UNIVERSIDADE FEDERAL DE VIÇOSA

Sustainable solutions for pulp mill sludge management

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Doctor Scientiae

VIÇOSA - MINAS GERAIS
2024

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Thesis submitted to the Forest Science Graduate Program of the Universidade Federal de Viçosa in partial fulfillment of the requirements for the degree of *Doctor Scientiae*.

Adviser: Claudio Mudadu Silva

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VIÇOSA - MINAS GERAIS
2024

Ficha catalográfica elaborada pela Biblioteca Central da Universidade Federal de Viçosa - Campus Viçosa

T
R696s
2024

Rodrigues, Bruna Virgínia Cunha, 1996-
Sustainable solutions for pulp mill sludge management / Bruna Virgínia Cunha Rodrigues. – Viçosa, MG, 2024.
1 tese eletrônica (129 f.): il. (algumas color.).

Texto em inglês.
Inclui apêndices.
Orientador: Cláudio Mudadu Silva.
Tese (doutorado) - Universidade Federal de Viçosa, Departamento de Engenharia Florestal, 2024.
Inclui bibliografia.
DOI: https://doi.org/10.47328/ufvbbt.2024.819
Modo de acesso: World Wide Web.

1. Industria de celulose. 2. Resíduos industriais. 3. Lodo residual. 4. Biocombustíveis. I. Silva, Cláudio Mudadu, 1960-. II. Universidade Federal de Viçosa. Departamento de Engenharia Florestal. Programa de Pós-Graduação em Ciência Florestal. III. Título.

GFDC adapt. CDD 22. ed. 634.989

Bibliotecário(a) responsável: Bruna Silva CRB-6/2552

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APPROVED: November 19, 2024.

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ACKNOWLEDGMENTS

To my mother, Lêda Ferreira, and my siblings Braz Junior and Ellem Cristiana for their unwavering support and for always giving me the strength and encouragement to keep moving forward.
To all my friends, who have become like family, always supporting me, regardless of the physical distance.
To Professor Claudio Mudadu, for his invaluable guidance, opportunities, patience, and lessons that extend far beyond the professional sphere.
To Professor Cássia Carneiro, for showing me that achieving both professional and personal excellence is always possible, and for all the valuable lessons that have greatly contributed to my growth.
To Caio Torres, for his friendship and invaluable professional and personal lessons he has shared with me.
To the colleagues of the Laboratório de Celulose e Papel (LCP) e Laboratório de Paineis e Energia da Madeira (LAPEM) for their daily friendship and for contributing to the successful execution of this work with excellence.
To the colleagues of MNEXT – AVANS Hogeschool, especially Jappe de Best, Qian Zhou, Alexander Compeer, Valentin Contin, and Moctar Coulibaly for warmly welcoming me and for all the essential support throughout my PhD studies.
To Dinos Clades for companionship, patience, love, and kindness.
To the Universidade Federal de Viçosa (UFV), particularly the Forestry Science Program, for their invaluable support and for providing all the necessary resources throughout the development of my research.
I am also thankful for the scholarships granted by CAPES (Coordination for the Improvement of Higher Education Personnel) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), which made the financing of this project possible.
I am grateful for the people around me and for the moments that bring me happiness and learning.
This study was financed in part by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior – Brasil (CAPES) – Finance Code 001.

“Practice isn’t the thing you do once you’re good. It’s the thing you do that makes you good”

(Malcolm Gladwell)

ABSTRACT

RODRIGUES, Bruna Virgínia Cunha, D.Sc., Universidade Federal de Viçosa, November, 2024. **Sustainable solutions for pulp mill sludge management**. Adviser: Claudio Mudadu Silva. Co-adviser: Angelica de Cassia Oliveira Carneiro.

The pulp and paper industry has a crucial role in the global market, impacting the economy, environment, and society. Chemical and mechanical processes are used for pulp production. The kraft method is the most common chemical process in the world. The mechanical process is commonly used to produce high-yield pulp. The high water consumption in pulp and paper mills results in a high generation of effluent. Due to the high organic and inorganic matter, the effluent is treated in the effluent treatment plant (ETP) before discharge in the river. During the effluent treatment, the sludge is generated. Usually, two types of sludge are generated in the pulp and paper mill: primary sludge (PS), which comes from primary effluent treatment, and secondary sludge (SS), generated during secondary or biological treatment. Because of its characteristics, secondary sludge is difficult to handle and is often mixed with primary sludge to make it easier to manage. The sludge generated in the pulp and paper industry accounts for the largest proportion of solid waste generated at the mill and its main destination is the industrial landfill. Due to major environmental, social, and economic problems, landfills have been restricted. The transition of the world market towards bioeconomy has required sustainable management of all co-products generated in the production chain. The sludge generated in the pulp mill is rich in lignocellulosic materials presenting the potential for energy generation. There is a growing demand for the reduction of fossil fuels and the implementation of new renewable fuel sources. Energy generation using sludge from the pulp industry has emerged as an alternative for managing this biomass, also providing support for the Sustainable Development Goals (SDGs). This study aims to assess the technical and environmental potential of generating energy from PS and SS generated in pulp mills. The work is divided into four chapters. Chapter 1 provides a comprehensive review of current practices for sludge disposal, along with the latest innovations in sludge management found in the literature. The growing restrictions on landfills underscore the urgent need to explore and implement more sustainable methods for sludge treatment. Chapter 2 presents the characterization of PS, SS, and the mixture of PS and SS (MS) generated in a thermo-mechanical pulp mill, assessing their energy potential and the possible effects of their combustion in the boiler. The high moisture content in the sludges created a negative energy balance, requiring a pre-drying step prior to combustion. Chapter 3 assessed the technical feasibility of producing pellets using PS and SS. The effect of moisture content and the blends of PS and SS on the final quality of the pellets was also determined. The pelletizing process standardized the dimensions of sludge, increasing the energy efficiency in the boiler and reducing costs in transportation and storage. Chapter 4 presents the environmental viability through life cycle assessment (LCA) of the energy generation of sludge pellets. This study highlights and compares the environmental effects of burning and producing pellets with sludge disposal in landfills. The results of this chapter showed that energy generation using pellets sludge had a better environmental performance than landfilling. Optimizing energy generation at the mill with a new renewable fuel source contributes to more sustainable production in the pulp and paper industry.

Keywords: bioeconomy; pulp and paper industry; renewable fuel; sludge; waste management

RESUMO

RODRIGUES, Bruna Virgínia Cunha, D.Sc., Universidade Federal de Viçosa, novembro de 2024. **Soluções sustentáveis para a gestão dos lodos das fábricas de celulose**. Orientador: Claudio Mudadu Silva. Coorientadora: Angelica de Cassia Oliveira Carneiro.

A indústria da celulose tem um papel importante no mercado global, com impacto na economia, no ambiente e na sociedade. A polpa celulósica é obtida a partir de processos químicos ou mecânicos. O método kraft é o processo químico mais utilizado no mundo. O processo mecânico é normalmente empregado para produzir pasta de alto rendimento. O elevado consumo de água nas fábricas resultam na elevada produção de efluentes. Devido ao elevado teor de matéria orgânica e inorgânica, o efluente é tratado na estação de tratamento de efluentes (ETE) antes de ser descarregado no rio. Geralmente nas indústrias de celulose e papel são gerados dois tipos de lodos na ETE, o lodo primário (LP), oriundo do tratamento primário de efluente, e o lodo secundário (LS), gerado durante o tratamento secundário. O LP é rico em fibras enquanto o LS é rico em microrganismos, devido suas características, o lodo secundário possui difícil manuseio, sendo frequentemente misturado ao lodo primário para facilitar o seu gerenciamento. Os lodos gerados nas indústrias de celulose e papel corresponde a maior proporção de resíduos sólidos gerados na fábrica e tem como principal destino o aterro industrial. Devido aos grandes problemas ambientais, sociais e econômicos, o aterro tem sofrido cada mais restrições. A transição do mercado mundial em direção a bioeconomia urge a demanda de um gerenciamento sustentável de todos co-produtos gerados na cadeia produtiva. O lodo gerado nas indústrias de celulose e papel é rico em materiais lignocelulosicos apresentando potencial para geração de energia. Há uma crescente demanda na redução de combustíveis fosseis e a implementação de novas fontes de combustíveis renováveis. A geração de energia utilizando o lodo das indústrias de celulose surge como uma alternativa no gerenciamento dessa biomassa, proporcionado também suporte aos Objetivos de Desenvolvimento Sustentável (ODS). O presente trabalho teve como objetivo avaliar o potencial de geração de energia do LP e LS gerado nas indústrias de celulose. O trabalho foi divido em 4 capítulos. O Capítulo 1 oferece uma revisão abrangente das práticas atuais de descarte de lodo, juntamente com as últimas inovações na gestão de lodo encontradas na literatura. As crescentes restrições ao uso de aterros destacam a necessidade urgente de explorar e implementar métodos mais sustentáveis para o tratamento de lodos. O Capítulo 2 apresenta a caracterização do LP, LS e da mistura de LP e LS (MS) gerados em uma planta de polpa termo-mecânica, avaliando seu potencial energético e os possíveis efeitos de sua combustão na caldeira. O alto teor de umidade nos lodos criou um balanço energético negativo, exigindo uma etapa de pré-secagem antes da combustão. O Capítulo 3 avaliou a viabilidade técnica da produção de pellets utilizando LP e LS. Também foi determinado o efeito do teor de umidade e das misturas de LP e LS na qualidade final dos pellets. O processo de peletização padronizou as dimensões do lodo, aumentando a eficiência energética na caldeira e reduzindo os custos de transporte e armazenamento. O Capítulo 4 apresenta a viabilidade ambiental por meio da avaliação do ciclo de vida (ACV) da geração de energia a partir de pellets de lodo. Este estudo destaca e compara os efeitos ambientais da queima e produção de pellets com o descarte de lodo em aterros. Os resultados deste capítulo mostraram que a geração de energia usando pellets de lodo teve um desempenho ambiental melhor do que o aterramento. A otimização da geração de energia na fábrica com uma nova fonte de combustível renovável contribui para uma produção mais sustentável na indústria de celulose e papel.

Palavras-chave: bioeconomia; indústria de celulose e papel; combustível renovável; lodo; gerenciamento de resíduos

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LIST OF ABBREVIATIONS AND ACRONYMS

ADt	Air-Dried ton
AOX	Adsorbable Organic Halogens
AS	Activated Sludge
B	Bark
C2C	Cradle-to-Cradle
CF	Characterization Factors
CFCs	Chlorofluorocarbons
COP	Conference of the Parties
CRD	Completely Randomized Design
CV	Coefficient of Variation
D	Durability
DALY	Disability-Adjusted Life Years
DB	Dichlorobenzene
DTG	Derivative Thermogravimetry
EMAS	Eco-Management and Audit Scheme
ETP	Effluent Treatment Plant
F	Fines
FC	Fixed Carbon
FT	Fluid Temperature
FU	Functional Unit
GRI	Global Report Initiative
GHGs	Greenhouse Gases
HCFCs	Hydrochlorofluorocarbons
HHV	Higher Heating Value
HT	Hemispherical Temperature
HTC	Hydrothermal Carbonization
HTL	Hydrothermal Liquefaction
IDEMAT	Industrial Design and Engineering Materials
IDT	Initial Deformation Temperature
ISO	International Organization for Standardization
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory

LCIA	Life Cycle Impact Assessment
LHV	Lower Heating Value
MBBR	Moving Bed Biofilm Reactors
MS	Mixed Sludge
NHV	Net Heating Value
PS	Primary Sludge
REF	Reference
SB	System Boundaries
SC1	Scenario 1
SC2	Scenario 2
SCR	Selective Catalytic Reduction
SD	Standard Deviation
SDGs	Sustainable Development Goals
SS	Secondary Sludge
ST	Spherical Temperature
TG	Thermogravimetry
TMP	Thermo-Mechanical Pulp
UASB	Up-flow Anaerobic reactor and Sludge Blanket
UN	United Nations
VFA	Volatile Fatty Acids
VM	Volatile matter
WB	Wet Basis

SUMMARY

GENERAL INTRODUCTION

The pulp and paper industry has an important role in the bioeconomy of many countries and has a significant impact on society and the environment (COSTA et al., 2022). Brazil is the second-largest producer of pulp in the world. In 2023 the country produced 24.3 million tons of pulp and exported US$ 8.2 billion (IBÁ, 2024). Cellulose can be obtained through a mechanical or chemical process. Approximately 98% of the pulp produced in Brazil comes from the chemical process. The chemical process has a low yield and results in a highly resistant pulp, while the mechanical process presents a high yield and a pulp with lower resistance properties (COLODETTE; GOMES, 2015; COSTA et al., 2023).

The thermomechanical pulping process consists mainly of water impregnation of logs or wood chips before a shredding step resulting in a high-yield pulp (CALDEIRA et al., 2021).

Among the chemical processes, the kraft process corresponds to the main pulp production process in the world (SEWSYNKER-SUKAI; DAVID; KANA, 2020). The kraft pulp production occurs in high pressure and temperature conditions (160 °C - 180 °C). The wood chips are mixed with white liquor, which is composed of sodium hydroxide (NaOH) and sodium sulfide (Na_2S). This process results in 90-95% of the lignin degradation. The residual lignin and some modified carbohydrates have chromophore properties, resulting in a brown coloration of the pulp. The bleaching stage aims to remove these chromophores groups, increasing and stabilizing the brightness of the pulp (COLODETTE; GOMES, 2015; RAMOS et al., 2022).

The wash in the bleaching stages consumes a high amount of water. It is estimated that 80% of the effluent generated in a bleached pulp mill comes from this stage (RAMOS et al., 2022; SANTOS et al., 2020). Pulp mill effluent has organic and inorganic material extracted from the pulp during the washing steps (SANTOS et al., 2020). Prior to discharging the effluent into water bodies, a series of treatments are required, since the pollutants present in the effluent cause adverse impacts on the aquatic environment (KUMAR; KUMAR; BHARDWAJ, 2017).

The pulp and paper mill effluent treatment plant (ETP) usually consists of preliminary treatment, a primary clarifier, and a biological treatment normally composed of an activated sludge process (CABRERA, 2017). The preliminary treatment aims to remove coarse solids and the adjustment of pH, and temperature (PÖYKIÖ; WATKINS; DAHL, 2018). The primary treatment is a physical process to reduce the suspended solid presented in the effluent. The sludge generated in this treatment is rich in fibers (TORRES et al., 2023). Secondary treatment can be accomplished by aerobic or anaerobic biological processes. Usually, pulp mills use aerobic processes which consist of aerated lagoons, activated sludge (AS), and moving bed biofilm reactors (MBBR) (VIDAL et al., 2021). The activated sludge process is the main method used in the mills. The secondary sludge generated in this process consists mainly of microorganisms (CABRERA, 2017; TORRES et al., 2023).

The sludge generated at the ETP corresponds to the largest amount of solid waste from the pulp mill, for each air-dried ton (ADt) of pulp produced are generated approximately 40 kg of primary sludge and 15 kg of secondary sludge (LOPES et al., 2018; TORRES et al., 2023). The high content of microbial matter in the secondary sludge makes it difficult to manage. To facilitate handling and dewatering operations, secondary sludge is often mixed with primary sludge (FANG et al., 2017). The main disposal practice of the sludge generated in the pulp mill is landfilling, incineration, or composting (GRIMM et al., 2019; LOPES et al., 2018).

Pulp production involves the use of many resources, such as wood, chemicals, water, and energy (AMÂNDIO et al., 2022; RAMOS et al., 2022). To ensure the sustainability of the process, the industry has used some tools to develop environmental sustainability. International Organization for Standardization (ISO) 14001 (Environmental management systems), EMAS (The EU Eco-Management and Audit Scheme), Ecolabel, ISO 9001 (quality management system), ISO 45001 (Occupational health and safety management systems), ISO 50001 (energy management system), the UN Global Compact, circular economy framework are some of the tools used by the pulp industry reach high sustainability standard (SOUSA et al., 2023).

The Global Report Initiative (GRI) framework is a standard commonly used by pulp mills to prepare periodic reports, ensuring greater transparency in the impact of their processes on society, the environment, and the economy (GRI, 2022; SOUSA et al., 2023). The pulp sector faces the challenge of transitioning to a bioeconomy, innovating the process and waste management (PÄTÄRI et al., 2016). This trend leads to waste becoming a co-product of the process. The traditional linear economic approach based on "take, make, and dispose” has been replaced by the circular economic model, which proposes reusing and recycling all products and co-products (AMÂNDIO et al., 2022).

With increasingly stringent environmental laws, landfills have become a less attractive disposal option. The European Union aims to recycle and reuse 70% of all waste generated by 2030. Incineration is also a common practice for waste disposal for energy recovery. The heterogeneity, low density, and high moisture content of the waste result in a combustion process with low efficiency increasing the emissions of pollutants gases (AMÂNDIO et al., 2022; RODRIGUES et al., 2021).

A more sustainable sludge management is still challenging for the pulp and paper industry. Understanding the properties of sludge is an important step towards its correct application. The sludge composition varies according to many factors, such as the pulping process, raw material used, and the effluent treatment plant (GRIMM et al., 2019). Before proposing a new solution to sludge management, a careful study of its properties must be carried out.

The sludge generated in the pulp and paper mills is a potential raw material for pellet production (RODRIGUES et al., 2021). Pellets are densified biofuel with a cylindrical shape that can be produced from most lignocellulosic material (ANUKAM et al., 2021; SANTANA et al., 2020). Compared to unpelletized biomass, the pellets have advantages such as a higher bulk density, a lower moisture content, a regular geometry, and a standardized size.

These properties enable easier handling, more cost-effective transportation and storage, and enhance energy efficiency within the boiler system (BARBOSA et al., 2022; MOSTAFA et al., 2019). Pellets are widely used for energy generation and heat in many countries since they are considered a sustainable alternative to fossil fuels (PRADHAN; MAHAJANI; ARORA, 2018). The primary consumers of pellets globally are European countries, accounting for 68% of consumption, followed by Asia, which represents 24% (BIOENERGY EUROPE, 2024).

The pelletizing process is the densification of the biomass using a press. The ground biomass is pressed through a flat die by compacter rollers, formed into endless strands, and then cut by knives to the desired particle length. The quality of the pellets depends on the properties of the biomass, such as moisture content, composition, and particle size. The temperature and pressure in the pelletizing die also influence the characteristics of the final product (RODRIGUES, 2021).

Energy production using pulp and paper mill sludge seems a more sustainable practice when compared to landfills. However, scientific proof needs to be obtained to verify this hypothesis. The life cycle assessment (LCA) allows the evaluation of a process or product, based on emissions and the extraction of resources converted into a specific environmental impact score. The LCA of biomass to energy conversions has been widely applied to assess the environmental impact of the production steps (SREEKUMAR et al., 2020).

This study aims to conduct a complete evaluation of the management options of the primary and secondary sludge generated in the effluent treatment plant of the pulp mill. The work was divided into 4 chapters.

Chapter 1 provides a review of current practices for the disposal of pulp and paper mill sludge, and recent innovations in sludge management documented in the literature over recent years. These advancements are analyzed in terms of their potential to address the challenges associated with traditional methods while offering new opportunities for sustainable waste management. Additionally, Chapter 1 underscores the importance of tools for environmental evaluation, particularly the role of Life Cycle Assessment (LCA). This method serves as a cornerstone for assessing the environmental feasibility of new products and processes, offering a structured approach to measure and minimize environmental impact. The integration of LCA into sludge management practices ensures a holistic understanding of sustainability, from waste generation to final disposal or repurposing.

Chapter 2 evaluated the physical and chemical properties of three different sludges and bark generated in a thermomechanical pulp mill. This characterization allowed a critical analysis of different management routes for this biomass. The energy conversion potential and the impact of their combustion in the biomass boiler also were assessed.

Chapter 3 discusses the technical feasibility of producing pellets using primary and secondary sludge from a bleached kraft pulp mill. The work evaluated the moisture effect and the proportion of primary and secondary sludge in the pellet blends. The chemical and physical properties of the sludges were obtained to analyze the potential of energy production for each blend.

Chapter 4 presents and discusses the environmental feasibility through a life cycle assessment (LCA) of pellets production using primary and secondary sludge generated in a bleached kraft pulp mill. The LCA evaluated the environmental impact of the integration of pellets production and combustion within the kraft pulp mill and compared it with landfilling, the conventional practice of sludge disposal.

OBJECTIVES

General objective

This study aims to characterize and propose new management routes for the primary and secondary sludge generated in the effluent treatment plant of pulp mills, evaluating the technical and environmental aspects of energy generation.

Specific objectives

i. Evaluate the potential of energy generation of primary and secondary sludge and bark generated in a thermomechanical pulp industry, determining the elemental composition, heating value, thermogravimetric degradation (TG/DTG), and the composition and behavior of the ashes.

ii. Evaluate the technical feasibility of producing pellets using kraft pulp mill sludge. Determine the effect of moisture content and the proportion between primary and secondary sludge in the mechanical, chemical, and physical properties of the pellets.

iii. Evaluate the environmental feasibility of producing pellets for energy generation using primary and secondary sludge from the pulp mill through a life cycle assessment (LCA). Comparing the environmental impact of the proposed process and landfilling.

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CHAPTER 1

PULP MILL SLUDGE MANAGEMENT AND ENVIRONMENTAL IMPACT EVALUATION: A COMPREHENSIVE REVIEW

PULP MILL SLUDGE MANAGEMENT AND ENVIRONMENTAL IMPACT EVALUATION: A COMPREHENSIVE REVIEW

Abstract

The pulp and paper industry has a high energy demand. Although most of this energy comes from renewable sources, a small percentage still relies on fossil fuels. The sludge generated in the effluent treatment plants (ETP) is rich in fibers and presents a high potential for energy generation. This biomass, still considered waste, is typically sent to landfills. New methods for sludge treatment have been explored to promote more sustainable practices in pulp mills. This study aims to investigate innovations in sludge management and evaluate techniques for assessing the environmental viability of these new processes. Thermal treatment is a technology used to convert sludge into valuable products like biochar, bio-oil, and biogas. However, the process is energy-intensive due to the high moisture content of the sludge, which requires significant dewatering. In contrast, hydrothermal treatment offers a more efficient alternative, as it enables the conversion of sludge without the need for pre-drying, making it more suitable for wet biomass. Sludge can also be used for biofuel production through fermentation, in the creation of building materials and other value-added products. However, many of these processes are complex or challenging to scale up. Producing pellets, densified biofuels, is a simpler process. The production of pellets from pulp mill sludge offers a practical and efficient alternative for sludge management, as it can be integrated into the pulp mills. The application of the circular economy concept enables the reutilization of waste in new processes, transforming it into value-added products. To demonstrate the environmental benefits of implementing a new management route, a life cycle assessment (LCA) is an essential tool. It quantifies all inputs and outputs of the process and translates this data into measurable environmental impacts, providing a comprehensive evaluation of the potential environmental advantages. Evaluating new sludge treatment methods, combined with a life cycle assessment (LCA), offers critical information to support changes in the management of pulp mill sludge. This chapter provides a comprehensive review of pulp mill sludge management

Keywords: Pulp industry; Effluent treatment plant (ETP); Sludge; Waste management; Life cycle assessment (LCA).

1. INTRODUCTION

The pulp and paper industry has been expanding worldwide. The high energy demand makes this sector develop a low-carbon economy (OBRIST et al., 2022). In 2017, pulp and paper mills were responsible for 6% of the world's energy consumption in the industrial sector (LIPIÄINEN et al., 2022). Most of the energy used in the mill is derived from the black liquor recovery boilers and the biomass boilers (ORANG; TRAN, 2015). However, this sector still relies on fossil fuels, such as in the CaO kiln during the causticization process, and has considerable carbon dioxide emissions (LIPIÄINEN et al., 2022; SAARI et al., 2022). The industry must invest in research focused on developing and implementing sustainable practices through innovative processes.

The sludge generated at effluent treatment plants (ETP) of the pulp and paper mills can be considered a potential source of energy because they are rich in lignocellulosic materials (BAJPAI, 2015; STOICA; SANDBERG; HOLBY, 2009). The primary sludge, originating from the physical-chemical process in the primary treatment, is usually composed of fibers, lost in the pulping process (MALAISKIENE et al., 2018). Secondary sludge or biological sludge generated in the activated sludge systems, consists mainly of organic matter and water (BAJPAI, 2015).

Secondary sludge is often mixed with primary sludge to facilitate dewatering processes. It is estimated that in Brazil about 1.3 million tons of primary and secondary sludge are generated annually by the pulp sector. Often these wastes are sent to industrial landfills or are used in agriculture after composting (FAUBERT et al., 2016). Landfill is not an attractive practice, and many countries have already banned this form of disposal. Soil application brings some problems such as the need for large areas and long periods for composting and costs (PERVAIZ; SAIN, 2015; ZAMBARE; CHRISTOPHER, 2020).

The challenges associated with managing the high sludge generation lead to the need for new treatment methods to improve the valorization of this material. Extensive research is being carried out to find more suitable applications for sludge in the pulp and paper sector. The production of biofuel, activated carbon, fertilizers, ceramics and bricks are some examples of high-value-added co-products that can be obtained from sludge (FAUBERT et al., 2019; GOTTUMUKKALA et al., 2016; GOVARTHANAN et al., 2022; JARIA et al., 2017).

Most of the technologies investigated so far have a high degree of complexity or high costs, which make their implementation on a large scale challenging (GOTTUMUKKALA et al., 2016). The implementation of new methods of sludge management requires the evaluation of the technical, environmental and economic feasibility associated with this new process. In order to provide a comprehensive overview of the characteristics and current applications of pulp and paper mill sludge, and to identify gaps for improving its management, a review of the latest studies is necessary.

This work aims to review the literature on sludge generated in the pulp and paper industry, recent techniques developed for its management, and the tools used to assess the environmental impact of these processes.

2. PULP MILL SLUDGE GENERATION AND QUALITY

Sludge is generated in pulp and paper mill effluent treatment plants (ETP) (Figure 1.1). The high water demand in the pulping industry leads to substantial affluent generation, which must be treated following environmental regulations before being discharged into water bodies (BRINK; SHERIDAN; HARDING, 2018).

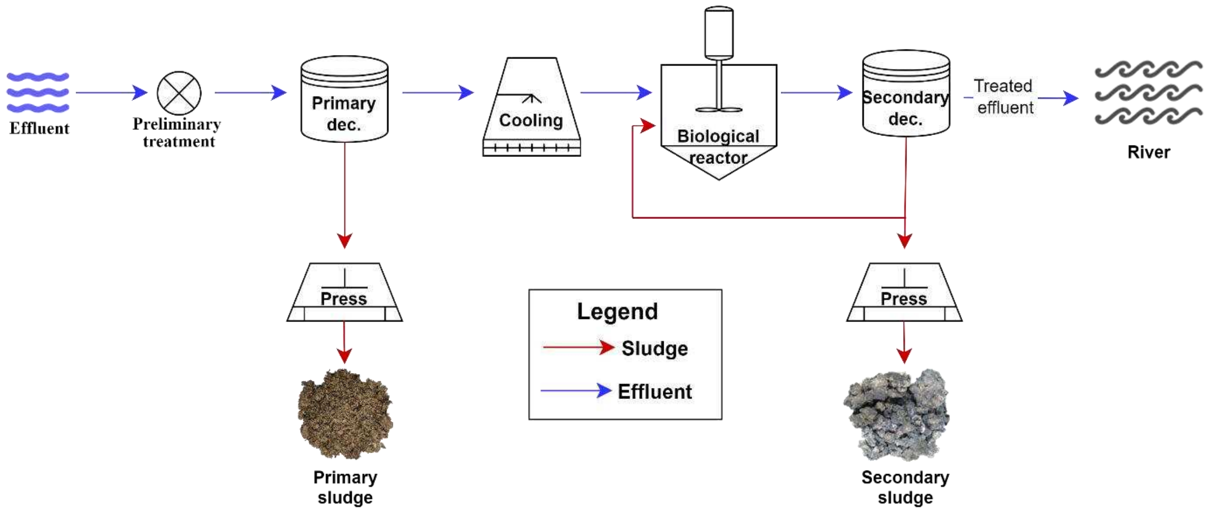

Figure 1.1 Effluent Treatment Plant (ETP) in the pulp and paper mill

The effluent of pulp and paper mills is rich in organic matter, chemicals, and suspended solids (VIDAL et al., 2021). The main sludge properties are shown in Table 1.1. Treatment of this effluent normally requires two main processes, primary and secondary or biological treatment. Generally, preliminary treatment is carried out to correct pH, remove larger solids, and reduce the effluent temperature (PÖYKIÖ; WATKINS; DAHL, 2018).

Table 1.1 Properties of the sludge generated in different pulp mills
Adapted by US EPA (2012), Bajpai (2015)

Production	Properties (%)						
	Total Solids	Ash	C	H	S	O	N
Paper Mill	45.0	3.0	48.4	6.6	0.2	41.3	0.5
Kraft	38.8	7.6	51.6	6.1	0.9	31.2	2.8
Bleached pulp	33.4	1.9	48.7	6.6	0.2	42.4	0.2

The primary treatment is a physical or a physical-chemical process that removes the suspended solid through sedimentation or flotation. Usually, this step removes approximately 80% of the suspended solids (CALDEIRA et al., 2021).

After the primary treatment, the effluent still has a variety of soluble organic compounds that are partially removed in the secondary treatment. At this stage, microorganisms are used to degrade organic matter. There are different technologies used in the secondary treatment, i.e., activated sludge process, aerated lagoons, anaerobic processes, or a combination of both (anaerobic-aerobic) (KAMALI et al., 2016; VIDAL et al., 2021). When required by the legislation, a tertiary treatment can also be applied to remove recalcitrant organic matter and color (RAMOS et al., 2022).

In the pulp mills effluent treatment plants, two distinct types of sludge are generated: primary sludge, which results from the primary treatment, and secondary sludge, which is generated during the biological treatment (GRIMM et al., 2019).

The amount and composition of the sludge are influenced by many factors, including the treatment technology used, the type of raw materials, the pulping process, and the characteristics of the final product (FAHIM et al., 2018; GRIMM et al., 2019). In kraft pulp mills, approximately 58 kg of sludge is generated for each ton of pulp produced. Of this total, around 70% constitutes primary sludge, while 30% is secondary sludge (RODRIGUES et al., 2021).

The efficiency of the fiber recovery system directly influences the volume of primary sludge generated in pulp mills. These systems play a crucial role in capturing valuable fibers from wastewater streams, thereby reducing waste and minimizing sludge production. Modern pulp mills are focusing on improving the efficiency of their fiber recovery systems. Advanced technologies and optimized processes allow for a higher recovery rate of fibers, leading to a significant reduction in both operational costs and the amount of primary sludge generated (KUMAR; VERMA, 2024).

The primary sludge is rich in lignocellulosic materials and can also contain calcium carbonate ($CaCO_3$), kaolinite, and silicon dioxide (SiO_2) quartz (MALAISKIENE et al., 2018).

After dewatered the consistency of the primary sludge is approximately 20-40% (FAHIM et al., 2018). The presence of $CaCO_3$ increases the pH and also affects the enzymatic hydrolysis of the cellulose fibers of the sludge (KUMAR; VERMA, 2024).

Secondary sludge, a byproduct of biological wastewater treatment processes in pulp mills, is a resource-rich material with a complex composition. Its primary constituents include bacterial cells, proteins, minerals, polysaccharides, nucleic acids, enzymes, adsorbable organic halogens (AOX), and ash. These components highlight its biochemical complexity and potential applications in various industrial processes. In addition to these components, secondary sludge also contains a high fiber content, which includes residual lignin, hemicellulose, and cellulose. These fibers are accompanied by essential macro- and micronutrients, and chemical compounds, enhancing their value (KUMAR; VERMA, 2024).

The secondary of this sludge makes it a promising material for resource recovery, such as bioenergy production, agricultural applications, and the development of bio-based materials. The consistency usually is about 0.5 – 2 % before the dewatering and 15 – 20 % after dewatering (BAJPAI, 2015; LOPES et al., 2018). Due to its lower consistency, secondary sludge is often mixed with primary sludge to facilitate handling and disposal (TORRES et al., 2023).

3. SLUDGE MANAGEMENT METHODS

The pulp and paper mills in Brazil generated approximately 1.3 million tons of sludge in 2023 (IBÁ, 2024). The main disposal of sludge has been landfilling, composting, and incineration. However, the current management approach has several drawbacks. Landfills are viewed as costly and environmentally harmful, and they have already been banned in many countries. The composting process requires long periods, it is expensive, and often it faces social challenges due to the strong odors it generates. Incinerating sludge without a pre-treatment step is not a viable option due to the high moisture content and particle heterogeneity, which result in low energy efficiency (RODRIGUES, 2021; SAARI et al., 2022).

The sludge represents 87% of the waste generated inside the pulp mill (TURNER; WHEELER; OLIVER, 2022). To ensure the sustainable development of pulp waste (co-products) use, new applications have been evaluated for sludge management as shown in Figure 1.2 (TORRES et al., 2023). The effective management of sludge can provide an alternative option to fossil resources and forest wood (DODDAPANENI; KIKAS, 2023).

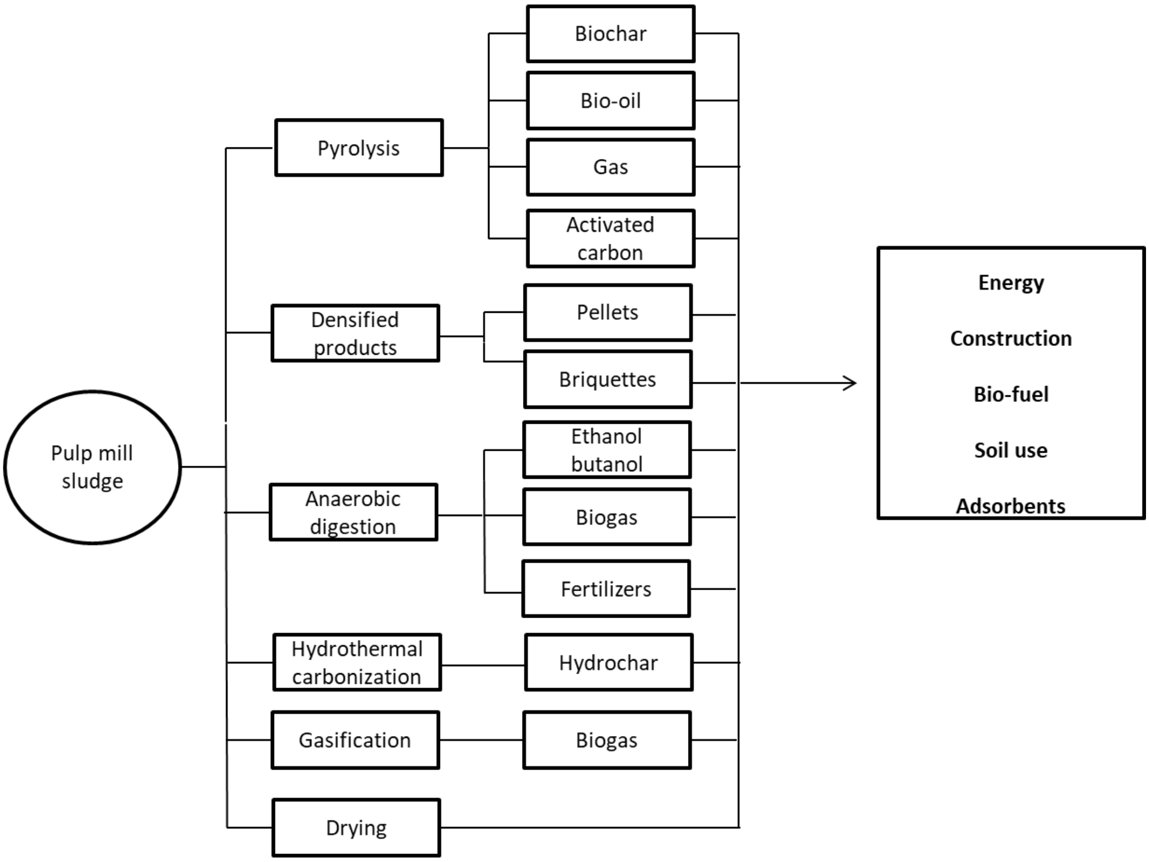

Figure 1.2. Sludge treatment methods and final products

Thermal treatment of the sludge can improve its properties and make it suitable for novel uses. Pyrolysis consists of an oxygen-deficient thermochemical process (400 – 900 °C) that results in the production of biochar, biogas, and bio-oil (RIDOUT; CARRIER; GÖRGENS, 2015). This process can be an alternative treatment to pulp and paper mill sludge, enabling energetic efficiency and chemical recovery (CHEN et al., 2024).

Slow pyrolysis is optimized to produce charcoal due to its relatively low heating rates, ranging from 1 to 50 °C per minute, and an extended residence time for the material. These conditions allow for a controlled decomposition process, yielding higher quantities of solid carbon-based products. In contrast, fast pyrolysis employs significantly higher heating rates, typically between 300 and 12000 °C per minute, coupled with shorter residence time. This rapid process maximizes the production of gaseous byproducts while favoring the formation of bio-oil, a valuable liquid energy source (GOTTUMUKKALA et al., 2016; OUADI et al., 2013).

The production of biochar from sludge using a chemical activator enhances its properties, making it an effective adsorbent for water treatment applications (REIS et al., 2022; WANG et al., 2023). The co-pyrolysis of pulp and paper mill sludge with other biomass is also an option that can show a positive influence on the pyrolysis process and the characteristics of the final product (LIN et al., 2024).

The gasification of biomass has emerged as a highly efficient process for energy conversion, offering the advantage of reduced environmental impact. However, certain challenges persist, particularly the high moisture and ash content of some biomass materials, which can hinder the efficiency of the conversion process (ROSHA; IBRAHIM, 2022). Co-gasification, where multiple types of feedstocks are combined, has been identified as a promising alternative to address these limitations. This approach enhances gas production and improves the overall versatility and adaptability of the gasification process by leveraging the complementary properties of different feedstocks (ROSHA et al., 2022).

Numerous studies have also explored the potential of sludge from pulp and paper mills as a feedstock for bio-oil production. Ridout et al. (2016) compared slow and fast pyrolysis techniques using two types of sludge: one from a kraft pulp mill with low ash content and another from a recycled paper mill with high ash content. Their findings revealed that fast pyrolysis achieved higher gross energy conversion, primarily due to the increased production of condensable organic compounds.

Torrefaction is a low-oxygen thermal process conducted at lower temperatures than pyrolysis (200 – 300 °C), resulting in the partial degradation of biomass (PRERADOVIC; PAPUGA; KOLUNDŽIJA, 2023). Torrefaction of pulp mill sludge makes it more suitable for energy generation. The degradation of compounds with lower heating values increases the total heating value and fixed carbon concentration of the sludge while also reducing its hygroscopic equilibrium moisture content (TORRES et al., 2023).

The main challenge to sludge management is the high water content. A drying step is required prior to the thermal treatment of sludge, which increases energy demand and costs (MARTINEZ; SERMYAGINA; VAKKILAINEN, 2021). Hydrothermal carbonization (HTC) is an alternative technology that allows the elimination of the pre-drying step (SALCEDO-PUERTO et al., 2024). The HTC process uses wet biomass to produce hydrochar in high-pressure (1–5 MPa) conditions and temperatures between 180 and 260 °C. To produce chemical and liquid biofuels in wet sludge, hydrothermal liquefaction (HTL) can be carried out. This process involves higher temperatures (280 – 370 °C) and pressure (10 – 25 MPa) than the HTC process and results in four different products bio-oil, aqueous phase, solid residue, and gas residue (KUMAR; VERMA, 2024).

The hydrochar presents a higher heating value and is more hydrophobic than the non-treated sludge. The advantage of this product is lower energy demand in the post-drying step and higher energy potential during the combustion process in the boiler, besides energy generation, the biochar also can be used as a soil conditioner (MARTINEZ; SERMYAGINA; VAKKILAINEN, 2021).

Anaerobic digestion is an alternative method for treating sludge. By adjusting the conditions during this process, it is possible to produce various types of biofuels (i.e. bioethanol, biohydrogen, biobutanol) (KUMAR; VERMA, 2024).

The high fiber content in the sludge makes this material suitable to be incorporated in the production of fiberboard and wood-plastic composites (KIZINIEVIC et al., 2022). The sludge can also be used as reinforcement in adobe (mud brick), improving the compressive strength and reducing the ecological footprint of the construction and buildings sector (MUÑOZ et al., 2020).

The composition of pulp mill sludge enables its use in different processes to create value-added products. Researchers have also explored its potential for producing chemicals, bioplastics, and nanomaterials (DHANDAPANI et al., 2021; MAJUMDAR et al., 2020; PEI et al., 2022).

Despite significant advancements in research, transitioning these innovations from laboratory settings to industrial-scale operations remains a complex and multifaceted challenge. One critical hurdle lies in scaling up. Processes that demonstrate promising results in controlled laboratory conditions often encounter unforeseen complications when adapted to industrial scales (SAARI et al., 2022). This includes maintaining process efficiency, consistency, and safety while managing increased throughput.

A thorough understanding of the environmental and economic implications of these processes is essential for their successful implementation. New treatment and conversion technologies must not only meet sustainability goals but also align with industry budgets and regulatory requirements. Comprehensive lifecycle analyses are often necessary to evaluate the environmental benefits and potential trade-offs, such as carbon footprint or water usage.

Furthermore, some advanced technologies rely on extreme operating conditions, such as high pressures and temperatures. While these conditions can improve the efficiency and yield of conversion processes, they also lead to increased energy demand, which can undermine the environmental benefits of the treatment. Additionally, the complexity and cost of the equipment required to operate under these conditions pose significant barriers to widespread adoption.

4. PELLETS PRODUCTION AND USE

Pellets are densified renewable fuel produced through the pelletizing process, which consists of densifying biomass using presses. This equipment allows pellets to be formed by compacting the raw material and increasing the temperature (70-100ºC) due to the friction generated during the process (WHITTAKER; SHIELD, 2017).

To obtain quality pellets, certain criteria must be followed during the pelletizing process, such as the temperature of the matrix, the pressure exerted on the biomass, and the geometry, while concerning factors intrinsic to the raw material, i.e., the moisture content, chemical composition, and particle size (SANTANA et al., 2020).

Assessing pellet quality is crucial for their effective application and is based on their chemical, physical, dimensional, and mechanical properties. These characteristics directly impact fuel performance and can be evaluated using established standards (PRADHAN; MAHAJANI; ARORA, 2018).

Pelletizing standardizes the shape and size of the material, increases its energy density, and reduces moisture content. These characteristics increase the energy efficiency in the boiler and lead to economic benefits by simplifying operations such as handling, transport, and storage (CASTRO et al., 2021; MOSTAFA et al., 2019). Biomass that presents low density requires bigger storage space and higher transportation costs (BAJWA et al., 2018).

Pellets can be produced from a wide range of biomass sources, including agricultural waste, woody biomass, livestock manure, marine biomass, food waste, and municipal waste (WEI; CHENG; SHEN, 2024). Primary and secondary sludge are attractive biomass sources for pellet production due to their high lignocellulosic content. Pelletizing addresses the main challenges associated with energy conversion from this biomass, such as heterogeneity, low density, and high moisture content (NOSEK et al., 2017; RODRIGUES, 2021).

Pellets have diverse applications across both residential and industrial sectors. In Europe, they are mostly used in residential cookers and heaters. In industry, pellets are used for energy generation, and biomass gasification (PRADHAN; MAHAJANI; ARORA, 2018). Pellets are also an excellent option for co-firing with fossil fuels, such as coal, which enables more cost-effective energy generation while reducing CO_2 emissions (WEI; CHENG; SHEN, 2024).

The torrefaction of pulp mill sludge pellets enhances their properties as a fuel and can be integrated into anaerobic digestion processes to produce volatile fatty acids (VFA) and biomethane. (DODDAPANENI; KIKAS, 2023).

In conventional pellet processes, biomass moisture levels are typically around 15%. However, the high moisture content of sludge presents a significant challenge. The pelletizing of sludge carried out with higher moisture content than the conventional, offers advantages, like increased pellets resistance, and reduces the need for extensive drying, reducing the energy demand of the process (RODRIGUES et al., 2021). Biomass has lower heat transfer efficiency; however, maintaining an optimal moisture level can significantly enhance this process. The presence of water in the right proportion contributes to the heat transfer, improving the mechanical properties of the pellets (UNGUREANU et al., 2018).

The ash content in sludge, particularly in secondary sludge, presents a significant challenge for energy conversion processes. High ash levels adversely impact the heating value of the material, reducing its efficiency as a fuel source. Additionally, the presence of ash contributes to operational issues, such as corrosion and incrustation within boilers. These issues not only compromise the performance and longevity of equipment but also lead to increased maintenance requirements, driving up operational costs (TORRES et al, 2023). Blending sludge with biomass that has a lower ash content helps regulate the overall ash composition, thereby enhancing energy conversion efficiency.

5. CIRCULAR ECONOMY

The concept of the circular economy emerged in the 1970s, but its practical application in Brazil has only gained attention in recent years. The circular economy, or restorative economy by nature, consists of a model in which the waste generated in the industries is reallocated and reused in different ways, without any loss (AZEVEDO, 2015). The implementation of this new system represents the opposite of the linear economic model, which is based on extraction, transformation, production, utilization and disposal (LEITÃO, 2015).

The main objective of the circular economy is to minimize the consumption of raw materials, energy, and water by closing the product life cycle, and reintegrating materials back into the production process (LEITÃO, 2015). In this model, what was once considered waste becomes a raw material for new products, maintaining a continuous flow of materials within a closed industrial cycle.

Within the context of the circular economy, William McDonough and Michael Braungart (2002) introduced a new concept called cradle-to-cradle (C2C), which aims to merge economic progress with environmental conservation. C2C is based on three fundamental principles: The first, "waste equals food," emphasizes the potential to reuse waste as nutrients for other processes; the second, "use of renewable energy” advocates for the use of renewable energy sources to power production systems; and the third, "celebrate diversity," rejects standard solutions in favor of adapted and diversified approaches (MCDONOUGH et al., 2003).

The implementation of the circular economy in production systems brings benefits in both economic and environmental aspects, promoting an increase in productivity, and a reduction in the exploitation of primary resources and the generation of waste (MOLINA-MORENO et al., 2017).

6. LIFE CYCLE ASSESSMENT

The economic and environmental feasibility of a process can be assessed through the life cycle assessment (LCA). This method enables a comprehensive analysis of the entire production chain, from development to end-of-life, providing insights into both the economic and environmental impacts of the product (UBANDO et al., 2022). To conduct an LCA, all resources consumed, and pollutants emitted throughout the process are accounted. This step ensures a comprehensive assessment of the inputs and outputs associated with the process (SALCEDO-PUERTO et al., 2024).

The LCA is carried out following the International Organization for Standardization (ISO) standard as shown in Figure 1.3, presenting four fundamental steps: i) goal and scope definition; ii) life cycle inventory (LCI); iii) life cycle impact assessment; and iv) LCA result and interpretation (SRIKUMAR et al., 2024).

The first step, goal and scope definition aim to clarify the reason and the application of the LCA, in this phase also are presented the system boundary, functional unit and allocation of the study. The LCI involves collecting detailed data on all the inputs and outputs of the process, including the resources used and the emissions generated. In the third step, the environmental impacts of the process are assessed, the life cycle impact assessment (LCIA), and categorized into midpoint and endpoint indicators. The result interpretation step consolidates the information from the previous phases, providing conclusions and insights into the overall environmental impact (YU et al., 2022).

Several LCA studies have been conducted to assess the environmental impact of agricultural waste and biomass use in different sectors, such as energy use, soil application, landfill disposal, and other added-value products (MARTÍN-GAMBOA et al., 2020; SALCEDO-PUERTO et al., 2024; SREEKUMAR et al., 2020).

The LCA applied to sludge management was found in literature in different studies. In their 2017 study, Abuşoğlu et al. 2017 conducted an LCA to compare the environmental impacts of two processes utilizing digested sewage sludge: incineration for energy generation and its use as a secondary fuel in a cement kiln. The analysis revealed that the incineration process demonstrated superior environmental performance compared to the use of sludge in the cement kiln.

The management of biosludge generated at a pulp mill in Finland was assessed using an LCA, evaluating three distinct scenarios. The first scenario involved incineration for energy production with ash disposal in a landfill. The second scenario focused on hydrochar production for energy generation, while the third explored hydrochar production for composting and nutrient recovery. Among the evaluated options, the third scenario demonstrated the lowest environmental impact (SALCEDO-PUERTO et al., 2024).

LCA is a valuable tool for understanding the environmental impact of each stage of a given product's production, enabling the comparison of different processes and identifying opportunities for improvement. As it provides a comprehensive view of the production chain, LCA supports efforts to improve sustainability and reduce the environmental footprint of a product (SRIKUMAR et al., 2024).

The goal and scope of an LCA is an important step, as it identifies the evaluated product and defines the main focus of the research (LIEBSCH, 2019). The system boundaries (SB) are established in this step, according to ISO 14040 the SB is "*a set of criteria specifying which unit processes are part of the product system*". It defines the limit of the analyses inside the process, specifying which input and output should be taken into account (DECORTE; VAN DEN BOSSCHE; STEEMAN, 2023).

The functional unit (FU) is also defined during the goal and scope definition, and it is used as a unit basis for the calculations and in the comparisons between different scenarios or products. The FU can be energy-based (MJ; kWh; MW end products) and mass-based (kg; ton feedstock/product), is also possible to have FU in area or distance (km; m), but it is not commonly used in biomass conversion (YU et al., 2022).

When a process produces multiple products, the environmental impact must be proportionally allocated among them. This ensures that each product is assigned its fair share of the overall environmental burden (SCHRIJVERS; LOUBET; SONNEMANN, 2016).

Although the ISO standard recommends expanding system boundaries to avoid allocation, there are four main methods for addressing the allocation problem. The first method consists of no allocation, where a specific product is responsible for the entire environmental impact. Economic allocation is the second and one of the most used methods, in this case, the environmental impact distribution is according to the value of each product. The biophysical allocation (third method) considers the physicochemical properties (e.g. mass and energy content) of the products. System expansion, the fourth method, is calculated by subtracting the avoided emissions from the total emissions. This approach accounts for the environmental benefits of displacing the production of a co-product, thereby reducing the overall impact (YU et al., 2022).

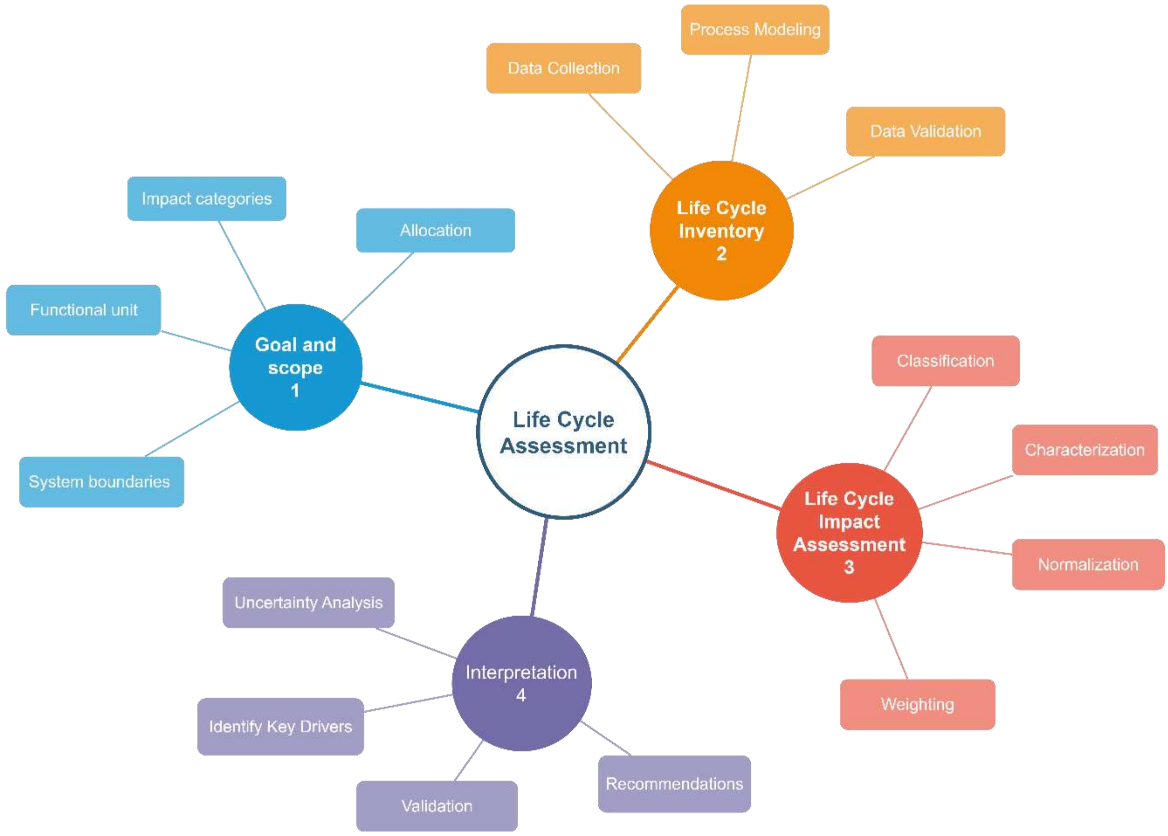

Figure 1.3. Steps to perform an LCA according to the International Organization for Standardization (ISO)

The LCI involves the documentation and quantification of all inputs and outputs associated with a product, process, or service. This includes gathering data on the resources consumed (such as materials, energy, and water) and the emissions or waste generated (such as pollutants and co-products) at each stage. The collected data is used to create an inventory that provides a detailed account of environmental resource use and emissions across all phases of the process, from raw material extraction to disposal (YU et al., 2022).

Data collection has a direct influence on the results of the environmental assessment. To have an accurate LCA model, it must ensure both the quality and availability of the inventory data (SALA et al., 2021). The data can be collected through different sources, such as commercial and public databases, laboratory tests, surveys, and literature reviews. For processes that are novel or have specific particularities, existing databases may be inadequate. In these cases, auxiliary tools like machine learning can be employed to generate the necessary information, helping to fill data gaps and improve the accuracy of the Life Cycle Inventory (YU et al., 2022).

There are two main ways to divide the LCIA, at the midpoint level and endpoint level. Midpoint indicators focus on specific environmental issues, while endpoint indicators reflect the broader effects on ecosystems, human health, and resource availability (MUENCH; GUENTHER, 2013).

Midpoint categories typically represent environmental impact indicators such as global warming potential, acidification potential, ecotoxicity potential, eutrophication potential, water depletion potential, and fossil energy use (HUIJBREGTS et al., 2016).

The endpoint categories results are obtained through the integration of the mid-point indicators and can be expressed in human health, ecosystem quality, and resource depletion (YU et al., 2022).

In the LCA results and interpretation phase, the results obtained in the previous steps are analyzed, identifying issues and opportunities. The conclusions drawn from the analysis serve as the basis for formulating recommendations (WANG et al., 2021).

7. CONCLUSIONS

The high production of sludge from pulp mills and the increasing restrictions on landfilling highlight the need for more sustainable management routes for this waste. Many researchers are exploring innovative uses for pulp mill sludge. The pelletizing process is a simple technology that allows the transformation of sludge into a densified fuel and can be used as an alternative option to its management.

Implementing a new process requires several steps and careful consideration. In addition to evaluating technical feasibility, it is essential to assess the complexity of the process, the environmental impact, and economic viability. The LCA is an important tool to evaluate the environmental impact of a product or a process.

Addressing the challenges of sludge management requires a multidisciplinary approach that integrates engineering, environmental science, and economics. Innovations in energy efficiency, process optimization, and equipment design, coupled with supportive policy frameworks, could play pivotal roles in overcoming these obstacles and unlocking the full potential of pulp mill sludge as a resource for sustainable production.

Assessing the technical feasibility of producing a new product using sludge, combined with an LCA analysis, supports the pulp and paper industries make more sustainable choices. This approach facilitates scaling up processes and developing effective solutions for managing pulp mill sludge.

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CHAPTER 2

POTENTIAL ENERGY GENERATION OF SLUDGE FROM A THERMOMECHANICAL PULP (TMP) MILL

POTENTIAL ENERGY GENERATION OF SLUDGE FROM A THERMOMECHANICAL PULP (TMP) MILL

Abstract

Pressure to increase the renewable energy matrix motivates the search for new energy sources. Pulp and paper mills have been constantly expanding in Brazil, and to ensure their competitiveness, new processes should be established both in terms of reducing the waste discarded in landfills, as well as substituting fossil fuels used in the mills. This study aims to evaluate the energy potential of primary sludge (PS), secondary sludge (SS), a mixture of primary and secondary sludge (MS), and bark (B), consisting of 80% eucalyptus and 20% pine, all generated in a Brazilian thermo-mechanical pulp (TMP) mill. Characterization of all biomasses was performed. Moisture content, higher, lower, and net heating value, elemental, structural, and immediate composition, thermogravimetric analysis and quantification of minerals and metals were determined. The bark presented the highest higher heating value (HHV) (18.90 MJ kg^{-1}), while the secondary sludge had the lowest HHV (12.45 MJ kg^{-1}). The high concentration of ash in the sludges PS (26.58%), SS (40.46%) and MS (25.78%) negatively affected their heating value. The sludges also showed high moisture content, which made their burning without a previous drying stage unattractive. Nevertheless, the sludges and the bark are rich in carbon which makes them attractive for use as biomasses for energy generation.

Keywords: Thermomechanical pulp, Pulp and paper mill sludge, Biomass, Circular economy, Waste-to-energy.

1. INTRODUCTION

The transition of the world energy matrix in replacing fossil fuels with renewable materials has generated new demands within the industrial sector (OLUJOBI et al., 2022). Following the trend of the so-called circular economy, pulp and paper mills have been seeking innovations in their processes to ensure their competitiveness in the market (BEN DAYA; NOURELFATH, 2019).

The production of pulp from wood chips can be accomplished through chemical, mechanical or thermal processes, or a combination of all these (DISLAIRE et al., 2021). The thermomechanical pulping (TMP) process is widely used for high-yield pulp production. The process consists of the impregnation of wood, in the form of logs or chips, with water (CALDEIRA et al., 2021; COLODETTE, 2015).

Before being submitted to the process, the bark is removed from the wood logs because it contains high levels of extractives and minerals and has a low cellulose content (NEIVA et al., 2016). In the past, the bark was also considered a waste generated in the process, but, currently, it is considered a co-product since it can be an energy source through the combustion in the biomass boiler or can also be used as a fertilizer to maintain soil nutrients (AMÂNDIO et al., 2021).

The treatment of the effluent from this production typically consists of conventional processes, primary treatment (physicochemical), and secondary treatment (aerobic, anaerobic, or a combination of both) (CALDEIRA et al., 2021; REDLINGER-POHN et al., 2016). The effluent treatment technologies normally result in the generation of primary and secondary sludge.

Primary sludge consists mainly of fibers and has a consistency ranging from 20-40% after dewatering (FAHIM et al., 2018). Secondary or biological sludge, on the other hand, has a high content of organic matter and microorganisms, with a consistency of 15-20% after centrifugation. The characteristics presented by the secondary sludge make its dewatering difficult, and for this reason, it is commonly mixed with the primary sludge to turn its handling and final disposal easier (HEINDL, 2024; LOPES et al., 2018).

Sludge management is a big issue in pulp mills, mainly due to its high generation. Many mills still consider this material to be waste, with its main destination being industrial landfills (GURRAM et al., 2015; NIJU; VIJAYAN, 2020). Landfills are responsible for anthropogenic emissions of pollutant gases, especially methane gas (CH_4), and present a potential risk of groundwater and soil contamination (KAROUACH et al., 2021; NJOKU; EDOKPAYI; ODIYO, 2019).

The valorization of sludge is an important agenda within the pulp and paper industrial sector since it meets several of the goals set at the Conference of the Parties to the United Nations Framework Convention on Climate Change (COP 29) in 2024. The energy use of sludge emerges as an interesting alternative because it makes it possible to both reduce the amount of waste sent to landfills and expand the energy matrix to include renewable sources (MONTE et al., 2009; NOSEK et al., 2017).

The use of bark and sludges directly in the boiler for energy generation presents a series of problems due to the high moisture content, heterogeneity of their particles and the low energy density related to a high volume (SETTE JR. et al., 2018). This highlights the importance of conducting a careful analysis of the main characteristics of these biomasses to evaluate the best conversion route.

Other researchers have already evaluated the energy potential of sludge. Pellets produced with paper sludge from a recycled paper mill were suitable to be used as fuel when mixed with sawdust (NOSEK et al., 2017). The co-fire using coal with 10% of primary or secondary sludge generated in a kraft pulp mill did not affect the burning behavior of the fuel and showed to be a good alternative to sludge management (COIMBRA et al., 2015). The gasification using 20% wood chips and 80% pellets made from deinking sludge and blends of rejects of secondary fiber paper mills produced biogas with the following properties 23.34% CO, 16.25% H_2, 5.21% CH_4, 12.71% CO_2, and 42.49% N_2 (OUADI et al., 2013). Chen et al. (CHEN et al., 2021) have investigated the thermal conversion characteristics of petrochemical wastewater sludge. The results indicated that the combustion process of the sludge had four stages: evaporation of moisture and light volatiles; degradation of organic components, degradation and oxidation of residues, and combustion of chars previously formed. Due to the wide variation in chemical and physical properties among different types of waste, the behavior during combustion and operating parameters of the boilers change according to the fuel characteristics (CHEN et al., 2021). The TMP sludge properties depend on some individual aspects of the mill, such as the wood type, the industrial process, and the effluent treatment plant (GRIMM et al., 2019). The properties of sludge influence the energy generation potential, and due to the wide variation between sludges, it is important to carry out a study evaluating these properties and their influence during the burning process,

The present work aims to evaluate and make a critical analysis of the physical and chemical characterization of primary sludge, secondary sludge, mixed sludge, and bark generated in a thermomechanical pulp mill. This evaluation focuses on determining the energy potential of the sludges and bark for use as fuel in the biomass boiler

2. MATERIALS AND METHODS

2.1. TMP process

The sludge and bark used in this study originated from a Brazilian high-yield thermomechanical pulp mill. The unit is located on Fazenda Levantina farm in the district of Monte Verde (in the municipality of Camanducaia, Minas Gerais state).

The wood used in the pulping process is *Eucalyptus sp*. (80%) and *Pinus sp*. (20%). To obtain the cellulose, the bark is removed, and the logs wood is processed. To the wood defibrillation mechanical action and heat are used. After the thermomechanical process, the rejects are removed through sieving and depuration processes, and then the pulp is bleached using hydrogen peroxide (H_2O_2), and sodium hydroxide (NaOH). and DTPA (Diethylenetriamine pentaacetate). The final brightness of the pulp depends on the final product and is approximately 75% ISO. After the bleaching process, the pulp is dried and packaged.

2.2. Effluent treatment plant and sample collection

The industrial process generates approximately 25-30 $m^3.h^{-1}$ of liquid effluents that are sent to an effluent treatment plant (ETP). The effluent from the TMP mill is generated in the processes of pulp dilution, bleaching, washing, and consistency adjustment. The effluent treatment plant consists of a preliminary treatment (sieves), physical primary treatment (sedimentation), physical-chemical primary treatment (flotation), and a secondary treatment consisting of an up-flow anaerobic reactor and sludge blanket (UASB) followed by an aerobic activated sludge process (AS) (Figure 2.1). During the primary physical-chemical treatment it is used coagulant, alkaliser, polymer, N and P. The sludge generated in the physical and physical-chemical primary treatment is mixed in a tank. The secondary sludge from the UASB and AS systems are densified in a gravity thickener, mixed with the primary sludge and sent to be dewatered in a centrifuge.

The bark generated after harvesting was collected for further analysis. The primary sludge (PS) was collected in the mixing tank, the aerobic secondary sludge (SS) was collected in the densifiers, and the mixed sludge (MS), a mixture of primary sludge with secondary sludge, was collected after the centrifuge step. To ensure representativeness, the samples were collected at three different points in the generation site, resulting in 3 samples of 200 liters each. The samples for this study were collected in October 2021.

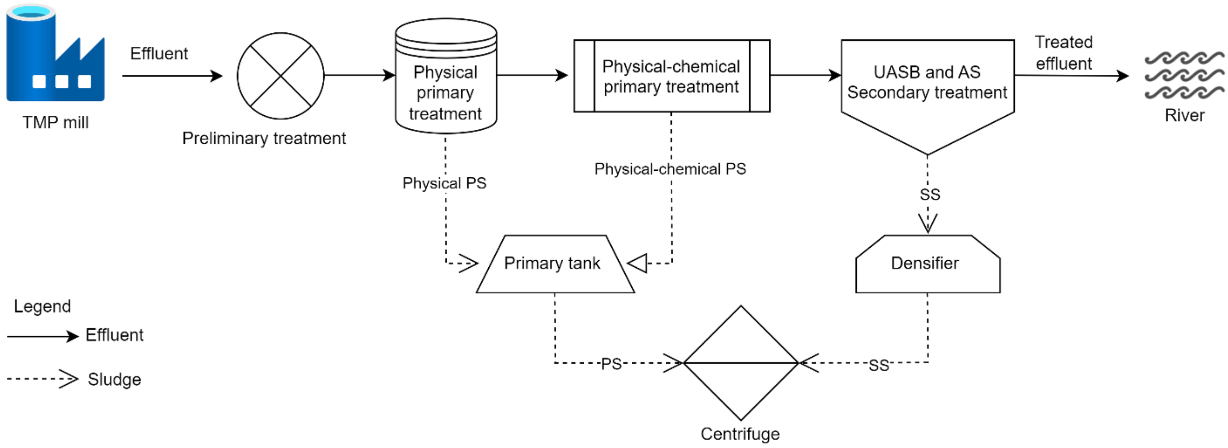

Figure 2.1. The effluent treatment plant (ETP) of the TMP mill

2.3. Sample preparation

The samples of primary sludge (PS), secondary sludge (SS), the mixture of PS and SS sludge (MS) and bark (B) were sent to the Pulp and Paper Laboratory of the Federal University of Viçosa, where they were conditioned at 7 ºC, in the cold storage room, to ensure their preservation.

Part of the PS, SS, MS and B was dried in an oven for 24 hours at 100 ºC, and after this period the MS and B had already reached a constant moisture content. This material was removed from the oven, conditioned in aluminum trays until it reached room temperature, and then stored in polyethylene bags. The PS and SS, due to the higher moisture content, remained in the 60 ºC oven for another 48 hours, totaling 72 hours in the oven, followed by the same cooling and storage process.

All samples were ground before performing the analyses. Due to the heterogeneity of the ground bark they were submitted to sieving on 40- and 60-mesh sieves. The material used for the analyses was that which was retained in the 60-mesh sieve.

2.4. Sample characterization

The energy potential of each biomass was evaluated through the analysis of moisture and solid content, according to Standard EN 14774-1 (DIN, 2009).

The elemental composition of the waste (carbon, hydrogen, nitrogen, and sulfur) of the sludge was determined using Elemental Analyzer equipment, model TruSpec CHN Micro, TruSpec S. The oxygen value was obtained by adding carbon, nitrogen, hydrogen, sulfur, and ash content decreased by 100, according to Standard EN 15296 (DIN, 2011).

The proximate analysis for the determination of ash, fixed carbon and volatile matter was performed according to Standard EN 14.775 and 15148 (DIN, 2010a, 2012). The fixed carbon was calculated with the sum of the ash and material content decreased by 100.

Total lignin was obtained by summing soluble and insoluble acid lignin following the TAPPI UM 250 (TAPPI, 2000a) and TAPPI T 222 om-02 (TAPPI, 2000b) standards, respectively. The sugar composition was determined according to SCAN-CM 71:09 (SCAN, 1993).

The contents of P, K, Ca, Mg, S, B, Fe, Cu, Zn and Mn were obtained through nitroperchloric digestion according to Sarruge and Haag (SARRUGE; HAAG, 1974) and read in an optical emission spectrometer with inductively coupled plasma (ICP-OES, Perkin Elmer Optima Model 8300 DV). The aluminum content was obtained in an atomic absorption spectrometer (SPECTRaA 55B, Varian Techtron, Victoria, Australia).

The higher heating value was obtained using the Parr 6300 adiabatic calorimeter pump method, based on the international test method ASTM D2015 (ASTM, 1982). The net heating value (NHV) was estimated using Equation 1, following Annex E of the European standard DIN EN 14918 (DIN, 2010b).

$$NHV = HHV - 212.2 \times H - \ 0.8 \times (O + N)) \times (1 - 0.01 \times M) - (24.43 \times M) \quad (1)$$

Where:

NHV (constant pressure): Net heating value at constant pressure, in J g^{-1};

HHV: Higher heating value, in J g^{-1};

H, O, N: hydrogen, oxygen, and nitrogen, respectively, in percent (%);

M: moisture, wet basis, in percent (%).

The lower heating value (LHV) was calculated according to Equation 2.

$$LHV = HHV - \left[600 * \left(\frac{9H}{100}\right)\right] \quad (2)$$

Where:

LHV: Lower heating value, in J g^{-1};
HHV: Higher heating value, in J g^{-1};
H: Hydrogen, in %.

The thermogravimetric analyses (TG/DTG) were performed using Shimadzu DTG 60H equipment with synthetic N flow (50 mL min^{-1}), temperature range of 25-1000 °C and heating rate of 10 °C min^{-1}, using sample masses of approximately 2 mg, absolutely dry.

2.5. Statistical Analysis

The tests were performed in duplicates for each sample, totaling eight tests. Descriptive statistics were performed, obtaining values of mean, standard deviation (SD) and coefficient of variation (CV).

3. RESULTS AND DISCUSSION

3.1. Biomass characterization

The evaluation of the energy potential of the biomass was performed using the results obtained from the elemental and immediate analyses along with the heating value and moisture content. Table 2.1 presents the results of the analyses. The higher heating value (HHV) consists of the maximum amount of energy that a biomass can release during combustion (XING et al., 2019).

The HHV value presented is similar to those found in the literature for materials from pulp and paper mills. The HHV of the PS (13.80 MJ kg^{-1}) was similar to that found by Demuner et al. (2021) in black liquor, which presented an HHV of 13.90 MJ kg^{-1}. When evaluating the mixed sludge from a kraft paper mill that uses bamboo species as feedstock, Goel (2017) reported an HHV of 12.7, which presented close results with the HHV found in the mixed sludge (13.05 MJ kg^{-1}) reported in this work (GOEL; KALAMDHAD, 2017).

Among the biomasses evaluated, the bark presented the highest HHV (18.90 MJ kg^{-1}) followed by the primary sludge (13.80 MJ kg^{-1}). The secondary sludge presented the lowest HHV, with a difference of 1.35 MJ kg^{-1} when compared to the PS. This fact influenced the reduction of the higher heating value of the mixture of PS and SS, which was 13.05 MJ kg^{-1} for this biomass.

The lower heating value (LHV) is a measure that is associated with the condensation of water vapor and takes into account the HHV and the hydrogen content present in the material. It always presents values lower than the HHV, as observed in the data presented in Table 2.1 (SILVA et al., 2015). For a material used for energy generation, it is desirable to have a higher heating value, since this property will influence the higher or lower steam generation in the boiler. HHV is directly related to the composition of the biomass. While the presence of organic compounds has a positive correlation with the HHV, the inorganic compounds will promote the reduction of the heating value (DEMUNER et al., 2021).

Table 2.1 Characterization of primary sludge (PS), secondary sludge (SS), sludge mixture (MS) and bark (B) for evaluation of energy potential

Properties (%)	**Sludge**									**Bark**		
	Primary			**Secondary**			**Mix**					
	PS			**SS**			**MS**			**B**		
Higher heating value - HHV (MJ kg^{-1})	13.80	±	0.00	12.45	±	0.07	13.05	±	0.07	18.90	±	0.14
Lower heating value - LHV (MJ kg^{-1})	12.90	±	0.00	11.35	±	0.07	12.00	±	0.00	17.80	±	0.14
Net heating value - NHV (MJ kg^{-1})	-1.65	±	0.00	-2.30	±	0.07	-0.09	±	0.00	9.16	±	0.14
Moisture - Wet basis (%)	94.77	±	0.10	98.99	±	0.01	83.68	±	0.08	42.49	±	11.31
Solids (%)	5.23	±	0.10	1.01	±	0.01	16.32	±	0.08	57.51	±	11.31
Fixed carbon (%)	13.47	±	0.13	10.18	±	0.23	14.54	±	0.11	22.59	±	0.92
Volatile matter(%)	59.95	±	0.31	49.36	±	0.51	59.68	±	0.11	66.97	±	0.61
Ashes (%)	26.58	±	0.33	40.46	±	0.29	25.78	±	0.10	10.44	±	0.31
Carbon (%)	35.5	±	0.14	32.30	±	0.28	37.70	±	0.00	50.25	±	0.07
Hydrogen (%)	4.57	±	0.01	4.23	±	0.11	4.83	±	0.01	5.40	±	0.07
Oxygen (%)	32.25	±	0.05	19.44	±	0.11	30.59	±	0.01	32.25	±	0.05
Nitrogen (%)	1.10	±	0.02	3.48	±	0.03	1.05	±	0.00	0.47	±	0.03
C/N	42.80	±	0.83	10.83	±	0.18	41.89	±	0.00	124.47	±	8.03
H/C	1.54	±	0.01	1.57	±	0.03	1.54	±	0.00	1.29	±	0.02
O/C	0.68	±	0.01	0.45	±	0.03	0.61	±	0.00	0.50	±	0.02

All the evaluated biomasses had a high moisture content. The moisture content on a wet basis (wb) of the SS (98.99%) and the PS (94.77%) indicated that the material consists largely of water, having a low solids content, of 1.01% for SS and 5.23% for PS. Even though it observed a reduction of 15.31% of the moisture content (wb) of MS when compared to the SS, it still has a solids content of only 16.32%.

The presence of water is undesirable for energy generation, since it promotes the reduction of the heating value, besides stealing energy from the system for the heating and evaporation of water. This fact promotes higher emission of gases and an increase in the consumption rate of the dry biomass to ensure energy efficiency, which results in higher requirements of the mechanical potential of the boiler (COSTA; TARELHO; SOBRINHO, 2019). The moisture content observed in the sludges of this study highlights the need for prior partial drying of the waste to augment its energy potential.

The fixed carbon (FC) contents found in the samples were 13.47%, 10.18%, 14.54% and 22.59% for the PS, SS, MS and B, respectively. The FC content identified in the bark was higher than that found in the literature, which is around 17.1% (NEIVA et al., 2018). Fixed carbon is a desirable property in biomass since it is directly correlated with the higher amount of energy and lower ash content of the fuel.

Volatile matter (VM) indicates the reactivity of the fuel with the increase in temperature. The VM content found in PS, SS, MS and B was 59.95%, 49.36%, 59.68% and 66.97%, respectively. A study conducted by Silva and Ataíde (2019) reported a VM content of 87.95% and FC of 11.59% in Eucalyptus grandis wood, results that differ from the values found for the biomasses evaluated in this work (SILVA; ATAÍDE, 2019). Fuels with a high content of volatile matter and a low fixed carbon content tend to burn faster (GARCIA et al., 2019; WALLIKHANI; ASAKEREH; FARROKHIAN FIROUZI, 2022).

The ash content is inversely proportional to that of the VM and FC, i.e., the lower the VM and FC, the higher the concentration of ash in the material. This fact can be observed in the samples of this study. The SS had the lowest VM and FC contents and the highest ash content (40.46%). In the case of the bark, it had the highest VM and FC contents, but it also was the biomass that had the lowest ash content (10.44%). The ash content of pellets produced with primary and secondary sludge from a paper mill was higher (44.33%) than the ash content found in the biomass evaluated in this study (NOSEK et al., 2017). The bark presented a higher amount of ash compared to the values in the literature for Pinus (5.21%) and Eucalyptus bark (9.03%) (LYKIDIS; KAMPERIDOU; MANTANIS, 2023; ZHENG et al., 2024).

The presence of ash in fuels is an undesirable property. It promotes the reduction of the heating value, can cause the formation of deposits, sintering, agglomeration, incrustation, slagging, corrosion and abrasion in equipment, and still becomes waste at the end of the process (BALRAJ et al., 2021). Due to the mineral composition and physical properties, the ashes can be used as a soil conditioner in the agricultural sector and also have a high potential for civil engineering industry applications, in cement production as an additive, road construction, and concrete production as an aggregate (ENNAHAL et al., 2024; TRIVEDI et al., 2016).

The main elements present in all the biomasses evaluated were carbon (C), oxygen (O) and hydrogen (H). The sum of these elements, together with nitrogen (N), sulfur (S) and ash, constitute the total composition of the four biomasses analyzed.

The bark presented the highest carbon content (50.25%), while in the PS (35.50%), SS (32.30%) and MS (37.70%), these values were lower. This fact can be explained by the loss of lignocellulosic materials from the original raw material, which is rich in carbon. Carbon, hydrogen and sulfur act positively in the release of energy during combustion, and oxygen and nitrogen act negatively, reducing the energy efficiency of the process.

These elements also contribute to the formation of undesirable nitrogenous compounds such as NO_x, N_2O and HCN (OBERNBERGER; THEK, 2004; SANTANA et al., 2020). SS showed the lowest C/N ratio. The high concentration of cells related to the addition of nitrogen, phosphorus and oxygen to the effluent during the secondary treatment to maintain the activity of microorganisms leads to a higher presence of nitrogen in this sludge and consequently a lower C/N ratio (10.83) (LOPES et al., 2018). This ratio is very important when sludge is used in the soil, because the lower the C/N ratio, the easier the decomposition of organic matter will be. Biomasses rich in carbon and low in nitrogen, such as bark (C/N: 124.47), tend to decompose more slowly in addition to promoting the immobilization of soil nitrogen (FAUBERT et al., 2016).

The chemical structure concerning the influence of the molar H/C ratio and the molar O/C ratio are presented in Figure 2.2 in the format of the Van Krevelen diagram for the four biomasses studied (PS, SS, MS and B). The sludges resemble the lignocellulosic biomass, presenting a molar H/C ratio with close values. This ratio was lower in the bark (1.29). The lower the H/C ratio, the higher the energy potential of the biomass (FARIA et al., 2021).

The bark presents a lower molar O/C ratio compared to PS and MS, justifying its high heating value (BURATTI et al., 2015; LOPES et al., 2018). However, the SS has a lower molar ratio than the bark and does not have a high heating value. This can be explained by the high ash content of this biomass.

All sludges evaluated presented high levels of moisture, resulting in a significant reduction in their heating values. The PS, SS and MS presented an HHV of 13.80 MJ kg^{-1}, 12.45 MJ kg^{-1} and 13.05 MJ kg^{-1}, respectively. When considering the moisture content of each material, these values were -1.65 MJ kg^{-1}, -2.30 MJ kg^{-1} and -0.09 MJ kg^{-1}, respectively. The negative values show that the energy released by the material is not enough to promote their drying, which affects the mass and energy balance of the system.

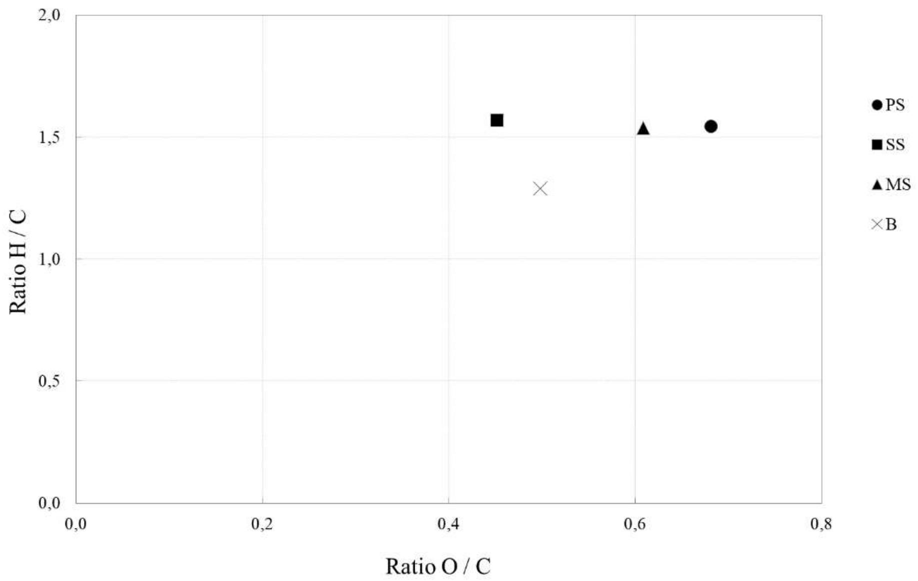

Figure 2.2 Van Krevelen diagram of the samples

The bark had the lowest moisture content among the biomasses evaluated, providing a lower reduction of its heating value. A value of 18.90 MJ kg^{-1} was found for the HHV of the bark and, when considering the moisture content, the NHV was 9.16 MJ kg^{-1}. While the PS, SS and MS reduced the heating value by more than 100%, the bark reduced it by 51.5%. This observation reiterates the need for pre-drying the sludge to ensure its energy potential; otherwise, the energy of the system will be negative.

3.2. Determination of minerals present in the biomasses

The presence of ash in biomass is one of the disadvantages of using waste for energy generation, as seen in Table 2.2. Biomasses are mainly composed of C, H, O, and ash. Ash percentages were 40.46%, 26.58%, 25.78% and 10.44% for SS, PS, MS and B, respectively. The formation of this undesirable component arises through reactions that occur during combustion between inorganic elements such as phosphorus (P), potassium (K), calcium (Ca), magnesium (Mg), sodium (Na), silicon (Si), sulfur (S) and chlorine (Cl) with other components, such as oxides, implying several problems not only in the burning equipment but also in the proper management of these ashes (ANTUNES; OLIVEIRA, 2013; BALRAJ et al., 2021).

The quantification and classification of the elements present in the biomass are important, and once these components are determined, it is possible to evaluate their behavior during burning. Table 2.3 presents the results of the inorganic element analyses. All the investigated inorganic elements were found in higher amounts in the PS, SS and MS wastes, except for iron (Fe), which was more present in the bark (0.308%). The Fe in the form of Fe_2O_3, together with other oxides such as Al_2O_3, CaO and MgO, will determine the composition of the ash and are considered to be more stable, which leads to a higher melting temperature of these ashes (LIU et al., 2018). The silica oxide (SiO_2) is also an important compound present in the ashes of bark and may lead to several problems of erosion of equipment (KUMAR et al., 2023). According to the literature the SiO_2 concentrations in the ashes of eucalyptus bark is around 5.8% (ZHENG et al., 2024), and in pellets made from pine bark is 9.8% (FILBAKK et al., 2011).

Table 2.2. Quantification of inorganics in primary sludge (PS), secondary sludge (SS), mixed sludge (MS) and bark (B)

Properties (%)	Sludge			Bark	Standard EN 14961-6 (%)
	Primary	Secondary	Mix		
	PS	SS	MS	B	
Al	5.24	3.52	5.34	0.641	-
P	0.287	0.454	0.280	0.040	-
K	0.578	2.08	0.311	0.160	-
Ca	0.498	0.802	0.540	0.210	-
Mg	0.223	0.406	0.190	0.120	-
S	0.058	0.223	0.050	0.030	≤ 0.2
Fe	0.052	0.052	0.060	0.310	-
Na	2.50	8.20	1.04	0.080	-
Cu	0.003	0.003	0.003	5×10^{-4}	≤ 0.002
Zn	0.005	0.006	0.005	0.003	≤ 0.010
Mn	0.039	0.083	0.042	0.008	-
B	0.001	0.003	6×10^{-4}	6×10^{-4}	-
Ni	4×10^{-4}	0.002	4×10^{-4}	6×10^{-5}	≤ 0.001
Cr	3×10^{-4}	3×10^{-4}	5×10^{-4}	nd	≤ 0.005
Pb	nd	Nd	nd	nd	≤ 0.001
Cd	nd	Nd	nd	nd	$\leq 5 \times 10^{-5}$
Total	**9.48**	**15.8**	**7.86**	**1.6**	-

The presence of heavy metals in the waste should be evaluated with caution. It can affect the characterization and management of the ash resulting from the burning, either for disposal in landfills or for soil application, pointing to the need for composting and leaching of heavy metals present in the biomass (TRIVEDI et al., 2016).

The DIN EN 14961-6 standard includes the specification of limit values for some inorganic components in pellet production. Pellets are densified fuels obtained from a wide variety of biomasses and can also be burned in boilers for power generation (BRAND et al., 2021; SANTANA et al., 2020). This standard was used for comparison purposes.

The standards specify the maximum concentrations of some harmful elements such as (S), copper (Cu), zinc (Zn), chromium (Cr), lead (Pb) and cadmium (Cd). The PS, SS and MS showed, respectively, Cu values of 29.8 mg kg-1, 31.4 mg kg-1 and 32.6 mg kg-1, which are higher than those recommended by the standard, which is ≤ 20 mg kg-1, while for the Ni element only the SS (18.9 mg kg-1) was higher than that recommended by the standard (≤ 10 mg kg-1). For the other elements reported in the standard, all biomasses evaluated showed values lower than those specified (DIN, 2012b).

The application of sludge to soil for agricultural purposes is a widely used form of management, as these wastes can be considered a source of many nutrients. However, the presence of heavy metals can be a limiting factor, as they can cause soil and plant contamination (FAUBERT et al., 2016; RODRIGUEZ et al., 2018). All heavy metals investigated in this study showed values lower than those specified by Brazilian National Environmental Council Resolution Nº. 498, which defines the maximum values of metals present in biosolids to be applied to the soil (CONAMA, 2020).

The HHV represents the maximum energy potential of biomass and does not take into account its moisture content. However, the presence of water negatively affects the combustion process, because this water requires energy to evaporate, which results in the reduction of the biomass heating value. When considering the presence of water in the material, one can calculate the net heating value (NHV), which will be the energy released by the biomass with a given moisture content. The NHV tends to increase with the reduction of the water present in the biomass. In Figure 2.3, it is possible to observe that the highest NHV occurs when there is the lowest moisture content since these measures are inversely correlated (SANTANA et al., 2020).

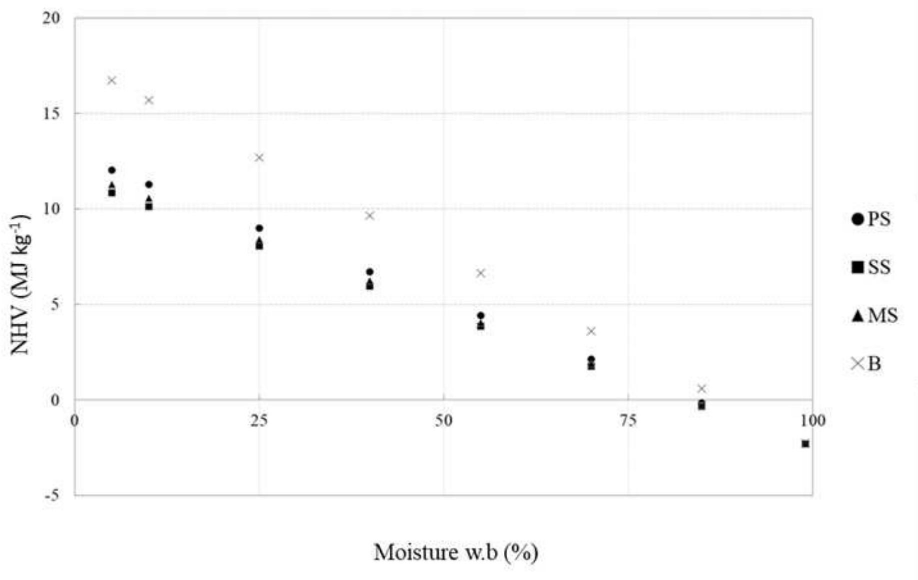

Figure 2.3. Ratio of the net heating value (NHV) to moisture w.b%

3.3. Structural Analysis

Lignin was the predominant component in all the biomasses evaluated, as can be seen in Table 2.3. The PS, SS, MS and B presented respectively 28.1%, 25.5%, 29.1% and 40.4% of total lignin. The heating value of biomass is directly related to the lignin content in it. Due to its high concentration of carbon and its complex bonds, lignin presents a high heating value (22.2-28.5 MJ kg^{-1}), which is higher than that found in cellulose, hemicellulose and even in the biomass itself (DEMIRBAS, 2017).

In this study, the bark shows the highest lignin content (40.4%) and was also the biomass that had the highest heating value, confirming this positive correlation. The lignin content varies with the species and the part of the tree, the bark has higher lignin content than the wood. Usually for eucalyptus bark, the lignin content in different species can range from 13.1% to 37% (LIMA et al., 2018). Miranda et al., 2017 reported 42.7% of lignin content in pine bark. The presence of silica in biomass can significantly influence the quantification of total lignin, often leading to an overestimation of lignin content during analysis.

The sugars in the biomass are mainly present in the holocellulose (cellulose + hemicellulose). While hemicelluloses are composed of many sugars (arabinose, galactose, glucose, xylose, mannose), cellulose is composed of only β-D-anhydroglycopyranose (glucose) units (WANG et al., 2015). A large amount of the wastewater, and consequently of the sludge generated in the WWTP, comes from washing the pulp between production stages. It contains high concentrations of lignocellulosic materials that are lost (Mounteer, 2015).

Table 2.3. Quantification of soluble, insoluble and total lignin and sugars present in PS, SS, MS and Bark

Properties (%)	Sludge			Bark
	Primary PS	Secondary SS	Mix MS	B
Soluble lignin	7.29 ± 0.071	5.60 ± 0.014	6.96 ± 0.092	2.60 ± 0.049
Insoluble lignin	20.8 ± 0.255	19.9 ± 1.01	22.2 ± 0.636	37.8 ± 4.17
Total lignin	28.1 ± 0.184	25.5 ± 1.03	29.1 ± 0.544	40.4 ± 4.12
Arabinose	0.2 -	0.2 -	0.2 -	2.4 -
Galactose	1.2 -	1.2 -	1.2 -	2.2 -
Glucose	9.3 -	3.6 -	7.9 -	15.7 -
Xylose	1.5 -	0.9 -	1.6 -	2.2 -
Mannose	0.7 ± -	1.4 -	0.7 -	2.3 -
Total	**40.1**	**32.8**	**40.7**	**65.2**

Glucose, as a component of both cellulose and hemicellulose, is the main sugar present in biomass (WANG et al., 2015). This was observed in the PS, SS, MS and B, which contained 9.3%, 3.6%, 7.9% and 15.7% of glucose, respectively. The composition of hemicelluloses will differ according to the genus of wood. Eucalyptus woods will normally have higher xylose units, while pine woods have higher mannose units. In the present study, the presence of both sugars was observed, because the mill where the sludge was obtained uses both Eucalyptus and Pinus wood.

The sugars present in the wood are associated with a lower heating value when compared to lignin or the biomass itself. Cellulose and hemicellulose have a heating value of around 17.5 MJ kg^{-1} (DEMIRBAS, 2017). During combustion, these carbohydrates are degraded at lower temperature ranges than those of lignin (CHEN et al., 2019; DEMUNER et al., 2021). Despite their lower heating value, they are considered to be rapidly released energy, which is interesting when burning them in the boiler, where fast energy generation is desired.

3.4. Thermogravimetric analysis (TG/DTG) of the biomasses

The TG/DTG curves of the PS, SS, MS and B samples are shown in Figure 2.4. The mass loss in the first temperature range of 23-146 °C for all samples is related to the evaporation of the physically adsorbed water (physisorbed water) on the surface, resulting from the dehydration of the material (KIM et al., 2022; TORRES et al., 2020).

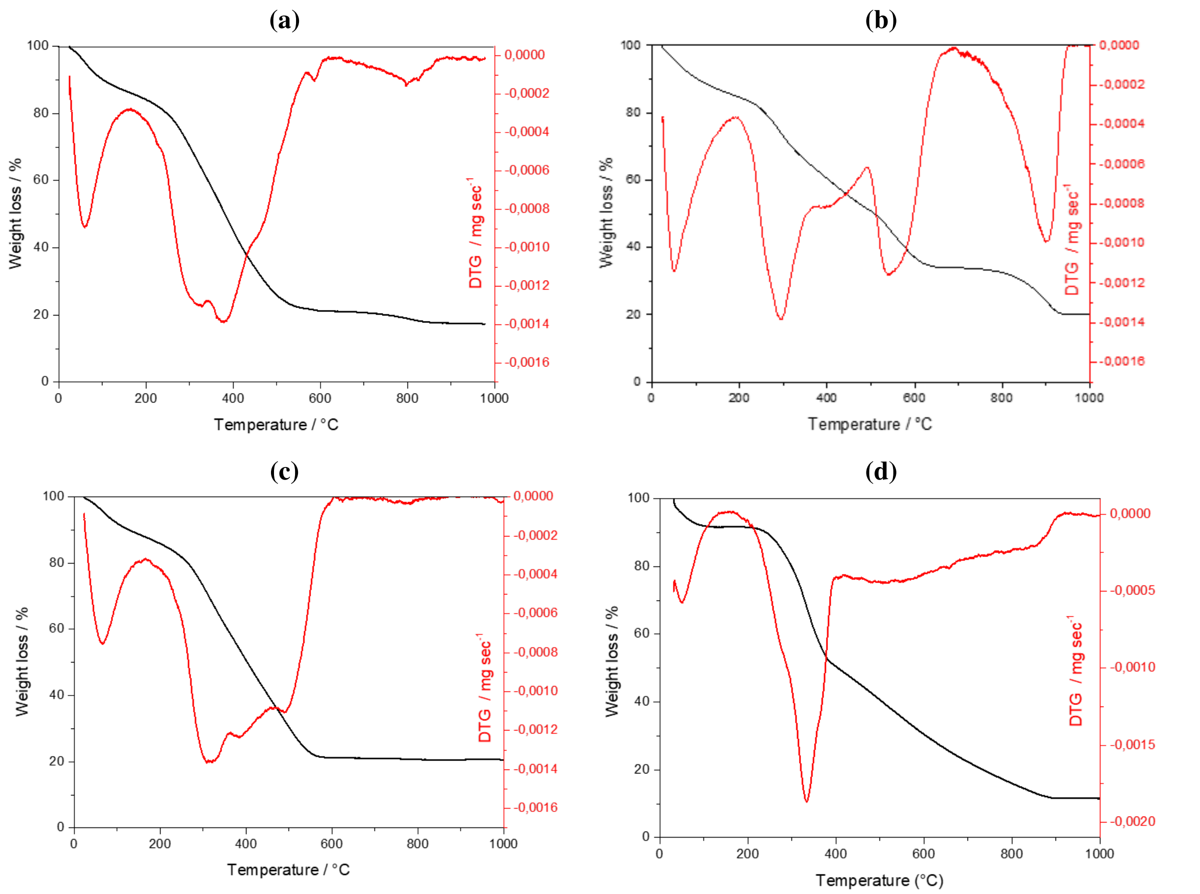

Figure 2.4. Thermogravimetric curves (TG/DTG) with synthetic nitrogen flow (50 mL min−1) and heating rate of 10 °C min−1 for primary sludge (a); secondary sludge (b); mixture (primary and secondary sludge) (c) and; bark (d)

It is observed that in the 135-648 °C range, the main mass loss of the four biomasses occurred, which is related to thermal degradation of the main material constituents (hemicellulose, cellulose and lignin). The thermal degradation of hemicellulose generally occurs first, in the temperature range of 220-315 °C, followed by cellulose (315- 400 °C), which has higher thermal stability (MALAIŠKIENĖ et al., 2018; RODRIGUES et al., 2021).

The final mass loss of the secondary sludge, in Figure 2.4 (b), in the temperature region between 711-977 °C, is characterized by the melting of salts, such as calcium and magnesium carbonate (DEMUNER et al., 2021; FANG; JIA, 2012; KIM et al., 2022; TORRES et al., 2020). The ash components may react with each other since inorganic elements are present in a high percentage of the SS (40.4%). This amount of ash is close to that of the black liquor (49.3%) from the kraft pulp and paper mill (DEMUNER et al., 2021; FANG; JIA, 2012).

4. CONCLUSIONS

After analysis of the sludge and bark samples, it can be concluded that the primary sludge presented moisture on a wet basis of 94.77%, an HHV of 13.80 MJ kg^{-1}, fixed carbon of 13.47% and an ash content of 26.58%. The secondary sludge had a wet basis moisture level of 98.99%, an HHV value of 12.45 MJ kg^{-1}, a fixed carbon level of 10.18% and an ash content of 40.46%. The mixture of primary and secondary sludge collected at the end of the dewatering press in the plant showed a moisture level on a wet basis of 93.68%, HHV of 13.05 MJ kg^{-1}, a fixed carbon level equal to 14.54% and an ash content of 25.78%.

The use of primary, secondary and mixed sludge for energy generation should only be considered after partial drying since the presence of water promotes a high reduction in the NHV. The bark presented a moisture level on a wet basis of 42.49%, HHV equal to 18.90 MJ kg^{-1}, a fixed carbon level of 22.59%, and an ash content of 10.44%. The use of bark for energy generation is feasible since this biomass has a high HHV and lower moisture content when compared to the sludges. However, to increase the NHV, a previous drying stage can be performed. The high ash content of all these biomasses, especially of the sludges, is a negative aspect of their energy potential.

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CHAPTER 3

TURNING CHALLENGES INTO OPPORTUNITIES: PELLETS PRODUCTION FROM HIGH-MOISTURE PULP MILL SLUDGE

TURNING CHALLENGES INTO OPPORTUNITIES: PELLETS PRODUCTION FROM HIGH-MOISTURE PULP MILL SLUDGE

Abstract

The pulp and paper industry plays an important role in the global economy. To ensure process sustainability, improvement in waste management is necessary. Sludge is the main solid waste generated within the mill. The effluent treatment plant usually generates primary (PS) and secondary sludge (SS). PS is rich in cellulose fibers and SS is rich in bacterial cells (biosludge). SS is more difficult to dewater and is often mixed with the PS to help drainage. Currently, the main way for disposal is landfilling or incineration without any pre-treatment. Seeking a more circular economy and maximum use of all residues (co-products), novel processes are required. An important challenge for sludge valorization is the removal of its water content. Pelletizing is a simple process that allows the standardization and increase of energy density of the biomass. This work aimed to produce pellets using various blends of PS and SS generated at a kraft pulp mill (100% PS, 75% PS, 50% PS, 25% PS, 0% PS) and testing different moisture content, on a wet basis (10%, 25%, 40%). The sludge was subject to elemental characterization, proximate analysis, heating value, thermal degradation evaluation, and ash melting behavior analysis. The chemical characterization of the sludge was performed to evaluate its energy potential and its impact on the boiler. The increase of secondary sludge increased the pellets ash content. The ash melting temperature of the sludges was higher than the typical operating temperatures of boilers. The sludge showed a high higher heating value (16.2 - 17.4 $MJ.kg^{-1}$). The pellets produced using the blend of 75% PS + 25% SS with a moisture content of 25% presented low fines generation (0.043%) and high durability (99.3%). The pellets produced with 100% SS at 40% moisture showed the lowest resistance. The pelletizing process has proven to be a suitable alternative for managing pulp mill sludge.

Keywords: Waste management, kraft pulp mill, Sludge treatment, Pellets, Sustainable energy.

1. INTRODUCTION

The circular economy is a concept that is gaining increasing prominence in the industrial sector (ZAMBARE; CHRISTOPHER, 2020). Reducing waste generation by transforming it into co-products allows materials to be recovered within the production chain, making the processes more sustainable (LEPPÄNEN et al., 2020; NEVES; MARQUES, 2022).

In an increasingly competitive market, industries face mounting pressure to align their practices with the principles of sustainable development. One critical strategy for achieving this is the optimal utilization of by-products, which are often still classified as waste. By transforming these by-products into valuable resources, companies can reduce environmental impacts, enhance resource efficiency, and meet sustainability targets, all while gaining a competitive edge in the marketplace. Embracing innovative approaches to by-product management is not only essential for compliance with sustainable development goals but also represents a significant opportunity for cost savings and new revenue streams (LEPPÄNEN et al., 2020).

The pulp and paper industry is important in the global market, supplying a variety of products that can be used for different purposes (HAILE et al., 2021). The kraft process stands as the predominant method for pulp production worldwide. This widely adopted process utilizes sodium hydroxide (NaOH) and sodium sulfide (Na_2S) as its primary chemical reagents. The success of the kraft process is attributed to several key advantages. It demonstrates a high tolerance for varying raw material qualities, enabling the use of diverse wood types and non-wood fibers. The pulp produced through the kraft process is known for its superior strength and quality (HENRIKSSON; GERMGÅRD; LINDSTRÖM, 2024).

The main raw material, wood, used for cellulose production is renewable and can be extracted from various lignocellulosic materials, coniferous and hardwood trees are the most common in the process (HAILE et al., 2021; RAMOS et al., 2022). However, pulp and paper mills are still responsible for high energy, water, and chemical consumption, as well as the high generation of solid and liquid wastes, causing a significant environmental impact (GOTTUMUKKALA et al., 2016; RAMOS et al., 2022).

The high quantities of solid waste generated in the process lead to great concern about its management since industrial landfills have become a restricted practice by several environmental authorities (JARIA et al., 2017; LEPPÄNEN et al., 2020; MOHAMMADI et al., 2019). The primary and secondary sludge from the effluent treatment plant (ETP) represents the largest portion of solid waste generated at a mill. It is estimated that for each ton of pulp produced, approximately 40-60 kg of primary sludge and 10-15 kg of secondary sludge are generated (LOPES et al., 2018; RODRIGUES et al., 2021). The global production of pulp and paper mill sludge was around 417.3 million tons in 2021 (POCHYBA et al., 2023).

Adequate management of sludge is still a challenge for the mills. The most common methods of sludge disposal are landfill and incineration, practices that are considered questionable due to their major environmental, economic, and social impacts (GOTTUMUKKALA et al., 2016; HAILE et al., 2021; NJOKU; EDOKPAYI; ODIYO, 2019). The valorization of sludge generated in effluent treatment plants is for the progressive sustainable development of industries, as it is in line with the guidelines of the circular economy (HAILE et al., 2021).

Primary sludge, generated during the primary (physical-chemical) treatment of the effluent, is mainly composed of fibers lost in the process and has a consistency of 60-80% (FAHIM et al., 2018; FAUBERT et al., 2016; ZAMBARE; CHRISTOPHER, 2020). Secondary sludge originates from the secondary (biological) effluent treatment process, consisting mainly of organic matter and minerals (ZAMBARE; CHRISTOPHER, 2020). Due to the low consistency (98-99.5%), secondary sludge is often mixed with the primary sludge to facilitate dewatering (BAJPAI, 2015; LOPES et al., 2018).

Sludge has a high content of lignocellulosic materials and a similar heating value to biomass already used as fuel (11.1-22.10 $MJ.kg^{-1}$) in biomass boilers (OLADEJO et al., 2019; RODRIGUES et al., 2021). These factors contribute to its potential for energy generation, although, the high moisture content and heterogeneity play a negative role (RODRIGUES et al., 2024).

The demand to replace fossil fuels with renewable energy sources is growing at an exponential rate (OLUJOBI et al., 2022), especially after the new targets set at the United Nations Climate Change Conference (COP 29) in 2024. In this context, sludge has emerged as an attractive alternative for the production of renewable biofuels. Nevertheless, strategies are still needed to overcome the undesirable characteristics mentioned above.

Pellets are densified granular fuels produced from different agroforestry biomasses (BRAND et al., 2021). Pellets are produced by compacting the raw material, which allows a higher level of homogeneity and an increase in energy density (MOSTAFA et al., 2019). The pelletizing of sludge can be an interesting option to overcome the main problems that can affect energy recovery (RODRIGUES et al., 2021).

The production of pellets using blends of wood sawdust combined with primary and secondary pulp mill sludge has demonstrated promising results in fuel performance. The inclusion of sludge in the pellet composition significantly increased the ash melting temperature of the resulting pellets. This enhancement makes them particularly suitable for use as a fuel in co-combustion processes alongside other fuels with lower ash melting temperatures (NOSEK et al., 2017).

Previous research has demonstrated the successful production of pellets with excellent physical and mechanical properties using primary sludge from a kraft pulp mill. The sludge, characterized by a moisture content of 31%, proved to be a viable feedstock for pellet manufacturing (RODRIGUES et al., 2021).

Despite advancements in pellet production research, a significant gap remains in the literature regarding the impact of blending primary and secondary sludge on pellet quality. Additionally, the influence of varying moisture content in these sludges on the physical, mechanical, and combustion properties of the pellets has yet to be thoroughly explored. Understanding these factors is crucial for optimizing pellet production processes and ensuring consistent fuel quality

This work aimed to produce densified biofuels (pellets) using different moistures and blends of primary and secondary sludge from a bleached kraft pulp mill. The chemical composition of the sludge was also assessed, predicting the possible impacts on the boiler.

2. MATERIALS AND METHODS

2.1. Material Collection

The primary and secondary sludge was collected from the effluent treatment plant (ETP) of a bleached kraft pulp mill located in Brazil with an annual production of 1.2 million tons of air-dried pulp. The ETP consists of a primary clarifier followed by an activated sludge biological process, where approximately 40 kg of primary sludge and 15 kg of secondary sludge are generated for every ton of air-dried pulp produced.

A sample of 50 kg of primary sludge was collected after the screw press with a consistency of 42% and 100 kg of secondary sludge with a consistency of 21%. The collected materials were stored in a cold room at a temperature of 5 ± 2 °C. The material was collected in April 2021.

2.2. Sludge characterization

2.2.1. Sample preparation

After passing through the press, the primary sludge (PS) has a moisture content of around 60-70% and the secondary sludge (SS) 80-90%. Before adjusting the moisture content, the primary and secondary sludge was mixed using different blends: 100% PS + 0% SS; 75% PS + 25% SS; 50% PS + 50% SS; 25% PS + 75% SS; 0% PS + 100% SS.

The blends were dried in the open air. To define the best operating conditions for pellet production in line with lower drying costs, tests were carried out on the input biomass with different moisture contents: 40%, 25% and 10%. To achieve the desired moisture content for pellet production, the sludges were spread on a ground surface protected by a tarp to prevent contamination. The moisture content was monitored daily using standard testing methods. Once the moisture content reached the target level for the specific treatment, approximately 3 kg of sludge from each blend was collected. The collected sludge samples were sealed in plastic bags to stabilize moisture levels before further processing. This preparation ensured uniformity in the material. Following stabilization, the sludge was used to manufacture pellets, enabling the assessment of the effects of moisture content and sludge blends on pellet properties.

2.2.2. Pelletizing process and drying

The pellets were produced in a laboratory pelletizing press with a horizontal rotating matrix Amandus Kahl, model 14-175, production capacity of 40 $kg.h^{-1}$. The dimensions of the compression press are as follows length of 30 mm, diameter of 7.5 mm at the inlet and 6.3 mm at the outlet. The knife that defines the size of the pellets is 20 mm from the matrix. 2 kg of pellets were produced for each treatment.

The moisture content of the pellets was checked at the entrance and exit of the pelletizer. After pelletizing, the pellets were placed on sieves and conditioned at room temperature until the moisture content stabilized.

2.3. Pellets characterization

2.3.1. Physical characterization

Bulk density (kg/m⁻³) was determined using the same standard mentioned above for sludge EN 15103 (DIN, 2010a). The moisture content was determined according to EN 14774-2 (DIN, 2010d). To determine the dimensions of the pellets, diameter and length, a digital caliper was used, and measurements were taken following the EN 16127 standard (DIN, 2012b).

2.3.2. Chemical characterization

The chemical characterization of the pellets was conducted following the same standards and methodologies detailed in Section 2.2.2 for the chemical characterization of the sludge.

2.3.3. Mechanical properties

The fines content (particles smaller than 3.15 mm) and the mechanical durability of the pellets were determined following standard EN 15210-1 (DIN, 2010e), using the Ligno-Tester equipment, Holmen®, which simulates the behavior of the pellets during transport and handling operations. The fines were generated using a controlled air flow with a pressure of 30 mbar and a residence time of 30 seconds. The samples free of fines went on to the mechanical durability test with an air flow of 70 mbar and a residence time of 60 seconds.

2.4. Experimental design

The experiment was carried out using a completely randomized design (CRD), arranged in a factorial design with two factors: five blends (100% PS + 0% SS; 75% PS + 25% SS, 50% PS + 50% SS; 25% PS + 75% SS and 0% PS + 100% SS) and three moisture content (10%, 25%, 40%) in three repetitions, totaling 75 sampling units.

Before the variance analysis. the data was submitted to the Lilliefors and Cochran tests to test the normality and homogeneity of the variances, respectively. An analysis of variance was carried out using the F test, with a significance level of 5% to compare the treatments, and when a significant effect was observed between the treatments (blends and moisture content), the Tukey test was used at 5% significance. Statistical analyses were performed using RStudio (R Core Team, 2020) software.

3. RESULTS AND DISCUSSIONS

3.1.Primary and secondary sludge properties

The chemical and physical properties of primary sludge (PS) and secondary sludge (SS) are shown in Table 3.1. The higher heating values (HHV) of PS and SS were 17.4 and 16.2 $MJ.kg^{-1}$ respectively, similar to the values reported by Torres et al., 2023 and higher than the HHV found in the literature for municipal sludge, which was in the range of 11.69-16.47 $MJ.kg^{-1}$ (GHODKE et al., 2021).

The main problem for sludge valorization is the water presence (RODRIGUES et al., 2021; ZAMBARE; CHRISTOPHER, 2020). The PS and SS presented 58.1 % and 79.0% moisture content, these values were lower than those found in the literature, which reported sludge moisture content ranging from 93% to 98% (ZAMBARE; CHRISTOPHER, 2020). The high moisture content negatively influences energy conversion, reducing the NHV of the material, increasing emissions, and affecting the energy balance of the process (RODRIGUES et al., 2021).

The carbon (C) and hydrogen (H) content of typical biomass is around 45% and 4.0-6.5% respectively (CAI et al., 2017). The primary sludge presented a C content of 45.3% and an H content of 6.25% while the secondary sludge showed values of 42.0% and 5.2% for the same properties. These values highlight the fact that pulp mill sludges have a similar composition to typical biomass. The C and H content of Eucalyptus species, the main wood used for cellulose production in Brazil, range from 43.84% - 54.47% and 5.47% - 7.40% respectively (EICHLER et al., 2017).

The bulk density of PS and SS was respectively 300 $kg.m^{-3}$ and 837 $kg.m^{-3}$. Usually, agricultural waste presents bulk density ranging from 80 to 100 $kg.m^{-3}$ (PRADHAN; MAHAJANI; ARORA, 2018). The higher bulk density in the sludge is due to the high moisture content since the water has a density of 975 $kg.m^{-3}$ (NAJAFI-MARGHMALEKI; KHOSRAVI-NIKOU; BARATI-HAROONI, 2016; RODRIGUES et al., 2021).

Table 3.1. Average values and standard deviation of primary and secondary sludge characterization

Properties	Sludge					
	Primary (PS)			Secondary (SS)		
HHV ($MJ.kg^{-1}$)	17.4	±	(0.064)	16.2	±	(0.141)
C (%)	45.3	±	(0.424)	42.0	±	(0.424)
H (%)	6.25	±	(0.057)	5.2	±	(0.049)
N (%)	0.13	±	(0.021)	5.27	±	(0.460)
S (%)	0.196	±	(0.006)	0.699	±	(0.000)
O (%)	46.4	±	(0.455)	24.3	±	(0.933)
Ashes (%)	1.7	±	(0.211)	22.5	±	(0.376)
Bulk density ($kg.m^{-3}$)	300	±	4.543	837	±	3.321

3.2.Thermal degradation

The thermal degradation of the PS and SS can be observed in Figure 3.1 in the thermogravimetric curve (TG) and its derivative (DTG). The mass losses observed at temperatures lower than 100 ºC usually correspond to the evaporation of the water present in the biomass and were not included in this work as they have no scientific relevance to the results (JIANG et al., 2014; KIM et al., 2022).

The initial thermal degradation temperature of the PS (312 ºC) was 18 ºC higher than that of the SS (294 ºC), while the final degradation temperature of the SS was 574 ºC, 111 ºC higher than the final degradation temperature of the PS (463 ºC). The sludge mass loss peaks correspond to the degradation of organic components such as hydrocarbons (200 - 370 ºC) and aliphatic components (400 - 600 ºC) (GAO et al., 2020; MALAIŠKIENĖ et al., 2018).

Primary and secondary sludge from pulp and paper mills are rich in fibrous material, mainly containing cellulose, hemicellulose and lignin (LOPES et al., 2018; RODRIGUES et al., 2021). Hemicellulose is thermally unstable and has a lower degradation temperature (225 - 325 ºC) compared to cellulose (315 - 400 ºC) and lignin (RODRIGUES et al., 2021; SALDARRIAGA et al., 2015). Lignin degrades over a wide temperature range, reaching up to 900 ºC, and is also the most thermally stable organic structural component of biomass (SALDARRIAGA et al., 2015; YANG et al., 2007; YARAS et al., 2021).

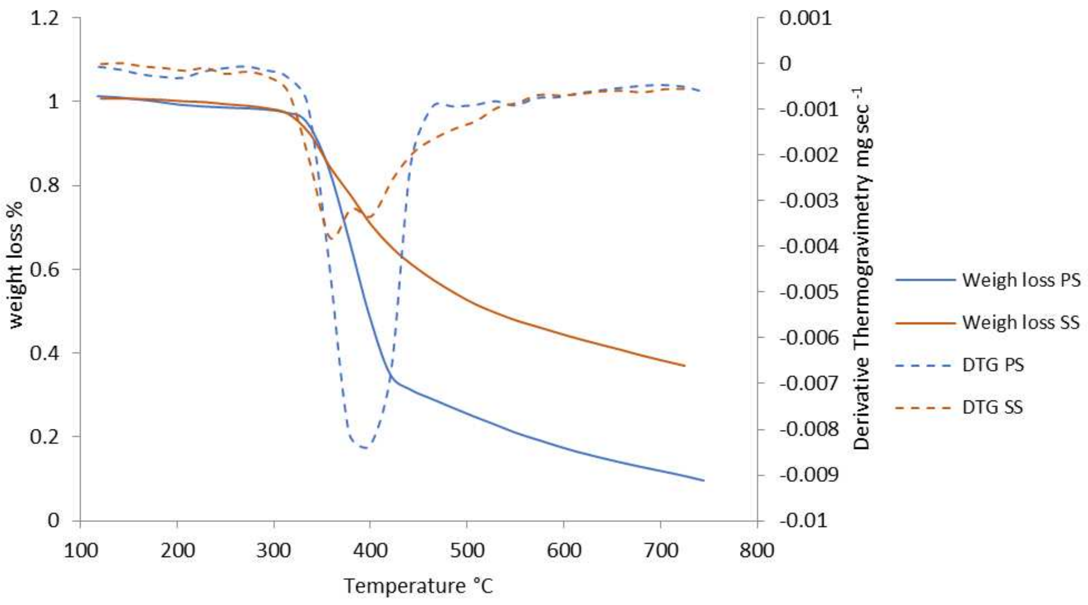

Figure 3.1. Thermogravimetric curves (TG/DTG) of primary and secondary sludge generated in a bleached kraft pulp mill

The mass loss of primary sludge (PS) and secondary sludge (SS), as shown in Table 3.2, was 49% and 20%, respectively, within the temperature range of 300 – 400 °C. This significant difference suggests a higher cellulose content in PS compared to SS. Between 400 °C and 500 °C, both sludges showed similar mass degradation (23%); however, the residual organic mass was higher in SS, indicative of its higher lignin content. The remaining mass at temperatures above 500 ºC may correspond to residual lignin and the presence of $CaCO_3$ and ashes (KUBONOVA et al., 2021). The lower mass loss in temperatures ranging from 200 - 300 °C is due to the lower hemicellulose content in the sludges.

Primary sludge from the pulp mills contains higher levels of cellulose (81%) and hemicellulose (12%) and only 5.7% lignin, while secondary sludge will have high levels of cellulose (31.2%) and lignin (30.5%) and a low concentration of hemicellulose (5.5%) (LOPES et al., 2018). This observation highlights the reduced degradation of secondary sludge at lower temperatures, which can be attributed to its higher content of lignin and ash.

The residual byproducts of pellet combustion consist primarily of inorganic compounds, with a smaller fraction consisting of carbon. This residual material has potential applications in soil management, including its use as a conditioner, a fertilizer, and as a contributor to soil carbon stock projects.

Table 3.2. Mass loss (%) of primary and secondary sludge across different temperature ranges

Sample	Mass loss (%)				Residual mass	
	100 - 200 ºC	200 - 300 ºC	300 - 400 ºC	400 - 500 ºC	Organic	Inorganic
PS	1.3	0.7	49	23	24.3	1.7
SS	0.3	1.7	20	23	32.5	22.5

3.3.Inorganic content and ash behavior

The inorganic material in the biomass forms the ash. Some compounds such as nitrogen (N), phosphorus (P), potassium (K), calcium (Ca), magnesium (Mg), sodium (Na), silicium (Si), sulfur (S) and chlorine (Cl) react during combustion forming some oxides that result in the generation of pollutants and the formation of deposits in the boiler increasing the maintenance and operation costs (ANTUNES; OLIVEIRA, 2013; WANG et al., 2022).

The inorganic elements on biomass are present in different forms, Na and K are usually in the form of ionic salts, Mg and Ca make large extent complexes with organic counter-ions, Fe, Cu, Ni, Cd, Cr, Co, Mn, Zn usually is found as small crystal structure (< 2 μm), Al and Pb are present in biomass in different forms and the non-metal elements P and S can be in organic and inorganic form, like sulfate, organic sulfur, proteins, and amino acids (LEIJENHORST et al., 2016).

The inorganic content and composition depend on several factors, including the raw material, the pulping method, the final product, and the effluent treatment technology (BAJPAI, 2015; KAUR; BHARDWAJ; LOHCHAB, 2019). The quantification of some inorganic elements in the sludge is shown in Table 3.3. The sludge of this study is from a kraft pulp mill, the main reagents for pulp production are sodium hydroxide (NaOH) and sodium sulfide (Na_2S). The content of Na and S in the PS were respectively 900 and 1300 mg kg^{-1} and in the SS 1700 and 11800 mg kg^{-1}. Other agroindustrial wastes such as wheat straw and barley straw had Na content of 37 and 661 mg kg^{-1} and an S content of 570 mg kg^{-1} and 983 mg kg^{-1} respectively (LEIJENHORST et al., 2016).

The European standard DIN EN 14961-6 presents the specification of limit values for some inorganic elements on pellets. All the values reported in this study were lower than the limit amount specified in the standard, except for S content in the SS, which was higher than 2000 mg kg^{-1}.

Table 3.3. Quantification of inorganics in primary sludge (PS), secondary sludge (SS), and in the blend 75% PS + 25% secondary sludge

Properties (%)		**Sludge**			**Standard EN 14961-6**
		Primary	**Secondary**	**Mix**	
		PS	**SS**	**MS**	
Al	mg kg^{-1}	460	9430	2720	ns
P	mg kg^{-1}	11200	58000	22900	ns
K	mg kg^{-1}	100	2900	800	ns
Ca	mg kg^{-1}	2200	39200	11500	ns
Mg	mg kg^{-1}	100	1400	430	ns
S	mg kg^{-1}	1300	11800	3930	≤ 2000
Fe	mg kg^{-1}	700	9900	3010	ns
Na	mg kg^{-1}	900	1700	1080	ns
Cu	mg kg^{-1}	5.1×10^{-4}	2.6×10^{-3}	1.0×10^{-3}	≤ 20
Zn	mg kg^{-1}	3.0×10^{-3}	0.01	4.1×10^{-3}	≤ 100
Mn	mg kg^{-1}	3.0×10^{-3}	0.02	7.7×10^{-3}	ns
B	mg kg^{-1}	0.00	8.0×10^{-4}	1.9×10^{-4}	ns
Ni	mg kg^{-1}	2.3×10^{-3}	2.2×10^{-3}	2.3×10^{-3}	≤ 10
Cr	mg kg^{-1}	5.8×10^{-3}	5.8×10^{-3}	5.8×10^{-3}	≤ 50
Pb	mg kg^{-1}	nd	Nd	nd	≤ 10
Cd	mg kg^{-1}	nd	Nd	nd	≤ 0.5
Total	**%**	**1.72**	**14.2**	**4.83**	ns

ns: Not specified in the standard
nd - Not detected

The determination of the fusibility temperature of the ash and the formation of slag helps to adapt the boiler for the use of different fuels in order to preserve the integrity of the equipment, reducing the need for maintenance and cleaning shutdowns (DODDAPANENI; PÄRN; KIKAS, 2022; LI et al., 2013).

The composition of the mineral phase of the fuel is the main property that influences the melting temperature. The oxides and alkaline salts form slag, deposition, and agglomeration of ash in the equipment leading to various problems that reduce the thermal efficiency of the boiler (LIU et al., 2018a; REINMÖLLER et al., 2015).

Figure 3.2 shows the initial deformation temperature (IDT), spherical temperature (ST), hemispherical temperature (HT), and fluid temperature (FT) of the ashes in the PS and SS, which ranged from 1185 to 1290 °C. The IDT, which marks the onset of ash melting, was observed to be 1185 °C for PS and 1259 °C for SS. These values signify the temperatures at which the ash samples begin to soften and lose their structural integrity. The values of IDT found in this study were higher than those reported in the literature for agro-industrial waste (590 – 730 °C) (NIU et al., 2010), and for sludge from the chemical-thermo-mechanical pulping process (741 °C) (DODDAPANENI; PÄRN; KIKAS, 2022).

The melting temperature of the SS ash was higher than that of the PS. One of the possible explanations is due to the higher Fe, Al, Ca, and Mg content of the SS compared to the PS. These compounds form oxides such as Fe_2O_3, Al_2O_3, CaO, MgO constituting the ash structure, promoting greater stability and inducing a higher melting temperature (LIU et al., 2018b).

Fluidized bed boilers, commonly used in large industrial applications, typically operate within a temperature range of 800–900°C (YLINIEMI et al., 2015). In contrast, the synthesis and deformation of ash from the sludge analyzed in this study occur at significantly higher temperatures, exceeding 1185°C. This thermal disparity indicates that the ash remains stable and unlikely to melt or cause slagging under the typical operating conditions of these boilers, ensuring smoother operation and reducing the risk of fouling or equipment damage.

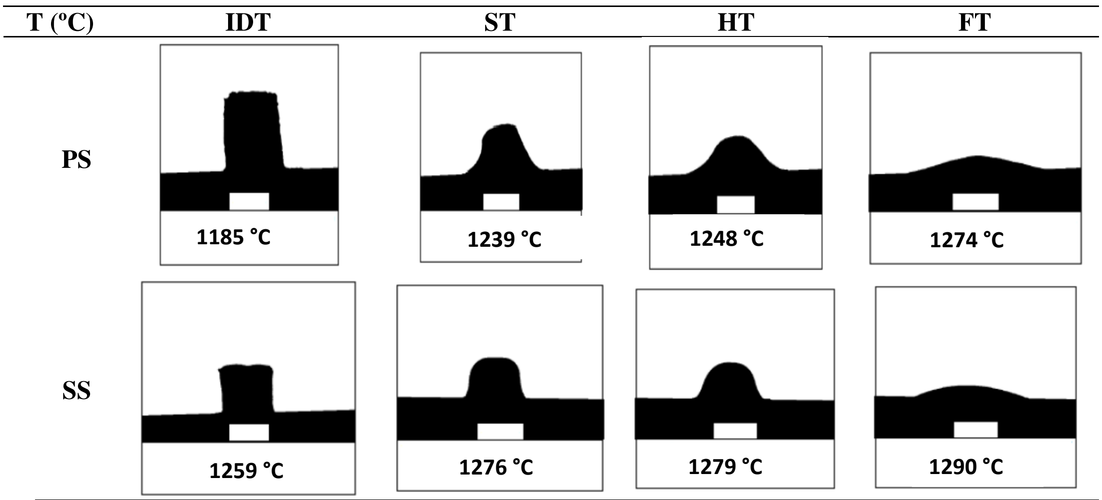

Figure 3.2. Ash melting temperature of primary (PS) and secondary (SS) sludge. IDT = initial deformation temperature, ST = spherical temperature, HT = hemispherical temperature, FT = fluid temperature

3.4.Proximate analyses and chemical composition of the pellets

Different blends of PS and SS were used to produce the pellets. Due to the variation in the sludge composition, the pellets produced had different properties. Table 3.4 shows the proximate analyses and higher heating value (HHV) of the sludge pellets.

Table 3.4. Proximate analyses and higher heating value (HHV) of the sludge pellets

Blends	VM (%)	Ashes (%)	FC (%)	MV:FC	HHV(MJ.kg^{-1})
100% PS + 0% SS	86.2 A	1.7 A	12.1 A	8.26 A	17.4 A
75% PS + 25% SS	84.0 B	5.3 B	11.5 A	8.16 A	17.1 A
50% PS + 50% SS	78.2 C	10.3 C	10.7 B	8.05 A	16.8 A
25% PS + 75% SS	74.0 D	17.4 D	8.6 C	7.93 A	16.5 A
0% PS + 100% SS	68.7 E	22.5 E	8.5 C	7.80 A	16.2 A

*The letters (A-E) in the same column represent statistical comparisons using the Tukey test (5% significance), while different letters indicate significant differences between the values

The higher heating value (HHV) represents the maximum energetic potential of a fuel (XING et al., 2019). Increasing SS in the pellets production, reduced the HHV. The HHV found in this work had similar values to those observed in other biomass pellets production, such as sugar cane bagasse (17.46 MJ.kg^{-1}), soybean wastes (16.77 MJ.kg^{-1}) (SCATOLINO et al., 2018), and elephant grass (18.51 MJ.kg^{-1}) (GARCIA et al., 2019).

The ash content presented in lignocellulosic biomass is usually between 1% and 5% (GARCIA-MARAVER et al., 2015; SCATOLINO et al., 2018). The blend of 75% PS + 25% SS showed an ash content of 5.3%, while the mixtures that added more than 25% secondary sludge presented an ash content higher than 10%. The international standard for non-woody pellets DIN EN 14961-2 establishes ash content lower than 10% (DIN, 2012c).

Materials with high values for volatile matter (VM) tend to burn easily, but also faster, causing greater heat loss when not managed properly (GARCIA-MARAVER et al., 2015). Agricultural waste usually has high VM contents. In this study, the VM of the pellets were between 68.7 % and 86.2%, which is similar to those found in other pellets made from elephant grass (79.09%), olive pruning (79.8%) (GARCIA-MARAVER et al., 2015) and coffee husks (73.15%) (DE SOUZA et al., 2020).

Fixed carbon (FC), unlike VM, makes the pellets more thermally stable and, as a result, slower and more controlled combustion (DA SILVA et al., 2020). The pellets produced using sludge showed values for FC between 8.5% and 12.1%. The pellets produced using different blends of elephant grass, sugarcane bagasse, and eucalyptus wood presented values for FC ranging from 11.75% to 15.05% (DA SILVA et al., 2020).

VM and fixed carbon FC have a positive influence on the HHV, while ashes content reduces the energetic potential of the fuel causing a lower HHV (RODRIGUES et al., 2021; SCATOLINO et al., 2018). The pellets produced using only primary sludge (100% PS + 0% SS) presented the highest value for VM (86.2%) and lowest ash content (1.7%), which resulted in a higher HHV (17.4 MJ.kg^{-1}). Pellets produced with 100% secondary sludge (0% PS + 100% SS) had 22.5% ash content, and 68.7% VM resulting in the lowest HHV (16.5 MJ.kg^{-1}).

The VM/FC ratio shows theoretical information about the fuel burning efficiency (DODDAPANENI; PÄRN; KIKAS, 2022). The higher the primary sludge content in the pellets, the higher the VM:FC ratio. A higher VM:FC ratio decreases the ignition temperature and increases the burning intensity (GARCÍA et al., 2013; SCATOLINO et al., 2018). The increase of SS increases ash content, resulting in a decrease in the HHV, FC and VM of the pellets.

The fuel quality also can be indicated by the oxygen-to-carbon (O:C) and hydrogen-to-carbon atomic ratio (H:C). Van Kreveken Diagram (Figure 3.3) is a tool that allows the visualization of the ratios O:C and H:C based on the chemical composition of the material (DODDAPANENI; PÄRN; KIKAS, 2022; TORRES et al., 2023). A lower O:C and H:C ratio usually is correlated with a higher heating value (DODDAPANENI; PÄRN; KIKAS, 2022). The increase of SS in the pellets reduces the O:C and H:C atomic ratios, because of the lower oxygen and hydrogen content in the SS (24.34% O and 5.20% H) compared to the primary sludge (46.24% O and 6.25% H).

Although the increase of secondary sludge in pellets resulted in a lower O:C and H:C ratio, pellets with higher secondary sludge proportion showed a lower HHV. This happens because the energetic potential of fuel depends on more than one property. Other factors like ash content, VM, and FC also influence the heating value (SCATOLINO et al., 2018; TORRES et al., 2023).

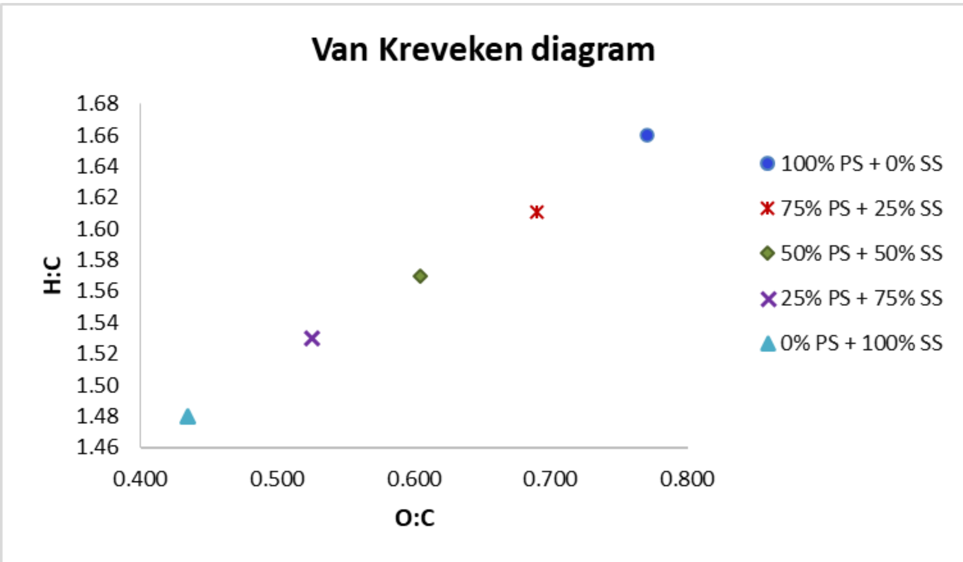

Figure 3.3. Van Krevelen Diagram of the different PS and SS blends

3.5. Physical properties of the pellets

3.5.1. Moisture content and dimensions

The pellets formed with PS and SS are shown in Figure 3.4, all treatments were able to form pellets. The main visual aspect is the color of the pellets which becomes darker with the increase of SS, due to the higher content of cellular materials from the microorganisms that usually is dark or brown (TORRES et al., 2023).

Table 3.5 shows the moisture content of the pellets after drying at room temperature. Pellets produced with higher moisture presented higher final moisture. The pelletizing process reduces by 13-14% the initial water content of the biomass (TUMULURU, 2018). According to the literature higher the moisture content in the pellets higher the diameter, due to the presence of water in the empty spaces of the pellets that disrupts bonds formed during the process (KAMPERIDOU, 2022; SCATOLINO et al., 2018).

Figure 3.4. Pellets produced with different blends of primary (PS) and secondary sludge (SS) at 10%, 25% and 40% moisture content

In this work, the moisture had the opposite effect on the diameter. The pellets with a lower final moisture content, presented higher diameter, except in the blends 100% PS + 0%

SS and 0% PS + 100% SS when increased pelletizing process moisture from 25% to 40%. The final moisture content was 10.2% and 23.0% and the diameter increased by 6.5% and 22.2% respectively. The opposite effect on diameter, when the moisture was changed from 10% to 25%, can be due to the increase of contact between the particles, since the water on biomass reduces the glass transition temperature, and allows stronger bonds (TUMULURU, 2018).

Table 3.5. Final moisture content of pellets after room-temperature drying (%)

Blends	**Initial moisture (%)**		
	10	**25**	**40**
100% PS + 0% SS	10.7 dA	10.2 aA	23.0 cB
75% PS + 25% SS	8.7 bA	10.1 aB	20.6 aC
50% PS + 50% SS	9.6 cA	12.4 bB	28.3 dC
25% PS + 75% SS	7.9 aA	13.2 cB	22.2 bC
0% PS + 100% SS	9.2 bcA	12.0 bB	37.1 eC

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

The lengths of the pellets, presented in Table 3.6, produced with 10% and 25% moisture content were not statistically different, except the pellets produced using just secondary sludge at 25% moisture content, which presented a higher length (22.1 mm). The diameters of the pellets (Table 3.7) ranged between 5.3 and 6.5 mm. Pellets produced with a moisture content of 40%, using blends of 100% PS + 0% SS and 0% PS + 100% SS, exhibited a larger diameter compared to those from other treatments. This increase in diameter can be attributed to the presence of water, which disrupts the bonds between particles, leading to pellet swelling and an expansion in size (UNGUREANU et al., 2018).

Pellets with more uniform dimensions tend to burn more homogeneously, which also improves their feeding efficiency into the ignition system (LIU; QUEK; BALASUBRAMANIAN, 2014). According to the standard DIN EN 14961-6 the values stipulated are 3.15 to 40 mm for length and 6 ± 1 mm for diameter (DIN, 2012b).

Table 3.6. Length **(mm)** of the pellets produced using PS and SS blends

Blends	**Moisture (%)**		
	10	**25**	**40**
100% PS + 0% SS	18.6 aB	19.0 aB	15.8 bA
75% PS + 25% SS	19.9 abB	19.9 aB	17.8 cA
50% PS + 50% SS	19.4 abAB	19.9 aB	18.4 cA
25% PS + 75% SS	19.9 abB	18.8 aAB	18.2 cA
0% PS + 100% SS	20.5 bB	22.1 bC	10.0 aA

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

Table 3.7. Diameter of the pellets **(mm)** produced using PS and SS blends

Blends	**Moisture (%)**		
	10	**25**	**40**
100% PS + 0% SS	6.1 aB	5.8 bA	6.2 cB
75% PS + 25% SS	6.0 aB	5.8 bA	5.8 bA
50% PS + 50% SS	5.9 aC	5.7 abB	5.4 aA
25% PS + 75% SS	5.9 aC	5.6 aB	5.3 aA
0% PS + 100% SS	6.1 aB	5.6 aA	7.2 dC

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

3.5.2. Bulk density and energy density

The bulk density of the pellets ranged from 304 to 731 kg.m^{-3} (Table 3.8). The standard DIN EN 14961-6 establishes a minimum bulk density of 600 kg.m-^{3} (DIN, 2012c). Pellets produced with lignocellulosic materials vary from 435 to 730 kg m^{-3} (DA SILVA et al., 2020; FARIA et al., 2016). Pellets produced with elephant grass and sugarcane bagasse had bulk density of 654 and 575 kg.m^{-3} respectively (DA SILVA et al., 2020). Typically, commercial pellets have a bulk density of around 600 - 750 kg.m^{-3} (KAMPERIDOU, 2022).

Table 3.8. Bulk density values of the pellets produced using PS and SS blends

Bulk density $kg.m^{-3}$			
Blends	Moisture (%)		
	10	25	40
100% PS + 0% SS	680 bC	622 bB	465 bA
75% PS + 25% SS	663 aC	623 bB	513 cA
50% PS + 50% SS	670 aC	598 aB	558 dA
25% PS + 75% SS	695 cC	608 aB	558 dA
0% PS + 100% SS	731 dC	628 bB	304 aA

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

The moisture content affected negatively the bulk density. Increasing the moisture content from 10% to 40% in the blend 75% PS + 25% SS the bulk density decreased by 22.6%. Almost all the pellets produced at 10% and 25% of moisture content had bulk density higher than 600 $kg.m^3$, except the blend 50% PS + 50% SS which had a bulk density of 598 $kg.m^{-3}$. Even though this value is statistically the same as the pellets produced with the 25% PS + 75% SS blend, it was lower than the value specified in the standard.

All the pellets produced with 40% moisture content had bulk density lower than 600 $kg.m^{-3}$. After drying, the water that fills the void spaces of the pellets is removed, and this space becomes empty causing a reduction in the bulk density (LABBÉ et al., 2020; SCATOLINO et al., 2018). The difference between PS and SS proportion in the blends had a lower influence on bulk density compared to the moisture content influence.

The bulk density and the HHV influence the energy density values. The higher the bulk density and the HHV higher the energy density (CASTRO et al., 2021; TORRES et al., 2023). As shown in Figure 3.5 the pellets that presented the highest energy density were those produced with 10% moisture content (values ranged from 9.25 to 9.84 $GJ.m^{-3}$). The pellets produced at 40% moisture content showed low values (ranging from 3.39 to 6.34 $GJ.m^{-3}$) compared to the pellets produced at 10% and 25% moisture content, but even in the pellets produced at 40% moisture content it was possible to increase around 4 times the energy density compared to the primary (1.59 $GJ.m^{-3}$) and secondary (1.03 $GJ.m^{-3}$) sludge *in natura.*

Biomass with higher density also has a higher amount of mass and energy per unit of volume, reducing the cost of transport and facilitating storage and use (LABBÉ et al., 2020; RODRIGUES et al., 2021).

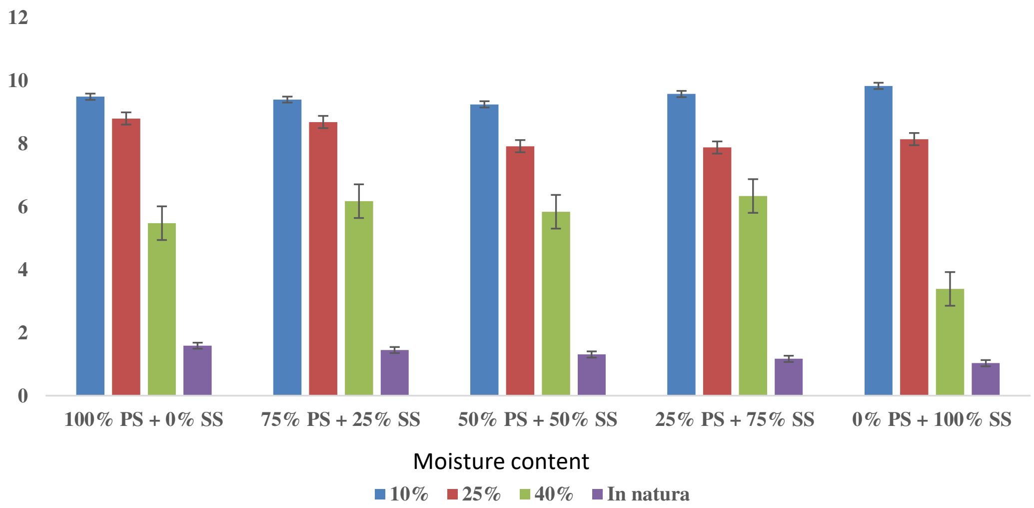

Figure 3.5. Energy density (GJ.m^{-3}) values of the pellets produced using PS and SS blends

3.6.Mechanical properties of the pellets

3.6.1. Fines generation and durability

The fines (F) generation (Table 3.9) and durability (D) (Table 3.10) are mechanical properties of the pellets and are directly associated with the resistance (CARVALHO et al., 2013; CASTRO et al., 2021). The pellets produced using 100% primary sludge at 10% moisture content had higher fines generation (0.533 %) and lower durability (87.6%) compared to the same blend at a moisture content of 25% (F 0.043% D 99.3) and 40% (F 0.363% D 96.4).

Table 3.9. Fines generation of the pellets produced using PS and SS blends

Fines (%)			
Blends	Moisture (%)		
	10	25	40
100% PS + 0% SS	0.533 bC	0.043 aA	0.363 aB
75% PS + 25% SS	0.078 aA	0.077 aA	0.340 aB
50% PS + 50% SS	0.043 aA	0.207 bcB	0.370 aB
25% PS + 75% SS	0.05 aA	0.263 cB	0.260 aB
0% PS + 100% SS	0.127 aA	0.103 abA	2.022 bB

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

Table 3.10. Durability of the pellets produced using PS and SS blends

Durability (%)			
Blends	**Moisture (%)**		
	10	**25**	**40**
100% PS + 0% SS	87.6 aA	99.3 aC	96.4 bB
75% PS + 25% SS	97.9 cA	99.3 aB	98.0 cA
50% PS + 50% SS	99.4 dB	98.7 aAB	98.1 cA
25% PS + 75% SS	99.4 dB	98.8 aAB	98.5 cA
0% PS + 100% SS	96.1 bB	99.0 aC	56.0 aA

Averages followed by the same capital letter between columns and lowercase between lines do not differ, at 5% significance, by the Tukey test
PS - primary sludge, *SS* - secondary sludge

The water content in the biomass during the pelletizing process improves the heating transference and reduces the glass transition temperature of some binder constituents making the bond between particles stronger (UNGUREANU et al., 2018; MOSTAFA et al., 2019). However, moisture levels higher than the optimum reduce mechanical strength, since there is a loss of the hydrogen bonds, and the exhaustion of water vapor causes fissures increasing the fines generation (CASTRO et al., 2021; SARKER et al., 2023).

The pelletizing temperature also affects the mechanical properties of the pellets. Higher temperatures tend to produce pellets with higher mechanical strength due to the stronger interaction between lignin and hemicellulose (SARKER et al., 2023). The pelletizing process typically occurs at temperatures ranging from 100 to 200°C (MA et al., 2021). However, in this study, the presence of higher moisture content in the feedstock resulted in a significant reduction in process temperature to below 100 °C. Specifically, pellets produced with 40% moisture content exhibited reduced durability, as the elevated water content not only lowered the pelletizing temperature but also impacted the structural integrity of the pellets.

The resistance of the pellets 100% PS + 0% SS increased with the increase of the moisture content of the sludge from 10% to 25% and decreased when the moisture content was increased from 25% to 40% during the pelletizing process. Other studies have shown the same effect, with higher mechanical properties in agro-industrial waste pellets produced at 25% moisture content than in pellets produced at 10% (JACKSON et al., 2016). This highlights the critical influence of moisture levels on both the thermal conditions and the mechanical resistance of the resulting pellets, underscoring the importance of optimizing moisture content for efficient pellet production.

The resistance of the pellets depends on several factors such as protein, fat, cellulose, hemicellulose, and lignin (TUMULURU, 2018). At 10% moisture content, the addition of secondary sludge increased the resistance. This fact can be due to the composition of secondary sludge, which is compounded mainly by microorganisms and complex biopolymers, i.e., lignin (TORRES et al., 2023).

The lignin is considered a natural binder, since during the pelletizing process the lignin increases the bond strength between particles, acting as a cement within the cells (CASTRO et al., 2021; PRADHAN; MAHAJANI; ARORA, 2018). The microorganisms in the SS are rich in proteins (LOPES et al., 2018). During the densification process, the proteins also act like a binder. The high temperature of the process denaturalizes the proteins, allowing the formation of new bonds with other compounds (JACKSON et al., 2016).

Pellets presenting high durability tend to generate less emissions during the combustion process. Stronger pellets are more difficult to break in fire beds, which reduces the formation of flying sparks, which, when drawn into the flue gas stream, suffer incomplete combustion, increasing CO emissions (LABBÉ et al., 2020).

According to the European standard for non-woody pellets DIN EN 14961-6, the fines generation values should be lower or equal to 3% and the mechanical durability higher or equal to 96% (DIN, 2012c). All the fines generation attended the values specified on the standard, pellets produced with the blend 100% PS + 0% SS at 10% moisture content and 0% PS + 100% SS at 40% moisture content showed mechanical durability lower than 96%.

4. CONCLUSIONS

Production of pellets using primary and secondary sludge generated in a kraft pulp mill proved to be technically feasible. The material evaluated in this study showed an HHV between 16.2-17.4 $MJ.kg^{-1}$. The SS has 22% ash content, but when mixed in different proportions with the PS, these values were proportionally reduced. The synthesis of the ashes starts at a temperature higher (1100 ºC) than the boiler operating temperature (800-900 ºC), reducing the impact caused by its fusibility on the equipment. Although it was possible to produce pellets at 40% moisture content, those presented low mechanical and physical properties, but still the energy density was four times higher than the *in natura* sludge. Pellets produced with 25% and 10% moisture content demonstrated high mechanical resistance, indicating their suitability for practical applications. Notably, increasing the moisture content from 10% to 25% resulted in a significant reduction in the generation of fines and enhanced the durability of the pellets.

The pelletizing process standardizes the dimensions of the sludge. The process proposed in this study improves sustainability in waste management and suggests a new source of renewable fuel, providing technical and scientific support to ensure the achievement of Sustainable Development Goals (SDG).

Future work involves assessing the environmental feasibility of the process through life cycle analysis (LCA) and the installation of a pellet pilot plant integrated into a pulp mill enabling to assessment of the challenges of scaling up the process and providing data to evaluate economic feasibility.

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CHAPTER 4

LIFE CYCLE ASSESSMENT OF PULP MILL SLUDGE MANAGEMENT: A COMPARATIVE STUDY BETWEEN LANDFILL AND PELLETS PRODUCTION

LIFE CYCLE ASSESSMENT OF PULP MILL SLUDGE MANAGEMENT: A COMPARATIVE STUDY BETWEEN LANDFILL AND PELLETS PRODUCTION

Abstract

The increasing need to replace fossil fuels requires the availability of new sources of renewable energy. Pulp mills generate a high amount of sludge, rich in lignocellulosic compounds, in the effluent treatment plants. Currently, landfilling is the main sludge disposal technique, which presents environmental, economic, and social drawbacks. The technical feasibility of producing pellets using pulp mill sludge has already been reported, however, there is a lack of studies about the environmental feasibility of this process. This study aimed to carry out a comparative LCA among three different scenarios of sludge management. Pellets production using external energy (SC1); pellets production using internal energy (SC2) and landfilling (REF). The LCA was performed following the ReCiPe 2016 methodology assessing 1 ton of sludge (dry matter 75%) as functional unit. The production and use of pellets had economic and environmental gains compared to landfilling. SC2 showed the best environmental performance, having a positive environmental impact in all the endpoint areas of protection. SC1 presented a positive impact on human health and 5 times less damage to ecosystems than the REF scenario. SC2 prevents the emission of 236 thousand CO_2eq and SC1 avoids 4.75 million m³ of fresh water consumption. The economic loss due to the social cost of the pollution in the REF scenario was approximately € 229 million.

Keywords: Life cycle assessment (LCA); Pulp and paper industry; Renewable energy; Waste management; Environmental impact.

1. INTRODUCTION

The 29th Conference of the Parties (COP29), held in Baku, Azerbaijan, highlighted the urgency of reducing greenhouse gas emissions. Combating the problems caused by climate change requires greater diversification of the global energy matrix. There is a high demand to replace fossil fuels with renewable energy sources, corroborating a low-carbon economy (CHU et al., 2023; TZEREMES; DOGAN; ALAVIJEH, 2023). The International Renewable Energy Agency has reported that by 2050 two-thirds of the energy generated in the world will come from renewable sources (TZEREMES; DOGAN; ALAVIJEH, 2023).

In the current global production scenario, a linear economy based on "waste" disposal is no longer viable. The implementation of new processes and the valorization of co-products that used to be classified as waste has emerged with full force in recent years, establishing the circular bioeconomy in the industrial market (PERIYASAMY et al., 2022; SUÁREZ-EIROA et al., 2019).

One of the options for producing renewable energy is the use of lignocellulosic waste such as sludge waste from pulp and paper mills (KUMARI; SINGH, 2018). Pulp and paper mills generate around 400 million tons of wet, lignocellulosic rich, sludge per year (FAUBERT et al., 2016; TURNER; WHEELER; OLIVER, 2022) with Brazil being the second largest pulp producer in the world. Currently, the main disposal method for this sludge is landfilling (RODRIGUES et al., 2021).

At pulp mill Effluent Treatment Plants (ETPs), two types of sludge are generated as byproducts of wastewater treatment: primary sludge and secondary sludge. Primary sludge is produced during the initial treatment stage, where suspended solids are removed through physical processes like sedimentation. This sludge is rich in cellulose, hemicellulose, and lignin, making it a potential resource for recovery or reuse. In contrast, secondary sludge arises from the biological treatment process, where microorganisms break down organic pollutants composed mainly of organic matter (FAHIM et al., 2018; RODRIGUES et al., 2021; TORRES et al., 2023).

One option to use pulp mill sludge as a renewable energy source is to produce pellets (RODRIGUES et al., 2021). Pellets are densified solid fuels produced from renewable raw materials that can be used for biogas production, co-firing, and pyrolysis (CLAUSER et al., 2021). The processes involved in pellets production are generally grinding, drying, and pelletizing at a pellet mill (TUMULURU, 2016).

Previous research demonstrated the technical feasibility of producing pellets using the primary and secondary sludge generated at the pulp mill (RODRIGUES et al., 2021). However, the environmental feasibility of the process must also be assessed.

Life cycle assessment (LCA) allows the evaluation of the environmental impacts involved in a given process, based on the resources used, energy demand and environmental releases. An LCA also identifies opportunities for environmental improvements in the process (CHAU; LEUNG; NG, 2015; SREEKUMAR et al., 2020).

There is a lack of literature about the environmental impacts of the pelletizing of pulp mill sludge for energy purposes, especially when it is compared with conventional methods of disposal, such as landfilling. This work aims to carry out a comparative study of the environmental impacts of energy generation and landfilling of the primary and secondary sludge generated in a kraft pulp mill in Brazil, through life cycle assessment (LCA) methodology.

2. MATERIAL AND METHODS

The LCA to evaluate the environmental impact of the process was performed following ISO 14040:2006 and ISO 14044:2006. The software used in the study was Sphera's LCA For Experts (FE) and openLCA 2.0.3. LCA inventory databases were obtained from different sources, such as the Ecoivent database, Sphera database, Industrial Design and Engineering Materials (IDEMAT) database, published literature, and data provided by pellets industries.

The data were gathered during the first half of 2024, providing a comprehensive foundation for the analysis and findings presented in this study. The LCA method involves 4 main steps: goal and scope definition, inventory analysis, impact assessment, and interpretation (DAYLAN; CILIZ, 2016).

The primary and secondary sludges used in this study for the pellets production are from a Brazilian kraft-bleached pulp mill.

2.1. Goal and Scope

This study aims to perform a comparative LCA analysis for pulp mill sludge management. Three different scenarios are being considered (Figure 4.1) a) Drying and pelletizing the sludge using external energy, pellets combustion in the boiler inside the mill to generate energy to be used as credit b) Drying and pelletizing the sludge using internal energy from energy losses at the mill, pellets combustion in the boiler inside the mill, part of the energy produced is returned to the process and partly used as energy credit c) The disposal of sludge in landfills. The scope of this work includes the life cycle of the sludge pellets production and its combustion in the biomass boiler, and the transportation and disposal of the sludge in landfills from a cradle-to-grave approach.

The system boundaries of the pelletizing sludge process. The primary and secondary sludge was generated at the ETP of the pulp mill. The pulp mill and the ETP are not included in the LCA analysis.

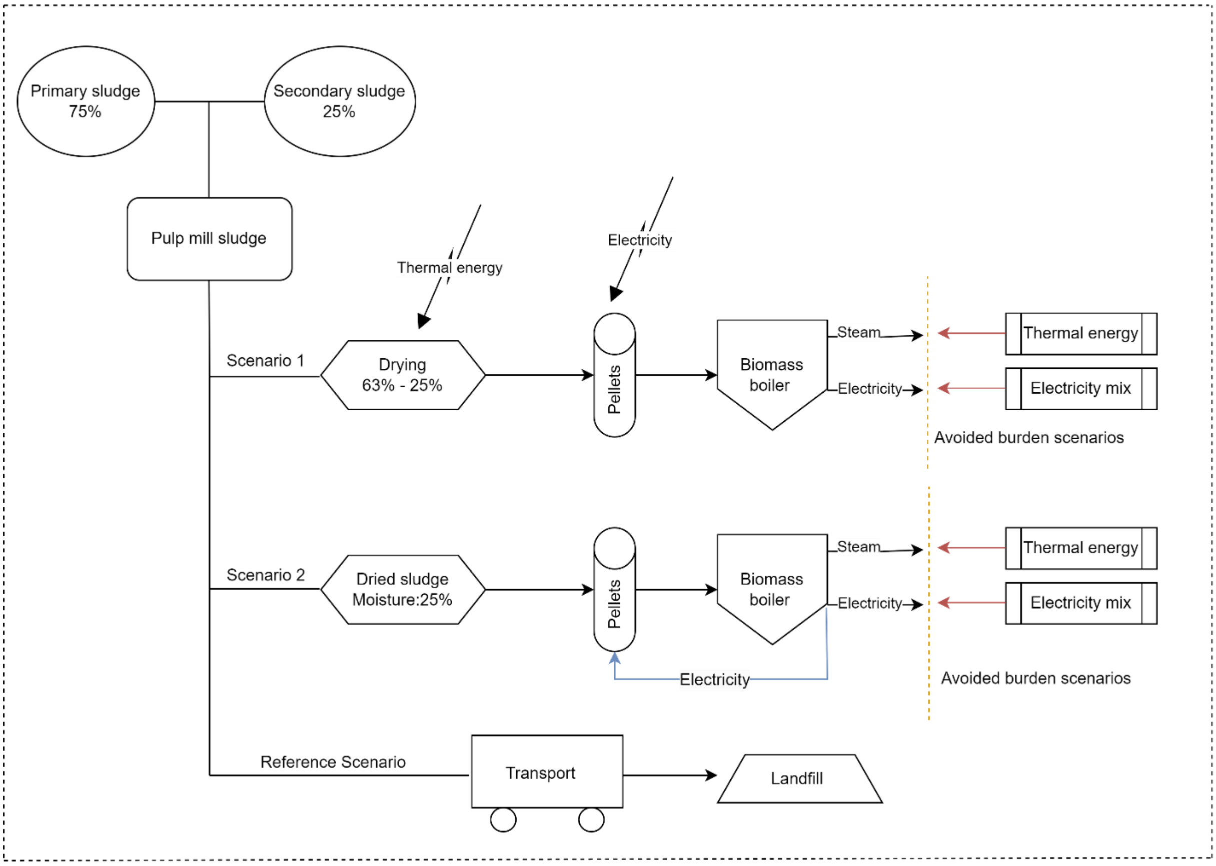

Figure 4.1. Systems boundaries for the LCA of pulp mill sludge management considering the pelletizing process and landfilling

2.2. Functional Unit (FU)

The functional unit (FU) used to compare the different scenarios is 1 ton of sludge (dry matter 75%) generated at a kraft-bleached pulp mill. The proportion of primary (750 kg) and secondary (250 kg) sludge used to form the sludge for the pelletizing process was based on the amount of generation of the respective sludge in the pulp mill (Torres et al., 2023).

2.3. Reference scenario

The reference scenario (REF) is the direct disposal of the sludge at a landfill. The data regarding landfills was taken from Sphera's LCA FE database. A simple paper waste landfill with the following specifications: 30 m height, 40.000 square meters, 100 years deposit. Included in this dataset: sludge treatment and deposition, gas and leachate treatment, sealing materials, and diesel for the compactor. The gas produced at the landfill is 22% flared, 28% used, and 50% emitted. Active carbon flocculation and precipitation processing are used to treat the leachate. The sealing is formed by gravel, sand, clay, and polyethylene film. The manufacturing processes of the sealing materials are considered.

Transportation of the sludge to the landfill is done by a diesel truck, Euro 3, 17.3 payload capacity over a distance of 15 km.

2.4. Studied scenario

The environmental impacts of using sludge for pellet production as a fuel in the biomass boiler were simulated using Sphera's LCA FE. The energy demand for the pellet plant was calculated using data extracted from the literature (TORRES et al., 2023; TUMULURU, 2016) and also data available in the Associação Brasileira das Indústrias de Biomassa Brasil (ABIB). The energy demand for the pelletizing plant used in this process was 694 $MJ.t^{-1}$ (ABIB BRAZIL, 2020). The initial sludge moisture content considered in this work was 63% as reported in the previous study conducted by (Rodrigues, Data not published). The energy demand to dry the sludge to 25% and the net heating value (NHV) of the pellets was calculated was estimated using Equation 1, following Annex E of the European standard DIN EN 14918 (DIN, 2010b).

$$NHV = HHV - 212.2 \times H - \ 0.8 \times (O + N)) \times (1 - 0.01 \times M) - (24.43 \times M) \quad (1)$$

Where:

NHV (constant pressure): Net heating value at constant pressure, in $J\ g^{-1}$;

HHV: Higher heating value, in $J\ g^{-1}$;

H, O, N: hydrogen, oxygen, and nitrogen, respectively, in percent (%);

M: moisture, wet basis, in percent (%).

Higher heating value was determined using the Parr 6300 calorimeter adiabatic calorimetric pump method based on ASTM D2015 (ASTM, 1982).

Elemental chemical analysis (C, H, N, S, O) was calculated using the analyzers TruSpec Micro CHN, and TruSpec S (LECO). The oxygen was determined using the sum of C, N, H, S, and ashes decreased by 100, following the standard EN 15296 (DIN, 2011).

The pellet production plant was not considered in this LCA.

Pellets from pulp mill sludge are not yet industrially used, so to simulate the combustion of pulp mill sludge pellets the dataset “Paper and board (water 22%) in waste incineration plant” from the Sphera database was used (SPHERA, 2023a). This dataset considers the thermal treatment of paper and board waste, with 22% moisture content in an average waste-to-energy plant with dry flue gas cleaning and SCR (Selective Catalytic Reduction) as NO_x removal technology. The main composition of the paper and board waste used in this process is shown in Table 4.1. The end-of-life inventory of this process includes the emissions and resources of the thermal treatment, the behavior of bottom ash, and air pollution control waste on a landfill. The bottom ash is reused as construction material after the metal recovery and stabilization. The facility model involves an incineration line equipped with a grate and a steam turbine. Part of the steam produced is internally used within the process and the rest is used to generate electricity or exported as heat or steam to industry or the domestic sector.

In the studied scenarios, two alternatives were evaluated. The first (SC1), considered that all the energy needed for drying and pelletizing comes from sources outside the mill, the source used in the model was energy from biomass. The energy from the biomass process includes biomass production, processing, transportation, and combustion in the boiler, ashes management, and biomass power plant construction. The electricity generated in the combustion of the pellets was used to replace energy from the Brazilian electricity grid mix and the generated steam replaced the production of steam from hard coal. The country reference in both situations was Brazil since the previous study used sludge generated in a Brazilian pulp mill to produce pellets (Rodrigues, Data not published). Hard coal energy was chosen to be replaced in this scenario, considering that coal is a solid fuel and the largest global source of energy, representing 10.4% of the thermal energy produced in Brazil (MME, 2024; MUNAWER, 2018).

The second alternative (SC2) involves utilizing the internal energy generated within the mill. In this scenario, the energy required for drying the sludge is not considered, as it can be supplied by surplus energy already available onsite. During the combustion stage of the pellets, a portion of the generated electricity is used internally to meet the energy needs of the pellet plant. As the amount of electricity produced exceeds the internal demand, the surplus electricity and the generated steam are utilized as an avoided burden, similar to the approach in SC1.

Table 4.1. Paper and board waste composition in the incineration plant

Paper waste composition					
C (%)	H (%)	N (%)	O (%)	Ash (%)	NHV $MJ.kg^{-1}$
29.7	4.45	0.364	32.9	16.9	10. 4

Adapted from: (SPHERA,2023)

2.5. Life cycle inventory and assumptions

The information and data sources used in this study are shown in Tables 4.2, 4.3, and 4.4.

In Scenario 2, the energy used to dry the sludge comes from the thermal energy that usually is generated (and not used) within the pulping process, i.e., biomass boiler, recovery boiler, and evaporation (CHANTASIRIWAN, 2023; RODRIGUES et al., 2021). This energy is excluded from the model since it is already produced in the mill as a byproduct of converting thermal energy into electricity.

The (maximum) moisture content of the sludge used for pellets production (25%) was based on a previous study (Rodrigues, Data not published).

Table 4.2. Data inventory for Scenario 1 of pellets production using sludge from the pulp mill, and external energy use from biomass

Process Steps	Description	Amount	Unit	Sources
INPUT	Sludge from a kraft-bleached pulp mill	1	ton	(Torres et al., 2023)
	Composition:			
	Primary sludge	75	%	
	Secondary sludge (biological sludge)	25	%	
Pelletizing	BR: Electricity from biomass	694	MJ	(ABIB BRAZIL, 2020)
Drying	BR: Thermal energy from biomass	7000	MJ	Calculated
Pellets	Sludge pellets	11200	MJ	Calculated
Combustion	NL: Paper and board (water 22%) in waste: Electricity	1550	MJ	(SPHERA, 2023a)
	NL: Paper and board (water 22%) in waste: steam	1640	MJ	(SPHERA, 2023a)
Credit	BR: Electricity grid mix Sphera: Electricity	-1550	MJ	(SPHERA, 2020a)
	BR: Thermal energy from hard coal: steam	-1640	MJ	(SPHERA, 2020b)

Table 4.3. Data inventory for Scenario 2 of pellets production using sludge from the pulp mill, and internal energy use from the pulp mill

Process Steps	**Description**	**Amount**	**Unit**	**Sources**
INPUT	Sludge from a kraft-bleached pulp mill	1	ton	(Torres et al., 2023)
	Composition:			
	Primary sludge	75	%	
	Secondary sludge (biological sludge)	25	%	
Pelletizing	NL: Paper and board (water 22%) in waste: Electricity	694	MJ	(ABIB BRAZIL, 2020)
Pellets	Sludge pellets	11200	MJ	Calculated
Combustion	NL: Paper and board (water 22%) in waste: Electricity	1550	MJ	(SPHERA, 2023a)
	NL: Paper and board (water 22%) in waste: steam	1640	MJ	(SPHERA, 2023a)
Credit	BR: Electricity grid mix Sphera: Electricity	-854	MJ	(SPHERA, 2020a)
	BR: Thermal energy from hard coal: steam	-1640	MJ	(SPHERA, 2020b)

Table 4.4. Data inventory for paper sludge disposal in landfills

Process Steps	Description	Amount	Unit	Source
INPUT MATERIAL	Paper sludge	1	ton	(SPHERA, 2023b)
Transport	Truck, Euro 3, 20 - 26t gross weight / 17.3t capacity	1	ton	(SPHERA, 2023c)
	Diesel mix at refinery	2.44	kg	(SPHERA, 2023c)
Landfill	Paper waste on landfill	1	ton	(SPHERA, 2023b)

2.6. Life cycle impact analysis

The method ReCiPe 2016 was chosen to convert the life cycle inventory to a limited number of environmental impact scores. In this methodology, the inventories of the emissions and raw materials are expressed into a more limited set of environmental effects.

The environmental impact per unit of stressor is indicated by the characterization factors (CF). This property can be derived at the midpoint level and endpoint level. In the first case, the CF are located somewhere along the impact pathway. In the endpoint, the CF is summarized into three areas of concern: human health, ecosystem quality and resource scarcity (Figure 4.2).

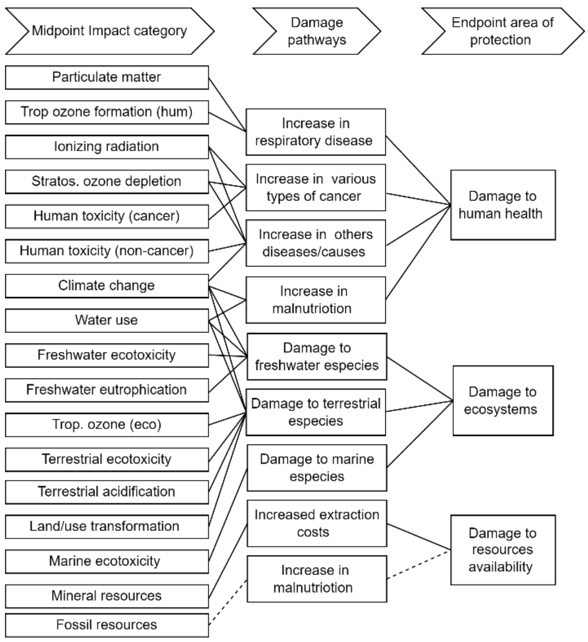

Figure 4.2. Impact categories according to the methodology ReCiPe2016
Adapted from (HUIJBREGTS et al., 2016)

2.7. Allocation procedure

The technology in this study proposed the production of energy from pellets using sludge from the pulp mill instead of disposal of this sludge in landfills. The avoided burden allocation approach was used to show the comparative net assessment of environmental impacts.

3. RESULTS AND DISCUSSION

3.1. Midpoint level impacts

The impact assessment of the different scenarios is presented at midpoint level. This level shows the impact caused by the environmental changes resulting from the process (HUIJBREGTS et al., 2016). Table 4.5 shows the results of the midpoint level impact assessment of some important categories. The complete table presenting each impact category for SC1, SC2, and REF can be found in Appendix A.

Table 4.5. Absolut values of the midpoint level impact of scenario 1 (SC1), scenario 2 (SC2) and reference (REF)

Comparative results			
Midpoint impact categories	**SC1**	**SC2**	**REF**
Climate change excl biogenic carbon [kg CO_2 eq.]	-160	-172	1190
Fossil depletion [kg oil eq.]	-35.3	-40.6	26.6
Freshwater Consumption [m^3]	-3.46	-1.59	0.156
Stratospheric Ozone Depletion [kg CFC-11 eq.]	8.13E-04	-3.46E-06	3.66E-05
Terrestrial Acidification [kg SO_2 eq.]	-0.563	-2.02	0.248
Terrestrial ecotoxicity [kg 1,4-DB eq.]	-209	-181	48

Climate change including biogenic carbon was not taken into consideration, once this study focused on fossil emissions reduction. Sludge is a waste product from the pulp mill, whose main raw material is eucalyptus, so a neutral biogenic carbon cycle can be assumed due to carbon fixation during the growth of the trees (MARTÍN-GAMBOA et al., 2020).

The REF scenario was the one that presented the highest contribution to climate change per ton of sludge treatment excl biogenic carbon (1190 kg CO_2 eq.) while the SC2 presented the lowest (-172 kg CO_2 eq.). The greenhouse gases (GHGs) are produced during waste biodegradation and in the leachate treatment system, generating CH_4, N_2O, and CO_2 (BOGNER; SPOKAS; CHANTON, 2011; VAVERKOVÁ, 2019). The amount of GHG released during the total landfill life is estimated to be about 1.37 E+09 kg of CO_2 equivalent (VAVERKOVÁ, 2019). Globally, landfilling is responsible for about 5% of GHG emissions (ZHANG et al., 2019).

The dataset used for landfilling in this study originates from European landfills, where the collection and utilization of part of the landfill gas was considered. In Brazil, many landfills still fail to collect and utilize these gases (SANTOS et al., 2018). When analyzed in openLCA 2.0.3, an organic waste landfill without CH_4 prevention had an impact of 2822 CO_2 eq per ton of disposed sludge. No dataset of Brazilian waste paper landfills was found in the inventory databases used in this study.

In SC2 all the midpoint impacts shown in Table 4.5 were negative while in the REF scenario, these values were positive. In SC1 just the stratospheric ozone depletion presented a positive value. The negative values indicate avoided emissions and thus a positive impact on the environment. The REF scenario presented higher environmental impact in most of the categories presented in this study.

The terrestrial acidification per ton of sludge caused by the REF was 0.248 kg SO_2eq, for SC1 and SC2 this impact was, -0.563 kg SO_2eq and -2.01 kg SO_2eq respectively. Terrestrial acidification is caused by the emissions of nitrogen oxides (NO_x), sulfur dioxide (SO_2), and ammonia (NH_3) which are produced in electricity generation, industries, transport operations, some agricultural activities, and also in the leachate generated in landfill (KYLILI; CHRISTOFOROU; FOKAIDES, 2016). Although leachate treatment was applied in the REF scenario, its great variation and complexity make it difficult to treat efficiently (COSTA; ALFAIA; CAMPOS, 2019). In SC2 all the energy demanded in the process is internally obtained. The external energy used to supply the energy demanded in SC1 increases the terrestrial acidification compared to SC2 although this impact remains with negative values.

In SC1 and SC2 the generated energy from the combustion of the pellets replaces energy that otherwise should have been generated from a fossil thermal process, resulting in a lower impact on fossil depletion when compared to the REF.

The Stratospheric Ozone Depletion for SC1 has a higher value (8.13E-04 kg CFC-11 eq/ton of treated sludge) than for SC2 (-3.46E-06 kg CFC-11 eq/ton of treated sludge). This is caused by the external thermal energy used in SC1. The power plant construction and the emissions of material to the ozone layer in the process are the main reasons for this increased value (HE et al., 2019). REF scenario presented the highest impact in the Stratospheric Ozone Depletion. The gas released by the landfills contains traces of chlorinated and fluorinated species, which produce stratospheric ozone depleting substances, like chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) (MANHEIM et al., 2024).

Terrestrial ecotoxicity refers to the release of chemical substances that impact soil-dwelling organisms and affect the soil ecosystem (SYDOW et al., 2020). Terrestrial ecotoxicity per ton of treated sludge of SC1 and SC2 were -209 and -181 kg 1,4- dichlorobenzene (DB) eq, respectively, showing a positive environmental impact due to the avoided energy production in the Brazilian electricity grid mix. The REF scenario presented a negative impact (48 kg 1,4-DB eq/ton of disposed sludge). This is caused by the leachate produced by landfills which is considered toxic, as it contains high amounts of organic compounds, salt, and heavy metal ions, representing a potential threat to the surrounding ecosystem (TENG et al., 2021).

3.2. Endpoint area of protection

The grouped and normalized midpoint results give the origin to a second and final assessment, the end-point level, which reflects the damage in three higher aggregation areas of protection i) human health, ii) biodiversity, and iii) resource scarcity (HUIJBREGTS et al., 2016). Appendix B shows the damage pathways results per scenario evaluated in this study.

3.2.1. Damage to human health

The unit for damage to human health (Figure 4.3) is DALY (disability-adjusted life years), which refers to the years lost or a person's disability due to an illness or early death (VAN ZELM et al., 2016).

Among the evaluated scenarios, the REF (2.23E-03 DALY/ton of treated sludge) was the only scenario presenting a negative impact on human health. Both SC1 and SC2 showed negative values (-4.47E-04 and -7.41E-04 DALY/ton of treated sludge), indicating avoided impact or a positive environmental impact. The REF scenario is the most harmful to human health because of the higher exposure to chemicals, and inhalation of toxic fumes and dust of people living near landfills as in the geographical scope of this study.

People living closer to landfill sites tend to suffer more medical conditions, like asthma, cuts, diarrhea, stomach pain, reoccurring flu, malaria, cough, skin irritation, cholera, diarrhea, and tuberculosis (NJOKU; EDOKPAYI; ODIYO, 2019). The positive environmental impact for SC1 and SC2 can be explained by the reduction of hard coal use. Burning coal releases toxic gases that are directly and indirectly associated with health problems such as skin, cardiovascular, brain, blood, and lung diseases, and different types of cancer (MUNAWER, 2018). The use of fossil fuels for energy generation is responsible for increasing the concentration of GHGs in the atmosphere, causing health problems in the population (LIJÓ et al., 2019).

The positive environmental impact of SC1 was 40% lower than the impact of SC2. because SC2 aims to be self-sufficient in energy, bringing more gain to this scenario.

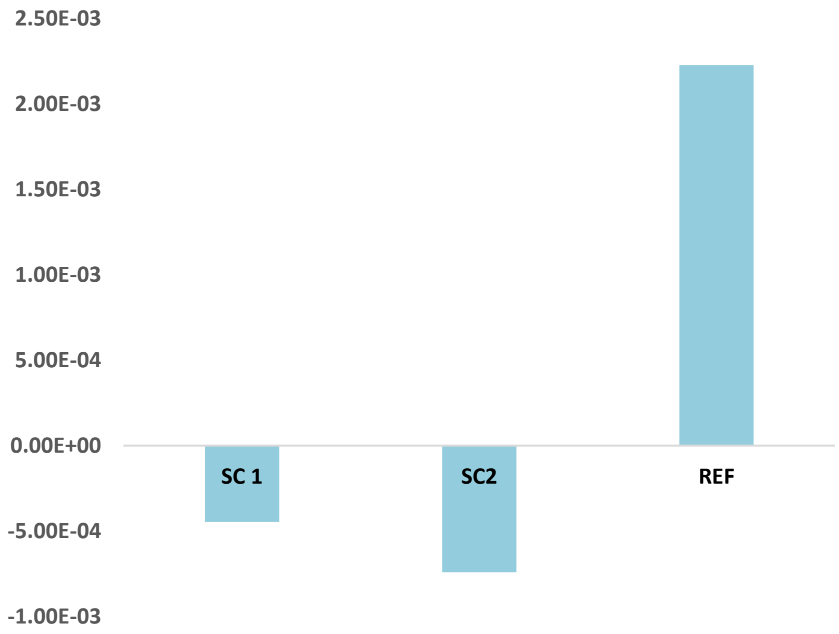

Figure 4.3. Damage to human health (DALY) at the endpoint level impact of scenario 1 (SC1), scenario 2 (SC2) and reference scenario (REF)

3.2.2. Damage to ecosystems

The damage to ecosystems in species.yr (Figure 4.4) aggregates the damage to terrestrial, fresh water and marine species over space and time. Both water and land use are important midpoints linked to damage to ecosystems since they have a major influence on the maintenance of biodiversity (HUIJBREGTS et al., 2016).

Both SC1 and REF have a negative impact on ecosystems, with REF showing a 45% higher impact than SC1. In contrast, SC2 demonstrated a positive effect on the ecosystem (-9.99E-07 species.yr/ton treated sludge).

Landfilling (REF) is harmful to the ecosystem in many ways. The circulation of heavy vehicles during the construction and operation stages compacts the soil affecting the local vegetation, causing damage to animal life. The gas generated from the decomposition of the organic matter can seep through the soil, reducing the oxygen available causing damage to the roots of plants, and reducing the animal population (IRAVANIAN; RAVARI, 2020).

The impacts of SC1 are partially caused by biomass energy which is used to dry the sludge. Biomass energy is renewable but not completely clean, as there are other processes associated with its use i.e. transport and manufacturing steps (LIJÓ et al., 2019). In SC1 the dependency on external energy sources also increases land use which causes damage to the ecosystem.

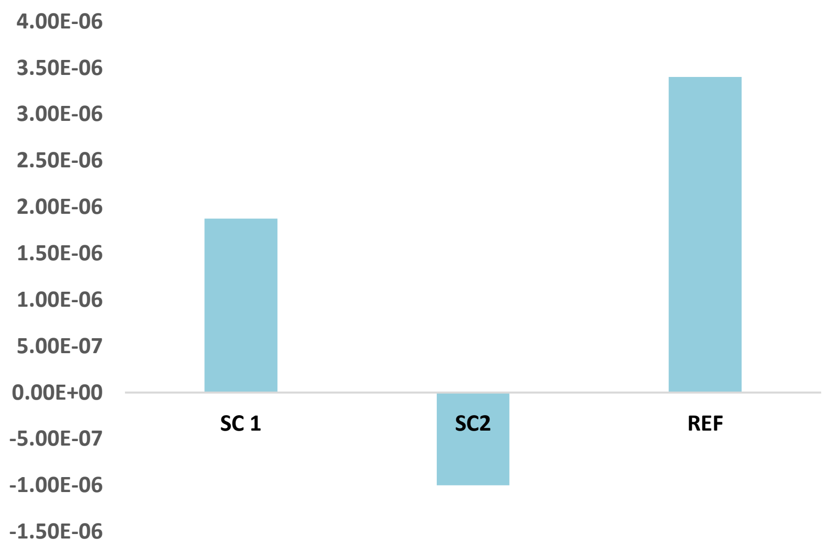

Figure 4.4. Damage to ecosystems (species.yr) at the endpoint level impact of scenario 1 (SC1), scenario 2 (SC2) and reference scenario (REF)

3.2.3. Damage to resources availability

Figure 4.5 shows the damage to resource availability, in dollars ($), and represents the future surplus costs involving mineral and fossil extraction resources (CANAJ et al., 2020). The midpoint indicators influencing this damage are metal and fossil depletion. The REF scenario shows SC1 the highest damage to resources. The infrastructure, diesel and cement used in landfill creation and operation increase the demand for fossil and mineral resources (HONG et al., 2017).

Looking at both scenarios, SC1 shows minor damage to ecosystems whereas SC2 has a positive impact on ecosystems. This can be explained by the energy use, the thermal energy used in SC1 has a higher impact on resource availability. In SC2, part of the energy produced returns to the system to supply the energy demand of the pelletizing process. In this scenario, it was also assumed that the energy for drying the sludge is obtained from energy losses inside the mill, with no extra impact on the availability of resources.

In SC1 and SC2 the energy which is produced with the combustion of the pellets is used as credit since it avoids energy production from the hard coal and the Brazilian electricity grid mix. When the avoided burden is not considered in SC1, the damage to resource availability per ton of sludge treated is 9.31 \$, which is similar to landfilling (9.77 \$). In SC1 and SC2 the energy which is produced with the combustion of the pellets is used as credit since it avoids energy production from the hard coal and the Brazilian electricity grid mix. When the avoided burden is not considered in SC1, the damage to resource availability per ton of sludge treated is 9.31 \$, which is similar to landfilling (9.77 \$).

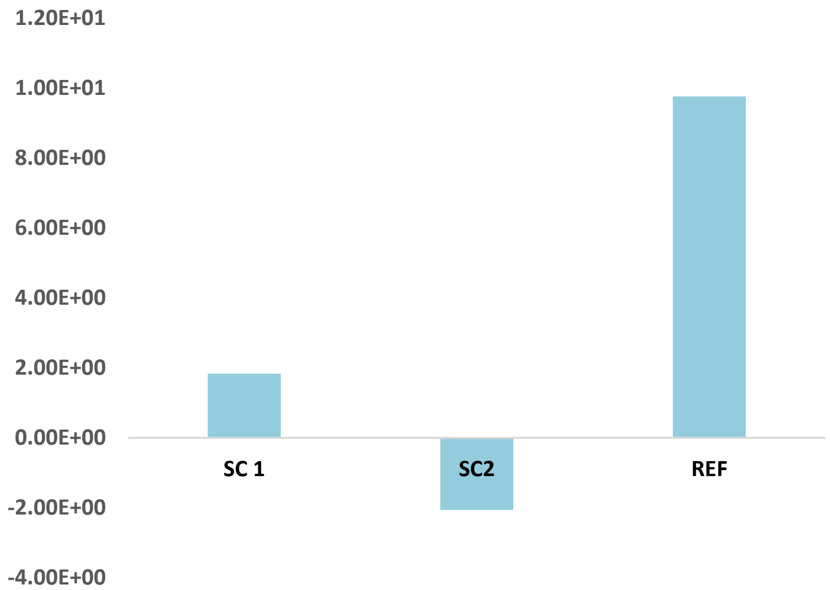

Figure 4.5. Damage to resource availability (\$) at the endpoint level impact of scenario 1 (SC1), scenario 2 (SC2), and reference (REF)

3.3. GENERAL REMARKS

The model functional unit (FU) considered is 1 ton of sludge. In 2022, sludge production in Brazil's pulp mills was approximately 1.37 million tons of primary and secondary sludge (TORRES et al., 2023). If instead of landfilling all the sludge produced at pulp mills in Brazil were processed according to the proposed scenarios the reduction of CO_2eq in the atmosphere would be 236 thousand tons (SC1) and 220 thousand tons (SC2) per year.

During the Paris Agreement (UNFCCF COP21) the country committed to reducing GHG emissions by 43% below 2005 levels, which was 2.7 billion tons. To achieve this goal, Brazil must reduce 802 million tons of GHG from the 2022 amount (2.3 billion CO2eq) (SEEG, 2023; TEODORO et al., 2024). Processing the sludge from pulp mills according to SC2 could contribute up to 0.029% to this reduction. For SC1 this would be 0.027%.

The pulp and paper industry in Brazil has a high water consumption, it is estimated that 25 m³ of water is used per ton of bleached pulp produced. In 2022 the pulp mills in Brazil consumed about 624 million m³ of water (IBÁ, 2023; SANTOS et al., 2020). The results of this study, analyzed at the midpoint level and extrapolated to encompass all sludge generated in pulp industries, reveal significant water conservation potential. SC1 and SC2 demonstrate the ability to avoid the consumption of approximately 4.75 million m³ and 2.18 million m³ of water per year, respectively, the water saved in SC1 alone is sufficient to meet the annual water needs of a city with a population of 81 thousand people (SOUZA et al., 2021).

In addition to the impact on the environment and society. The damage caused by emissions can also be quantified in economic losses. According to the CE Delft report, the impact of each midpoint has a social cost due to the damage caused by the pollution which is given in euros per kilogram of pollutant (CE DELFT, 2023). The scenarios proposed in this study have negative prices due to the prevention of environmental damage. The production of pellets of the sludge generated in Brazil in one year, according to SC1 and SC2 can save € 22.3 million and € 145 million respectively. In contrast, the REF scenario results in an economic loss of € 229 million.

4. PELLETS USE ADVANTAGES

The standardized shape and high density of the pellets bring advantages in logistical operations such as storage, transport, handling, and automatic boiler feeding and also increase the energy efficiency of the conversion process (PRADHAN; REUSSER; KROPP, 2013). The combustion of the sludge pellets in this work was represented by a paper and board incineration plant, once this model had a fuel with chemical properties similar to the sludge generated at the mill. However, the advantages of having sludge pellets as fuel instead of the non-pelletized sludge were not highlighted in the LCA results.

During the burning process, pellets are an auxiliary fuel of high performance, since the controlled size, high energy density, and lower moisture content lead to better combustion behavior and more control in the boiler operation resulting in homogeneous combustion and less particulate emissions (PRADHAN; REUSSER; KROPP, 2013; RODRIGUES et al., 2021). Due to their high density, pellets can burn for a long period, which heat the boilers to higher temperatures, being environmentally friendly and cost-effective (ZHOU et al., 2016).

The results presented in the LCA showed that the energy generation using primary and secondary sludge of the pulp mill presented environmental gains compared to the landfill, although it was not possible to evaluate the pellet form of the sludge.

Future advancements in the current model for utilizing pellet sludge hold significant potential for optimizing sustainability and efficiency. One promising direction is the integration of real-world data from biomass boilers that use pelletized sludge as an auxiliary fuel. By incorporating this operational data, the combustion step in the model can be refined to simulate and enhance the process more accurately.

Another critical avenue for improvement lies in the post-combustion phase, specifically, the evaluation of ash generated from pellets combustion. These ashes may serve as valuable resources due to their composition, which often includes organic compounds. By investigating their application as a soil conditioner, the potential for improving soil quality and fertility can be assessed. Additionally, the ashes could contribute to carbon stock, offering a dual benefit of waste recycling and climate change mitigation.

5. CONCLUSIONS

This study assessed the environmental impact of three different scenarios regarding pulp and paper sludge management in Brazil based on an. LCA which followed the ReCiPe 2016 provided 19 midpoint categories and three endpoint categories of environmental impact.

The results showed that producing pellets for energy generation using sludge from kraft pulp mills has environmental advantages compared to landfilling. Even when external energy was required for the process (SC1), the results in the endpoint levels showed a positive impact on human health, 45% less damage to ecosystems, and 5 times less impact on resource availability compared to the REF scenario.

In SC2, the energy used to produce the pellets comes from the energy produced by the combustion of the pellets. The energy to dry the sludge comes from the low/medium pressure steam, lost during energy conversions inside the mill. As a result, SC2 showed the best environmental performance, showing a positive environmental impact in all the endpoint levels.

According to SC2, 236 thousand tons of CO_2 eq emissions can be avoided in one year if all the kraft pulp mill sludge produced in Brazil is converted into pellets for energy use. The water saved in SC2 is enough to supply a city of around 81 thousand inhabitants. The economic gains with the avoided emission in SC2 are € 145 million. For SC1 the positive impacts are less but still considerable with 220 thousand tons of CO_2 eq emissions avoided, 2.18 million m^3 water saved, and € 22.3 million of economic gains.

Future work will involve implementing a pilot pellet unit integrated into the pulp mill and testing pellet combustion in the biomass boiler. The data collected from this scaled-up scenario will enable more accurate model adjustments, validation of assumptions, and improved reliability of the LCA.

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SPHERA. **Electricity grid mix; AC, technology mix; consumption mix, to consume**. Brazil: [s.n.].

SPHERA. **Thermal energy from hard coal; technology mix regarding firing and flue gas cleaning; production mix, at heat plant; 100% efficiency (en)**. Brazil: [s.n.].

SPHERA. **Diesel mix at refinery; from crude oil and bio components; production mix, at refinery; 12.36 wt.% bio components; 9.25 wt.% biogenic Carbon; 10.87 wt.% Share bio C to C-Total; for use in Transport Process share_CO2_bio: 0.1087; 500 ppm sulphu**. [s.l: s.n.].

SPHERA. **Paper and board (water 22%) in waste incineration plant; average waste-to-energy plant, without collection, transport and pre-treatment; production mix (region specific plants), at plant; SCR 67% / SNCR 33% / Heating value 10,4 MJ/kg / Electrical efficiency 14,9 % / Heat efficiency 15,7 % (en)**. Netherlands: [s.n.].

SPHERA. **Landfill of paper waste; landfill including landfill gas utilisation and leachate treatment and without collection, transport and pre-treatment; production mix, at landfill site; 22 % flare, 28 % used, 50 %**. [s.l: s.n.].

SPHERA. **Truck, Euro 3, 20 - 26t gross weight / 17.3t payload capacity; diesel driven, Euro 3, cargo; consumption mix, to consumer; 20 - 26t gross weight, 17.3t payload capacity (en)**. [s.l: s.n.].

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GENERAL CONCLUSIONS

The sludge generated at the pulp mill is rich in lignocellulosic content and has a high availability and low or negative costs. These characteristics make this material suitable for use as raw material in new processes. Energy production using this sludge has been technically and environmentally proven in this work. The main challenges to energy conversion are the high moisture and ash content, low density, and the non-homogeneity of the material.

The composition of pulp mill sludge varies significantly based on numerous factors, including the specific processes employed in pulp production and the methods used for effluent treatment. Consequently, it is essential to conduct a comprehensive characterization of the sludge. This detailed analysis allows for the identification of its physical, chemical, and biological properties, enabling the selection of the most appropriate applications or management strategies based on its unique characteristics.

The production of pellets using the sludge improved its energy potential, increased the energy density, and transformed this material into a homogeneous biomass. It was able to produce pellets with a high moisture content (25% and 40%). The production and use of sludge pellets presented environmental and economic advantages when compared to landfilling.

The utilization of pulp mill sludge for energy production offers a significant opportunity to diversify the renewable energy matrix while directly contributing to several Sustainable Development Goals (SDGs), including SDG 7 (Affordable and Clean Energy), SDG 9 (Industry, Innovation, and Infrastructure), SDG 12 (Responsible Consumption and Production), and SDG 13 (Climate Action).

The sludge management strategies evaluated in this work present a practical alternative for pulp mills since this material can be used efficiently to produce energy on-site as a supplementary fuel. The results obtained in this study present an innovative character and contribute to the transition to a more circular economy in forest-based industries.

APPENDIX A – ABSOLUT VALUES OF IMPACT CATEGORIES AT THE MIDPOINT LEVEL

Midpoint impact categories	SC 1	SC 2	Landfill
Climate change, default, excl biogenic carbon [kg CO2 eq.]	-160	-172	1190
Climate change, incl biogenic carbon [kg CO2 eq.]	892	884	1480
Fine Particulate Matter Formation [kg PM2.5 eq.]	-0.248	-0.677	0.0843
Fossil depletion [kg oil eq.]	-35.3	-40.6	26.6
Freshwater Consumption [m3]	-3.46	-1.59	0.156
Freshwater ecotoxicity [kg 1,4 DB eq.]	0.086	-0.0104	0.0111
Freshwater Eutrophication [kg P eq.]	0.0188	-0.000496	0.00665
Human toxicity, cancer [kg 1,4-DB eq.]	0.271	-0.116	0.036
Human toxicity, non-cancer [kg 1,4-DB eq.]	8.05	-14.8	4.22
Ionizing Radiation [kBq Co-60 eq. to air]	-0.309	-0.165	0.275
Land use [Annual crop eq.·y]	261	-8.01	1.37
Marine ecotoxicity [kg 1,4-DB eq.]	-0.0265	-0.138	0.0347
Marine Eutrophication [kg N eq.]	0.37	-0.00996	0.0288
Metal depletion [kg Cu eq.]	3.9	0.621	9.43
Photochemical Ozone Formation, Ecosystems [kg NOx eq.]	0.685	-0.411	0.407
Photochemical Ozone Formation, Human Health [kg NOx eq.]	0.657	-0.41	0.401
Stratospheric Ozone Depletion [kg CFC-11 eq.]	0.000813	-3.46E-06	3.66E-05
Terrestrial Acidification [kg SO2 eq.]	-0.563	-2.02	0.248
Terrestrial ecotoxicity [kg 1,4-DB eq.]	-209	-181	48

APPENDIX B - ABSOLUT VALUES OF IMPACT CATEGORIES AT THE DAMAGE PATHWAY AND ENDPOINT LEVEL

Midpoint Impact categories	Damage pathways	Comparative results			Endpoint area of protection	Comparative results		
		SC1	SC2	REF		SC1	SC2	REF
Fine Particulate Matter Formation [DALY]	**Increase in respiratory disease**	-1.59E-04	-4.26E-04	4.65E-05	**Damage to human health [DALY]**	-4.47E-04	-7.41E-04	2.23E-03
Photochemical Ozone Formation, Human Health [DALY]								
Ionizing Radiation [DALY]	**Increase in various types of cancer**	1.33E-06	-3.89E-07	1.23E-07				
Stratospheric Ozone Depletion [DALY]								
Human toxicity, cancer [DALY]								
Ionizing Radiation [DALY]	**Increase in other diseases/causes**	-1.48E-04	-1.62E-04	1.09E-03				
Stratospheric Ozone Depletion [DALY]								
Human toxicity, non-cancer [DALY]								
Climate change Human Health, default [DALY]								
Climate change Human Health, default [DALY]	**Increase in malnutrition**	-1.41E-04	-1.52E-04	1.09E-03				
Freshwater Consumption, Human Health [DALY]								

Midpoint Impact categories	Damage pathways	Comparative results			Endpoint area of protection	Comparative results		
		SC1	SC2	REF		SC1	SC2	REF
Climate change Freshw Ecosystems [species.yr]	**Damage to freshwater species**	1.26E-08	-3.59E-10	4.53E-09	**Damage to ecosystems [species.yr]**	-4.36E-07	-9.99E-07	3.40E-06
Freshwater Consumption, Freshw Ecosystems [species.yr]								
Freshwater ecotoxicity [species.yr]								
Freshwater Eutrophication [species.yr]								
Climate change Terrest Ecosystems [species.yr]	**Damage to terrestrial species**	-4.49E-07	-9.98E-07	3.40E-06				
Freshwater Consumption, Terrest Ecosystems [species.yr]								
Photoch. Ozone Formation, Ecosystems [species.yr]								
Terrestrial ecotoxicity [species.yr]								
Terrestrial Acidification [species.yr]								
Land use [species.yr]								
Marine ecotoxicity [species.yr]	**Damage to marine species**	-2.79E-12	-1.45E-11	2.82E-12				

Midpoint Impact categories	Damage pathways	Comparative results			Endpoint area of protection	Comparative results		
		SC1	SC2	REF		SC1	SC2	REF
Metal depletion [$]	**Increased extraction costs**	9.02E-01	1.44E-01	2.18E+00	**Damage to resource availability [$]**	1.83E+00	3.75E+00	9.77E+00
Fossil depletion [$]	**Oil/gas/coal energy cost**	9.29E-01	-2.21E+00	7.59E+00				