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Environmental Assessment of Cellulose Pulp Production from Wood Waste using Organosolv Treatment

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Abstract* – In the upcoming years, the paper industry is expected to rely on containerboard for packaging. Since 2020, containerboard has been made from 80 % recycled fibres. Creating a new supply chain for fibres could help mitigate possible shortages. In this context, wood waste emerges as a valuable resource with the potential to serve as a plentiful and cost-effective reservoir for generating new materials, such as cellulose fibres. This study presents an assessment of the environmental impact associated with the organosolv extraction of cellulose pulp from wood waste based on a previous in vitro study conducted by the authors. The increasing demand for sustainable materials has prompted exploration into alternative methods for cellulose pulp production, with a focus on minimizing environmental footprint. Organosolv extraction, a promising technique, involves the use of organic solvents and acid catalysts to break down lignocellulosic biomass, resulting in high-quality cellulose pulp production. To evaluate the environmental implications of this process, a comprehensive life cycle assessment (LCA) approach was adopted. The LCA framework considers all stages of the organosolv extraction process, from wood waste collection to cellulose pulp manufacture, using a gate-to-gate approach. The functional unit for assessment is set as one metric ton of cellulose pulp. The dataset utilized for the LCA comprises primary data obtained from in vitro experiments that have been scaling-up, complemented by secondary data sourced from literature and the *Ecoinvent* life cycle inventory database. Furthermore, sensitivity analysis was conducted to assess the influence of key parameters on the environmental performance of the organosolv extraction process, with a focus on variations in energy consumption to identify potential areas for optimization and improvement. In summary, this study emphasizes the environmental aspects of utilizing organosolv extraction for cellulose pulp production from wood waste. Further research is warranted to explore energy consumption during the organosolv process for obtaining more precise data and optimizing the process. This could be achieved through pilot-scale experiments or utilizing process simulation software.

***Keywords* – Delignification, LCA (life cycle assessment); pulping; recycling.**

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1. INTRODUCTION

Wood is a valuable and sustainable material that can be used in many different fields: construction, biorefineries, pulp and paper industry, furniture, packaging, and many others [1]. Some of these products become wood waste at the end of their life cycle. Wood waste can be recycled or utilized to generate energy, such as electricity and heat. Currently, the primary recycling method for wood is the production of particleboard, as seen in Italy, where almost 100 % of wood waste is transformed into this product [2].

There are numerous pathways for recycling wood, including its use as animal bedding, in pellet production, and more. Among these options, pulping stands out as one of the possible alternatives [3], [4].

Pulp serves as a crucial raw material in the paper industry, functioning as the primary component for both paper and containerboard. Moving forward, the paper industry is expected to experience significant growth driven primarily by the increasing demand for containerboard, particularly in packaging applications [5]. Exploring alternative sources for pulp production could prove to be a promising option. According to data provided by the Confederation of European Paper Industries (Cepi), the trade balance for market pulp in Cepi's area shows a negative value of approximately 2.16 million metric tons [6]. Cepi represents nearly 80 % of total European pulp production through the national associations of eighteen member states. In 2022, pulp production in Cepi's area is predominantly carried out through the sulfate (or kraft) process, accounting for 73.6 % of the total production. The kraft method entails treating wood with a heated blend of water, sodium hydroxide, and sodium sulfide. This process could produce malodorous air emissions and associated environmental and health risk [7]. Another promising pulping technique that could avoid this problem is organosolv [8], [9]. It involves a process that uses organic solvents to dissolve hemicellulose and lignin, resulting in pulp enriched in cellulose.

The organosolv method, pioneered by Theodor Kleinert [10] in 1968 as an eco-friendly alternative to kraft and sulphite pulping, offers several advantages. Notably, it produces high-quality lignin from wood waste, transforming a typically discarded stream into a valuable resource. Additionally, organosolv solvents can be efficiently recovered through distillation, reducing water pollution and consumption. Although small-scale organosolv pulp mills primarily focus on non-wood fiber sources, such as straw and bagasse, softwood fractionation has also been successfully achieved at pilot scales. However, commercial-scale operations face challenges due to process costs and fluctuating product prices. To address this, efforts are needed to optimize energy balance and reduce overall costs by effectively separating fractionated components [11]. Ethanol-based organosolv processes, utilized in pulping various wood types, offer environmental benefits over traditional methods by eliminating sulfur emissions and reducing particulate matter emissions [12].

Given these considerations, the objective of this study is to assess the feasibility of organosolv pulping for producing unbleached pulp from wood waste, while juxtaposing its environmental impact with that of unbleached pulp derived from kraft pulping. Employing the life cycle assessment methodology, we undertake a comparative analysis of these two processes. The organosolv process is meticulously modelled using a combination of laboratory data, literature sources, and the *Ecoinvent* database. This paper represents a foundational step in a broader exploration aimed at elucidating the critical parameters of the organosolv process and refining them for future investigations.

2. METHODOLOGY

The research adheres to the standards outlined in ISO 14040 and ISO 14044 [13], [14]. Utilising *SimaPro* 9.0 software developed by Pre-Consultants and *Ecoinvent* v.3.5 database, the study constructed the Life Cycle Assessment (LCA) model and conducted the impact assessment computations.

2.1. Process Overview

A LCA of ethanol organosolv process using wood waste is modelled based on laboratory and literature data. This process has been developed in the CIRIAF laboratory (Perugia, Italy) following Pazzaglia *et al.* [15]. Here it is briefly explained.

Wood waste has been collected from Umbria's waste collection centres by *Gesenu Spa*, the local waste management company. The wood waste is then transported to a mechanical treatment plant located in Ponte Rio, (Perugia, Italy) where it is shredded and sent to a wood waste treatment plant in Northern Italy to be processed into medium-density fibreboard. This constitutes the current pathway. This study aims to assess an alternative process starting from shredding wood waste. The proposed model utilises ethanol organosolv pulping to produce unbleached pulp from wood waste in Ponte Rio, based on mass balance data from laboratory studies and energy balance information from the literature.

2.2. Goal and Scope Definition

The aim of this study is to carry out a gate-to-gate LCA of organosolv process on a large scale, using 1 metric ton of unbleached pulp as the functional unit for comparison. This evaluation is compared with existing literature on the production of 1 metric ton of unbleached pulp via the sulphate (kraft) pulping method, which is the conventional process for this product [6], [16]. Sulphate pulp production data were taken from the *Ecoinvent* database under the category "sulfate pulp production, from softwood, unbleached". To compare the two types of pulp, the same pulp factory has been modeled, representing a pulp mill with an annual consumption of 400.000 metric tons of lignocellulosic matter and a total lifetime of 50 years. The pulp factory data comes from the following ecoinvent category: "Pulp factory {RER}pulp factory construction cut-off, S". The unbleached pulp has a 10 % moisture content. This research is part of the CellWood project, which aims to explore alternative solutions for recycling wood waste. The target audience includes the scientific community, Fondazione Perugia (the project promoter), and *Gesenu Spa*, the collaborating company. The modelling framework involves an attributional LCA using mass allocation to evaluate the footprint of unbleached pulp.

2.3. Wood Waste Characterization and Component Mass Balance

The wood waste primarily consists of wood from municipal waste (87.3 %), followed by wooden packaging (6.8 %), construction and demolition wood (>0.1 %), and wood generated by the bulky waste treatment (5.5 %). The wood waste treated in this mechanical treatment plant contains an average metal content of 0.71 % by weight relative to wood waste after shredding operation. Table 1 shows the dry solids mass balance for the raw material, cellulose pulp, and in the filtrate post organosolv. These data originate from Pazzaglia *et al.* [15], refer to this work for detailed information on the process and instrument used in laboratory.

TABLE 1. CHEMICAL COMPOSITION OF DRY SOLIDS FOR WOOD WASTE, ORGANOSOLV PULP, AND FILTRATE MEASURED IN METRIC TON RELATED TO A 1 METRIC TON ORGANOSOLV PULP (FUNCTIONAL UNIT)

Cellulose	**Lignin**	**Hemicellulose**	**Extractives**	**Ashes**	**Others**	**Total**
Wood waste						
0.74	0.55	0.30	0.14	0.05	0.14	1.92
Organosolv pulp						
0.68	0.17	0.05	Negligible	Negligible	Negligible	0.90 (1 metric ton with 10 % moisture content)
Filtrate post organosolv						
0.06*	0.37	0.25*	0.14	0.05	0.14	1.01

*Note: these values represent the sum of derived sugars from cellulose (glucose) and hemicellulose (mainly xylose and arabinose) after organosolv treatment

2.4. System Boundary

The process began with collecting wood waste from designated centres in Umbria. It was then transported to a mechanical treatment plant in Ponte Rio, Perugia, Italy, where it was shredded into smaller pieces. At this stage, the moisture content of the wood waste increases from 8 % to 18 % by adding water to prevent the dispersion of wood dust into the air. Up to this point, data have been provided by the waste management company. Subsequently, the metallic fraction of the waste was separated using an overbelt electromagnetic separator. The metal-free wood waste was chipped and milled to achieve a final dimension of 1 mm. The milled wood waste was then introduced into a reactor where the organosolv process took place, utilizing an ethanol/water mixture (80:20 w/w) as a solvent and sulfuric acid (2 % w/w) as a catalyst, maintained at 160 °C for 15 minutes. The reactor has been dimensioned based on an annual consumption of 400 000 metric tons of wood waste. Considering an operating time of 8000 hours per year [17], and a reactor volume 10 % larger than the reaction mixture volume [18], the reactor's dimension results in approximately 280 cubic meters. The liquid-to-solid ratio has been set on 4.5:1 [19]. The resulting solution mixture underwent filtration and washing using an ethanol/water solution (80:20 w/w) in a ratio 2:1 to the pulp weight to recover ethanol soluble lignin. This was followed by a rinse with pure water with at a ratio of 2:1 to the pulp weight [20] to recover ethanol from the pulp. This process resulted in wet unbleached pulp and a filtrate mainly composed of water, ethanol, sugars, and lignin. The wet pulp has 77.54 % moisture content, measured in laboratory through a HB43-S Halogen Classic Plus (Mettler Toledo, Columbus, OH, USA). It was dried to a moisture content of 10 % [21], resulting in unbleached pulp. To enhance the sustainability of the process, the filtrate underwent distillation to obtain a mixture of ethanol and water, which was then reused in the process. The ethanol recovery is >99 % following Moncada studies [22]. The distillation residue mainly contained water, sugars, and lignin. Upon filtration, the wet lignin (with the same moisture content as the wet pulp) was dried to a moisture content of 10 % [17], yielding a by-product, and a water solution containing primarily sugars derived from hemicellulose that undergoes wastewater treatment. As for the emissions of the process, they are negligible according to the Moncada study [22]. In terms of infrastructure, a pulp factory with dimensions identical to those of the sulfate pulp plant, as per the *Ecoinvent* database, has been hypothesized. Fig. 1 shows the process and its system boundary.

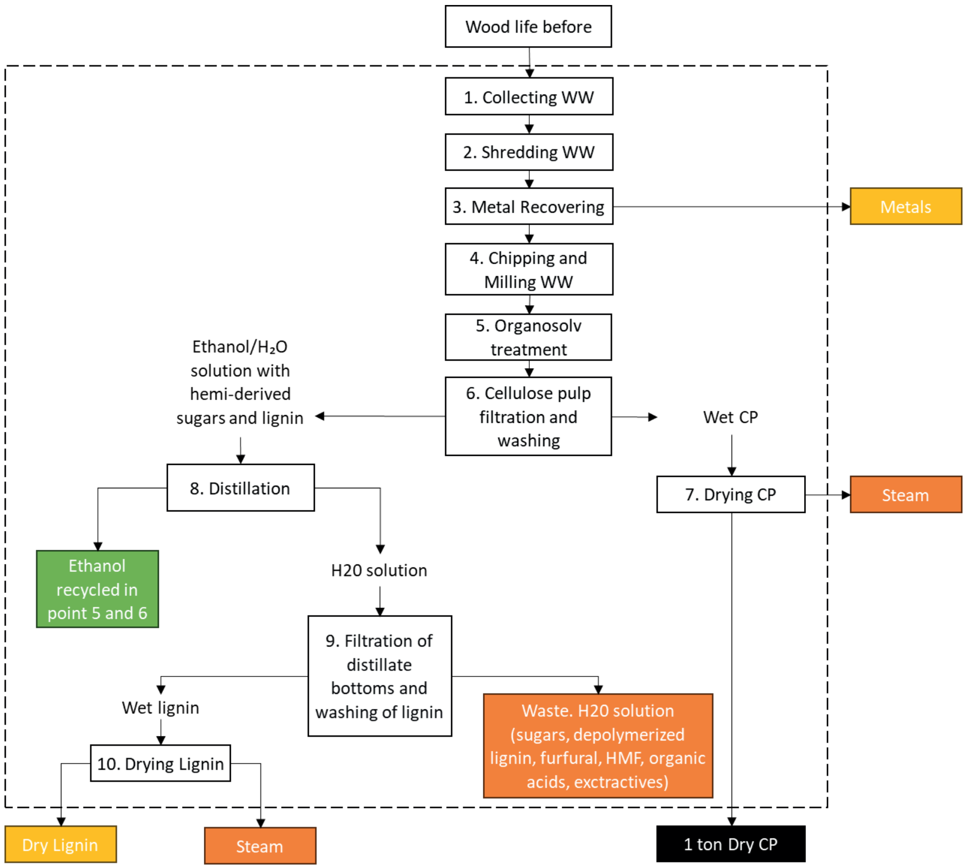

Fig. 1. System boundaries: black square is the functional unit, yellow squares are by-products, orange squares depict emissions and waste, green square represent recovered ethanol, white squares the process steps, no square items are intermediate product before other processes.

2.5. *Environmental Impact Assessment*

To assess the environmental impact of the process the *ReCiPe* 2016 Endpoint (H) V1.08/World (2010) H/A, GWP100, and cumulative energy demand method (CED) have been used. The first method presents outcomes across three distinct areas of focus: “human health”, “ecosystems”, and “resources”. GWP100 has been chosen instead of GWP100 (including CO_2) because *SimaPro* does not account for the CO_2 uptake from the wood wastes used as raw materials in the manufacturing process of organosolv pulp. Therefore, GWP100 could be more appropriate for this specific case.

2.6. *Life Cycle Inventory (LCI)*

In LCI, data are standardized and denoted with respect to the functional unit of 1 metric ton of unbleached pulp and mass allocation. Specifically, the allocation was performed based on the production of 1 metric ton of pulp, 0.42 metric tons of lignin, and recovery of 0.02 metric tons of metals. Pulp accounts for 69.82 % of the environmental impact. Most of the data is derived from laboratory analysis [15], with the remainder sourced from literature. The implementation of inventory data utilized the *Ecoinvent* 3.9.1 database to incorporate information regarding transport, chemicals, and energy. Table 2 presents the LCI, depicting inputs and outputs categorized by process.

TABLE 2. LIFE CYCLE INVENTORY

	Category	Material/component	Quantity	Unit	Data source
1. Collecting wood waste					
Input	Transport, freight, lorry 16-32 metric ton, EURO5 {RER} market for transport, freight, lorry 16-32 metric ton, EURO5 Cut-off, S	Freight lorry	25.1	tkm	Primary data
2. Shredding wood waste					
Input	Diesel {Europe without Switzerland} market for diesel Cut-off, S	Diesel	4.82	kg	Primary data
	Water, decarbonised {FR} market for water, decarbonised Cut-off, S	Water	0.17	metric ton	Primary data
3. Metal recovering					
Input	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity	5.87	MJ	[23]
4. Chipping and milling wood waste					
Input	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity	107.45	MJ	[24]
	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity	723.19	MJ	[25]
5. Organosolv treatment					
Input	Sulfuric acid {RER} market for sulfuric acid Cut-off, S	Sulfuric acid	0.10	metric ton	Primary data
	Water, decarbonised {FR} market for water, decarbonised Cut-off, S	Water	0.46	metric ton	Primary data
	Heat, central or small-scale, biomethane {Europe without Switzerland} market for heat, central or small-scale, biomethane Cut-off, S	Heat reaction	2821.36	MJ	[18]

	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity stirring	0.05	MJ	[18]
6. Cellulose pulp filtration and washing					
Input	Ethanol, without water, in 99.7 % solution state, from ethylene {RER} market for ethanol, without water, in 99.7 % solution state, from ethylene | Cut-off, S	Ethanol	0.03	metric ton	[18], [22]
	Water, decarbonised {FR} market for water, decarbonised Cut-off, S	Water	3.13	metric ton	Primary data
	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity filtration	67.78	MJ	[18]
7. Drying cellulose pulp					
Input	Heat, central or small-scale, biomethane {Europe without Switzerland} market for heat, central or small-scale, biomethane Cut-off, S	Heat	6772.67	MJ	[18]
Output	Emission to air	Water	2.10	metric ton	Primary data
8. Distillation					
Input	Heat, central or small-scale, biomethane {Europe without Switzerland} market for heat, central or small-scale, biomethane Cut-off, S	Heat	14912.27	metric ton	[18]
9. Filtration of distillate bottoms and washing of lignin					
Input	Electricity, high voltage {IT} market for electricity, high voltage Cut-off, S	Electricity filtration	9.39	MJ	[18]
	Water, decarbonised {FR} market for water, decarbonised Cut-off, S	Water	0.52	metric ton	[20]
Output	Wastewater average {Europe without Switzerland} market for wastewater, average Cut-off, S	Wastewater	2.05	m^3	Primary data
10. Drying lignin					
Input	Heat, central or small-scale, biomethane {Europe without	Heat	2815.04	MJ	[18]

	Switzerland} market for heat, central or small-scale, biomethane Cut-off, S				
Output	Emission to air	Water	0.87	Metric ton	Primary data
Pulp factory					
Input	Pulp factory {RER} pulp factory construction Cut-off, S	Pulp factory	39.08 E-13	p	*Ecoinvent*

3. RESULTS AND DISCUSSION

Life Cycle Impact Assessment (LCIA) represents an essential phase in LCA, wherein the amassed data on input and output flows are rigorously evaluated to gauge the environmental effects across the entirety of a product or process lifecycle.

3.1. ReCiPe 2016 Endpoint Level

The *ReCiPe* impact assessment method version 1.08 (H) was utilised to identify environmental hotspots within the studied process, particularly in comparison to sulfate unbleached pulp production. This method encompasses three aggregated damage assessments: human health, ecosystem, and resources. The endpoint level has been selected due to its ease of understanding for decision-makers [26].

Table 3 shows the result consider the single score of the *ReCiPe* method.

TABLE 3. DAMAGE ASSESSMENT

Damage category	Organosolv unbleached pulp, Pt	Sulfate unbleached pulp, Pt	Organosolv unbleached pulp, %	Sulfate unbleached pulp, %
Total	24.45	17.76	100.00	100.00
Human health	23.30	12.78	95.31	71.97
Ecosystem	0.78	4.76	3.19	26.82
Resources	0.37	0.21	1.51	1.2

Table 3 illustrates the greater global impact of the organosolv process compared to the sulfate process. This is primarily due to the human health damage assessment, which is nearly twice as high in organosolv as in sulfate. However, the organosolv process achieves a favorable outcome in terms of ecosystem impact, being four times lower. The resources endpoint category shows similar results for both processes. Through the *ReCiPe* method, it is also possible to observe the primary contributors to the environmental impact of 1 metric ton of organosolv pulp. Heat impact is particularly noteworthy, accounting for 62 %, followed by sulfuric acid (15.8 %), electricity (12.3 %), and ethanol (4.96 %). The human health category shows a similar contribution from the same contributors. Considering the processes involved, distillation has the highest impact (33.9 %), followed by organosolv treatment (22.2 %), drying cellulose pulp (15.4 %), and chipping-milling operations (11.2 %).

3.2. GWP100

Addressing climate change stands as one of the paramount challenges humanity confronts in the 21st century, as underscored by the United Nations in 2015 [27]. The Global Warming

Potential (GWP) serves as a crucial metric in assessing the impact of climate change, as indicated by the IPCC in 2021 [28]. In this study, GWP100 has been employed, utilizing a time frame of 100 years. Table 4 presents the results for GWP100 pertaining to fossil, biogenic, land transformation, and total emissions from the processes under investigation.

TABLE 4. GLOBAL WARMING POTENTIAL

Impact category	Unit	Organosolv unbleached pulp	Sulfate unbleached pulp
GWP100 – fossil	kg CO_2–eq	482.39	293.31
GWP100 – biogenic	kg CO_2–eq	67.46	0.51
GWP100 – land transformation	kg CO_2–eq	0.51	1.46
GWP100 – total	kg CO_2–eq	550.36	295.28

The total organosolv process emissions are nearly double than sulfate pulping. It is due to using mainly black liquor as internal fuel in sulfate pulping. Black liquor is a by-product of kraft process and it is commonly used to produce heat through a recovery boiler. Moreover, from *Ecoinvent* documentation it is evident that the emission from this recovered energy is not included in the emissions calculations because no specific emissions profiles were available. As a result, these emissions were accounted for solely within the key emission factors provided by the industry, without being directly measured by a third party. For this reason, there is the possibility of an underestimation of the sulfate pulping emissions.

3.3. Cumulative Energy Demand

The CED method permits discovering the total amount of energy needed to develop a process. In this case, it is useful to compare organosolv pulping of waste with sulfate pulping methods. Table 4 shows the results of the application of the CED method.

TABLE 4. CUMULATIVE ENERGY DEMAND COMPARISON BETWEEN ORGANOSOLV AND SULFATE PULPING

Impact category	Unit	Organosolv unbleached pulp	Sulfate unbleached pulp
Total	GJ	9.94	38.37
Non-renewable, fossil	GJ	7.47	3.64
Non-renewable, nuclear	GJ	0.97	0.68
Non-renewable, biomass	GJ	0.2 E-3	1.15 E-3
Renewable, biomass	GJ	0.75	33.77
Renewable, wind, solar, geothe	GJ	0.27	0.09
Renewable, water	GJ	0.46	0.17

The most evident difference between the two processes is the larger amount of energy required by the sulfate process. This is primarily attributed to tree harvesting, which accounts for nearly all of the renewable biomass energy consumed. This is typical due to the utilization of wood residue for heat production during the harvesting process. Unlike in organosolv pulping, wood harvesting is not a factor, as wood waste serves as the primary raw material. However, organosolv pulping consumes a greater amount of energy from fossil sources. Therefore, a sensitivity analysis regarding variations in energy consumption and potential changes in energy sources has been conducted.

3.4. Sensitivity Analysis

The purpose of this subsection is twofold: firstly, to ascertain the necessary reduction in energy consumption by organosolv pulping to equalize its environmental impact with sulfate pulping in terms of GWP100 – fossil, and secondly, to evaluate the disparities resulting from switching the electricity source from predominantly fossil-based to renewable.

A first sensitivity analysis explores scenarios involving reductions in heat and electricity consumption by 10 %, 20 %, and 30 %. By analysing this data, a trend line has been determined, revealing that a decrease of approximately 46.42 % in heat and electricity consumption in the organosolv process is necessary to achieve an equivalent GWP100-fossil value to that of the sulfate process. The selected reductions of 10 %, 20 %, and 30 % in heat and electricity consumption aim to model realistic, incremental improvements in energy efficiency, reflecting potential advancements in process optimization and energy recovery technologies. For instance, this preliminary study does not include heat recovery options, which could be implemented in future work using appropriate plant design software. Energy recovery is a crucial aspect, as indicated by many other studies [29], [30]. These reductions are linked to factors such as the efficiency of heat exchangers, chemical recovery systems, and the integration of renewable energy sources. The values provide a framework for assessing the environmental impact of these potential improvements, particularly in terms of GWP100-fossil.

Another sensitivity analysis has been conducted, following the same concept of reducing heat and electricity demand. In this analysis, the use of renewable energy for electricity is considered, while biomethane is utilized for heating. Fig. 3 shows the results.

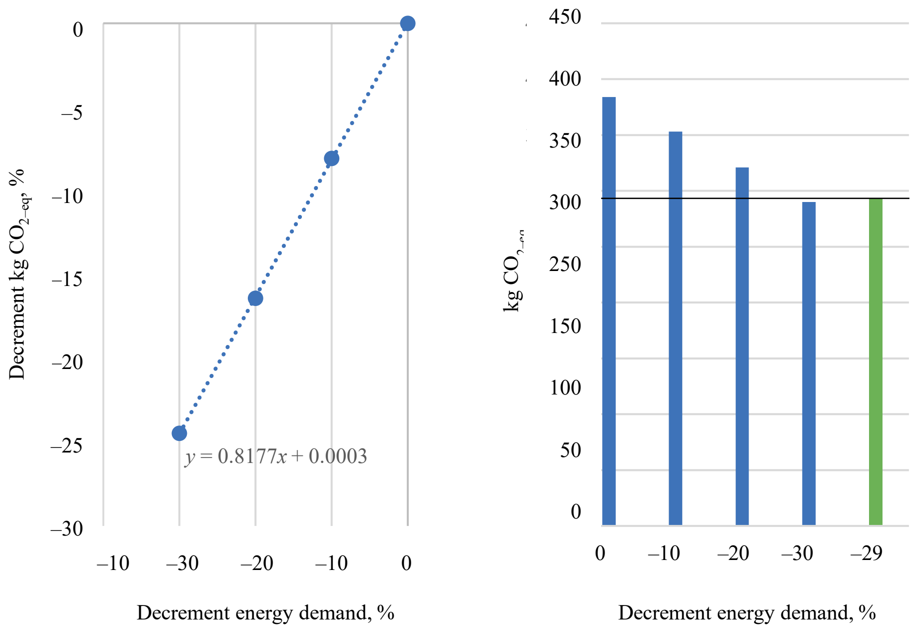

Fig. 3. On the left: depicts the relationship between the decrease in energy demand and the percentage reduction in kg CO_2-eq. On the right: identifies the required reduction in energy demand to match the kg CO_2-eq emissions of the sulfate process. The black line indicates the sulfate process kg CO_2-eq.

From this analysis, it is noteworthy that a reduction of 28.61 % in energy demand allows reaching the same level of CO_2–eq – fossil emissions as the sulfate process. An energy saving of this magnitude also reduces the single score in the Recipe method: a reduction of 30 % in energy demand results in a single score of 17 points for organosolv compared to 17.76 points for sulfate. Therefore, it becomes more significant to prioritize future studies on the energy source and energy-saving processes.

5. CONCLUSION

The present work displays a gate-to-gate LCA comparing the production of 1 metric ton of unbleached pulp via organosolv and sulfate process. The LCI has been built using laboratory and literature data. Recipe method, at endpoint level, GWP100 and CED method have been used to assess the processes' environmental impact.

All methods demonstrate better results for the sulfate process compared with organosolv, primarily due to the internal energy production in the sulfate treatment using black liquor. Through sensitivity analysis on GWP100-fossil, energy-saving measures and alternative energy sources were tested to determine a combination of factors affecting the GWP100-fossil value of the sulfate process. The results indicate that using biomethane for heating, renewable electricity, and achieving a global energy saving of 28.61 % allow for reaching the same value of kg CO_{2-eq}.

This study aims to assess the use of wood waste in a biorefinery concept through a cleaner process as organosolv. This study is a starting point to evaluate the environmental performance of this process comparing to the most usual techniques (as sulfate or sulfite pulping). Furthermore, in future research, we will concentrate on utilizing distillate bottoms, recovered lignin, and cellulose pulp. Another area for future study will involve comparing the utilization of wood waste in the organosolv process with its current application in particleboard production.

However, the primary focus for further research would be to investigate the heat and energy sources and their recovery during the process, starting from design software and potentially scaling up to a pilot-scale plant if the results prove encouraging.

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