPB waste. The aim was to investigate which alternative treatment methods for repulping reject is the most environmentally friendly considering climate change. A functional unit needs to be established to provide a reference for the input and output of the system. In this study, 1 tonne of reject from the repulping process was selected as the functional unit to satisfy the treatment of the reject materials.

2.1.1. System boundary

The investigated treatment options were incineration, material recovery by mechanical recycling and material recovery by chemical recycling process (pyrolysis). This study did not consider the production phase of LPB because the LPB treatment would be the same for all the compared treatment options. In other words, the environmental impact of the repulping process and the separation of the polymer-Al fraction from the fibre fraction were excluded. The system boundary included repulping reject treatment by the incineration process, the mechanical and chemical recycling systems. The system boundary also included the avoided emissions obtained by replacing the materials and energy produced from different treatment options. Additional functions were necessary to operate the main processes, such as the electricity grid mix and thermal energy production; therefore, they were included in the system boundary. Details about the system boundary are shown in Fig. 1.

Scenario analysis

Based on the repulping reject treatment options, this study consisted of three scenarios: (1) system as usual (incineration), (2) material recovery by mechanical treatment and (3) material recovery by chemical treatment (pyrolysis). Scenario 1 represents incineration of the repulping reject with energy recovery since it is one of the most commonly used treatment processes for this residue. Scenario 2 includes material recovery through mechanical treatment because mechanical treatment is already applied for separately collected plastic waste, and its possibilities for this type of plastic should be evaluated. Chemical treatment of the repulping reject can be done by pyrolysis, depolymerisation or gasification. Pyrolysis of the repulping reject was considered more convenient for this study because the pyrolysis process produces oil fuel that can be used for virgin plastic production. Since this study includes plastic production from the chemical recycling process, therefore, pyrolysis system was used in the chemical recycling process.

This study aimed to investigate the potential environmental impact of the processing of repulping reject residues; its findings are expected to be used to support decision making. Thus, a system expansion method was applied in LCA, meaning that additional functions arising from the treatment of plastic residues, such as energy and material recovery, were considered. This study used the 0:100 EoL method with a credit system. Credit was gained by avoiding the environmental impact when the recovered energy and material replaced the virgin materials and energy from the production mix. Electricity is recovered in each scenario, replaces the electricity production of Finland in 2017 (peat 4.13%, hard coal 8.73%, coal gases 0.87%, natural gas 4.92%, fuel oil 0.27%, biomass 16.22%, biogas 0.62%, waste 1.53%, nuclear 33.49%, hydro 22.01%, wind 7.14%, photovoltaics 0.07%). Thermal energy is also recovered and substitutes thermal energy produced from biomass (sub-scenario X.1) or natural gas (sub-scenario X.2). In addition to electricity and heat, the chemical recycling routes also recycle plastic, which replaces virgin polyethylene (PE).

System expansion

This study followed the system expansion approach to calculate the impact of each scenario's overall global warming potential (GWP). The following formula was used for the calculation:

Total environmental impact = Environmental impact from process – avoided impact from primary production

In this study, the recycled plastic from the chemical recycling process was used to replace virgin plastics. This procedure prevented the production of virgin plastic. Therefore, the greenhouse gas (GHG) emissions from producing virgin plastic were avoided. The energy recovered from incinerating plastic and fibres can be considered as a by-product of repulping reject waste treatment. In this study, credits were given for the produced thermal energy and electricity by subtracting the emission data obtained from producing thermal energy from natural gas or biomass and electricity from the electricity production. The data on avoiding GHG emissions by substituting thermal energy was collected from the Sphera database. The avoided emissions from substituting thermal energy from the heat generated from natural gas and biomass, electricity generated from the Finnish production mix and virgin plastic were also collected from the Sphera database. The background of the datasets is given in the supplementary materials Table 1. The emission factors for these processes are presented in Table 1.

2.1.2. Assumptions and limitations

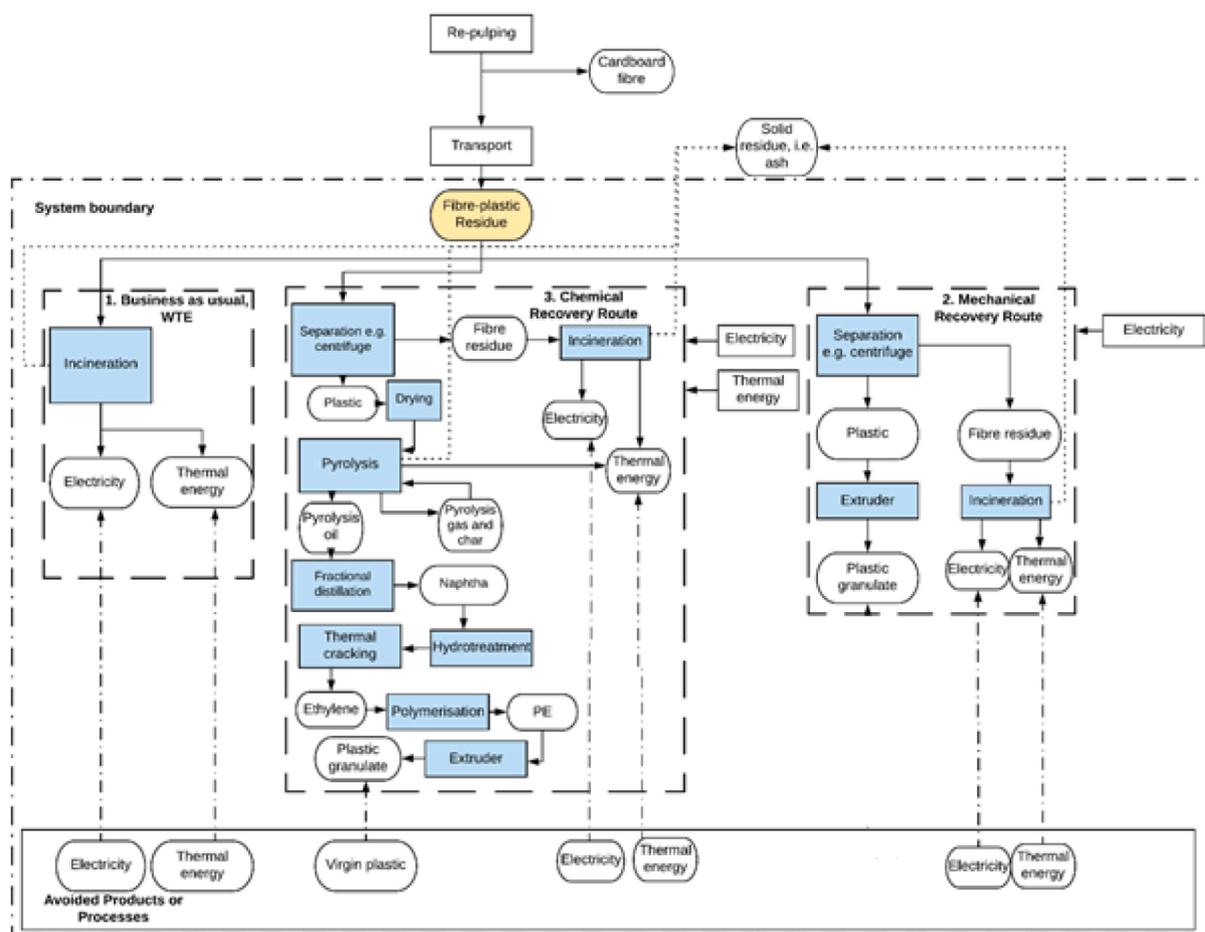
This study has some assumptions and limitations. After the repulping

Table 1

Emission factors of the substituted processes (Sphera, 2020).

Substituted product	Emissions factor	Unit
Electricity from the grid	0.2	kg CO ₂ /kWh
Heat from natural gas	0.06	kg CO ₂ /MJ
Heat from biomass	0.002	kg CO ₂ /MJ
Production of virgin PE	1.63	kg CO ₂ /kg plastic

process, the share of dry matter in the reject was 63%. According to Stora Enso personnel, the dry matter consists of 70% plastic, 25% fibre and 5% Al (Rämö, 2020); thus, these percentages were considered in this study. The plastic in the reject was considered to be PE since it is the most commonly used plastic in the LPB (Arvanitoyannis, 2005). PE and aluminium were separated from fibre in the laboratory. The separation efficiency was found with a range of 55%-85%; thus, the separation efficiency was assumed 80% in this study. There is no empirical evidence that recycled plastic from repulping reject of LPB by mechanical recycling process can replace virgin plastic. Therefore, it was assumed that recycled plastic from mechanical recycling did not replace any



*WTE = Waste to Energy; PE = polyethylene.

Fig. 1. The system boundary of the study. *WTE = Waste to Energy; PE = polyethylene.

virgin PE. It was assumed that the type of transportation was not changed significantly between the scenarios; therefore, transportation was excluded in this study. The ash treatment remained the same in all the scenarios, although the amount varied to some extent. Moreover, ash treatment has been shown to have a minor contribution to the results of the waste-to-energy (WtE) and recycling process (Liikanen et al., 2017). Consequently, ash treatment was also excluded from the present study.

2.2. Inventory analysis

The inventory data were collected from different studies in the literature, by interviewing experts and from the Sphera database. The substituted energy and virgin PE production data were collected from the Sphera database. The incineration, electricity and heat production data are representative of Finland. However, it is possible to use this model from a global perspective by changing the incineration, heat and electricity processes in GaBi. The details of the used processes are given in the supplementary materials Table 1.

2.2.1. Incineration

In the incineration process, repulping reject residue is the input; the outputs are electricity, thermal energy and emissions. Although the incineration process data were collected from the Sphera database, the efficiency is adjusted to represent the efficiencies of the Finnish waste incineration plants. According to Reimann (2012), combined heat and power (CHP) incineration plants in Europe have an average efficiency of 76%, but this varies between regions. According to that report, thermal efficiency in Northern Europe is around 72.6%, and electrical efficiency is 11%. In that report, the energy needed for pre-treatment of feed was taken into account while calculating the efficiency of the power plant (Reimann, 2012). Since the total efficiency of the incineration technology is 83.6% (72.6% heat efficiency and 11% electric efficiency), it can be considered as a waste recovery option according to Waste Framework Directive 2008/98/EC (Europa, 2018; European Commission, 2020).

GaBi uses a lower heating value as received (LHV_{ar}) of 10.4 MJ kg⁻¹ for paper and board by considering moisture content of 22% and LHV_{ar} of 41.2 MJ kg⁻¹ for plastic waste. Based on the information obtained from a cardboard recycling facility, the moisture content of repulping reject is around 37% (Rämö, 2020). Therefore, based on the moisture content of the repulping reject, the heating values in the present study were adjusted to 8 MJ kg⁻¹ and 26 MJ kg⁻¹ for fibre and plastic, respectively, and they were used in every scenario. Furthermore, the effect of Al was considered as a mass that does not generate energy.

2.2.2. Mechanical recycling

The mechanical treatment scenario starts with the separation of fibre and plastic through centrifugation. PE and Al were separated from fibre through the separation process. Subsequently, the separated PE and Al were extruded into granulates. It was assumed that Al does not cause any problem in the PE extrusion process. The plastic and Al fraction that could not be separated from the fibre was treated using the incineration process to recover energy. The upstream process for producing the required electricity and the downstream process of the reject treatment

Table 2
The mechanical recycling parameters.

Parameter	Value used	Unit	Reference
Centrifuged plastic separation efficiency	80	%	
Centrifuge energy demand	1.4	kWh tonne ⁻¹ of residue	Alfa Laval (2020)
Extrusion energy demand	1800	kJ kg ⁻¹	Väntsi and Kärki (2015)
Replacement ratio of the virgin plastic granulate	0	%	

were accounted for.

In modelling mechanical recycling, pre-set processes from GaBi were used, such as electricity production and incineration. The incineration process used in this scenario was the same as the process was used in the incineration scenario. The data from the separation and extrusion processes were obtained from the literature and by contacting companies that recycle plastics mechanically or provide suitable technology. Table 2 presents the mechanical recycling scenario parameters. The mechanical recycling process substitutes thermal energy (either heat from biomass (scenario 2.1) or natural gas (scenario 2.2) and electricity.

2.2.3. Chemical recycling (pyrolysis)

In the chemical recycling scenario, the repulping reject was directed to the separation process, where the plastic-Al and fibre were separated by centrifugation. Similar to the mechanical recycling scenario, the fibre residue was directed to the incineration system, but the plastic-Al residue was directed to the dryer for pyrolysis. Plastic decomposes back to oil and other possible products in the pyrolysis process, such as gas, char, and wax. The oil fraction was refined to produce the plastic granulates, while the gas fraction was used to produce energy, which is later used in the pyrolysis process.

In this scenario, it was assumed that the pyrolysis temperature was around 400–450 °C. It was also assumed that a catalyst (silica-alumina) was used to enhance the yield of oil instead of wax. Lee (2012) and Mianadad et al. (2017) found that pyrolysis of PE without any catalysts primarily yields wax. The present study assumed that the production has oil but no wax by using the catalyst. However, the production of a catalyst or its regeneration was not included in this study. The yields from the pyrolysis are presented in Table 3.

Since the residue contains a large amount of moisture (37%), a dryer was used before feeding the plastics into the pyrolysis process. After drying the plastics, the Lower heating value as received (LHV_{ar}) of the

Table 3
Parameters and yield from the pyrolysis process.

Parameter	Min.	Max.	Av.	Value used	Unit	Reference
Dryer energy demand				2.3	MJ kg ⁻¹ plastic	Ingrao et al. (2014)
LHV _{ar} of the waste plastics	22	43	32.5	26	MJ kg ⁻¹	Aguado et al. (2002) Horttanainen et al. (2013)
LHV _{ar} of the plastic pyrolysis oil				38	MJ kg ⁻¹	Kalargaris et al. (2017)
Energy demand	148.5		250	250	kWh tonne ⁻¹ of plastic	Aguado et al. (2002); Ecomation (2016)
Shares of the plastic pyrolysis products:						
Gas	8.5	32.3	17.11	17	w-%	Achillas et al. (2007); Shah et al. (2010); Uddin et al. (1997)
Oil	67.4	80.2	73.24	73		
Char	0.53	19.4	9.6	10		
CO ₂ emissions from burning gas	2.34	3.05	2.87	2.87	kg CO ₂ kg ⁻¹ gas	Calculated
CO ₂ emissions from burning char	0.116	0.180	0.148	0.148	kg CO ₂ kg ⁻¹ char	Calculated

LHV_{ar}* = Lower heating value as received

Table 4
Parameters for plastic production from pyrolysis oil.

Parameter	Value used	Unit	Reference
Mass of oil to produce 1 kg naphtha	1.1	kg	Boustead (2005)
Mass of natural gas to produce 1 kg naphtha	0.031	kg	Boustead (2005)
Mass of naphtha to produce 1 kg ethylene	1.151	kg	Calculated
Mass of natural gas to produce 1 kg ethylene/kg naphtha	0.202	kg	Calculated
Mass of ethylene to produce 1 kg PE	1.012	kg	Calculated
Mass of natural gas to produce 1 kg PE/kg ethylene	0.125	kg	Calculated
Extrusion energy demand	1800	kJ/kg	Väntsi and Kärki (2015)
Emissions from processes			
Naphtha production	0.13	kg CO ₂ -eq kg ⁻¹ feed in	Calculated
Hydrogenation	0.018	kg CO ₂ -eq kg ⁻¹ feed in	Estimated, Bezergianni and Chrysikou (2016)
Cracking	0.93	kg CO ₂ -eq kg ⁻¹ feed in	Calculated
Polymerisation	0.35	kg CO ₂ -eq kg ⁻¹ feed in	Calculated

plastic increased from 26 MJ kg⁻¹ to 41.2 MJ kg⁻¹. The heat for drying was obtained from the surplus energy of the pyrolysis process. The LHVar of the pyrolysis gas and char were calculated by the difference in the plastic's energy content and the pyrolysis oil.

Emissions from burning the gas were calculated using the gas compositions presented in Williams and Williams (1999) and Achilias et al. (2007). Emissions from burning the char were calculated using the emissions based on the ultimate analysis of char by Jamradloedluk and Lertsatitthanakorn (2014) and Syamsiro et al. (2013). Although, these studies were performed for char from high-density polyethylene, they were used as a reference in the present study due to the lack of data.

Plastic production from pyrolysis oil requires thermal energy from natural gas. Moreover, natural gas was used in the reactions. Electricity was used in the extrusion process. The emission data of thermal energy from natural gas and electricity from the electricity production mix were collected from the Sphera database. The parameters used for plastic production are presented in Table 4. The values were calculated using Boustead (2005), Plastic Europe (2016) and Plastic Europe (2012) as a reference. The hydrogenation emissions were evaluated using Bezergianni and Chrysikou (2016) as a reference.

2.2.4. Uncertainty analysis

Uncertainty analysis is conducted in LCA when a lack of data and data inaccuracy is observed in the LCI (Guo and Murphy, 2012). The repulping reject composition that was used in this study is not a constant value. In this study, the composition was: plastic (70 w-%), fibre (25 w-%) and Al (5 w-%). However, the compositions of plastic: fibre: Al could be 80:15:5 or 60:35:5. In addition to the LHVar of plastic (26 MJ kg⁻¹) and fibre (8 MJ kg⁻¹), several other LHVar has been reported, such as 22 MJ kg⁻¹ and 30 MJ kg⁻¹ for plastic (Aguado et al., 2002; Horttanainen et al., 2013) and 6 MJ kg⁻¹ and 10 MJ kg⁻¹ for fibre (calculated based on the moisture content).

In addition to the repulping reject composition and LHVar, uncertainties in the data were also observed in the yield from pyrolysis. In the pyrolysis process, the gas and oil yield is interconnected. It was found that the oil yield could be increased to 80 w-% and decreased to 60 w-% (Achilias et al., 2007; Shah et al., 2010; Uddin et al., 1997).

Table 5
Data for the uncertainty analysis.

Uncertainty 1 (Changing plastic, fibre and aluminium ratio)		
Plastic:Fibre:Aluminium 60:35:5	Plastic:Fibre:Aluminium 70:25:5 (Base)	Plastic:Fibre:Aluminium 80:15:5
Uncertainty 2 (Separation efficiency)		
60%	70%	80% (Base) 90%
Uncertainty 3 (Virgin plastic replacement ratio)		
0% (base)	30%	50% 80% 100%
Uncertainty 4 (Change in LHVar)		
	Low	Base High
Plastic	22	26 30
Fibre	6	8 10
Uncertainty 5 (Pyrolysis yield)		
Yield 1 (oil: gas: char) 60:30:10	Base (oil: gas: char) 73:17:10	Yield 2 (oil: gas: char) 80:10:10

Consequently, the gas yield is changed to 10 w-% if the oil yield increases and 30 w-% if the oil yield decreases. The separation efficiency of the centrifuge is not a constant figure, and thus different separation efficiency was analysed in the uncertainty analysis. Virgin plastic replacement by recycled plastic from the mechanical recycling process depends on the separation efficiency and the presence of impurities. Technological improvements might improve the removal of impurities from PE. By assuming technological improvement in the future, the uncertainty analysis investigated the impact of mechanical recycling with different virgin PE replacement ratios. However, the virgin plastic replacement ratio is always 100% for the chemical recycling scenario since the pyrolysis process produces pyrolysis oil from which the plastic granulates are produced.

Since the considered LCI data varies depending on different factors, this study conducted an uncertainty analysis based on ISO 14040:2006. The data shown in Table 5 were used for the uncertainty analysis.

2.3. LCIA

The environmental impact of the treatment of plastic residue was classified according to the ReCiPe 2016 v1.1 Midpoint (H) method. This method is widely used in LCA due to its robustness and low uncertainty (Sphera, 2020; Abdulkareem et al., 2021). Only the GWP excluding biogenic carbon was employed when assigning the LCI results since this impact is relevant for assessing the emissions from the studied repulping reject treatment processes. The other impact categories from the ReCiPe 2016 v1.1 Midpoint (H) method were excluded due to the lack of inventory data for the chemical recycling process.

3. Results

The results of the three reject treatment scenarios illustrated in Fig. 2 depended on several factors, such as the substitution of biomass-based heat source and natural gas-based heat source, avoided emission by substituting energy and virgin plastic, and energy consumption in the recycling process. It was found that waste incineration scenarios (1.1 and 1.2) had the maximum GWP impact compared to mechanical recycling and chemical recycling scenarios. In contrast, chemical recycling scenarios had the lowest GWP impact than mechanical recycling scenarios and waste incineration scenarios. The chemical recycling scenarios replaced 300 kg of virgin plastic. On the contrary, the mechanical recycling process did not replace any virgin plastic. As a consequence, scenarios 3.1 and 3.2 had a better environmental impact than scenarios 2.1 and 2.2. The details of the results are presented in the supplementary materials Table 2.

Repulping reject treatment through the incineration process resulted in the maximum GWP impact due to the incineration of the plastic portion, which is also evident from the findings reported in other research (IEL, 2019; Korhonen and Dahlbo, 2007; Lazarevic et al., 2010). In this scenario, 700 kg of plastic was incinerated, which caused

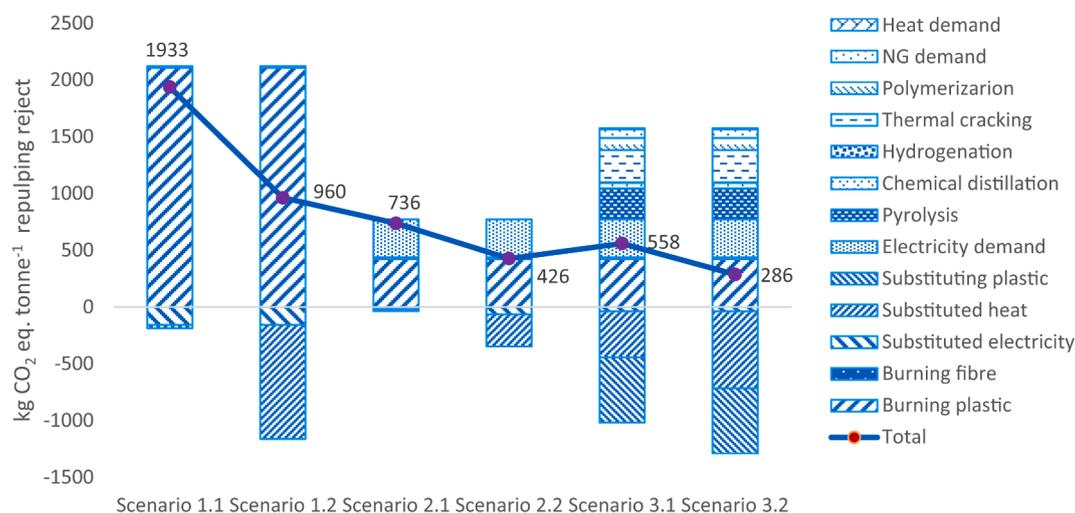


Fig. 2. Impact of GWP, excluding biogenic carbon from different scenarios.

2100 kg CO₂ eq. tonne⁻¹ repulping reject with an emission factor of 123 kg CO₂ eq. MJ⁻¹ plastic. In Eriksson and Finnveden (2009), the emissions from plastic incineration were 2400–2500 kg CO₂ eq. tonne⁻¹ plastic. In Shonfield (2008), the emissions were 1800 kg CO₂ eq. tonne⁻¹ plastic. A clear difference can be seen between the present study's findings and the results reported in other studies, which is due to the differences in the types of plastic and the LHVar.

The substituted heat source influenced the total emissions from different scenarios. It was found by comparing scenario 1.1 with 1.2, 2.1 with 2.2, and 3.1 with 3.2 that the total emissions from scenarios (1.1,2.1,3.1) with the substituted biomass-based heat source were higher compared to the scenarios (1.2,2.2,3.2) with the substituted natural gas-based heat source. The CO₂ emission factor is lower for biomass than natural gas. Therefore, the biomass-based heat source has a lower amount of avoided emissions than the natural gas-based heat source, resulting in an increase in the total emissions seen in the incineration scenario.

Compared to the mechanical recycling scenarios, chemical recycling scenarios had a lower GWP impact due to the avoided emissions by substituting energy and virgin plastic. The total avoided emission in scenarios 3.1 and 3.2 was more than 1000 kg CO₂ eq. and 1280 kg CO₂ eq., respectively. On the contrary, scenario 2.1 avoided 35 kg CO₂ eq. and 2.2 avoided 345 kg CO₂ eq. It is difficult to compare the total emissions from the mechanical recycling processes in the present study with the other studies since the process chain, and system boundary varies from study to study. Without the substituted emission, the total emission from the mechanical recycling scenarios was 770 kg CO₂ eq. tonne⁻¹ repulping reject. Of this, 438 kg CO₂ eq. was generated from burning the plastic and fibre portion and 333 kg CO₂ eq. from electricity consumption during the recycling process.

In the chemical recycling scenarios, the GWP impact resulted from (i) incineration of the fibre and plastic that was separated from the centrifugal process, (ii) the pyrolysis process and (iii) the substituted virgin plastic, heat and electricity. Excluding the avoided emission by replacing virgin plastic, heat and electricity, the total emission generated from the chemical recycling scenarios was 1570 kg CO₂ eq. tonne⁻¹ repulping reject. Of this, nearly one-third of the emission was related to incineration of the plastic portion, 21% resulted from electricity consumption in the pyrolysis, 17% was due to thermal energy consumption in the pyrolysis process, 18% was from thermal energy consumption in the thermal cracking of naphtha. The remaining 16% was generated from the distillation process, polymerisation, natural gas acquisition and incinerating the fibre portion of the repulping reject. Jeswani et al. (2021) showed that the pyrolysis process emits 1162 kg CO₂ eq. tonne⁻¹

of mixed plastic waste treatment without considering system credits is slightly higher than what was observed in the present study (1130 kg CO₂ eq.) due to the consideration of different types of plastic and the system boundary. In that study, one-third of the emission was generated from the pyrolysis process, 26% was from waste collection and sorting, and 8% was from the purification of the pyrolysis oil.

3.1. Uncertainty analysis

The uncertainty analysis results presented in Fig. 3 showed that the composition of the repulping rejects significantly impacted the overall emissions. The details of the results are given in the supplementary materials Table 3-7. The total emission increased in the incineration scenarios due to the increased amount of plastic in the incineration process. In contrast, the total emission decreased when the plastic share decreased from 70% to 60%. Resembling the incineration scenarios, the GWP impact increased when the plastic share increased in the chemical recycling scenarios. Because, the amount of plastic that was separated from the centrifugal system was increased compared to the base scenario. When the increased amount of plastic was incinerated, it produced more GHG emissions and thus had higher GWP than the base scenario. The GWP impact decreased as the share of plastic decreased. Similar to the waste incineration and chemical recycling scenarios, the GWP increased with the increase of plastic share in the reject, and GWP decreased with the decrease of plastic share in the reject.

Separation efficiency had a significant impact on the mechanical and chemical recycling scenarios. The emission decreased as the separation efficiency increased because more plastic was recovered, and a lower amount of plastic was incinerated (scenario 2.1,2.2,3.1,3.2). In addition, more virgin plastic was substituted (scenario 3.1,3.2). The impact of the virgin replacement ratio had a significant impact on the overall result of the study. An increase in the replacement ratio by mechanical recycling process resulted in a decrease in the total emission due to the increase in the avoided emission by replacing virgin plastic. Mechanical recycling had a lower GWP impact than chemical recycling only when it replaced virgin plastic. However, replacing virgin plastic with the mechanical recycling process is a highly optimistic view which is impossible with current separation technologies.

It was also found that changing the LHVar impacted the overall GWP of the studied scenarios. The overall emissions increased with the decrease of the LHVar of the repulping reject and decreased with the increase of the LHVar. The emission changing ratio was higher for the replaced biobased energy than for the replaced natural gas-based energy. Changes in the pyrolysis yields did not significantly impact the

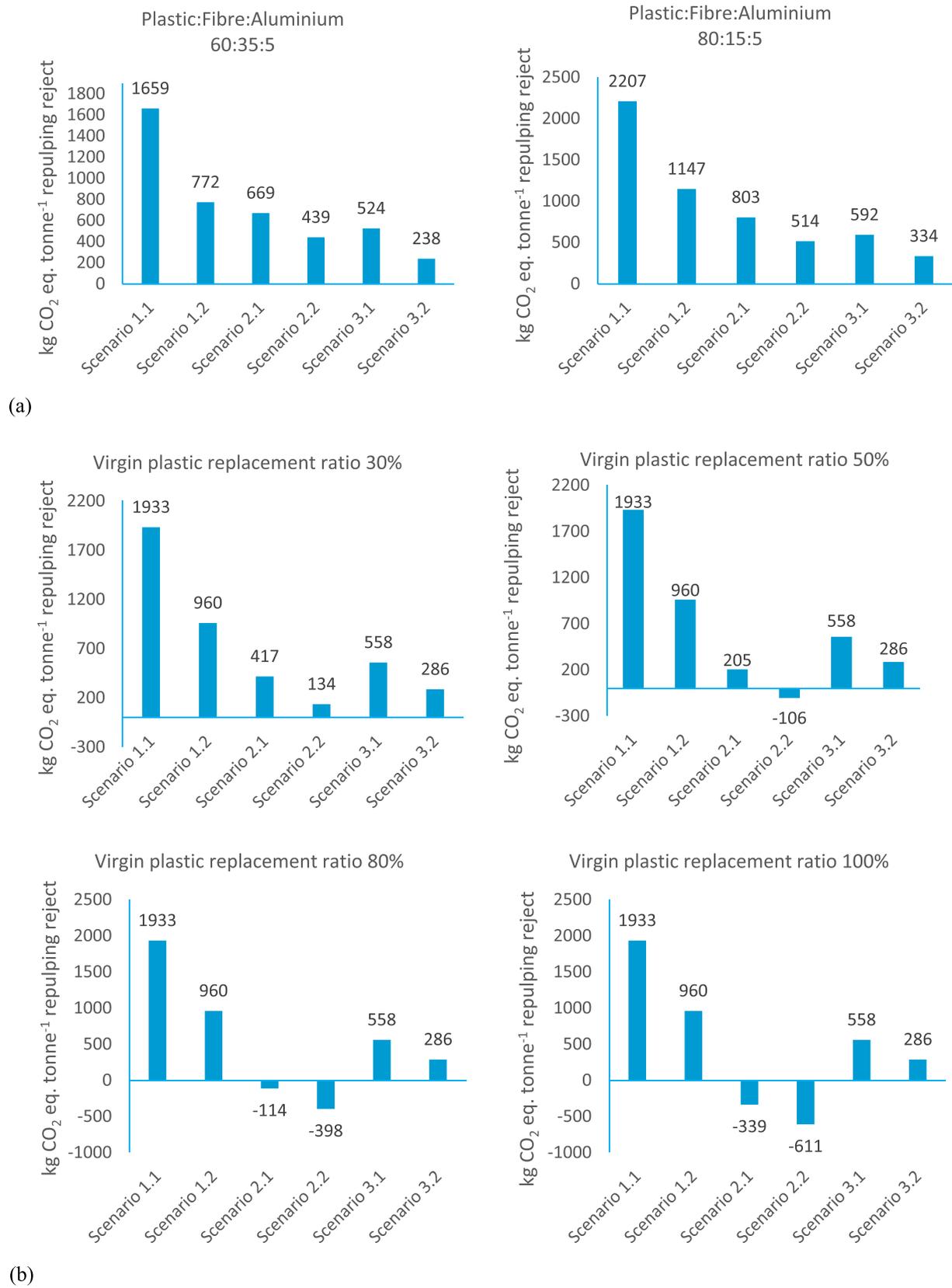


Fig. 3. Results of the uncertainty analysis.

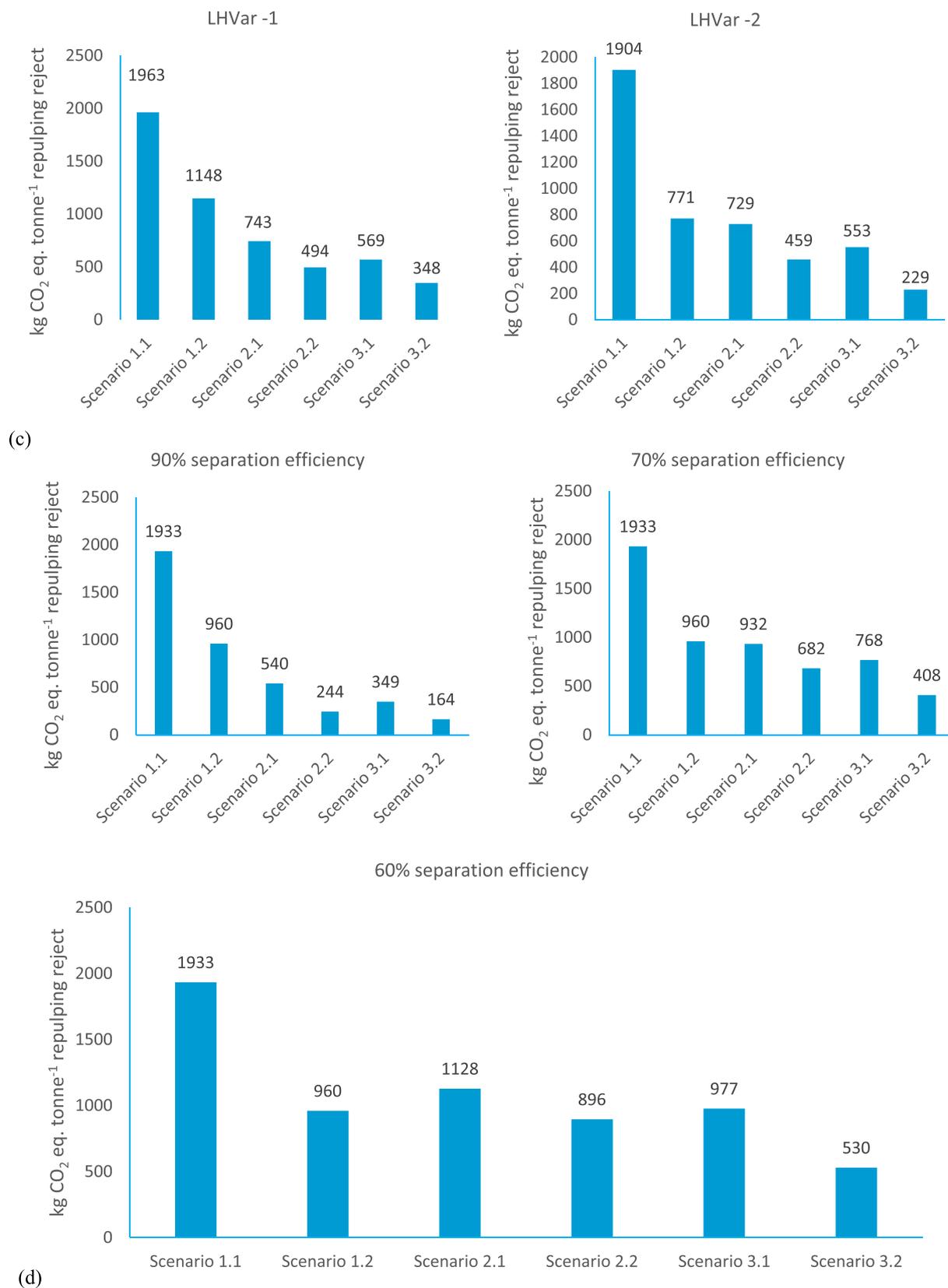


Fig. 3. (continued).

total GWP impact of the chemical recycling scenarios. When the yield of oil increased 9.6%, the emissions decreased by 2–4%. When the yield of oil was decreased by 18%, the emissions increased by 4–8%.

4. Discussion

Due to its composite nature, LPB cannot be recycled by paper mills that recycle paper-based packaging. Recycling LPB requires a different

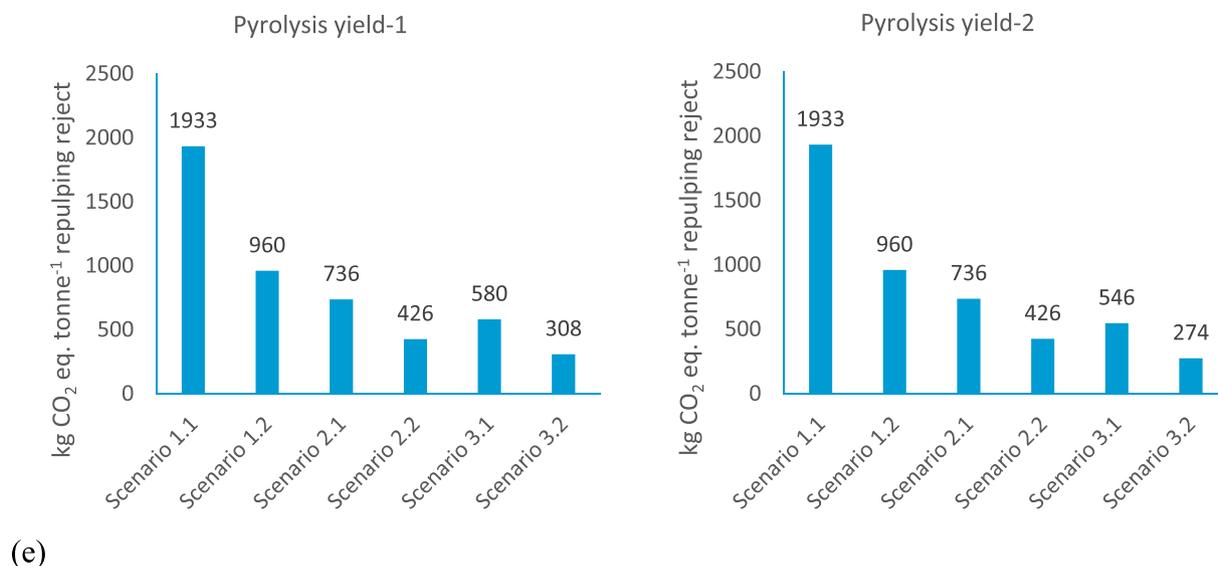


Fig. 3. (continued).

and more intense pulping process than with normal paper and board. In the EU, there are 20 mills available where fibres are separated from plastic and Al (Lahme et al., 2020). Recycling the repulping rejects is more challenging than recycling the fibre fraction because of the immature technology. To address this problem, the ACE has provided some recommendations for its member states in the EU; this includes, for example, appropriate collection and sorting of LPB at a national level, easy separation of non-fibre materials from fibre, introducing high tech technologies that can separate polymer and Al streams more effectively, and manufacturing composite products (Carpentier, 2020; Lahme et al., 2020).

Collection and separation are prerequisites for an efficient recycling system. In Finland, LPBs are collected with other cardboard waste. After collection, either LPB is separated in the sorting facilities before sending it to the paper mills or sent to the recycling centre without separation. Efficient collection and sorting increase the volume of recyclable materials and creates high-quality waste streams. The commingled collection of LPB with other cardboard waste impacts the purity of the repulping reject materials. Therefore, a separate collection of LPB waste would increase the purity of the waste. However, separate collection of LPB will probably result in lower collection rates than co-collection systems, which may discourage the recycling company from recycling LPB waste.

Over the last couple of years, the share of LPB that has been recycled has been steadily increased to recover more fibres. This involved significant investments in a volatile market where the price of related collected paper and board feedstocks has significantly varied in the last decade, which greatly affected these enterprises' profitability. There are several plants in Germany, Finland, and Brazil where the plastic and Al are separated from the fibre and used for different purposes such as making composite products, paraffin, and gas for heating by using mechanical and chemical recycling processes (Pretz and Pikhard, 2010). However, the repulping reject materials are mainly used for energy recovery options because the technology is not advanced enough to efficiently separate the plastic part from the fibre and Al fractions. The recycled PE from mechanical recycling contains impurities such as polypropylene (PP), fibre, and Al. As a consequence, the recycled product from a mechanical recycling system does not have the same quality as virgin materials. This limitation reduces the economic viability of recycling repulping reject. For example, several recycling companies in Germany and Spain started to separate plastic and Al but stopped after a few years of operation because it was not financially feasible (Lahme et al., 2020). In this case, the technology should be

improved to increase repulping reject recycling.

The data used in this study originate from various sources, which may not reflect the real-time situation in the industry. However, based on the available data, this study showed that a significant reduction in GHG emissions could be possible using the chemical recycling process for repulping reject materials. Compared to the incineration process, the chemical recycling process could reduce 70%-71% of GHG emissions, and the mechanical recycling process could reduce 55%-60%. The chemical recycling process (pyrolysis) requires a reasonably pure PE, PP or polystyrene (PS) (Selina et al., 2021; Ragaert et al., 2017). Since LPB reject does not contain O, Cl, N, S or other heteroatoms, it is an ideal feedstock for the pyrolysis process. The chemical recycling process can produce mono plastic; therefore, it can retain the quality of the recycled product. However, chemical recycling is an energy-intensive, and complex process. The other option for addressing repulping reject recycling could be producing composite material, which ACE also recommends. The composite material can be used to make garden furniture, pallets, flower pots, etc. However, further analysis is required to investigate the environmental impacts of the composite material produced from the recycling of repulping reject material.

In summary, this study found that the chemical recycling process had the lowest GWP impact, while the incineration process resulted in the maximum GWP impact. To increase the recycling rate of the repulping reject, it is important to design LPB so that a consumer can easily sort it from other waste streams, and non-fibre materials can be easily separated from the fibre material. Moreover, greater emphasis should be given to technological improvement to increase the separation efficiency of non-fibre materials from fibre materials.

5. Conclusions

In this study, a detailed investigation of the treatment of plastic residue from LPB waste treatment was conducted using LCA. Three different scenarios based on the treatment methods were considered: incineration, mechanical recycling and chemical recycling. On top of the main scenarios, two sub scenarios based on the heating sources were also investigated in this study. Furthermore, five uncertainty analyses were performed.

By conducting LCA, it was found that the incineration process had the maximum GWP impact followed by the mechanical recycling process. The chemical recycling process showed the best environmental performance among all the treatment options, mainly due to avoiding GHG emissions by substituting virgin plastics. By investigating sub-

scenarios, it was found that substitution of bio-based thermal energy resulted in more GWP in all of the main scenarios than natural gas-based thermal energy. The uncertainty analysis showed that the total GWP impact of the treatment options was significantly influenced by the residue composition, separation efficiency, replacement ratio and LHV of plastic and fibre, while pyrolysis yield had a minor impact.

Chemical recycling could be a better option in a circular economy-based approach because it produces substantially lower GHG. The chemical recycling process also helps retain the value of plastic residue for a more extended period by producing material, which can be used as a raw material in other products. On the contrary, mechanical recycling produces mediocre quality plastic. In this case, a more sustainable way to recycle repulping rejects would be using the chemical recycling process. However, the technology related to the pyrolysis process should be increased to reduce energy consumption and increase pyrolysis oil yield.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This study was conducted as part of the PLASTin - All-in for Plastics Recycling project (8907/31/2019). Funding for the project was received from Business Finland and companies.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2021.10.003>.

References

- Abdulkareem, M., Havukainen, J., Nuortila-Jokinen, J., Hortalainen, M., 2021. Environmental and economic perspective of waste-derived activators on alkali-activated mortars. *J. Clean. Prod.* 280, 124651. <https://doi.org/10.1016/j.jclepro.2020.124651>.
- ACE, 2020. Sustainability - Beverage Carton [WWW Document]. URL <https://www.beveragecarton.eu/policy-areas/sustainability/> (accessed 12.14.20).
- Achillas, D.S., Roupakias, C., Megalokonomos, P., Lappas, A.A., Antonakou, E.V., 2007. Chemical recycling of plastic wastes made from polyethylene (LDPE and HDPE) and polypropylene (PP). *J. Hazard. Mater.* 149 (3), 536–542. <https://doi.org/10.1016/j.jhazmat.2007.06.076>.
- Agudo, R., Olazar, M., San José, M.J., Gaisán, B., Bilbao, J., 2002. Wax formation in the pyrolysis of polyolefins in a conical spouted bed reactor. *Energy Fuels* 16 (6), 1429–1437. <https://doi.org/10.1021/ef020043w>.
- Alfa Laval, 2020. Alfa Laval - P1 [WWW Document]. URL <https://www.alfalaval.com/products/separation/centrifugal-separators/decanter/p1/> (accessed 4.13.21).
- Arvanitoyannis, I., 2005. Food packaging technology. Edited by R Coles, D McDowell and MJ Kirwan. Blackwell Publishing, CRC Press, Oxford, 2003. 346 pp ISBN 0-8493-97788-X. *J. Sci. Food Agric.* 85, 1072–1072. <https://doi.org/10.1002/jsfa.2089>.
- Bezergianni, S., Chryssikou, L.P., 2016a. Sustainability Assessment of Fuels Production via Hydrotreating Waste Lipids and Co-processing Waste Lipids with Petroleum Fractions. Springer, Cham.
- Bezergianni, S., Chryssikou, L.P., 2016b. Sustainability assessment of fuels production via hydrotreating waste lipids and co-processing waste lipids with petroleum fractions. *Green Energy Technol. Part F2*, 387–400. https://doi.org/10.1007/978-3-319-30127-3_30.
- Bisinella, V., Albizzati, P., Astrup, T., Damgaard, A., 2018. Life Cycle Assessment of management options for beverage packaging waste, Danish Ministry of Environment and Food.
- Boustead, I., 2005. European Plastics Industry.
- Carpentier, A., 2020. Best Practices Best Practices.
- Ecomation, 2016. The making of a circular economy.
- Eriksson, O., Finnveden, G., 2009. Plastic waste as a fuel - CO₂-neutral or not? *Energy Environ. Sci.* 2, 907–914. <https://doi.org/10.1039/b908135f>.
- Europa, 2018. DIRECTIVE 2008/98/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL [WWW Document]. URL <https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:02008L0098-20180705&from=EN#toclid59> (accessed 8.19.21).
- European Commission, 2020. Waste Framework Directive [WWW Document]. URL https://ec.europa.eu/environment/topics/waste-and-recycling/waste-framework-directive_en (accessed 10.7.21).
- Eurostat, 2021. More than 40% of EU plastic packaging waste recycled [WWW Document]. URL <https://ec.europa.eu/eurostat/de/web/products-eurostat-news/-/ddn-20210113-1> (accessed 4.1.21).
- Guo, M., Murphy, R.J., 2012. LCA data quality: Sensitivity and uncertainty analysis. *Sci. Total Environ.* 435–436, 230–243. <https://doi.org/10.1016/j.scitotenv.2012.07.006>.
- Harst-winttraecken, E.J.M. Van Der, 2015. Robustness of Life Cycle Assessment Results - Influence of data variation and modelling choices on results for beverage packaging materials.
- Hortalainen, M., Teirasvuo, N., Kapustina, V., Hupponen, M., Luoranen, M., 2013. The composition, heating value and renewable share of the energy content of mixed municipal solid waste in Finland. *Waste Manag.* 33 (12), 2680–2686. <https://doi.org/10.1016/j.wasman.2013.08.017>.
- IEL, 2019. Plastic & Climate: The Hidden Costs of a Plastic Planet.
- Ingrao, C., Lo Giudice, A., Tricase, C., Rana, R., Mbohwa, C., Siracusa, V., 2014. Recycled-PET fibre based panels for building thermal insulation: Environmental impact and improvement potential assessment for a greener production. <https://doi.org/10.1016/j.scitotenv.2014.06.022>.
- ISO, 2006. ISO 14040:2006(en), Environmental management — Life cycle assessment — Principles and framework [WWW Document]. URL <https://www.iso.org/obp/ui/#iso:std:iso:14040:ed-2:v1:en> (accessed 7.4.18).
- Jamradloedluk, J., Lertsatitthanakorn, C., 2014. Characterisation and utilisation of char derived from fast pyrolysis of plastic wastes. *Procedia Eng.* 69, 1437–1442. <https://doi.org/10.1016/j.proeng.2014.03.139>.
- Jelse, K., Eriksson, E., Einarson, E., 2009. Life Cycle Assessment of consumer packaging for liquid food 46.
- Jeswani, Harish, Krüger, Christian, Russ, Manfred, Horlacher, Maike, Antony, Florian, Hann, Simon, Azapagic, Adisa, 2021. Life cycle environmental impacts of chemical recycling via pyrolysis of mixed plastic waste in comparison with mechanical recycling and energy recovery. *Sci. Total Environ.* 769, 144483. <https://doi.org/10.1016/j.scitotenv.2020.144483>.
- Kalargaris, I., Tian, G., Gu, S., 2017. The utilisation of oils produced from plastic waste at different pyrolysis temperatures in a DI diesel engine. *Energy* 131, 179–185. <https://doi.org/10.1016/j.energy.2017.05.024>.
- Kaza, S., Yao, L., Bhada-Tata, P., Van Woerden, F., 2018. What A Waste 2.0 - A Global Snapshot of Solid Waste Management to 2050. <https://doi.org/10.1596/978-1-4648-1329-0>.
- Korhonen, M., Dahlbo, H., 2007. Reducing Greenhouse Gas Emissions by Recycling Plastics and Textiles into Products. Helsinki, Finland.
- Lahme, V., Daniel, R., Marsh, P., Molteno, S., 2020. Recycling of multilayer composite packaging: the beverage carton.
- Lazarevic, David, Aoustin, Emmanuelle, Buclet, Nicolas, Brandt, Nils, 2010. Plastic waste management in the context of a European recycling society: Comparing results and uncertainties in a life cycle perspective. *Resour. Conserv. Recycl.* 55 (2), 246–259. <https://doi.org/10.1016/j.resconrec.2010.09.014>.
- Lee, K.-H., 2012. Effects of the types of zeolites on catalytic upgrading of pyrolysis wax oil. *J. Anal. Appl. Pyrolysis* 94, 209–214. <https://doi.org/10.1016/j.jaap.2011.12.015>.
- Liikanen, M., Havukainen, J., Hupponen, M., Hortalainen, M., 2017. Influence of different factors in the life cycle assessment of mixed municipal solid waste management systems e A comparison of case studies in Finland and China. *J. Clean. Prod.* 154, 389–400. <https://doi.org/10.1016/j.jclepro.2017.04.023>.
- Miandad, R., Barakat, M.A., Aburizaiza, A.S., Rehan, M., Ismail, I.M.I., Nizami, A.S., 2017. Effect of plastic waste types on pyrolysis liquid oil. *Int. Biodeterior. Biodegrad.* 119, 239–252. <https://doi.org/10.1016/j.ibiod.2016.09.017>.
- Mohan, A.M., 2015. Outlook for global liquid paperboard market is bright | Packaging World [WWW Document]. URL <https://www.packworld.com/design/materials-containers/news/13368355/outlook-for-global-liquid-paperboard-market-is-bright> (accessed 12.14.20).
- Plastic Europe, 2016. Eco-profiles and Environmental Product Declarations of the European Plastics Manufacturers.
- Plastic Europe, 2012. Ethylene, Propylene, Butadiene, Pyrolysis Gasoline, Ethylene Oxide (EO), Ethylene Glycols (MEG, DEG, TEG). Brussels.
- Plastics Recyclers Europe, 2018. Chemical Recycling | Plastics Recyclers Europe [WWW Document]. accessed 2.19.21. <https://www.plasticsrecyclers.eu/chemical-recycling>.
- Pretz, T., Pikhart, O., 2010. Beverage carton recycling.
- Ragaert, K., Delva, L., Van Geem, K., 2017. Mechanical and chemical recycling of solid plastic waste. *Waste Manag.* 69, 24–58. <https://doi.org/10.1016/j.wasman.2017.07.044>.
- Reimann, D.O., 2012. CEWEP Energy Report III.
- Robertson, Gordon, 2021. Recycling of aseptic beverage cartons: A review. *Recycling* 6 (1), 20. <https://doi.org/10.3390/recycling6010020>.
- Ruttenborg, V., 2017. Life Cycle Assessment of fresh dairy packaging at ELOPAK. Norwegian University of Science and Technology.
- Selina, Möllnitz, Markus, Bauer, Daniel, Schwabl, Renato, Sarc, 2021. Wet-mechanical processing of a plastic-rich two-dimensional-fraction from mixed wastes for chemical recycling. *Waste Manag. Res.* 39 (5), 731–743. <https://doi.org/10.1177/0734242X21996435>.
- Shah, Jasmin, Jan, M. Rasul, Mabood, Fazal, Jabeen, Farah, 2010. Catalytic pyrolysis of LDPE leads to valuable resource recovery and reduction of waste problems. 51 (12), 2791–2801. <https://doi.org/10.1016/j.enconman.2010.06.016>.
- Sphera, 2020. Life Cycle Assessment (LCA) Software - Sphera [WWW Document]. URL <https://sphera.com/life-cycle-assessment-lca-software/> (accessed 4.6.21).

- Storaenso, 2020. Liquid packaging - Paperboard Materials | Stora Enso [WWW Document]. URL <https://www.storaenso.com/en/products/paperboard-materials/liquid-packaging> (accessed 12.10.20).
- Syamsiro, M., Hu, W., Komoto, S., Cheng, S., Noviasri, P., Prawisudha, P., Yoshikawa, K., 2013. Co-production of liquid and gaseous fuels from polyethylene and polystyrene in a continuous sequential pyrolysis and catalytic reforming system. *Energy Environ. Res.* 3 <https://doi.org/10.5539/eer.v3n2p90>.
- Uddin, Md Azhar, Koizumi, Kazuo, Murata, Katsuhide, Sakata, Yusaku, 1997. Thermal and catalytic degradation of structurally different types of polyethylene into fuel oil. *Polym. Degrad. Stab.* 56 (1), 37–44. [https://doi.org/10.1016/S0141-3910\(96\)00191-7](https://doi.org/10.1016/S0141-3910(96)00191-7).
- Väntsi, O., Kärki, T., 2015. Environmental assessment of recycled mineral wool and polypropylene utilised in wood polymer composites 104, 38–48. <https://doi.org/10.1016/j.resconrec.2015.09.009>.
- Verghese, K., Lockrey, S., Clune, S., Sivaraman, D., 2012. Life cycle assessment (LCA) of food and beverage packaging. *Emerg. Food Packag. Technol. Princ. Pract.* <https://doi.org/10.1016/B978-1-84569-809-6.50019-1>.
- von Falkenstein, Eva, Wellenreuther, Frank, Detzel, Andreas, 2010. LCA studies comparing beverage cartons and alternative packaging: Can overall conclusions be drawn? *Int. J. Life Cycle Assess.* 15 (9), 938–945. <https://doi.org/10.1007/s11367-010-0218-x>.
- Wellenreuther, F., Falkenstein, E. Von, Detzel, A., 2012. Comparative Life Cycle Assessment of beverage cartons combiblocSlimline and combiblocSlimline EcoPlus for UHT milk.
- Williams, Paul T, Williams, Elizabeth A, 1999. Fluidised bed pyrolysis of low density polyethylene to produce petrochemical feedstock. *J. Anal. Appl. Pyrolysis* 51 (1-2), 107–126. [https://doi.org/10.1016/S0165-2370\(99\)00011-X](https://doi.org/10.1016/S0165-2370(99)00011-X).