Energy efficiency and environmental assessment of papermaking from chemical pulp - A Finland case study.

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Abstract

Pulp and paper manufacturing sector constitutes one of the largest industry segments in the world in terms of water and energy usage as well as of significant use and release of chemicals and combustion products. Since its chief feedstock –wood fiber– is renewable, this industry can play an important role in sustainable development, becoming an example of how a resource can be managed to provide a sustained supply to meet society's current and future needs. This calls for a thorough assessment of environmental costs and impacts associated to pulp and paper operations, including both direct and indirect inputs supporting the whole papermaking process as well as the main outputs, co-products and by-products. By means of Life Cycle Assessment (LCA) methodology, this paper aims at assessing the environmental sustainability of the pulp and paper production so as to identify those phases across the whole supply chain that entail the highest environmental loads, thus requiring improvements. To determine the environmental impacts as accurately as possible, the manufacturing stages performed in the pulp and paper mill complex of Stora Enso Oyj Veitsiluoto Mills at Kemi, Northern Finland, were taken as a model and assessed by means of the SimaPro 8 LCA software, utilizing ReCiPe Midpoint (H) method for the impact assessment. As expected, most of the resulting impacts are caused by the industrial production phase. The production processes of pulp and paper jointly affect all the investigated impact categories with the highest shares, ranging from 50% of generated impacts on water depletion up to 88% on freshwater eutrophication. Generally, the main contributions to environmental loads come from the electricity and heat requirements and, only at a minor extent, from the use of chemicals such as the sodium hydroxide and sodium chlorate. In particular, pulp production process generates the main loads on global warming (46% of the total impacts), ozone depletion (39%), freshwater eutrophication (55%), human toxicity (46%), metal depletion (42%) and fossil depletion (46%). In the remaining investigated impact categories, namely terrestrial acidification, photochemical oxidant formation and terrestrial ecotoxicity, most of impacts derive from the use of optical brighteners and fillers in the final steps of paper production and from the intensive consumption of water in the recycling step of end-of-life affecting water depletion. Moreover, the implementation of measures for material and energy efficiency in the assessed system, such as the use of renewable energy generated *in situ* from black liquor and residual biomass to support the requirements of the integrated pulp and paper mills and the waste paper recycling, resulted to be crucial in lowering the environmental burdens. In particular, the partial fulfillment of electricity and heat requirements by means of a circular use of residues within the system leads to a noteworthy reduction of impacts in all the investigated impact categories, up to more than 70% in global warming and fossil depletion potentials, thus contributing to higher process sustainability compared with other averaged European systems for paper production.

The obtained research results are a valuable source of management information for the decision makers, at both company and national levels, with the aim to improve the environmental performance of pulp and paper industry.

1. Introduction

A reduction in the industrial usage of energy is a valuable means of countering the threat of increased global warming, caused by human use of fossil fuels (IPCC, 2014). The results of modeling simulations by the IEA (2011) for the year 2035, suggest that about half of the cumulative emission reductions, required to meet the 2°C target, can be achieved through improved energy-efficiency. In the industry sector, this share is even higher, amounting to 60% (IEA, 2011). More efficient production processes throughout a reduced consumption of energy and of natural resources are a key goal for energy-intensive industries worldwide, in order to strengthen the overall performance against the increasing energy prices, the resource scarcity and the global environmental concerns (Gaudreault et al., 2010). In particular, the pulp and paper industry ranks fourth in terms of energy consumption among industries, accounting for almost 6% of total industrial energy consumption and 2% of direct industrial carbon dioxide (CO_2) emissions (IEA, 2016). Not only energy (fossil fuels, electricity) but also natural resources (water, wood) as well as chemicals are intensively depleted by the pulp and paper sector (Avşar and Demirer, 2008). Nonetheless, it is important to stress the occurred decoupling of growth in energy use and production: despite an increase of 23% in paper and paperboard production in the last two decades, the sector's energy use has grown only 1%, due to the high share of biomass as substrate and the use of by-products, such as black liquor, sawdust, wood chips and other wood residues, for steam and electricity generation (EC, 2015; IEA, 2017). Since the most significant emissions associated with the pulp and paper industry are generated by the off-site production of steam and electricity, the increasing rate of combustion of by-products on-site to this aim is helpful in lowering the indirect emissions. Actually, it has been reported that, since 1990, CO_2 emissions intensity of the European paper industry have decreased by approximately 25% (Worrell, 2011) and the share of wood pulp in paper production has decreased from 52% in 2000 to 43% in 2014 (FAO, 2016), thanks to improved waste paper recovery and recycling. Therefore, the improvement efforts of this industrial sector are undeniable. However, the demand for household, sanitary and office papers is projected to steadily grow, due to rising populations and incomes, and rising packaging material needs for shipping of consumer goods (FAO, 2016). Major reductions in energy use and CO_2 emissions are still needed, with energy use and direct non-biomass CO_2 emissions declining by 0.8% and 17%, respectively, by 2025 from 2014 levels to meet the 2°C scenario (2DS) (IEA, 2017). This calls for the development and deployment of technological innovations, such as better processes and materials for pulp and paper production or technologies that can economically capture and store the CO_2 emissions, and the application of appropriate assessment tools for ensuring both energy and material efficiency (Kong et., 2016). Thus, although among the top industrial energy consumers, the pulp and paper sector can play an important role in the transition to a low-carbon energy system throughout emerging technologies and proper assessment procedures as key elements in the mitigating strategies against energy and environmental costs.

In such a context, the majority of the studies available in the scientific literature analyses the environmental impacts related to the pulp and paper industry and their potential reduction by means of specific measures, such as the use of cleaner energy, green chemicals, non-virgin materials as well as the recycling of pulping and papermaking by-products (Counsell and Allwood, 2007; Corcelli et al., 2017; Corcelli et al., 2018; Kong et al., 2016; Kong et al., 2017). In line with other studies (Wiegard, 2001; Dias et al., 2002; Holmgren and Hening, 2005; Murphy and Power, 2007; Schmidt et al., 2007; Merrild et al., 2008), Poopak and Reza (2012) calculated the potential environmental benefits of using non-virgin material (i.e. bagasse) instead of wood in a paper and pulp factory in Iran. Moreover, Krishna Manda et al. (2012) proved that the use of new coatings (micro or nano TiO_2), in combination with the different pulp types, brings savings in wood, energy, GHG emissions and other environmental impacts in comparison with conventional paper. Other studies are focused on improvements in energy efficiency in the paper industry. Hong et al. (2011) and Fleiter et al. (2012) evaluated that cleaner process technologies (such as heat recovery in paper mills and the use of innovative paper drying technologies) can significantly improve energy efficiency in the pulp and paper industry, leading in turn to lower carbon emissions. However, as recently noted by Silva et al. (2015), few Life Cycle Assessment (LCA) studies have focused on the pulp and paper sector, although LCA is one of the main techniques for quantitatively assessing environmental impacts during the life cycle of products and processes (ISO, 2006 a,b). LCA is an helpful tool to identify the most relevant environmental impacts and hotspots and to underpin decision-making strategies for environmental improvements in a life cycle perspective (Baumann and Tillmann, 2004). Lopes et al. (2003) performed an LCA of pulp and paper industry in Portugal and reported that the substitution of heavy fuel oil by natural gas in the pulp and paper production processes is environmentally beneficial. Dias et al. (2007) studied the offset paper made from *Eucalyptus globulus* in Portugal and evaluated the effect of differences in the market where the product is consumed: German market vs. Portuguese market. The paper consumed in Portugal showed lower environmental impacts in the distribution phase, but higher impacts in the final disposal phase, due to CH_4 emissions from landfills significantly impacting the categories of global warming and photochemical oxidant formation. Jawjit et al. (2007) highlighted several environmental impacts caused by the eucalyptus-based Kraft pulp industry in Thailand, such as impacts on global warming, acidification, eutrophication, photochemical smog, toxicity and the production of solid waste. In a comparative LCA of paper produced from eucalyptus (*Eucalyptus globulus*) and hemp (*Cannabis sativa*), Vieira et al. (2010) found that paper made in Portugal from industrial hemp generates higher environmental burdens than from eucalyptus. The main differences between the life cycles were in terms of global warming, acidification, eutrophication, photochemical oxidant formation, and land use impacts during the fiber and pulp production stages. This was mainly due to the fact that the cultivation of hemp requires larger amounts of fertilizers and more mechanical operations in crop production, and also consumes larger amounts of chemical additives in pulp production. Gonzalez et al. (2011) presented a life cycle inventory of pine and eucalyptus pulp production in Chile, which indicated that pine-based pulp generates greater chemical and environmental loads than eucalyptus, mainly because the liquid effluents it generates may seriously affect the quality of receiving waters. In a more recent study, Silva et al. (2015) provided an LCA of offset paper production in Brazil and suggested alternatives options for thermal energy generation and chemical recovery in the pulp and paper industry.

In spite of the existing previous analyses concerning LCA of papermaking, the present study represents a novelty and a more comprehensive assessment in that, to the best of our knowledge, no LCA studies have been reported for papermaking in Finland, although Finland is one of the main producer and exporter of pulp and paper. Indeed, Finland is the second producer country in Europe (CEPI, 2016), with 10.1 million tons of paper and paperboard produced (Finnish Forest Industry Federation, 2017). Among the energy intensive industries, the pulp and paper industry accounts for nearly 50% of the annual industrial energy use (Official Statistics of Finland), something which in turn implies that this sector in Finland will be a crucial target for the implementation of energy efficiency policies. Furthermore, since Finnish technology for papermaking is exported to many countries worldwide, the Finnish solutions are considered internationally relevant (Lodenius et al., 2009).

Thus, the present study fills this gap by performing a comprehensive LCA of paper (office and magazine) produced in Finland. The life cycle of the paper production process was assessed by means of LCA, with special focus on those steps and hotspots that present the highest environmental impacts and proposing improvement scenarios for minimizing such impacts. The novelty value of this study consists in the presentation of the LCA results split over different stages of the production process, including the forestry phase, the pulping and papermaking phases, the production of energy *in situ*, the final distribution and the end-of-life (EoL). This was possible thanks to high-quality local data provided from a leading company producer of pulp and paper in Finland for the core processes (pulping and papermaking). These data (referred to year 2015) were thoroughly analyzed (further details are given below), with the main objectives of: i) assessing the environmental loads generated across the whole supply chain of pulp and paper; ii) quantifying the potential benefits deriving from energy and material efficiency measures; iii) proposing the most efficient options for improving the overall environmental performance of paper production from virgin pulp up to EoL. In so doing, transparent and disaggregated information for each process stage are beneficial both to assess the impacts of the papermaking process and to provide detailed life cycle inventory data potentially useful for other studies on the LCA of papermaking.

2. Case study

2.1 Description of the mill

Stora Enso Oyj Veitsiluoto Mills is located at Kemi (65°41'28"N, 24°37'23"E), Northern Finland, in the province of Lapland and it is one of the world’s leading companies for paper and pulp production (Brogaard et al., 2014).

Detailed information about Stora Enso company can be found in the Sustainability Report (http://assets.storaenso.com/se/com/DownloadCenterDocuments/Sustainability_Report_2015.pdf) and in

Veitsiluoto Mill-EMAS Environmental Statement 2010 (http://docplayer.net/20703600-Veitsiluoto-mil-emas-environmental-statement-2010.html).
The pulp mill is fully integrated into the paper mill (i.e. pulp and paper manufacturing taking place within the same site). The annual production of Stora Enso Oyj Veitsiluoto Mills consists of 420,000 tons of bleached softwood and hardwood pulps, 580,000 tons of uncoated office paper, 280,000 tons of coated magazine paper and 200,000 m^3 of sawn goods (Stora Enso, 2015. Personal communication). In order to minimize the amount of waste to be disposed of in landfills and to utilize the thermal energy value of produced waste, wood wastes from wood handling plant, sawmill and groundwood mill are incinerated for energy production in a fluidized bed combustion boiler (246 MW). The biosludge deriving from the biological wastewater treatment plant of the pulp and paper mill is also incinerated, in order to reduce its volume and avoid the need for disposal in a landfill (Nurmesniemi et al., 2007). The remaining energy requirements are satisfied by means of gas, fossil fuels and electricity purchased from the national grid. The ashes (i.e., bottom ash and fly ash) derived from the combustion process are totally reused, either as a hardener in filling mine cavities at a mine located nearby or at the mill area for landscaping the ash basin. Furthermore, most of the green liquor dregs from the pulp mill are utilized as neutralizing agents for acidic wastewaters (Pöykiö et al., 2006), whereas all the paper mill sludges (i.e. fibre clay) from the chemical wastewater treatment plant are used as hydraulic barrier material for landfills and for landscaping (Nurmesniemi et al., 2007).

2.2 Description of the system

The investigated system, described in terms of flows of lignocellulosic material, is shown in Figure 1. Five main subsystems (forestry, pulp production, paper production, distribution, end-of-life), briefly described below, as well as wastewater and solid waste treatment plants were included in the assessment, whereas phases such as the transportation to the customer and the use of the paper products were not evaluated, according to Lopes et al. (2003) and to Ghose and Chinga-Carrasco (2013). Although the role of paper as a service provider in society is extremely important (e.g. for food packaging), the focus of this study is on the production of paper itself, rather than on the service provided. Furthermore, the production and maintenance of capital goods (buildings, machinery and equipment) has been included within the system boundaries.

Forestry phase: it includes all the operations carried out in Spruce and Pine stands: silviculture operations (site preparation, stand establishment and tending) and logging operations (harvesting and forwarding). A more detailed description of these activities can be found in Karjalainen and Asikainen (1996). Spruce and Pine plantations were considered as they are the only raw material processed in the pulp mill. Seedling production was excluded from the system boundaries, due to the lack of data, in agreement with other forest-related LCA studies (Gonzalez-García et al., 2014).
Transport of wood from forest lands –located in Finland, Sweden, Baltics, Russia– to the pulp and paper production mill is also included. Three modes of transport, i.e. via truck, rail and freight vessel, were accounted for. The average distance travelled was 125 km by truck (53%), 292 km by trains (40%) and 1156 km by freight vessel (7%) (Stora Enso, 2015. Personal Communication). In particular, the inland road transport was via >32t trucks for raw material input and final product output. The rail and ship contributed to most of the transport abroad.

Pulp production phase: it includes all the industrial activities related to pulp production, which take place in the pulp mill, i.e. timber debarking, chipping into regular size, digesting (or cooking), pulp washing, pulp screening and primary refining (I), bleaching processes.
This phase starts with the woodchips production lines, which involve debarking and scrubbing of harvested Spruce/Pine wood. The scrubbed wood then goes into chippers and the resulting woodchips are sent to digesters (Pokhrel and Viraraghavan, 2004), while the bark is used as biomass to produce thermal energy. The next step is the cooking process using the Kraft method, in which the wood is mixed with chemicals (e.g., sodium hydroxide/caustic soda) and steam-heated in bath digesters to dissolve lignin and separate the wood fibers. The main products resulting from the cooking process are cooked pulp and black liquor (Hynninen, 1998). The next steps are washing, screening and refining, in which the diluted cooked pulp is washed and then passed through a scrubber to eliminate fiber agglomerates and impurities. The crude fibers are then separated from the black liquor and washed to remove residual chemicals. The unbleached Kraft pulp goes on to the bleaching plant, where residual lignin is removed by treating the pulp with oxygen and hydrogen peroxide at alkaline pH

(Hynninen, 1998). While bleached pulp is produced, the black liquor from the cooking process is used as fuel for industrial boilers to generate thermal energy used in the industrial production subsystem. At the same time, black liquor is rich in alkali and dissolved organic matter and allows the recovery of cooking chemicals (i.e. white liquor) in the chemical recovery step. Bleached pulp, the final product of this phase, is then transferred to the paper production phase.

Paper production phase: it includes all the industrial activities related to paper production, which take place in the paper mill, integrated within the pulp mill, i.e. pulp screening and secondary refining (II), forming, pressing, drying and finishing.
During this final phase, the bleached pulp enters papermaking process, where a suspension of fibers in water with a suitable consistency is formed and the remaining fibrous and non-fibrous components are added to it – precipitated calcium carbonate, adhesive agents and additives (e.g. fungicides, algicides, antifoam). In the papermaking process, after going through refining, purification, pressing and drying processes, the paper sheet is formed. Lastly, paper coating and finishing operations are performed and the product is packaged.

Energy production *in situ*: it involves the production *in situ* of heat and electricity to be used in the pulp and paper mills by means of combustion of biomass waste, black liquor and biosludge from wastewater treatment plants in cogeneration units. The additional energy requirements are purchased from the national grid.

Distribution phase: it includes the transportation of produced paper from the pulp and paper mills (Kemi) to the delivery point (Paris). The average distance from Kemi (Finland) to Zeebrugge (Belgium) is 2617 km and it was assumed to be travelled by freight vessel; the average distance from Zeebrugge (Belgium) to Paris (France), i.e. 300 km, was supposed to be travelled by truck.

End-of-life phase: it includes the management of waste paper, collected through the usual treatment of municipal waste. In this investigation, the recycling option was preferred to other ways of handling the used paper (incineration or landfilling) in a closed loop perspective. Indeed, according to the European Paper Recycling Council (EPRC), the European paper recycling value chain has already made significant strides on the paper recycling rate in the EU, having reached a near theoretical maximum of 71.5% in 2015 (EPRC, 2016). Thus, this study was performed under the assumption that recycled paper replaces primary fibres in the production of virgin paper. Recycling waste materials into new (“secondary”) products may require significant inputs of energy for collection/treatment and certainly generates additional emissions. Therefore, this phase was included in the present study in order to identify both costs and benefits from paper recycling. The datasets for recycled paper, adjusted from the Ecoinvent database (Haschier, 2007), consist of the following steps: collection and sorting of waste paper, deinking of pulp, recycled paper production, energy production on-site, internal waste water treatment and transports of the auxiliaries to the paper mill. For comparison purposes, the options of incineration and landfilling of waste paper were analyzed as well.

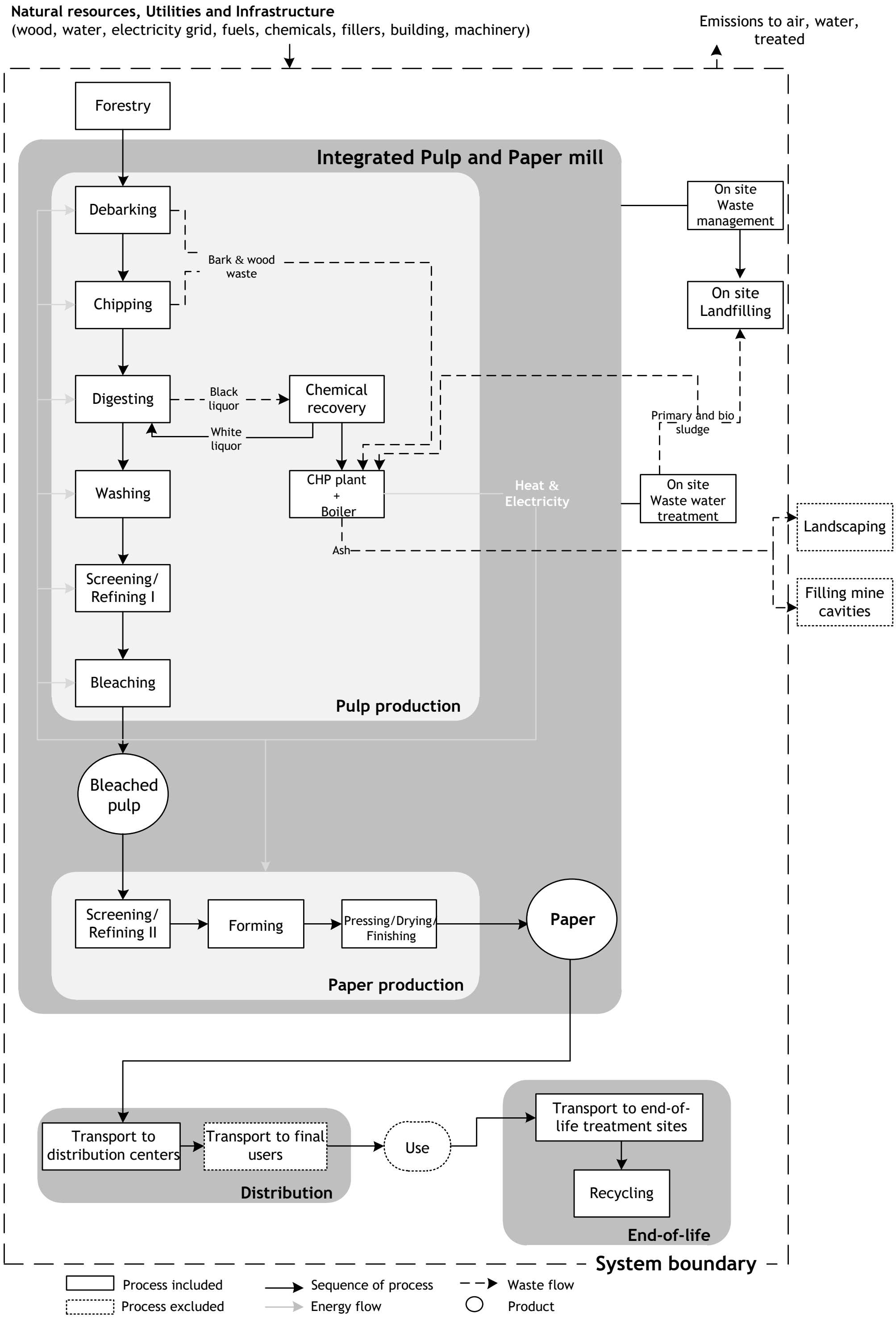


Figure 1. System boundaries and process chain under study.

3. Method

The methodological framework used in this paper was the LCA as defined by ISO standards (ISO 2006, a, b) and ILCD Handbook guidelines (EC 2010, 2011). LCA is a method that attempts to quantify the environmental impacts associated with a product or service throughout its lifecycle. It is defined as a technique for the compilation and evaluation of inputs, outputs and potential environmental impacts of a product system throughout its life cycle – from the extraction of resources, through the production of materials, parts and the product itself, the use of the latter, and the management after it is discarded, either by reuse, recycling or final disposal of (from 'cradle to grave' or 'cradle to cradle', according to a very common definition of LCA). According to the ISO standards (ISO 2006, a, b) and ILCD Handbook guidelines (EC 2010, 2011), LCA is compiled of several interrelated components: i) goal and scope definition; ii) inventory analysis (LCI); iii) impact assessment (LCIA) and iv) interpretation of results for explanation of conclusions and recommendations. The same scheme is followed in this paper.

3.1 Goal and scope definition

The goal of this study was to analyze and quantify the environmental impacts associated to the production of paper, so as to identify those steps along the production process chain that entail the highest environmental impacts. To determine the environmental impacts as accurately as possible, the phases of the production process were modelled on the basis of the manufacturing processes used by the pulp and paper mill complex of Stora Enso Oyj Veitsiluoto Mills in Finland. Decision-makers can refer to this study to identify the aspects influencing the environmental performance of papermaking and hence to plan strategies for the achievement of effective industrial energy efficiency and optimized use of resources. The innovation of applying the LCA to this specific case study consists in relying on high quality local data for the core processes (pulping and papermaking) and in splitting electricity and heat requirements, chemicals demand and emission levels over all the production steps, overcoming potential methodological limitations.

According to the ISO standards (ISO 2006, a, b), a functional unit should represent qualitative and quantitative aspects of the function that a product proposes to fulfill. The functional unit (FU) must be measurable and it is established to provide a base of reference to which inventoried input and output data are referred (ISO 2006, a). The FU selected in this study was the production of 1 ton of produced paper (67% of office paper and 33% of magazine paper).In particular, the investigated paper is produced from virgin fibre chemical pulp. Indeed, Finnish paper production is primarily based on the use of virgin fibre, since recycled fibre accounts only for about 5% of all the fibre raw material used by the paper and paperboard industry (www.forestindustries.fi).

According to the ILCD Handbook (EC, 2010), the analyzed context can be identified as a micro-level decision support (so-called situation A) and an attributional LCI modeling framework was therefore applied.

Concerning the definition of system boundaries, in the present study focused on paper production, the *cradle-to-cradle* approach was adopted, assuming that the end of life disposal step for produced paper was a recycling process (incineration of waste paper was evaluated only for comparative purposes). In order to focus on the performance of the investigated mills and compare it with other systems, a *cradle-to-gate* approach was also assumed. Generally, allocation of environmental impacts between products and coproducts in multi-output systems is one of the most critical issues in LCA methodology. The ISO 14044 (ISO 2006, b) standard recommends avoiding allocation (e.g. based on physical or economic relationships), whenever possible, either through subdivision of processes or by expanding the system boundary to include the additional functions related to them. For this reason, a system boundaries expansion (or avoided burden approach) was performed, based on average data (i.e. market mix) for crediting energy recovery and on virgin production processes for paper recycling.

In the case of forestry system under study, the total wood-biomass production was considered as a whole. The residual biomass generated in the harvesting step (such as leaves, stools and branches) were not computed in the analysis as co-products since they remain in the plantation, contributing to soil quality, in agreement with other forest-related LCA studies (Berg and Lindholm, 2005; Dias and Arroja, 2012; Gonzàlez- García et al., 2009). In the case of pulp and paper production phases, it was assumed that recycled paper displaces paper of equivalent grade made from virgin fibre. The environmental burdens of the production of virgin paper were subtracted from the environmental burdens of the recycling process. Moreover, in the pulp mill, besides the main product (sulphate pulp), tall oil and turpentine are usually produced as further products. Also in this case, no allocation was applied, since, accordingly to the Ecoinvent database (Wernet et al., 2016), it was considered that tall oil and turpentine are by-products (a pulp mill is aimed at pulp production, but cannot avoid these other substances).

3.2 Inventory data analysis and assumptions

Data used for an LCA must have a high level of quality and reliability, as being dependent on its capacity to accommodate the system and reflect reality will ultimately provide more credibility to the LCA (Björklund 2002). In this study, data were obtained from multiple sources. Personal interviews and company visits were integrated with environmental declarations, internal company reports and bibliographic sources. Primary and site-specific data, supplied by the Stora Enso company, were used for the foreground system, in particular for processes specifically related to the transport (from forest to mill gate and from the latter to delivery point) and to pulping and papermaking. Moreover, the input data, related to the use of water and chemicals, are inclusive of water and chemicals recovered in the pulping process. It is noteworthy that the production process applied in the investigated mill can be considered representative of the most advanced technology in office paper production in Finland.

The total amounts of consumed energy and water are updated and site-specific (data from Stora Enso company), but data related to split of energy demand and water consumption over the investigated steps of the production process were not available and secondary data were used to this aim. Specifically, the distribution of total amounts of consumed energy and water was performed according to Giraldo & Hyman (1996) and Brown et al. (1996), whereas forest operations and emissions were obtained from Doherty (1995) and Karjalainen and Asikainen (1996). Although forestry technologies improve overtime in terms of energy and material efficiency, these secondary data are site-specific and, even if referred to 1995 and 1996, were considered reliable for a conservative assessment.

Some background data, related to energy generation, use of energy, auxiliary materials and impacts of the waste management (wastewater treatment, hazardous, dregs, sludge wastes, airborne/waterborne emissions, EoL treatments of waste paper) have been derived from the Ecoinvent v.3.1 database (Wernet et al., 2016). The latter is the database generically used for background processes, being the most comprehensive and acknowledged database in Europe. Averaged European data were used for materials and chemicals provided from companies located in EU. In particular, for the supply of electricity, the Finnish medium-voltage electric mix was selected. The production of energy (electricity and heat) *in situ* throughout the combustion of residual biomass (i.e. bark and wood waste, black liquor), was modelled on the combined heat and power production (CHP) from the Ecoinvent database. The production of heat with wood chips by a cogeneration plant and the average Finnish production mix of medium voltage electricity was used for crediting energy supply; for crediting paper recovery, the avoided production of virgin paper was assumed, with a substitution ratio of 1:0.8, due to degradation in the recycling process (Rigamonti et al., 2009). When analyzing the incineration option, the avoided environmental burdens from the production of energy by means of conventional routes were included, crediting European average mix for heat and electricity production on the base of the calorific value for waste paper reported in Ecoinvent (Wernet et al., 2016).

The summarized inventory data managed for the pulp and paper supply chain associated with the selected functional unit (1 ton of produced paper) is shown in Table 1.

Table 1. Life cycle inventory. Values are referred to the functional unit of 1 ton of produced paper.

Inputs	Unit	Value	Outputs	Unit	Value
			FORESTRY PHASE		
Energy[b]			***Emissions to air***[b]		
Fuel	GJ	3.12E-01	CO_2	kg	2.89E+01
			N_2O	kg	7.25E-04
Transport from forest land to pulp & paper mill[a,c]			CO	kg	2.41E-01
Transport by road	t-km	9.00E+01	CH_4	kg	2.07E-03
Transport by railway	t-km	1.59E+02	NO_X	kg	3.64E-01
Transport by sea	t-km	1.10E+02	NMVOC (Non-Methane Volatile Organic Compounds)	kg	5.22E-02
			Products[a]		
			Wood Logs*	kg	1.36E+03
			PULP PRODUCTION PHASE[a]		
Resources			***Emissions to air***		
Water	m^3	3.42E+01	SO_2	kg	2.49E-02
			TRS (Total Reduced Sulphur Compound)	kg	3.79E-02
Energy			Hydrogen sulphide	kg	1.90E-02
Electricity	kWh	2.25E+02	Methyl mercaptans	kg	1.90E-02
Steam	GJ	6.30E+00	NOx	kg	6.79E-01
Fuels	GJ	7.24E-01	Particulates	kg	2.08E-01
Materials			***Emissions to water***		

Wood Logs*	kg	1.36E+03	Solids	kg	2.50E+00
Main Chemicals**:			BOD_7	kg	8.82E-01
Sodium hydroxide	kg	2.43E+01	CODcr	kg	1.03E+01
Sulphuric acid	kg	1.61E+01	Phosphrous	kg	1.37E-02
Oxygen	kg	9.90E+00	Nitrogen	kg	1.77E-01
Sodium chlorate	kg	1.29E+01	AOX (Adsorbable Organic Halogen)	kg	3.92E-02
Hydrogen peroxide	kg	5.89E+00			
Sodium sulfate	kg	3.48E+00	***Products***		
Quicklime (CaO)	kg	4.90E-01	Bleached pulp	kg	4.88E+02
ENERGY PRODUCTION *IN SITU*					
Resources			***Main emissions to air***[d]		
Biomass residuals from pulp & paper mills (used on-site as fuel) *,[a]	kg	1.74E+02	CO, biogenic	kg	1.16E+00
Water[d]	m^3	1.00E-02	CO_2, biogenic	kg	1.12E+03
Materials			Particulates	kg	5.81E-02
Sludge waste from pulp & paper mills (used on-site as fuel) *,[a]	kg	3.19E+01	NO_x	kg	2.67E-02
Dregs waste from pulp & paper mills (used on-site as fuel) *,[a]	kg	4.45E+00	Hydrocarbons	kg	3.60E-02
Chemical, organic[d]	kg	6.80E-02	***Solid waste to treatment***[d]		
Sodium chloride[d]	kg	4.77E-02	Wastewater	kg	9.15E-03
NO_x retained, by selective catalytic reduction[d]	kg	9.30E-01	Municipal solid waste	kg	3.81E-02
			Products[a]		
			Electricity	kWh	8.19E+02
			Heat	GJ	9.61E+00
PAPER PRODUCTION PHASE[a]					
Resources			***Emissions to air***		
Water	m^3	7.50E+00	SO_2	kg	5.12E-04
			NOx	kg	6.93E-04
Energy			Particulates	kg	3.60E-06
Electricity	kWh	3.11E+02			
Steam	GJ	4.58E+00	***Emissions to water***		
Fuels	GJ	6.66E-02	Solids	kg	5.81E-10
			BOD_7	kg	2.21E-09
Materials			CODcr	kg	7.67E-09
Bleached pulp	kg	4.88E+02	Phosphrous	kg	4.65E-12
Main Chemicals:			Nitrogen	kg	8.14E-11
Calcium carbonate (lime)	kg	2.58E+02			
Kaolin	kg	4.84E+01	***Solid wastes from pulp and paper mills to treatment***		
Starches	kg	2.83E+01	Green Liquor Dregs (to landfill)	kg	1.02E+00
Latexes	kg	1.23E+01	Lime Mud (to landfill)	kg	3.79E-01
Optical brightness	kg	5.85E+00	Municipal solid waste (to landfill)	kg	1.28E+00
			Hazardous Wastes (to landfill)	kg	1.20E-01
			Products		
			Virgin paper	kg	1.00E+03
DISTRIBUTION PHASE[a]					
Transport from pulp & paper mill to delivery point[c]			***Emissions from Ecoinvent database***		
Transport by sea	tkm	2.36E+03			
Transport by road	tkm	3.00E+01			
Materials					
Packaging paper	kg	3.69E+01			
Plastic	kg	5.00E-01			
END-OF-LIFE PHASE (Paper recycling)[d]					
Transport			***Main emissions to air***		
Transport by road	tkm	1.42E-01	Ethylbenzene	kg	1.84E-04
			2-Propanol	kg	4.14E-03
Resources			Propylene oxide	kg	3.45E-04
Water	m^3	1.38E+02	Heptane	kg	1.80E-03

Energy			***Main emissions to water***		
Electricity	kWh	1.36E+00	Suspended solids	kg	1.32E-02
Heat	GJ	3.00E-03	Hydrocarbons	kg	9.25E-04
Fuel	GJ	5.39E-05			
			Main solid waste to treatment		
Materials			Hazardous waste	kg	1.77E-03
Main chemicals:			Municipal solid waste	kg	9.39E-03
Sodium hydroxide	kg	1.76E-02	Fly ash and scrubber sludge	kg	1.81E-02
Sodium percarbonate	kg	1.21E-03			
Chlorine	kg	1.89E-03	***Products***		
Sulphuric acid	kg	5.48E-02	100% Recycled paper***	kg	8.00E+02
Aluminium sulfate	kg	7.52E-03			

[a]Data from Stora Enso company.
[b]Data about silvicultural and logging operations modified from Doherty (1995) and Karjalainen & Asikainen (1996).
[c]Emissions to air are included in the Ecoinvent datasets for each transport mode.
[d]Data from Ecoinvent v.3.1 database (Wernet et al., 2016).
*Wood logs and biomass residuals were considered in dry mass.
**Chemicals were reported in 100% mass solids.
***A substitution ratio of 1:0.8 was assumed according to Rigamonti et al. (2009), meaning that 1 unit of waste paper converts into 0.8 unit of recycled material. In the recycling scheme, for simplification of analysis, the pulp was assumed to be 100% waste paper (without virgin fibres inputs).

3.3 Life Cycle Impact Assessment

The environmental assessment of the process was modelled by means of LCA Professional software SimaPro v.8.0.5 (Pre-Consultants, 2014), integrated with Ecoinvent v3.1 database (Wernet et al., 2016). The impact assessment was performed by means of one of the most recent and up-to-date LCA methods, the ReCiPe method (Goedkoop et al., 2009; Vezzoli, 2018). The ReCiPe Midpoint (H) v.1.10 (http://www.lcia-recipe.net/) was chosen, considering that it includes both upstream categories (i.e. referred to depletion of natural resources, such as fossil, metal and water depletion categories) and down-stream categories (i.e. referred to impacts generated on natural matrices, such as terrestrial, marine or freshwater acidification) (Frischknecht et al., 2007). Moreover, the ReCiPe Midpoint (H) method assesses the environmental impacts in different impact categories of interest, such as global warming, abiotic depletion, acidification, eutrophication, human toxicity, among others. The ReCiPe method provides characterization factors to quantify the contribution of the different flows to and from a process to each impact category and normalization factors to allow a comparison across indicators (Goedkoop et al., 2009). Characterization quantifies the extent of the contribution of flows to each impact category (for example, expressing the contribution of CH_4, N_2O and CO_2 to the Global Warming category, by means of CO_2 equivalence factors). Normalization is a procedure used to express the characterized impact indicators in a way that allows comparison to each other. Normalization standardizes the indicators by dividing their characterized values by a selected reference value, translating into an assessment of how much the investigated process contributes to a given category with reference to a value considered acceptable or unavoidable in a given point in space and time (e.g. the average worldwide value in the year 2000). There are numerous methods of selecting a reference value, including, for example, the total emissions or resource use for a given area that may be global, regional or local (Sleeswijk et al., 2008).

In this study, in order to support decision makers by means of a simplified overall assessment across areas of environmental concern (EC, 2016), the following impact categories were analyzed: Global Warming Potential (GWP, in kg CO_2 eq), Ozone Depletion Potential (ODP, in kg CFC-11 eq), Terrestrial Acidification Potential (TAP, in kg 1,4-DB eq), Freshwater Eutrophication Potential (FEP, in kg P eq), Human Toxicity Potential (HTP, in kg 1,4-DB eq), Photochemical Oxidation Formation Potential (POFP, in kg NMVOC), Terrestrial Ecotoxicity Potential (TEP, in kg 1,4-DB eq), Water Depletion Potential (WDP, in m^3), Metal Depletion Potential (MDP, in kg Fe eq), Fossil Depletion Potential (FDP, in kg oil eq).

Furthermore, a sensitivity check and uncertainty analysis were performed to test the robustness of results. Sensitivity check allows to determine what level of accuracy is necessary for a flow to make the analyzed system sufficiently reliable and valid. To this aim, alternative scenarios were proposed, based on progressive reductions or substitution of the most sensitive input flows (both energy and material flows) and the effects of these changes on final results were examined. In addition, a Monte Carlo analysis was carried out to address the uncertainty related to data collection and processing.

4. Results

The performed analysis has two main objectives: (1) to identify the flow(s) and process steps that are "responsible" for the major environmental impacts generated by the production of paper in a life cycle perspective, by modelling physical flows, resources consumption and emissions to the environment, with reference to 1 ton of produced paper; (2) to quantify the environmental benefits deriving from the implementation of material and energy efficiency, throughout an enhanced recovery of resources and the production of heat and electricity *in situ*. The latter issue required a system expansion, in order to consider the avoided costs deriving from the possible recovery of energy and materials (Ekvall and Andrae, 2005): in the present study, environmental savings of goods and energy (i.e. heat, electricity, virgin paper) were subtracted from the accounting of the system's impacts, considering that their production by means of conventional routes is avoided. When the calculated impacts show negative values (see below), they suggest potential savings in the production of virgin materials and energy and hint the amount of environmental benefits that can be achieved.

4.1 Environmental performance in a life cycle perspective

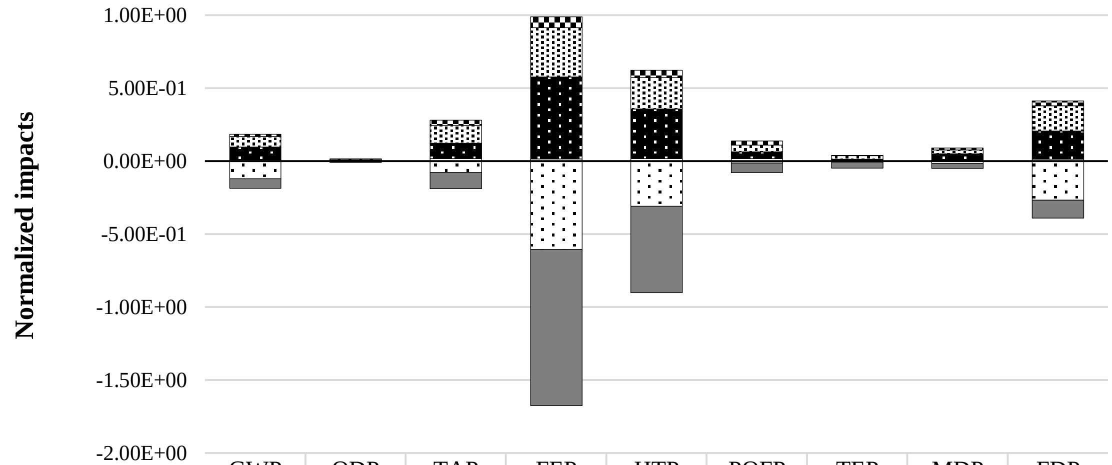
Table 2 summarizes the characterized impacts of the paper production calculated by applying the ReCiPe Midpoint (H) method to the investigated pulp and paper mill complex of Stora Enso Oyj Veitsiluoto Mills, with reference to a functional unit of 1 ton of produced paper. The impact assessment results are distributed among all the investigated phases of the life cycle: the totals calculated for each impact category provide an overall assessment, but the values for each individual phase may help identify the needed improvements. The contributions coming from forestry, pulp and paper production and distribution phases are positive, whereas the phases of energy production *in situ* and of EoL contribute negatively to the total impacts (except for WDP), since the benefits gained from the avoided production of electricity and heat and of virgin paper overcome the environmental loads caused by the cogeneration plant and by the recycling process. Summing the contributions from each phase, the totals are negative values indicating a net environmental advantage in GWP, FEP, HTP and TEP categories, respectively amounting to - 11.1 kg CO_2 eq, - 0.284 kg P eq, - 176 kg 1,4-DB eq and - 0.0723 kg 1,4-DB eq. In the remaining impact categories, the total values are positive, thus indicating that the impacts generated by forestry, pulp and paper production and distribution phases are higher than the benefits gained due to the recovery of energy and recycling of paper. Among the impacting phases, the production processes of pulp and paper jointly affect all the impact categories with the highest shares, ranging from 50% of generated impacts on WDP up to 88% on FEP: in particular, pulp production phase generates the main loads on GWP (46% of the total impacts), on ODP (39%), on FEP (55%), on HTP (46%), on MDP (42%) and on FDP (46%). The remaining impact categories, namely TAP, POFP and TEP, are mainly affected by the paper production phase, with shares of respectively 38% (*versus* 33% from pulp production phase), 28% (*versus* 27% from pulp production phase) and 50% (*versus* 25% from pulp production phase). Impacts generated from forestry phase do not go beyond 10% on POFP. The latter impact category is also affected at a significant level by the distribution and energy production phases (16% of total impacts in both cases), whereas 24% of total impacts on ODP are generated by energy production *in situ*. The recycling process included in EoL phase is responsible for 47% of total impacts on WDP, but at the same time the avoided production of virgin paper leads to benefits in most impact categories, ranging from 49% in TAP up to 87% in WDP itself. In GWP, ODP and FDP the main environmental advantages come from the production of heat by burning the residual biomass in the CHP plant (totaling 50%, 47% and 56% of gained benefits, respectively).

Table 2. Recipe Midpoint (H) characterized impacts calculated for the pulp and paper mill, referred to a functional unit of 1 ton of produced paper, broken down into the different treatment phases.

Impact category	Unit	Forestry phase	Pulp production phase	Paper production phase	Energy production *in situ*	Distribution phase	EoL phase	Total
GWP	kg CO_2 eq	8.80E+01	9.82E+02	8.32E+02	-1.36E+03	1.71E+02	-7.26E+02	-1.11E+01
ODP	kg CFC-11 eq	9.95E-06	1.57E-04	1.12E-04	-9.06E-05	2.21E-05	-9.24E-05	1.18E-04

TAP	kg SO_2 eq	5.77E-01	3.67E+00	4.22E+00	-2.62E+00	1.15E+00	-3.87E+00	3.14E+00
FEP	kg P eq	6.35E-03	2.33E-01	1.40E-01	-2.51E-01	3.14E-02	-4.44E-01	-2.84E-01
HTP	kg 1,4-DB eq	1.15E+01	2.13E+02	1.36E+02	-1.95E+02	3.14E+01	-3.72E+02	-1.76E+02
POFP	kg NMVOC	9.58E-01	2.61E+00	2.69E+00	-7.98E-01	1.56E+00	-3.70E+00	3.32E+00
TEP	kg 1,4-DB eq	1.30E-02	9.65E-02	1.91E-01	-6.22E-02	2.60E-02	-3.36E-01	-7.23E-02
WDP	m^3	1.82E-01	1.42E+01	1.56E+01	-3.69E+00	1.05E+00	1.36E+00	2.87E+01
MDP	kg Fe eq	3.11E+00	3.32E+01	1.80E+01	-1.16E+01	1.06E+01	-2.51E+01	2.82E+01
FDP	kg oil eq	1.96E+01	3.00E+02	2.70E+02	-4.16E+02	5.07E+01	-1.92E+02	3.15E+01

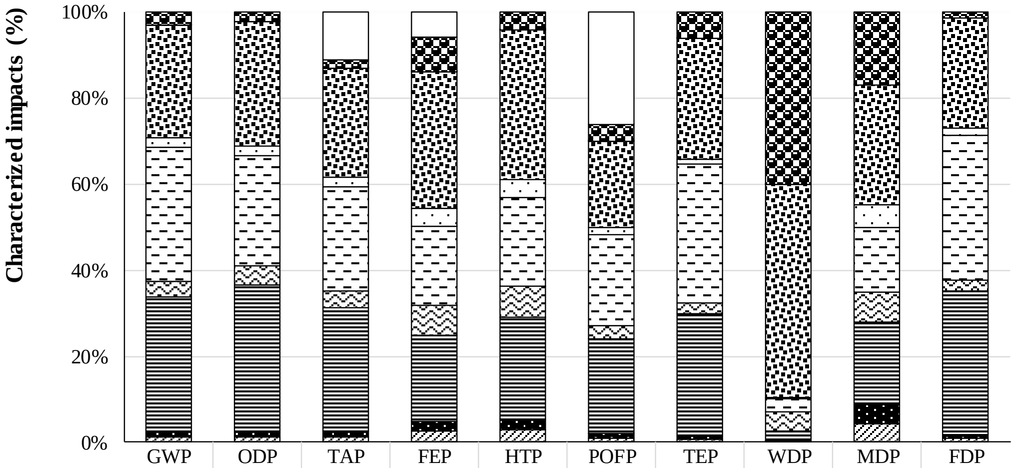
If normalized values of impacts are taken into account (Figure 2), according to Europe ReCiPe Midpoint (H) method normalization factors, a comparison across impact categories becomes possible (water depletion category is not detectable at all, due to the normalization factor equal to zero, and it is not shown in the Figure). Considering the environmental loads, the most highly impacted category results to be FEP, followed by HTP, with impacts generated mostly by pulp and paper production phases. Nevertheless, the benefits deriving from energy production *in situ* and EoL phases in these impact categories are such as to produce net benefits and make these categories the least impacted (with total normalized impacts amounting to - 0.68 and - 0.28, respectively). Conversely, the highest values of normalized impacts are recorded for TAP and POFP (0.091 and 0.058, respectively). In these cases, the benefits are minor than the loads.



	GWP	ODP	TAP	FEP	HTP	POFP	TEP	MDP	FDP
EoL phase	-6.47E-02	-4.20E-03	-1.13E-01	-1.07E+00	-5.92E-01	-6.50E-02	-4.07E-02	-3.52E-02	-1.23E-01
Distribution phase	1.52E-02	1.00E-03	3.36E-02	7.56E-02	5.00E-02	2.74E-02	3.15E-03	1.48E-02	3.26E-02
Energy production in situ	-1.21E-01	-4.11E-03	-7.63E-02	-6.04E-01	-3.10E-01	-1.40E-02	-7.52E-03	-1.63E-02	-2.68E-01
Paper production phase	7.43E-02	5.08E-03	1.23E-01	3.37E-01	2.16E-01	4.74E-02	2.31E-02	2.53E-02	1.73E-01
Pulp production phase	8.76E-02	7.13E-03	1.07E-01	5.62E-01	3.39E-01	4.59E-02	1.17E-02	4.65E-02	1.93E-01
Forestry phase	7.85E-03	4.52E-04	1.68E-02	1.53E-02	1.82E-02	1.69E-02	1.58E-03	4.36E-03	1.26E-02

Figure 2. Recipe Midpoint (H) normalized impacts calculated for the pulp and paper mill, referred to a functional unit of 1 ton of produced paper, broken down into the different treatment phases.

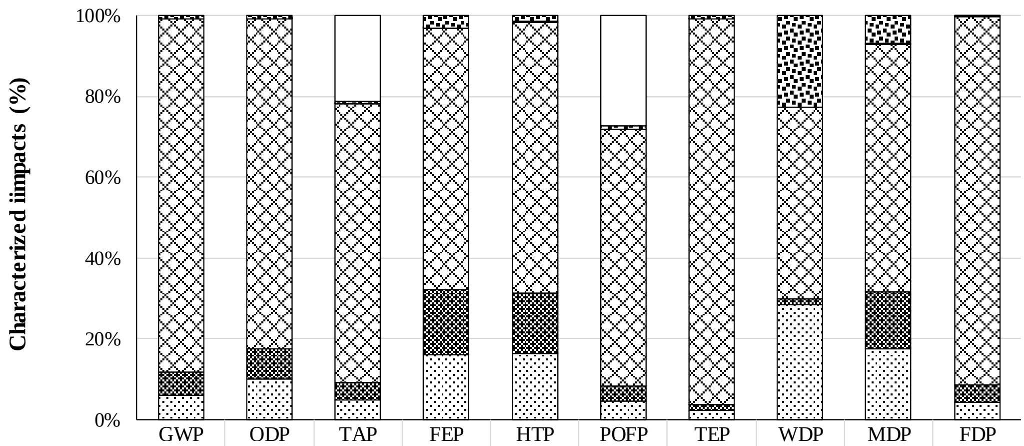
The phases of pulp and paper production are undoubtedly highly impacting on all the investigated impact categories. Therefore, it is worth deepening the insight into the pulp and paper production phases throughout a breakdown of impacts into the different steps of production processes. Figures 3 and 4 respectively show the percentage contributions to the total impacts deriving from the different steps of pulp and paper production phases. Concerning the pulp production phase (Figure 3), all the impact categories are mostly affected by digesting, chemical recovery and bleaching steps, that together are responsible for around 90% of impacts generated on GWP, ODP, TEP and FDP and around 80% of impacts generated on TAP and HTP, generally due to the amount of energy required (in particular heat). In deeper detail, the digesting step generates 34% of the total impacts on ODP and 31% of the total impacts on GWP, respectively releasing 5.38E-5 kg CFC-11 eq and 309 kg CO_2 eq. A similar impact on GWP is attributable to the chemical recovery step, that also affects FDP with about 100 kg oil eq (corresponding to 34% of total impacts) and TEP with 9.65E-2 kg 1,4-DB eq (32% of the total impacts), whereas the impact categories FEP, HTP, WDP, MDP are mainly affected by the bleaching step, in the amounts of 0.074 kg P eq, 73.7 kg 1,4-DB eq, 35.4 m^3 and 9.29 kg Fe eq. The shares of the impacts on TAP and POFP are almost equally distributed among the three abovementioned processing steps. Local emissions (namely, emissions from the foreground system) concern only TAP, FEP and POFP at a limited extent (always less than 26%), while solid waste and wastewater treatments entail an higher contribution only to the impacts on WDP (corresponding to 40%).



	GWP	ODP	TAP	FEP	HTP	POFP	TEP	WDP	MDP	FDP
Local emissions	ND	ND	4.05E-01	1.37E-02	ND	6.81E-01	ND	ND	ND	ND
Wastewater treatment	3.00E+01	3.45E-06	7.29E-02	1.88E-02	8.71E+00	1.00E-01	5.73E-03	2.86E+01	5.62E+00	3.62E+00
Solid waste treatment, landfill	8.38E-01	3.07E-08	9.12E-04	1.41E-04	3.80E-01	1.03E-03	2.03E-05	1.88E-03	8.23E-03	4.87E-02
Bleaching	2.56E+02	4.54E-05	9.36E-01	7.37E-02	7.37E+01	5.26E-01	2.73E-02	3.54E+01	9.29E+00	7.75E+01
Screening & Refining I	2.02E+01	3.38E-06	7.76E-02	9.74E-03	9.25E+00	4.48E-02	1.12E-03	1.72E-01	1.69E+00	4.73E+00
Chemical recovery	3.08E+02	4.04E-05	8.91E-01	4.30E-02	4.40E+01	5.52E-01	3.10E-02	2.16E+00	4.98E+00	1.01E+02
Washing	3.41E+01	6.84E-06	1.38E-01	1.59E-02	1.53E+01	7.92E-02	2.52E-03	3.14E+00	2.34E+00	8.11E+00
Digesting	3.09E+02	5.38E-05	1.06E+00	4.68E-02	5.02E+01	5.73E-01	2.74E-02	1.67E+00	6.42E+00	9.98E+01
Chipping	1.14E+01	1.88E-06	4.56E-02	5.73E-03	5.85E+00	2.62E-02	6.93E-04	9.76E-02	1.44E+00	2.68E+00
Debarking	1.16E+01	1.92E-06	4.64E-02	5.83E-03	5.94E+00	2.67E-02	7.04E-04	9.95E-02	1.44E+00	2.73E+00

Figure 3. Breakdown of Recipe Midpoint (H) characterized impacts for different steps of the pulp production phase, referred to a functional unit of 1 ton of produced paper. (ND=Not detectable).

Concerning the paper production phase, shown in Figure 4, the very last processing step (including pressing, drying and finishing operations) determines the largest share of impacts in all investigated categories, ranging from 48% in WDP up to 91% in FDP. The contribution from forming, screening and refining steps together is slightly higher than 30% only for impacts on FEP, HTP, WDP and FDP. As highlighted also for the pulp production phase, the impacts generated from solid waste and wastewater treatments as well as from local emissions are negligible, except for TAP and POFP (affected by local emissions at 21% and 27%, respectively) and WDP (affected by wastewater treatment at 23%). Generally, the impacts generated in this phase are due not only to the energy requirements, but also to the optical brighteners and fillers used in the finishing step.



	GWP	ODP	TAP	FEP	HTP	POFP	TEP	WDP	MDP	FDP
Local emissions	ND	ND	9.00E-01	4.65E-09	ND	7.35E-01	ND	ND	ND	ND
Wastewater treatment	6.74E+00	7.74E-07	1.64E-02	4.22E-03	1.95E+00	2.25E-02	1.29E-03	6.41E+00	1.26E+00	8.13E-01
Solid waste treatment, landfill	8.38E-01	3.07E-08	9.12E-04	1.41E-04	3.80E-01	1.03E-03	2.03E-05	1.88E-03	8.23E-03	4.87E-02
Pressing/Drying/Finishing	7.27E+02	9.16E-05	2.91E+00	9.03E-02	9.09E+01	1.71E+00	1.82E-01	1.35E+01	1.11E+01	2.45E+02
Forming	4.89E+01	8.28E-06	1.82E-01	2.28E-02	2.03E+01	1.05E-01	2.52E-03	4.13E-01	2.53E+00	1.14E+01
Screening & Refining II	4.89E+01	1.12E-05	2.09E-01	2.23E-02	2.21E+01	1.19E-01	4.44E-03	8.09E+00	3.17E+00	1.18E+01

Figure 4. Breakdown of Recipe Midpoint (H) characterized impacts for different steps of the paper production phase, referred to a functional unit of 1 ton of produced paper. (ND=Not detectable).

4.2 Benefits of energy and material efficiency

The implementation of measures for material and energy efficiency in the assessed system leads to environmental benefits that can be quantified by accounting for the avoided costs of conventional production of heat, electricity and virgin paper. The integrated pulp and paper mill under investigation, whose impacts have been shown above, already includes the production of energy *in situ* by means of combustion of available residual biomass, aimed at partially fulfilling the energy requirements of the plant. Such a measure allows for a considerable saving in the withdrawal of electricity and heat from the national grid, namely 819 kWh of electricity and 9.61 GJ of heat (Table 1), thus accomplishing a noteworthy improvement in the environmental performance of the production process. In order to appreciate such improvement, a comparison between the production process of 1 ton of paper, with and without the option of energy production *in situ*, is shown in Figure 5. This comparison was performed excluding the distribution and EoL phases, since the focus was pointed on the performance of the integrated mill, in a perspective from *cradle-to-gate*. Reduced impacts were observed in all the investigated impact categories, in particular more than 70% of impacts on GWP and FDP were cut down, whereas a less important reduction (around 30%) was obtained for MDP and POFP.

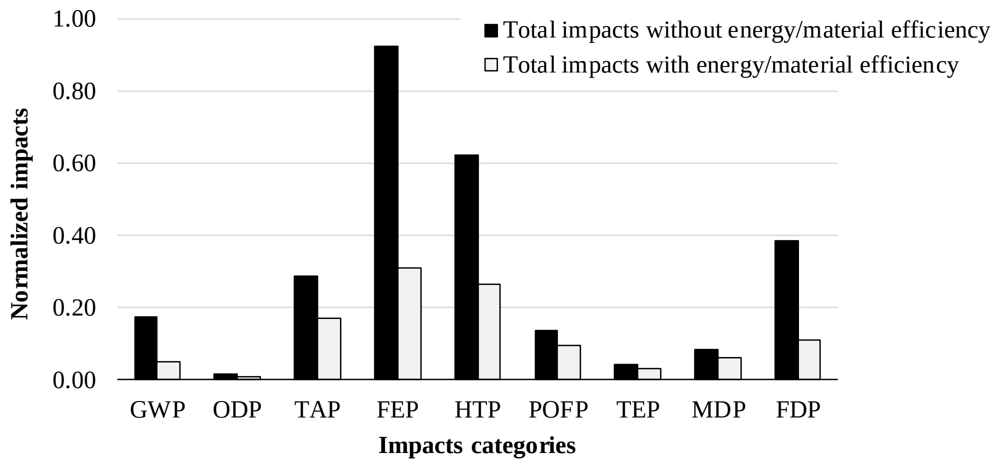


Figure 5. Recipe Midpoint (H) normalized impacts with and without energy and material efficiency implementation, referred to a functional unit of 1 ton of produced paper (distribution and EoL phases are not included).

The environmental benefits deriving from the implementation of measures for energy and material efficiency, such as those adopted in the Stora Enso Oyj Veitsiluoto Mills, can be also validated by comparing the paper production process under investigation with similar processes elsewhere. Pulp and paper industry can usually rely on different pulping processes: chemical (such as Kraft pulping), mechanical (or thermo-mechanical) or a combination of both (Avşar and Demirer, 2008). In Table 3 and Figure 6, respectively, the characterized and normalized impacts of 1 ton of paper produced within the investigated system (Column A, without including distribution and EoL phases) are set over against the impacts generated from other averaged European processes (Hischier, 2007; Wernet et al., 2016): the first compared case (B) was based on the same technology (chemical pulping), the second one (C) on an alternative technology (such as the mechanical pulping) and the third case (D) on the production of 100% recycled paper (i.e., no virgin fibres are included in the recycling process).

Table 3. Recipe Midpoint (H) characterized impacts referred to a functional unit of 1 ton of paper, according to different production technologies, i.e. chemical pulping, mechanical pulping, recycled paper (distribution and EoL phases are not included).

Impact category	Unit	(A) Paper from chemical pulp (Stora Enso company, Finland)	(B) Paper from chemical pulp (Ecoinvent – CEPI* European mix)	(C) Paper from mechanical pulp (Ecoinvent – CEPI European mix)	(D) 100% recycled paper (Ecoinvent, European mix)
GWP	kg CO_2 eq	5.44E+02	9.31E+02	1.51E+03	1.87E+01
ODP	kg CFC-11 eq	1.88E-04	1.18E-04	1.99E-04	1.97E-06
TAP	kg SO_2 eq	5.85E+00	4.99E+00	7.64E+00	1.26E-01
FEP	kg P eq	1.29E-01	5.65E-01	9.86E-01	7.59E-03
HTP	kg 1,4-DB eq	1.66E+02	5.13E+02	7.25E+02	3.79E+01
POFP	kg NMVOC	5.46E+00	4.77E+00	4.57E+00	1.24E-01
TEP	kg 1,4-DB eq	2.38E-01	4.28E-01	2.10E-01	6.46E-03
WDP	m^3	2.63E+01	3.33E+01	3.09E+01	2.80E+01
MDP	kg Fe eq	4.28E+01	4.20E+01	6.14E+01	8.46E+00
FDP	kg oil eq	1.73E+02	2.46E+02	4.42E+02	4.88E+00

*CEPI= Confederation of European Paper Industries

The paper produced by mechanical pulping resulted to be the most energy intensive and impactful on all the investigated impact categories, in line with previous studies (Das et al., 2004; Bajpai, 2016). As far as the chemical pulping is concerned, the assessed production process showed an overall better performance than the average process from Ecoinvent, thus confirming the effectiveness of the option of producing energy *in situ* by burning the residual biomass. In particular, if referring to the characterized impacts, only 544 kg CO_2 eq were released from the investigated process *versus* 930 kg CO_2 eq from the paper produced through chemical pulping and over 1500 kg CO_2 eq from mechanical pulping. Similar trends were observed also for FEP, HTP and FDP, whereas no evident differences among the compared processes were noted for ODP. The impacts from Stora Enso mills were slightly higher than the impacts from one of the average production processes in the case of TAP, TEP and MDP and the highest in the case of POFP. The impacts generated from the production of recycled paper are much more favorable than those generated from the other processes from virgin sources, being one or two orders of magnitude lower (for instance, only 18.7 kg CO_2 eq are released).

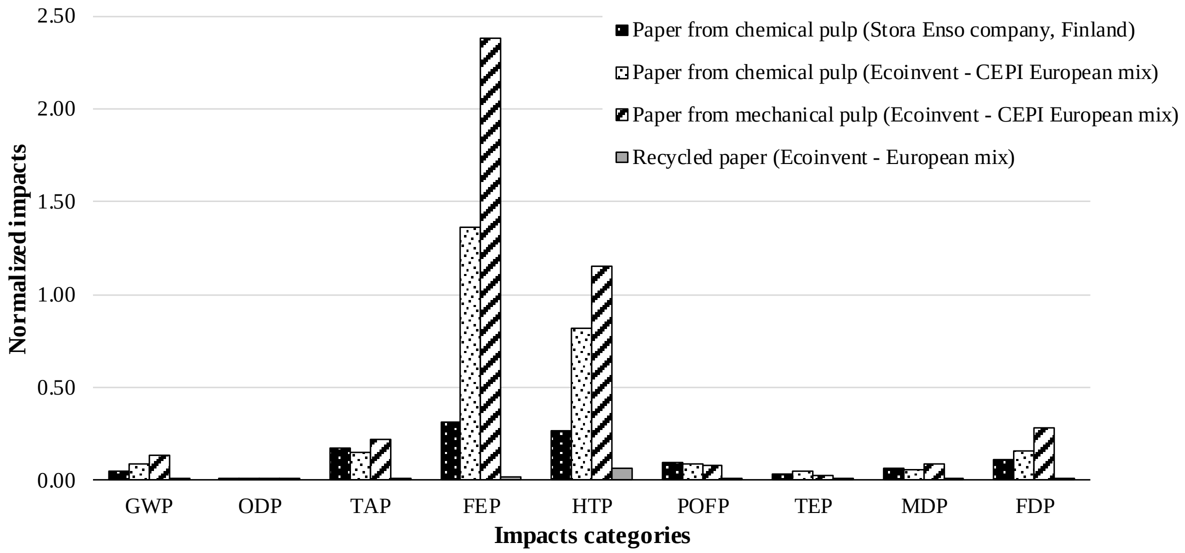


Figure 6. Recipe Midpoint (H) normalized impacts referred to a functional unit of 1 ton of produced paper, according to different production technologies, i.e. chemical pulping, mechanical pulping, recycled paper (distribution and EoL phases are not included).

Finally, due to the significant contributions to the decrease of impacts deriving from the EoL phase (see Figure 2), the options of incinerating or landfilling waste paper rather than recycling were also assessed (dataset from Hischier, 2007), in order to ascertain the relevance of material efficiency throughout an improved recovery of material resources. The comparison between the systems including recycling, incineration and landfilling options is shown in Figure 7: according to previous literature (Schmidt et al., 2007; Merrild et al., 2008), the impacts generated by the system when including the recycling option as EoL are definitely lower than if incineration and landfilling are accounted for. The avoided production of virgin paper leads to net savings (negative values of impact) in GWP, FEP, HTP and TEP, as highlighted before (see Figure 2). In the case of incineration, only FEP resulted to be benefited by the generation of heat and electricity from waste paper. Landfilling is the worst option in all included environmental impact categories.

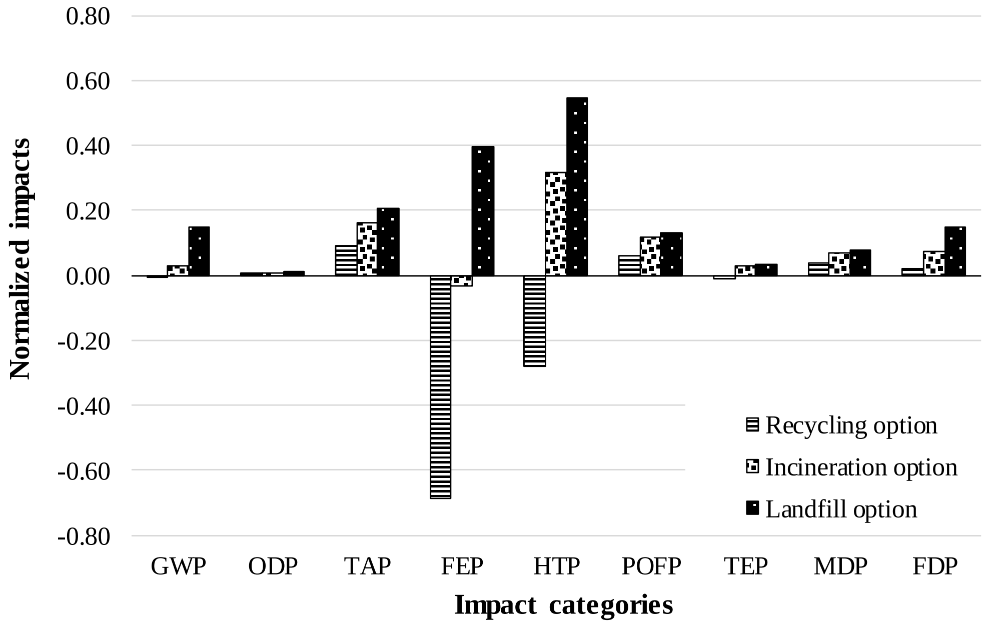


Figure 7. Recipe Midpoint (H) normalized impacts referred to a functional unit of 1 ton of produced paper, according to different EoL options.

4.3 Sensitivity analysis

In order to check the robustness of LCA results and their sensitivity to changes in the input flows included in the study, a sensitivity analysis was performed by assuming a reduction or substitution of the inputs correlated with the highest environmental loads (i.e. electricity, heat and chemicals). The interaction between the variation of these inputs and the achieved LCA results was thus pinpointed.

Firstly, the sensitivity to energy consumption (both electricity and heat) was assessed. Three different possible situations were analysed and compared to the original scenario analysed in this study (scenario S0): S1 – reduced energy consumption by 3%; S2 – reduced energy consumption by 5% and S3 – reduced energy consumption by 10%. Reductions of energy requirements up to 10% were assumed to be possible by limiting the inefficiencies of the process, especially in the pulping and papermaking phases (Piekarski et al., 2017).

The normalized results achieved considering the variations of energy consumption follow in Figure 8. Reducing the energy input flows promoted small improvements in all the nine investigated impact categories, with significant reductions in GWP and FDP. In the case of GWP category, the reduction of impact ranged from 80% in S1 up to 93% in S3. In the case of FDP category, in particular, S3 showed a net environmental benefit (negative value of the impact).

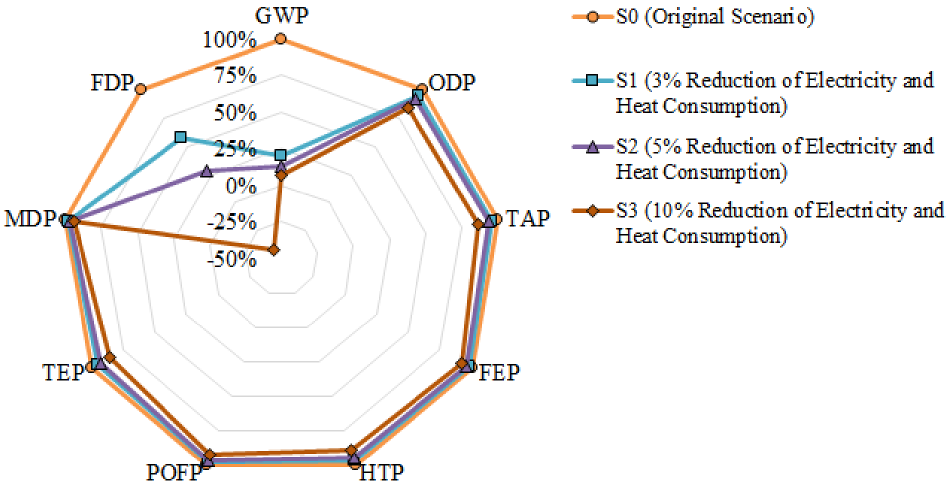


Figure 8. Sensitivity analysis for changes to energy input flows
(WDP is not shown, due to the normalization factor equal to zero).

A strong sensitivity of FDP and GWP categories to changes in electricity and heat consumption, coupled to minor changes in the other categories was expected, although important. Therefore, a second step of the sensitivity analysis consisted in a careful sensitivity check related to the variability of major chemical inputs in the pulping phase. Indeed, as described in section 4.1, the latter resulted to be the most impacting in all the considered impact categories, especially for the digesting and bleaching steps, due to the use of chemicals. Therefore, the sensitivity analyses were performed considering alternative possible situations in comparison with the original pulping process analysed in this study (Pulping_0): Pulping_1 – in the digesting step, soda ash was assumed to substitute sodium hydroxide (or caustic soda) at a ratio 1:1, according to Shivhare et al. (2013); simultaneously, a partial replacement of sodium chlorate (-50%) by oxygen was supposed in the bleaching step, in accordance with Jawjit et al. (2007); Pulping_2 – a substitution of sulphuric acid, used in the digesting step, with biodegradable enzymes for extracting the lignin was assumed according to Fu et al. (2005). In particular, in the latter case, the enzyme manufacturing process was modelled based on existing data for current industrial production reported in Agostinho et al. (2015).

As shown in Figure 9, the substitution/reduction of chemicals use in Pulping_1 generated a non-negligible decrease of impacts on the investigated categories, ranging from 1% of reduced impacts in TEP to 12% in ODP. Nevertheless, the Pulping_2 option produced more environmental loads than the original pulping phase in all explored impact categories, except for TAP, HTP and MDP.

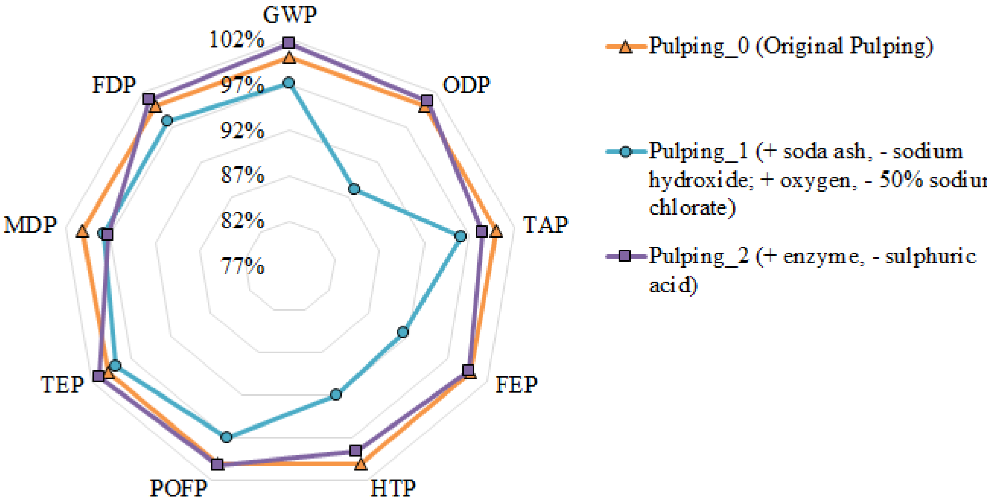


Figure 9. Sensitivity analysis for changes to chemical input flows (WDP is not shown, due to the normalization factor equal to zero).

4.4 Uncertainty analysis

The results of an LCA study can be affected by several uncertainty sources (initial assumptions, system boundaries, data quality, methodological choices, etc) (Björklund, 2002). The accuracy of foreground data collection and background database update are key factors in determining the uncertainty level and affect the final values of impact categories. Moreover, it is important to know to which extent the results of an LCA are affected by uncertainty, because this could be helpful for decision makers in judging the significance of the differences in product comparisons and options for product improvements (Cellura et al., 2011). To this aim, a Monte Carlo simulation within SimaPro 8.0.5 software was performed, in order to propagate the uncertainty linked to key foreground input/output and background processes/emissions along the pulp and paper supply chain (e.g., direct emissions data, activity data, or emission factors). Monte Carlo is a well-known method that substitutes point estimates with random numbers obtained from probability density functions and then builds models of possible results (Huijbregts, 1998). It recalculates thousands of times, each time applying a different set of random values before it is complete. In this study, the Monte Carlo simulation approach was followed after assigning the proper distributions (triangular or log-normal) to all the input parameters, and the impact results were obtained in form of ranges of values instead of single values. In the case of foreground data, distributions were mostly assumed to be triangular, with a range of minimum and maximum values (± 5% for primary data or ±10% for secondary data), whereas a log-normal distribution was given in the Ecoinvent database for the background inputs. The function was implemented in the SimaPro software, considering a sufficiently large number of trials (1,000). The obtained distribution functions gave the results in terms of expected values and lower and upper bounds of the 95% confidence interval for each of the 10 midpoint impact categories considered.

Table 4 shows the obtained results for the impacts generated in the production of 1 ton of paper over the entire life cycle. For each midpoint impact category the following statistical values are reported: mean, median, standard deviation (SD), coefficient of variation (CV, defined as the ratio between the SD and the mean), standard error of the mean (SEM, defined as the standard deviation of the sampling distribution of the mean). It is possible to observe that the variations of the values are moderately low (coefficient of variation in the range of 5% – 45%) for almost all the midpoint indicators (GWP, ODP, TAP, FEP, POFP, MDP, FDP), thus confirming the reliability of data. The impact categories most affected by uncertainty with much larger values of coefficient of variation are ecotoxicity related categories (HTP and TEP) and WDP, due to uncertainty of both Ecoinvent background data and characterization/normalization factors of the selected method (Benini et al., 2014). This calls for additional studies about the assessment method for these categories, to decrease the global uncertainty related to their impact calculation.

Table 4. Results of Monte Carlo uncertainty analysis related to 1 ton of produced paper (overall lifecycle).

Impact category	Unit	Mean	Median	SD	C_v	SEM
GWP	kg CO_2 eq	2.13E+03	2.12E+03	2.16E+02	10.1%	3.20E-03
ODP	kg CFC-11 eq	4.00E-04	3.81E-04	1.07E-04	26.7%	8.44E-03
TAP	kg SO_2 eq	1.12E+01	1.11E+01	7.93E-01	7.1%	2.24E-03
FEP	kg P eq	4.30E-01	3.84E-01	1.93E-01	44.8%	1.42E-02
HTP	kg 1,4-DB eq	2.02E+03	3.52E+03	3.26E+04	1620%	5.11E-01
POFP	kg NMVOC	9.49E+00	9.46E+00	5.22E-01	5.5%	1.74E-03
TEP	kg 1,4-DB eq	4.15E-01	4.19E-01	9.74E-01	235%	7.42E-02
WDP	m^3	5.52E+01	2.60E+02	1.58E+03	2860%	9.04E-01
MDP	kg Fe eq	7.74E+01	7.58E+01	1.20E+01	15.5%	4.90E-03
FDP	kg oil eq	6.54E+02	6.47E+02	8.07E+01	12.3%	3.90E-03

SD = Standard deviation; C_v = Coefficient of variation; SEM = Standard error of mean.

5. Discussion

When assessing a production process, the sources of impacts and criticalities have to be carefully identified in order to define potentials for an improved environmental performance. The LCA of the investigated system pinpointed the expenses in environmental terms related to the production process and to its energy and material requirements, but at the same time ascertained the benefits that can be achieved throughout an efficient use of resources. The unveiling of the LCA results in a disaggregated prospect allowed to breakdown the contribution of the production process in its different phases and to identify the most impacting steps during the manufacturing process. Regarding the generated impacts, this study confirmed earlier cradle-to-gate (Gonzalez-García et al., 2009), cradle-to-costumer (Dias and Arroja, 2012), and cradle-to-grave LCA studies (Lopes et al., 2003; Dias et a., 2007; Vieira et al., 2010), emphasizing that most of environmental impacts of paper derive from the industrial production stage. In deeper detail, the digesting, chemical recovery and bleaching steps of the pulp production phase resulted to be responsible for the main environmental loads. Only in few impact categories, most of impacts could not be ascribed to the pulping deriving from the use of optical brighteners and fillers in the ultimate steps of paper production (thus affecting TAP, POFP and TEP) and from the intensive consumption of water in the recycling step of EoL (thus affecting WDP). Generally, the main contributions to environmental loads arised from the electricity and heat requirements and, only at a minor extent, from the use of chemicals such as the sodium hydroxide and the sodium chlorate, in accordance with previous studies (Hong and Li, 2012; EC, 2015; Kong et al., 2016). Conversely, the use of energy generated *in situ* from black liquor and residual biomass to support the requirements of the integrated pulp and paper mills came out to be a key issue in lowering the overall impacts. In addition, the recovery of water and chemicals in the pulping phase (see Figure 1) implies further material savings, that are accounted for in the assessment but not separately quantifiable, due to the lack of disaggregated data. The results calculated for the investigated system showed an overall high-ranking environmental performance in comparison with other averaged European systems for paper production.

For a deeper understanding of the whole paper system, the system boundaries were expanded to include the disposal phase of used paper at its end-of-life. Different options were envisaged through the scenarios of recycling (with material recovery), incineration (with energy recovery) and landfilling and net advantages were gained if recycling was assumed as final disposal of waste paper. Recycling of waste paper into new products was thus confirmed to constitute an environmental benefit in all the impact categories (except for water depletion), requiring less energy and causing fewer emissions than manufacturing the same amount of paper from virgin resources, as already highlighted by Schmidt et al. (2007), Villanueva and Wenzel (2007) and Merrild et al. (2008), among others. Nevertheless, the recycling process also hides some non-negligible constraints. Recycling cannot be intended to completely substitute the production processes based on virgin fibres, as a ‘downcycling’ occurs leading to changes in the inherent properties and affecting the quality of the produced paper (Bala-Gala et al., 2015). Indeed, paper can be successfully recycled no more than 3.6 times on average, according to EPRC (2016). Afterwards, the cellulose fibres are too degraded for use in papermaking and other waste management options have to be considered, using waste paper as energy source in the case of incineration, or as a filler for insulation, or landfilling as the worst option. Incineration allows much lower energy and material savings than recycling and, although it may constitute a viable option in regions that lack recycling facilities, the transport of waste paper to the nearest recycling facility has to be evaluated. Therefore, the huge benefits deriving from the production of recycled paper may somehow be constrained, in that the production of recycled paper is dependent on the production of virgin paper. Recycling is a likely option for the EoL phase in the overall transition to a circular economy but cannot be an alternative production pattern in itself.

Facing resource scarcity, such as wood, energy and chemicals, as well as reducing emission intensities is a chief challenge for the pulp and paper industry and various technological improvements, especially aimed at energy savings, have been developed and, in some cases, deployed at industrial scale. Recently, the biorefinery concept applied to pulp and paper mills has been recommended as an innovative industrial transformation and upgrading towards a more profitable and efficient production. All types of biomass available, including forest residues, waste wood chips, paper mill residues and sludge generated from the pulping and papermaking process, can be used to produce a wide variety of materials, from chemicals to fuels, in addition to paper products (CEPI, 2009; Kong et al., 2016). The increasing recovery of potentially usable resources, in order to provide a feedback mechanism into the process or an even larger scale, falls within the concept of circularity, producing two potential benefits, namely decrease of impacts and recovery of resources.

Nevertheless, emerging and advanced technologies as well as circular approaches need to be evaluated on a case by case basis and quantitative assessments of environmental impacts are required for a comprehensive judgement of sustainability (Fiorentino et al., 2017). The enzymatic bleaching of the pulp is a clear exemplification, as it uses biodegradable enzyme to supplement, or eventually to replace, chemicals for extracting the lignin in the bleaching process and is therefore considered a progress towards cleaner industrial production and environmental sustainability. However, it cannot be excluded that the production of enzymes consumes more energy and raw materials than it saves (Jegannathan and Nielsen, 2013). In the investigated case study, the proposed use of enzyme actually favours some impact categories, even if at a restricted degree, while TAP, HTP and MDP are more damaged than with the conventional pulping process.

For the sake of clarity, it should be also noted that the magnitude of generated environmental loads and of attained benefits is firmly susceptible to a range of factors, such as the uncertainties in inventory data, the definition of system boundaries, the modelled sources of electricity and heat and the quality of the applied technology. Although desirable, a direct comparison of the results achieved in this study with previous LCA literature is hardly possible: even if the functional unit selected in these studies is most often 1 ton of paper, the paper grade can widely vary (newsprint, super-calendered, office, among others), thus implying different production technologies (mechanical or chemical pulping) at stake. The choice of system boundaries, of impact assessment methods and of raw material (e.g. Eucalyptus, Spruce or Pine wood) affect the results that can be obtained. For instance, the production of paper in Norway (Ghose and Chinga-Carrasco, 2015) is assessed from a *gate to gate* perspective, not including the forestry phase, whereas Silva et al. (2015) used CED, EDIP 1997 and USEtox 2008 to quantify the impacts generated from the production of 1 ton of office paper by chemical pulping in Brazil. Evaluations of paper production in Portugal (Lopes et al., 2003; Dias et al., 2007; Vieira et al., 2010) are based on characterization factors from different literature sources and comparisons of results would be meaningless. This calls for an urgent need for further standardization of LCA procedures, so that different production processes can be effectively compared. Therefore, at the state of the art, room is left to potential and non-negligible improvements. The chance to rely on high quality local data for the core processes (pulping and papermaking) and to split electricity and heat requirements, chemicals demand and emission levels over all the production steps provided an added value to this study, overcoming potential methodological limitations. Moreover, the LCA approach also allows a sensitivity and uncertainty check to provide an insight in the robustness of selected impact indicators. The outcome of the fluctuation in input flows, performed by reducing energy supplies and substituting key chemicals, seems to be slightly affected in terms of identification of hotspots and drawbacks as well as in terms of calculated impact indicators, showing a pattern that remains constant and hence corroborating the soundness of the assessment and the conclusions that can be drawn.

6. Conclusions

The intensive energy consumption and the addiction to large amounts of resources, chemicals, fuels and water make the pulp and paper industry a key part of the strategy towards a more sustainable and cleaner industrial production. In this study, the LCA methodology, in a double perspective from *cradle-to-gate* and from *cradle-to-cradle*, was applied to a paper manufacturing process in Finland in order to identify those process steps that entail the highest environmental loads and require improvements. Such a geographical contextualization is due to the fact that the Finnish technologies in this sector are acknowledged to be the most advanced worldwide.

The achieved results demonstrated that activities related to wood pulp manufacturing phase, such as digesting, chemical recovery and bleaching steps, are the main contributors to the environmental impacts across the entire supply chain. In particular, the main burdens are generated on global warming, ozone depletion, freshwater eutrophication, human toxicity, metal depletion and fossil depletion (more than of 40% of total contributions). In the remaining investigated impact categories, namely terrestrial acidification, photochemical oxidant formation and terrestrial ecotoxicity, most of impacts derive from the final steps of paper production and from the intensive consumption of water in the recycling step of end-of-life affecting water depletion. In addition, compared with incineration technology (with energy recovery) and landfilling, recycling of paper demonstrated high environmental benefit because of the avoided production of paper from virgin sources, although the limited possibilities of recycling used paper has to be taken into account. The use of renewable sources of energy (such as residual biomass power) to support the requirements of the pulp and paper mills resulted to be crucial in lowering the overall environmental impact and, therefore, further benefits may derive from an enhancement of material and energy efficiency of the process thanks to circular recycling patterns.

The pulp and paper industry is currently in a transitional situation, where it is no longer only producing pulp and/or paper but also additional products which can increase both the mill profitability and efficiency. The concept of pulp and paper mills as integrated biorefineries that produce low-carbon energy commodities, including biofuels for transport, is increasingly gaining attention and in the longer term, the sector can also contribute to sustainable energy supply, for example, by feeding excess heat and electricity into the grid.

Looking at future perspective, the sector should continue to focus on improving energy and material efficiency, deploying Best Available Technologies (BATs) and optimizing the materials recycling rate. On the basis of achieved results, private- and public-sector stakeholders and policy-makers are expected to make efforts to develop future processes, technologies and suitable assessing procedures to provide valuable information and encourage the integration of pulp and paper industry in a circular perspective.

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