

# UNIVERSIDADE ESTADUAL DE CAMPINAS FACULDADE DE ENGENHARIA DE ALIMENTOS

# SAMUEL FONTENELLE FERREIRA

# ANÁLISE TÉCNICO E AMBIENTAL DA DIGESTÃO ANAERÓBIA DE RESÍDUOS ORGÂNICOS

# TECHNICAL AND ENVIRONMENTAL ANALYSIS OF ANAEROBIC DIGESTION OF ORGANIC RESIDUES

Campinas 2021

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## TECHNICAL AND ENVIRONMENTAL ANALYSIS OF ANAEROBIC DIGESTION OF ORGANIC RESIDUES

Thesis presented to the Faculty of Food Engineering of the University of Campinas in partial fulfillment of the requirements for the degree of PhD in Food Engineering

Tese apresentada à Faculdade de Engenharia de Alimentos da Universidade Estadual de Campinas como parte dos requisitos exigidos para a obtenção do título de Doutor em Engenharia de Alimentos

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#### **RESUMO GERAL**

Diversos estudos mostram que os resíduos provenientes da indústria de celulose e papel e das principais culturas agrícolas (cana de açúcar, açaí, laranja, soja, milho, dentre outras) têm um alto potencial para a produção de biocombustíveis. O Brasil possui grandes fontes de biomassa para aplicação de tecnologias renováveis de conversão que podem agregar valor à cadeia de suprimentos alimentícia e se alinhar ao desenvolvimento de uma bioeconomia. O objetivo deste trabalho foi analisar o impacto técnico-econômico e ambiental da valorização de resíduos orgânicos por meio da digestão anaeróbia. Além disso, estudouse o potencial de geração de energia do biogás e rotas tecnológicas para a gestão de resíduos no conceito de economia circular. Os resultados obtidos no estudo de adoção de reatores UASB (Upflow Anaerobic Sludge Blanket) pela indústria de celulose mostraram, por meio de uma análise financeira, que o investimento necessário para uma plataforma de biorrefinaria seria devolvido em 6,4 anos com um alto retorno sobre o investimento e poderia evitar  $1,06 \times 10^5$  CO<sub>2eq</sub> toneladas, contribuindo efetivamente para a descarbonização da economia do Brasil. O estudo de avaliação de quatro categorias de impactos ambientais (acidificação, eutrofização, oxidação fotoquímica e mudanças climáticas) ao substituir o uso de um combustível fóssil tradicional por biometano mostraram que a substituição do óleo Diesel em veículos pesados foi benéfica em todas as categorias de impacto, a substituição de gás liquefeito de petróleo em fornos a gás impactou positivamente apenas as mudanças climáticas e a substituição de gasolina-C em veículos leves foi desvantajosa. A análise de contribuição mostrou que a queima do combustível foi o processo mais relevante para todas as categorias de impacto e forneceu dados para uma análise mais aprofundada do ciclo de vida completo do biometano, em uma abordagem do poço à roda completo. O estudo da gestão de resíduos da indústria brasileira açaí mostrou uma nova rota tecnológica para a esses resíduos, visando a valorização por meio da bioenergia, com base no conceito de economia circular. A partir de 1 tonelada de açaí alimentada na unidade de processamento, produz-se 1,2 tonelada de resíduos sólidos e efluentes e o processo industrial completo exige 25 kWh por tonelada de polpa de açaí produzida. Esses resíduos quando tratados em reatores anaeróbios podem produzir 2,77 m<sup>3</sup> de biogás e a energia gerada poderia ser reciclada e aproveitada no processo, no qual, cerca de 61% da necessidade de eletricidade externa pode ser reposta a partir do biogás produzido, estabelecendo uma economia energética circular para o setor e contribuir para a descarbonização. Conclui-se que a implementação de reatores anaeróbios para o tratamento de resíduos orgânicos poderia apoiar a transição para uma economia circular, com benefícios ambientais, sociais e econômicos para o desenvolvimento sustentável.

**Palavras chaves:** Fábrica de celulose e papel; Reator UASB; Biogás; Mitigação de GEE; Avaliação de Ciclo de vida; Economia circular.

#### ABSTRACT

Several studies showed that residues from paper and pulp and several agricultural cultures (sugarcane, açaí, orange, soybean, corn, etc.) have a high potential for producing biofuels. Brazil has several sources of biomass for application in renewable conversion technologies that can add value to the food supply chain and align with the development of a bioeconomy. The main objective of this work was to analyze the technical-economic and environmental impacts of the valorization of organic wastes through anaerobic digestion. Besides that, it studied the potential for energy generation from biogas e technological routes for waste management within the concept of a circular economy. The results obtained in the study about the adoption of UASB (Upflow Anaerobic Sludge Blanket) reactors by the pulp and paper industry showed, using financial analysis, that the needed investment to build a biorefinery platform would be returned in 6.4 years, with a high return on investment and could avoid  $1.06 \times 10^5$  CO<sub>2eq</sub> metric tons, effectively contributing to the decarbonization of Brazilian economy. The study about the assessment of four environmental impact categories (acidification, eutrophication, photochemical oxidation and climate change) when replacing a standard fossil fuel by biomethane showed that the replacement of Diesel oil in heavy-duty vehicles was beneficial in all impact categories, the replacement of liquefied petroleum gas had a positive impact only in climate change and the replacement of gasoline-C was disadvantageous. The contribution analysis showed that the burning of fossil fuel was the most relevant process to all impact categories and supplied data for deeper life cycle assessment of the full biomethane life cycle, with a full well-to-wheel approach. The study assessing the management of wastes in the açaí industry presented a new technological route for these residues, aiming for the valorization using bioenergy, within the concept of a circular economy. From one ton of açaí fed in the processing facility, 1.2 tons of liquid and solid wastes are produced and the whole industrial process demands 25 kWh per ton of acaí

pulp produced. These residues, when treated in anaerobic reactors, can produce 2.77 m<sup>3</sup> of biogas, and the energy generated could be recycled and used in the process, which can have 61% of its external electricity needs can be replaced by the biogas produced, stablishing an energetic circular economy for the sector and contributing the decarbonization. It can be concluded that the implementation of anaerobic reactors for the treatment of organic residues can underpin the transition to a circular economy, with environmental, social, and economic benefits for the sustainable development.

*Keywords:* Pulp and Paper Mill; UASB Reactor; Biogas; GHG mitigation; Life Cycle Assessment; Circular Economy.

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Capítulo 1 - Introdução geral, Justificativa, Objetivos e Estrutura da Tese

## 1.1. Introdução Geral

Avaliações mostram alto potencial de resíduos provenientes da indústria de celulose e papel e das principais culturas agrícolas (cana de açúcar, açaí, laranja, soja, milho, dentre outras) para produção de biocombustíveis. O Brasil, possui grandes fontes de biomassa vegetal para aplicação de tecnologias renováveis de conversão de biomassa em subprodutos, que podem agregar valor à cadeia de suprimentos alimentícia e se alinhar ao desenvolvimento de uma bioeconomia (Forster-Carneiro *et al.*, 2017; Dos Santos *et al.*, 2018).

De acordo com a Indústria Brasileira de Árvores (Ibá, 2017) o Brasil é o segundo produtor de celulose o oitavo produtor de papel no mundo. A celulose é produzida a partir de madeira (no Brasil a principal fonte é o eucalipto), que passa for um tratamento usando uma substância fortemente alcalina, o licor branco, que separa a celulose da lignina. A celulose assim separada precisa passar por diversos tratamentos, principalmente secagem e branqueamento, até ser transformada em seu produto final, papel ou derivados (Souza, 2008). O setor de papel e celulose é um setor considerado energointensivo, pois demanda muita energia tanto na parte florestal, na forma de combustível na colheita e transporte do eucalipto, quanto na parte industrial na forma de calor força motriz. As usinas de celulose possuem uma caldeira de biomassa, onde são queimados os rejeitos de madeira, essa caldeira gera energia para a fábrica, o que torna algumas autossuficientes em energia (Bajpai, 2016). O processamento do açaí (*Euterpe Oleracea Martius*), cuja fruta tem baixa quantidade de polpa (parte usada para o consumo), correspondendo a apenas 10% da massa do açaí, sendo o resto semente e as fibras. No Brasil, ainda não há um sistema de recuperação em larga escala desses resíduos que possuem alto poder calorífico (Matos et al., 2011; Virmond et al., 2012; Yuyama et al., 2011).

A digestão anaeróbia é uma tecnologia usada no tratamento de resíduos e no tratamento secundário de águas residuárias. É uma tecnologia bastante atraente pois não necessita aeração, o que diminui o gasto de energia, possui baixa geração de lodo se comparada com o tratamento aeróbio e gera biogás, que pode ser usado para geração de energia (Wellinger *et al.*, 2013; Bernal *et al.*, 2017).

O biogás vem sendo explorado como fonte de energia renovável, pois possui alto poder calorífico devido a sua alta concentração de metano (aproximadamente 60%). Antes de ser utilizado, o biogás precisa passar por um processo de dessulfurização, visando remover o ácido sulfídrico, que pode corroer os equipamentos. O biogás pode ser queimado diretamente ou enriquecido para aumentar seu conteúdo de metano. incluindo dessulfurização (aproximadamente 94%), o chamado biometano. Nessa forma pode substituir o gás natural em praticamente todas as suas aplicações, tais como substituto do diesel em veículos pesados, gasolina em veículos leves e injeção na rede de gás natural (Deublein e Steinhauser, 2011; Becher, 2016). O biogás também pode ser utilizado para a geração de eletricidade graças à Resolução Normativa nº 482/2012 da ANEEL (Agência Nacional de Energia Elétrica) que permite a geração distribuída de energia elétrica. Esse sistema permite a micro geração de energia elétrica e compensação financeira através de sistemas de crédito (Gomes *et al.*, 2017).

Existem diversas metodologias de análise de impacto ambiental, porém para o seguinte trabalho foi considerada a Análise de Ciclo de Vida (ACV), principalmente por ser adequada para a comparação de produtos e tecnologias e por ser padronizada pela International Organisation for Standardisation (ISO) (Hauschild, 2015) É uma metodologia que considera os produtos ou serviços desde sua extração no ambiente ("berço") até seu descarte ("túmulo"). (Guinée *et al.*, 2004). Esta metodologia considera os impactos ambientais resultantes de uma série de etapas do ciclo de vida do produto, a partir de uma unidade funcional, logo diretamente relacionada a uma dada função do produto. Para o uso adequado da ACV é necessário montar um inventário de emissões de cada etapa e a partir de uma série de fatores de impacto, calcular o impacto de cada categoria. (Guinée *et al.*, 2004). São poucos os trabalhos na literatura tratando da obtenção da avaliação de sustentabilidade dos resíduos procedentes da indústria de papel e celulose e resíduos agrícolas, tais como açaí.

Resíduos agroindustriais também podem ser uma matéria-prima potencial para bioenergia. A produção de biocombustíveis renováveis a partir de biomassa pode ser realizada usando diferentes rotas tecnológicas, como a conversão material lignocelulósico para produzir açúcares fermentáveis, bioetanol, ácidos orgânicos, compostos bioativos e biogás. Os resíduos procedentes da indústria de celulose e papel, do processamento de açaí e resíduos orgânicos em geral foram avaliados e possíveis rotas tecnológicas foram sugeridas, contemplando o conceito de economia circular e redução da pegada de carbono.

Este trabalho apresenta rotas de valorização de resíduos agroindustriais (focado nas indústrias de papel e celulose e nas indústrias de processamento de açaí) visando produção de biogás para geração de energia, bem como o impacto ambiental de diversas rotas de utilização do biogás como fonte de energia.

## 1.2. Objetivos

## 1.2.1 Objetivo Geral

O objetivo principal deste trabalho foi analisar o impacto técnico-econômico e ambiental da valorização de resíduos orgânicos através da digestão anaeróbia. Adicionalmente, analisar o potencial de geração de energia do biogás em diversos cenários e analisar rotas tecnológicas para a gestão de resíduos no conceito de economia circular.

#### 1.2.2. Objetivos específicos

Avaliar o impacto ambiental e financeiro da adoção de um reator UASB em uma usina de papel e celulose: estudo de caso;

Avaliar os impactos ambientais do uso de biogás na forma de biometano em 3 cenários (uso doméstico, veículos leves e veículos pesados, usando metodologia de Análise de Ciclo de Vida;

Avaliar a produção brasileira de açaí, balanços de massa e energia decorrentes de seu cultivo, extração, processamento e resíduos disposição;

Analisar uma nova rota tecnológica para a gestão de resíduos do açaí visando recuperação de bioenergia com base no conceito de economia circular.

#### 1.3.Estrutura da Tese

Esta tese se encontra dividida em capítulos. Os capítulos apresentam os resultados experimentais correspondem artigos que estão publicados ou estão sendo revisados em revistas científicas da Área de Engenharia de Alimentos.

O Capítulo 1 é composto pela introdução, que descreve o tema central da tese, fazendo uma exposição, de forma sucinta dos pontos mais relevantes, os objetivos e estrutura da tese.

O capítulo 2 se trata de um estudo de caso da utilização de um reator UASB em uma planta de celulose e papel e descreve os resultados obtidos em uma planta de celulose e papel localizada em Lençóis Paulistas, após a instalação de um reator UASB para tratamento secundário de efluentes e a análise ambiental e financeira dessa utilização. O desafio referente à indústria brasileira de celulose e papel, estava relacionado aos efluentes que não são usados para gerar energia, resultando na queima de biogás em flares. Desta forma o trabalho desenvolveu uma estrutura conceitual "sistêmica" para a engenharia de uma biorrefinaria, acoplada a uma estação de tratamento de efluentes de uma fábrica de celulose e papel, projetada com um reator UASB para tratamento secundário de efluentes e posterior recuperação de energia em caldeiras de biogás para geração térmica e eletricidade.

O Capítulo 3 aborda um estudo dos usos finais do biogás na forma de biometano e análises de impacto ambiental de 3 rotas para o uso de biometano (fogão a gás, veículos leves e veículos pesados) aplicando a metodologia de avaliação do ciclo de vida). Desta forma, quatro categorias de impacto foram avaliadas: acidificação, mudança climática, eutrofização e oxidação fotoquímica, ao substituir um uso tradicional de combustível fóssil por aqueles usos finais de biometano.

O Capítulo 4 abordou uma avaliação de impacto ambiental da produção brasileira de açaí, com foco na geração de resíduos, balanços de massa e energia decorrentes de seu cultivo, extração, processamento e disposição final. Uma nova rota tecnológica para a gestão de resíduos do açaí foi introduzida para recuperação de bioenergia com base no conceito de economia circular.

O Capítulo 5 apresenta uma discussão dos principais resultados obtidos neste trabalho, enquanto o capítulo 6 apresenta as conclusões gerais e sugestões para trabalhos futuros.

O Capítulo 7 apresenta uma memória do período de doutorando com todos os trabalhos acadêmicos realizados paralelamente ao desenvolvimento desta tese.

Por fim, o capítulo 8 contém uma lista com todas as referências utilizadas nesta tese, e em Anexos consta Copyright dos artigos.

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# Capítulo 2 – Estudo de caso: Utilização de um reator UASB em uma planta de celulose

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# An integrated approach to explore a UASB reactor for pulp and paper industry energy recycling: case study in Brazil

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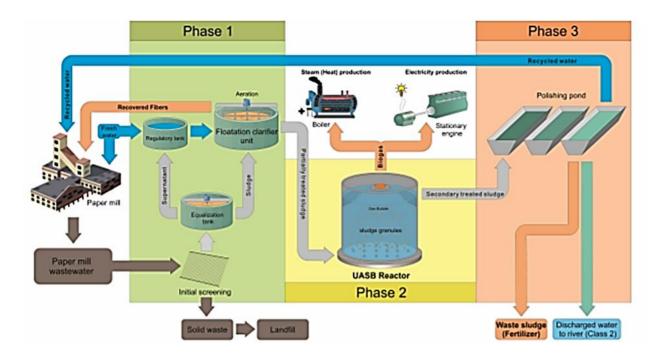
#### Abstract

Brazil is currently focused on its energy matrix transition in favor of increasing of the share of renewable energy carriers for both enhanced energy security and mitigation of greenhouse gas emissions. In this context, the country's pulp and paper industry whose different wastes teams are not generally exploited, could play a critical role. Accordingly, the main objective of this work is to develop a conceptual 'systemic' biorefinery framework integrating the treatment of pulp and paper mill wastewater using upflow anaerobic sludge blanket (UASB) reactor with energy recovery through biogas production and its conversion into heat and power in stationary engines and boilers, respectively. Based on the results obtained through the present case study, it was revealed that the adoption of UASB reactors by the paper mill industry could properly addresses the environmental concerns

faced while could contribute to the national agenda favoring an increasing share of renewable energies in the country's energy matrix. The financial analysis showed that the investment required for the implementation of UASB reactors within a biorefinery platform would be minor vs. the investment in the whole mill and would be returned in 6.4 yr with a high return on investment even when operated at half of operational capacity. Moreover, through the developed UASB reactor-based biorefinery, the Brazilian pulp and paper industry, as a whole, could avoid  $1.06 \times 105$  CO2eq tons, effectively contributing to the decarbonization of the country's economy.

Keywords: Paper Mill, Wastewater, Anaerobic Digestion, Biogas, Energy Recovery.

# **Graphical Abstract**



## 1. Introduction

Bioenergy produced from biological resources and different biomasses, rich in carbon structures, including wood and agricultural wastes can be applied to the production of heat, electricity or fuels contributing to the global decarbonization. In developing countries, the large scale adoption of energy recycling strategies for several industrial wastewater and residues is still a challenge when compared to developed countries that have already embraced technological solutions to properly deal with byproducts, i.e. side products derived of the processing of the main product, and wastes (dos Santos et al., 2018). Energy matrix transition in developing countries, such as Brazil, lacks an effective strategy to provide extra value to wastewater and residues in a virtuous energy recycling, mainly because of the great dependence on fossil fuels (57% of the whole domestic energy supply) and/or hydro and thermoelectric energy (65.2% and 27.8%, respectively, of the domestic electricity supply including renewable and non-renewable sources) (EPE, 2018). Meanwhile, Brazil presents an expressive availability of biomass-to-energy from several sources (Welfle, 2017).

Brazilian efforts to increase the production and use of renewable energy were strongly accelerated after the 21<sup>st</sup> Conference of the Parties to the United Nations Framework Convention on Climate Change (COP 21) and after the establishment of RenovaBio - a state policy that aims to draw up a joint strategy to recognize the strategic role of all types of bioenergy in the Brazilian energy matrix, both for energy security and for mitigation of greenhouse gas (GHG) emissions (MME, 2018b). At COP 21 Brazil has committed to reduce GHG emissions to 37% less than 2005 by 2025 and to 43% less by 2030 (MMA, 2018).

The challenge extends to Brazilian wood-based industry, including pulp and paper production, which is locally important and reaches more than 6% of the National Industrial Gross Domestic Product (IBÁ, 2017). In this industry, wastewater sludge, produced in high quantities, is difficult to handle because of its high water content (55 to 85%) and low dewaterability (Berni et al., 2014; Bayr, 2014; Foekel, 2010). Pulp and paper mill wastewater usually contain inks, dyes, very high level of Biochemical Oxygen Demand (BOD) (20 to 40mg/L) or Chemical Oxygen Demand (COD) (300 to 500 mg/L), due to the presence of lignin and its derivatives from the raw cellulosic materials, chlorinated compounds, suspended solids (mainly fibers), fatty acids, tannins, resin acids, sulphur and sulphur compounds, etc. (Ali and Sreekrishnan, 2001; Souza, 2008). The discharge of insufficiently treated wastewater into the rivers or streams leads to serious side effects to aquatic life, to flora and fauna nearby the facilities, as long as well, the discharge in domestic wastewater treatment systems requires additional steps for the proper accomplishment of country's standards. Thereby solutions for effluent organic matter removal before any discharge pathways are necessary (Kesalkar et al., 2012). Moreover, in Brazil, pulp and paper industry effluents are not usually destined to generate energy, even with the presence of wastewater pretreatment systems, resulting in biogas burning in flares.

In order to better address these concernments, the adoption of technologies more efficient in organic matter removal and, in addition, focused on biogas conversion into energy is required. Anaerobic digestion (AD), the biological degradation of organic matter into methane (50–75%), carbon dioxide (25–50%), hydrogen (5–10%), and nitrogen (1–2%) (Maghanaki et al., 2013) is been widely adopted worldwide for sewage sludge treatment and, on the other hand, its dissemination for industrial effluents faces limitations in anaerobic reactors configuration and operating conditions that makes the technology feasible for large scale application. The Upflow

Anaerobic Sludge Blanket reactor (UASB), among other types of anaerobic reactors, is been commonly adopted by pulp and paper industry since 1980's when its main weakness, hydraulic retention time (HRT), was overcame (Kamali et al., 2016). The biogas produced trough AD can be converted into bioenergy by means of combined heat and power systems. Despite this, in Brazil, UASB reactors are not disseminated in paper and pulp industry because there are limited studies on integrated biorefinery in the paper industry, as well as, economic and environmental assessments that clearly reinforce its advantages. Anaerobic treatment in the pulp and paper industry began in the 1970s, the first system being the anaerobic lagoon type, later UASB reactors emerged in the 1980s, and from the 1990s the real-scale anaerobic treatment predominates in the treatment of effluents from pulp mills around the world (Savant et al., 2006). Brazil presents a leading role in the utilization of UASB reactors and has the largest park of anaerobic reactors in the world for sewage treatment with and without of post-treatment and the trend is that their utilization will continue to grow (Maghanaki et al., 2013).

Notwithstanding, Brazilian biogas market is still in initial development phase, but steadily growing. The Brazilian National Policy on Solid Residues (PNRS), approved by Law 12.350/2010 (Brasil, 2010) and

planned to be fully enforced by the end of 2022, should help the expansion of biogas plants, even those designed for wastewater treatment. The PNRS's goal is to avoid and prevent generation of solid residues by promoting sustainability, increasing recycling and proper final disposal while sharing responsibilities with the whole society, namely government, producers, sellers and consumers (Esparta, 2016). The Brazilian National Electric Energy Agency (ANEEL) has a record of biogas plants producing electricity connected to the distribution system. However, this record does not include information on other biogas plants potential energy suppliers. Despite the large size of the Brazilian territory, collection of biogas plant information from other plants not registered at ANEEL would demand a major effort. In Brazil there are 22 biogas power plants connected to the electric grid. The majority of biogas plants are installed on agricultural properties, processing residues, and at landfills (Persson and Baxter, 2015). In addition, ANEEL is responsible for the regulatory aspects of the electricity sector. This agency has the power to decide and encourage research & development projects in the main areas of social interest, among them; those related to wider and better energy use of nonconventional energy sources, such as the ones derived from liquid effluents (ANEEL, 2008, MME, 2018b). According to the ANEEL,

biomass is considered one of the main alternatives for diversification of energy sources, thereby reducing dependence on fossil fuels.

The first economic aspect to be considered is the use of a clean and low-cost fuel in substitution of electric and thermal power, such as the biogas and its possible energy applications (electricity and thermal energy generation). Distributed generation is currently seen as the solution to several problems, as well as the source of several benefits related to reliability, power quality and environmental issues. For this reason, future electrical power grids tends to be decentralized and to include a large concentration of small and medium sized distributed plants (Ramos et al., 2014). Systemic important aspects, including economic, social and environmental approaches, should be considered for biomass-to-energy projects: i) the rational strategies pertaining to the protection of natural resources; ii) the potential to promote the replacement of non-renewable energy for clean energy; iii) economic viability for energy recovery (Novato and Lacerda, 2017).

The main objective of this work is to develop a conceptual 'systemic' framework for the engineering of a biorefinery, based on data obtained from a study case, coupled to a wastewater treatment plant of a pulp and paper mill,

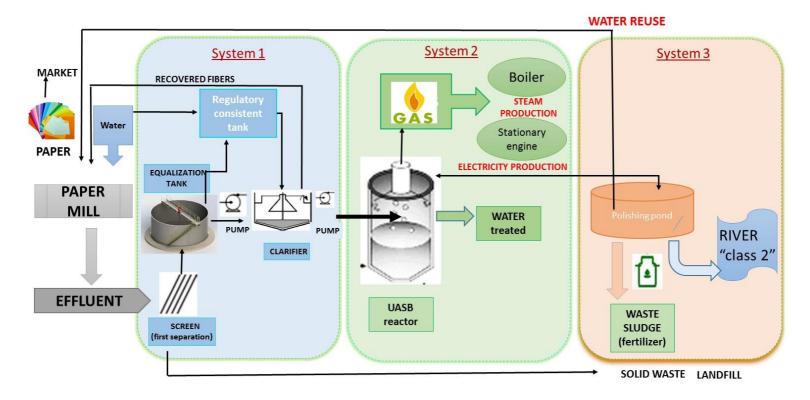
designed with a UASB reactor for the secondary wastewater treatment and subsequently biogas energy recovery in boilers for thermal (TP) and electricity power generation (EP).

#### 3. Materials and Methods

#### 3.1. Study case in a paper mill industry in São Paulo State-Brazil

The study case was developed for a paper mill located in São Paulo state, Brazil, which has already invested USD 3.4 millions in an advanced wastewater treatment (WWT) from 2004 to 2014. The first UASB reactor was incorporated into the existing effluent treatment system at the mill in 2004. In 2014, a second UASB reactor was incorporated. The primary treatment consists in an equalization tank and effluent flotation. The secondary treatment consists of UASB reactors (UASB 1 and 2) and finally the tertiary treatment is done in an aerobic polishing pond.

The three wastewater treatment (WWT) systems and the biogas output for the study are case presented (Figure 1). System 1 is the conventional aerobic system with primary settling and sludge digestion after treatment. System 2 is a combined anaerobic digestion (AD) and two UASB reactors, the main structure of the biorefinery, where biogas is destined to a boiler (to produce steam for heating) and to a stationary engine (to electricity generation). System 3 is an additional sludge digestion post-treatment with a polishing pond. This system removes contaminants in accordance with environmental regulations, offering the possibility of using the treated effluent as reuse water in the production. Furthermore, UASB reactor bottom sludge could be applied as soil fertilizer or could be further processed by pyrolysis to produce biochar which can act as soil conditioner (Buller et al., 2014; Elkhalifa, 2019) and activated carbon (Alhashimi, 2017).



**Figure 1.** Wastewater treatment (WWT) systems developed for a paper mill industry located in São Paulo State-Brazil.

The integration of the processes here presented can be classified as a biorefinery, according to an integrated approach as follows:

- Effluent treatment plant (primary treatment), followed by the production of biogas through UASB reactor;
- 2) Biogas producing steam and electricity for the industrial plant. This approach regards to the use of untreated biogas as a fuel in boiler for steam and electricity generation. The use of such biogas can decrease both Natural Gas (NG) consumption and overall CO<sub>2</sub> emissions.

The paper mill of this study case is settled for white top line (WTL) or newsprint only. The paper mill produces 12.9 ton per hour of WTL or newsprint, according to Equation 1, considering the following data: paper grade 150 g/m<sup>2</sup>, paper machine speed of 550 meters per minute and sheet width of 2.6 meter. For each ton of white top line (WTL) or newsprint produced there is a total water consumption of 30 m<sup>3</sup>.

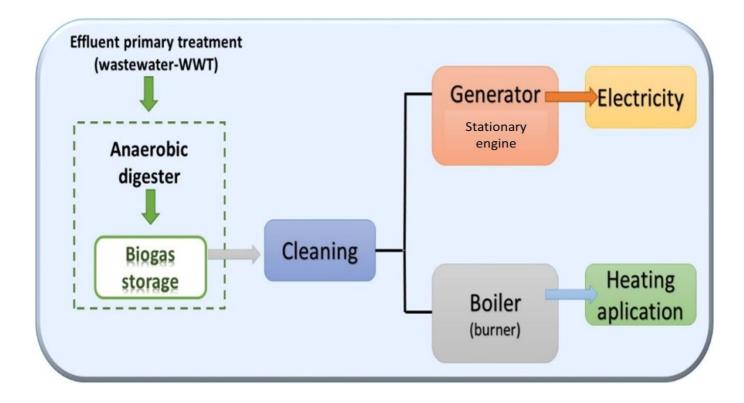
$$\frac{Production}{hour} \left(\frac{Kg}{h}\right) = \frac{\left[velocityPM\left(\frac{m}{min}\right)*paperweight\left(\frac{g}{m^2}\right)*sheetwidth\ (m)*60\ (min)\right]}{1000}$$

Eq. (1)

#### **3.2. Biogas production**

The energy value of biogas was determined by the average concentration of methane, which has the LHV of 35.59 MJ/Nm<sup>3</sup> (Salomon and Lora, 2009). The primary equipment commonly employed in the use of biogas as a fuel to produce electricity are internal combustion engines, mainly because of the costs of acquisition, operation and maintenance. The conversion process to the use of biogas in paper mill, with 70% of methane, as fuel is easier because most gas engines already use natural gas (NG). For the correct sizing of the moto generator group, the following parameters are considered: i) efficiency of the engine generator; ii) calorific value of biogas; and iii) generation plant working time, and iv) daily availability of biogas in the unit (m<sup>3</sup>/day).

Two routes for biogas destination that fit a biorefinery approaches are presented in **Figure 2**. One of them, the technical arrangement 'AD – UASB' for electricity production by using biogas in stationary engine, was built to treat effluents - that are released into the river - from the pulp preparation and for the paper machine (PM). The other possible route is to occupy the technical arrangement proposed for steam production by burning biogas in the boiler.





The volume of effluent to the primary treatment is  $9,288 \text{ m}^3/\text{day}$  and, the volume of effluent destined to the reactor AD - UASB is  $7,200 \text{ m}^3/\text{day}$ . The difference ( $2,088 \text{ m}^3/\text{day}$ ) remains in a closed loop, returning to pulp preparation and paper machine.

With 16 hours of hydraulic retention time (HTR) in AD-UASB, considering the measured efficiency of 20% in the floaters, the affluent Chemical Oxygen Demand (COD) is 1,800 mg/L (mean value of the physicochemical analyses of the unrefined effluent that reach the floaters (primary treatment) in 12 months). Meanwhile, Brazilian legislation discharge limit for COD of the final treated wastewater (affluent) is less than 340.0 mg/L. The pH remains from 6.7 to 7.8, the temperature is kept in 30°C (mesophilic reactor) and sediment residue (SR) is 100 mL/L.

Under such conditions the Organic Load Rate (OLR) for AD - UASB system is obtained from **Equation 2**:

$$OLR = V\left(\frac{m^3}{day}\right) * COD\left(\frac{kg COD}{m^3}\right)$$
 Eq. (2)

Considering 0.3 cubic meters of methane gas (CH<sub>4</sub>) for each kilogram of removed organic load (Torkian et al. 2003), as well as COD removal efficiency of 80%, the volume of biogas generated (Q) can be calculated as follows.

$$Q_{CH4} = 0.80 * OLR \left(\frac{kg COD}{day}\right) * 0.300 \left(\frac{m^3}{kg \, removal \, (OLR)}\right)$$
 Eq. (3)

As the biogas here produced has an average of 70 % of CH<sub>4</sub>, the production of biogas is given by **Equation 4**.

$$Q_{biogas} = \frac{Q_{CH4}}{0.70} \qquad \qquad \text{Eq. (4)}$$

## 3.3. Potential of electricity generation by using stationary engine

The electricity generation (EG) from a stationary engine fed by biogas can be calculated (Lymberopoulos, 2004, Lobato et al., 2012), according to **Equation 5**. The parameters values are presented in **Table 1**.

$$EG = Q_{biogas} * LCV_{CH4} * C_m * n_e * n_g * F_c \qquad \text{Eq. (5)}$$

Where,

EG - Potential of Electricity Generation (MJ/day);

- $Q_{biogas}$  Amount of biogas ( $m^3/day$ );
- LCV Lower calorific value of methane  $(MJ/m^3)$ ;
- $C_m$  Percentage of methane in biogas (%);
- $n_e$  Engine efficiency (%)
- $n_g$  Generator efficiency (%)

 $F_c$  - Correction factor due to uncertainties (%). This factor take into account the losses in the pipes, mechanical couplings, the presence of other gases not fully quantified and other factors that lead to losses in the final energy.

**Table 1-** Values of the parameters to obtain the electricity generation (EG) and total energy produced (EP).

<i>LCV</i> - Lower calorific value of methane (MJ/m <sup>3</sup> )	n <sub>e</sub> (%)	С <sub>т</sub> (%)	n <sub>g</sub> (%)	F <sub>c</sub> (%)
35.59	34	70	95	90

#### 3.4. Replacement of Natural Gas by Biogas

The use of biogas for the sole purpose of heat generation is simpler than the previous alternative and less complex requirements are observed.

The total energy produced (EP) per day is calculated following Equation 6.

$$EP = Q_{biogas} * C_m * LCV_{CH4}$$
 Eq. (6)

Where,

EP - Energy produced (MJ/day); Q<sub>biogas</sub> - Amount of biogas (m<sup>3</sup>/day); C<sub>m</sub> - Percentage of methane in biogas (%); LCV - Lower calorific value of methane (MJ/m<sup>3</sup>).

## 3.5. Avoided Emissions of UASB reactor and biogas use

-Avoided GHG emissions from Electricity Generation (AGHG<sub>EG</sub>)

The electricity generated by burning all the biogas produced in a stationary engine was obtained according to **Equation 7**. The parameters were based on data from the Ministry of Science, Technology, Innovation and Communication (MCTIC, 2018). For the emission factor of electricity in the matrix, the mean of all the 12 month values, from January 2018 to December 2018, was applied, aiming to account for the energy supply seasonality. The resulting emission factor was 0.0786 ton of  $CO_2eq/MWh$  of electricity.

$$AGHG_{EG} = 0.0786 * EG * T$$
 Eq. (7)

Where, EG is the electricity generated converted to MWh and T is the time of operation of the plant during the year, in hours.

#### -Avoided GHG emissions from Heat Generation (AGHG<sub>HG</sub>)

For heat generation avoided emissions it was assumed that the total volume of biogas would be burnt in the existing boiler as a renewable substitute of the Brazilian natural gas. Thus, the avoided emissions would be the amount of tons of  $CO_{2eq}$  emitted by the natural gas burned in the boiler. Emissions were calculated according to **Equation 8** (ECOPART, 2009).

$$AGHG_{EG} = 0.056 * HG * T \qquad \qquad \mathbf{Eq.} (8)$$

Where, 0.056 is the emission factor in tons of  $CO_{2eq}$  per GJ burnt, obtained from a Brazilian pulp and paper mill company environmental report, HG is the heat generated by burning all of the biogas and T is the time of operation in hours. The operation is 4,350 hours per year.

#### 3.6. Avoided Costs of UASB reactor and biogas use

#### -Avoided Cost from Electricity Generation ( $AC_{EG}$ )

When biogas is destined to electricity generation, part of the power needed for the paper mill operation can be replaced and an avoided cost can be calculated.

The price of electric power in Brazilian grid was based on data from the Electrical Energy Trade Chamber (CCEE, 2018) according to the following specifications: data from the Southeast Region, the region where the study case plant is located; data for the 52 weeks, one year period, from January 2018 to December 2018, to account for seasonality influence in market prices and prices refer to heavy-demand electricity sector. The mean value results resulted in 74.30 USD/MWh. The avoided cost from the electricity generation was calculated from **Equation 9**.

 $AC_{EG} = 74.30 * EG * T$  Eq. (9)

Where, EG is the potential of electricity generation converted to MW (refer to eq. 5, section 3.3) and T is the time of operation for the plant during the year, in hours. In order to reach the value of the avoided costs a working time of 4,350 hours per year was considered. This value corresponds to the real occupation of the WWT system (50% of the whole capacity) in the study case plant.

## -Avoided Costs from Heat Generation (AC<sub>HG</sub>)

The avoided costs from natural gas (NG) replacement by biogas is calculated by considering the volume of natural gas that was displaced, since natural gas is charged by the consumed volume, according to data from the Mines and Energy Ministry (MME, 2018).

The variable cost of NG is related to the dimension of the consumed volume. The plant consumes 150 m<sup>3</sup> of NG, per ton of paper produced which is equivalent to 526.5 m<sup>3</sup>/h. For this magnitude of consumption, according to Comgás (a Brazilian gas supplier), the variable cost is 0.38 USD/m<sup>3</sup> (Comgás, 2019)

According to the Mines and Energy Ministry (MME) the lower heating value (LHV) of the NG consumed is 36.84 MJ/kg, or 27.26 MJ/m<sup>3</sup>, assuming the stated density of 0.74 kg/m<sup>3</sup> (MME, 2018a)

The avoided emissions were obtained from Equation 10.

$$AC_{HG} = VC_{gas} * \frac{HG*T}{LHV_{gas}}$$
 Eq. (10)

Where, Natural Gas (HG) is the heat generated by the biogas, T is the time of operation and  $LHV_{gas}$  is the lower heating value of natural gas. Refer to **Supplementary Materials** for more information on emissions and prices.

#### 3.7. Financial analysis

Economic and financial analysis applies to obtain viability, stability and profitability indicators for projects or investments. Balance sheets, assets and resources information, net income and cash flow statements are some of the necessary information to obtain indicators that can support decision making processes. The difference between financial and economic analysis relies on whose profitability the evaluation measures. Financial analysis focuses on the business profitability and provides information for possible investors, while the economic analysis deals with public profitability and it is linked to opportunity costs. For this study case, a financial analysis was done, where two profitability indicators (a) and (b) were obtained to assess the risk assessment along with the classic payback (Blank and Tarkin, 2008).

(a) Profit margin, the ratio of net incomes and revenues (net profits over sales). It measures the % of sales that incorporates company's earnings. It is very useful to compare different companies' performance;

(b) Return on assets (ROA) indicates the profitability (relative to the total assets). It is calculated as annual earnings over total assets;

(c) Payback is the period, in years, required to pay for the original investment.

The analysis was done considering the mean paper price in the market of USD 450/ton of paper, the following financial parameters for the study case:

- UASB acquisition cost=USD 3,4millions;
- Financing time of 120 months (10years);
- Payment = Main stream + Interest rate;
- Interest rate = Financing cost + Bank Spread + Financial Agent;
- Spread Interest rate = 9.25% per year;
- Financing cost = Long Term Interest Rate = 6.85% per year;
- Bank Spread =1.40% per year;
- Financial Agent Spread =1.00% per year;
- Depreciation horizon for UASB = 25 years (financial depreciation rate of 3%);
- Intangible Assets =10% of acquisition cost (payment rate in installments fund, on intangible assets of 3%).

Refer to Supplementary Materials for more information on the calculations

## 4. Results and Discussion

## 4.1. Technical results

This study case was developed for a paper mill located in São Paulo state, Brazil and developed a conceptual 'systemic' framework for the engineering of a biorefinery, coupled to a wastewater treatment plant of a pulp and paper mill, with a secondary wastewater treatment and subsequently biogas energy recovery in boilers for thermal (TP) and electricity power generation (EP). There are three wastewater treatment (WWT) systems (**Figure 1**). There are a conventional aerobic system with primary settling and sludge digestion after treatment; a combined anaerobic digestion (AD) and two UASB reactors; and finally an additional sludge digestion post-treatment with a polishing pond.

The technical-analytical parameters calculated for the pulp and paper industry WWT systems with UASB reactors are: Organic Loading Rate (OLR), Amount of Methane (Q<sub>CH4</sub>), Amount of Biogas (Q<sub>biogas</sub>), Electricity Generation (EG) and Energy Produced (EP) (**Table 2**).

 Table 2- The analytical technical parameters calculated in the pulp and paper industry of the

 UASB reactor (Equations 1 to 6).

Result	Value	Unit
OLR- Organic Loading Rate	12,960	kgCOD/day
Q <sub>CH4-</sub> Amount of Methane	3,110	m <sup>3</sup> /day
Qbiogas-Amount of Biogas	4,443	m <sup>3</sup> /day

EG- Electricity Generation	32,178.25	MJ/day
EP- Energy Produced	8.94	MWh/day

The organic loading rate (OLR) must be analyzed constantly because it interferes significantly with the microbial ecology and the characteristics of the UASB reactor (Torkian et al., 2003). In the anaerobic treatment of wastewater, the loading rate was analyzed and the results for pulp and paper industry WWT system indicate a significant reduction of the Chemical Oxygen Demand (COD) (values of approximately 40.0 mg / L), even at high organic load rates (OLR of 12,960 kg COD / day) in short hydraulic retention time (**Table 3**). On the other hand, secondary sludge production, which is a byproduct of biological treatment, should be considered at higher concentrations due to increased COD loading and improved removal of suspended solids. According to (Mahmood and Elliott, 2006) secondary sludge is much more difficult to dehydrate than primary sludge, and most pulp and paper facilities extract a mixture of primary and secondary sludge.

Table 3- Chemical Organic Demand (COD) (mg/L) of effluent white top line (WTL) production
for systems treatment.

Month	System 1		System 2		System 3	
	(floaters)		(AD-UAS	Bs)	Lagoon	
	Before	After	Before	After	Before	After
January	2,672	2,138	2138	321	321	289
February	3,520	2,816	2816	422	422	380
March	2,260	1,808	1808	271	271	244
May	2,305	1,844	1844	277	277	249
June	2,064	1,651	1651	248	248	223
July	2,786	2,229	2229	334	334	301
August	2,150	1,720	1720	258	258	232

September	2,331	1,865	1865	280	280	252
October	2,387	1,910	1910	286	286	258
November	2,230	1,784	1784	268	268	241
December	2,290	1,832	1832	275	275	247

Brazil has two specific laws for the release of industrial and domestic effluents into water bodies, a Federal one (MMA, 2011) and other more restrictive for São Paulo State (CETESB, 1976). The Federal law establishes that the maximum biological oxygen demand (BOD) is 120 mg/L for final disposal on the water body, and that this limit can only be exceeded in the case of treatment system effluent with a minimum removal efficiency of 60% of BOD, or by means of a self-purification study of the water body that proves to meet the goals of the receiving body (MMA, 2011). Otherwise, in São Paulo State there is a specific legislation, which limits the BOD of 5 days, at 20°C, to a maximum of 60 mg/L (sixty milligrams per liter), and this limit may be exceeded in the case of effluent wastewater treatment system that reduces the pollutant load by at least eighty percent (80%). This goal of reduction can be extended to COD as well, since both of them are related to organic load.

**Table 3** shows the COD values of white top line (WTL) production effluents, and it is possible to observe a range between 223 to 380 mg/L throughout the year after the polishing pond (tertiary treatment), or System 3 (Figure 1). These values are acceptable considering that they were calculated through COD, which evaluates the amount of dissolved oxygen (**DO**) consumed in acidic environment that leads to the degradation of organic matter, being it biodegradable or not (Rajeshwari et al. 2000). In this way, their values are higher than those established by the legislation, which was set in terms of biological oxygen demand (BOD). According to (Meyer and Edwards, 2014) the rates of removal of COD in real-scale reactors vary between 30 to 90% and

CH<sub>4</sub> production ranges from 300 to  $400\text{m}^3\text{g}^{-1}$  of COD removed, with the highest COD removal rates being obtained with condensate flows from chemical pulp (75 to 90%) and paper mill effluent (60 to 80%).

Literature on UASB reactors has increased substantially and several international publications (Khan et al., 2011; Isola et al., 2018). Zhang et al. (2012) proposed five scenarios for wastewater treatment of pulp and paper industry with primary treatment, biological treatment (UASB), sequential batch reactor treatment and oxidation ditch reactor, anaerobic / oxide process, treatment tertiary water reuse and membrane technologies and the results show that this environmental goal can be met by joint efforts to implement any treatment technology following an integrated process prevention with pollution removal at the end of the process.

Von Sperling (2016) describes the major points of the Brazilian national standards for water quality and effluent discharge (MMA, 2005). These Brazilian national standards are used as a basis for licensing of new industrial activities or domestic sewage discharge, and are the reference for Environmental Impact Assessment (EIA) studies, which regulates all of the activities that have previously demonstrated that the legislation will be complied. The classification of water bodies has already been undertaken for many catchment areas in Brazil, but the majority remains classified as Class 2. The adoption of UASB reactor for the study case results in a very efficient wastewater treatment system that fully complies with Brazilian laws. Whereas, the classification of water bodies has already been undertaken for many catchment areas in Brazil the dissemination in paper mill industries fails in the adoption of such efficient system. In the whole country only a few players of this industry adopt UASB.

#### 4.2. Avoided GHG emissions, avoided costs and financial results

The integration of the processes here presented can be classified as a biorefinery, where the biomass energy recovery and/or recycling can be maximized through several conversion technologies that can be jointly applied to different feedstocks to produce a wide range of bioproducts (Cherubini, 2010). Anyway, the use of sustainable feedstock is not enough to ensure the sustainability of the conversion technologies. In the biorefinery concept, the optimization of the biomass use for biomaterials, fuels and energy applications require economic and environmental assessments to fully fulfill the sustainable status (Loftus et al., 2015).

According to the integrated approach of effluent treatment plant (primary treatment), followed by the production of biogas through UASB reactors and its further use for steam and electricity generation to supply energy to the production process (the paper mill of this study case is settled for white top line (WTL) or newsprint production only), two routes for biogas destination were proposed to fit the biorefinery.

The internal combustion engine was already in used in the paper mill to produce electricity and it can be easily converted to burn biogas. Another proposed route was for steam production by burning biogas in the boiler.

From the data obtained for biogas volume produced ( $Q_{biogas}$ ) and its potential for electricity generation (EG) and for energy production for heat generation (HG) the avoided emissions for both alternatives were separately obtained.

The results of the avoided emissions for the substitution of natural gas for biogas for heat generation are is expressively higher than for electricity generation, according to **Table 4**.

**Table 4-** Avoided emissions and costs from replacement of natural gas for biogas.

		Electricity Generation	Heat Generation
		(EG)	(HG)
Avoided (tCO <sub>2eq</sub> /year)	emissions	127.0	1,125.5
Avoided costs (M	II USD/year)	2.89	0.28

For the avoided GHG emissions, the worse environmental contribution of biogas destined to electricity generation reflects the Brazilian energy matrix, which is strongly based on hydroelectric power, approximately 40% of the national energetic balance, (MME, 2018), whose carbon footprint is low, and eventually considered 'green'.

On the other hand, despite both alternatives are avoiding the use of the common energy sources (grid electricity and natural gas) and can provide savings, the avoided costs for the replacement of electrical energy for biogas indicate that electricity generation from biogas is pretty beneficial, ten-fold higher than the substitution of natural gas for biogas.

For the avoided costs results, the explanation is also based on the country's energy matrix and in the seasonality of energy supply (dependent on rainfall regime), that relies in thermoelectric energy to fulfill the whole country in the dry seasons of the year. Thermoelectric energy is more expensive than hydroelectric energy and its cost is reflected in the mean market energy price considered here (dos Santos et al., 2018).

Considering the emission factor for pulp and paper industry for Brazil of 0.33 ton  $CO_{2eq}$ / ton of pulp and paper (MCTIC, 2017) and the whole country pulp and paper production, 19.5 and 10.5 millions of tons, respectively (IBÁ, 2017), the whole sector emissions are estimated to reach 9.9 x 10<sup>6</sup> ton  $CO_{2eq}$  per year. Applying the avoided emissions here obtained per ton of paper produced to the country's production, i.e. 10.5 millions of tons, the substitution of natural gas for biogas could avoid more than  $1.06 \times 10^5$  ton CO<sub>2eq</sub>/year.

From the financial analysis, considering only the investment on UASB reactors

1 and 2 with 50% of its capacity occupation, the payback of this equipment for a paper mill plant in full operation (i.e. an operating facility 493 already established) is 6.4 years, the ROA after UASB reactor acquisition is 14%. The profit margin is 35%, both for the paper mill full equipment original investment and after UASB acquisition; it means that UASB investment is not a factor that erodes the financial result. The investment on UASB reactors accounts for only 6.8% of annual sales and for 19.8% of the operational profit and the depreciation of the reactors is 25 years. Even more, the whole capacity occupation does not affect the financial results here indicated. The average ROA for two large Brazilian companies of this sector is 18%, the average profit margin is 38%. The financial analysis here presented does not consider the avoided costs related to biogas energy recovery in feedback loops for the boiler and stationary engine. It aimed solely to evaluate UASB insertion in the paper mill study case to demonstrate that this investment is minimal and brings other benefits from GHG avoided emission and energy recovery.

#### 5. Conclusions

Paper industry has a large amount of liquid and solid effluents that originate from wastewater process production, which can be used to generate energy. The adoption of UASB reactors for paper mill industries properly addresses environmental concernments (air and water quality) and contributes to the energy matrix transition for more renewable sources.

In addition, the financial analysis related to the investment in a UASB reactor presents outstanding results. UASB investment is very small and does not affect the financial industry balance, been paid in a short time with a high return on investment (>50%) even using half of the system's capacity. Moreover, the whole paper industry in Brazil could avoid 1.06 x  $10^5$  ton CO<sub>2eq</sub>per year, contributing to the decarbonization of the economy.

Considering the rising energy costs and the growing concerns about climate change, biogas is an economically attractive green energy source. It can be used to replace conventional energy sources such as electricity and natural gas, while also reducing carbon footprint. Biorefineries projects, like the biochemical conversion (anaerobic digestion) for biogas production, require systemic assessments including financial aspects, costs, markets, yield and environmental benefits, as presented in this study, to properly optimize the use of the available resources, to maximize the profitability and to minimize wastes.

Biorefineries hold significant promise for the production of various biofuels and other biomaterials. However, regulations and policies, at various levels of government, must be adapted to take into consideration the social, environmental, and economic impacts of an emerging bioeconomy. The framework presented in this study for paper mill wastewater systems energy recovery can strength the value chain of this industry while addressing environmental and social aspects related to air and water quality.

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Campinas 22<sup>th</sup> march 2019

## **Dear Editors of Biofuel Research Journal**

In the annex we are presenting an additional material that was used to prepare this manuscript. The following tables report data gathered from Brazilian official sources for prices and emission factors. In addition, financial information is supplied below.

 Table 1- Emission factors for an annual period.

2017						20	18					
December	January	February	March	April	May	June	July	August	September	October	November	December
0.0892	0.0640	0.0608	0.0635	0.0523	0.0607	0.0915	0.1076	0.1181	0.1182	0.0802	0.0366	

Source: Brazilian Science, Technology, Innovation and Communication Ministry (MCTIC, 2018).

Price on electric power auction in Southeast Region (USD/MWh) Year Month Week Heavy Medium Light 2018 50.26 50.26 49.14 1 1 2018 2 45.29 45.29 44.04 1 3 40.27 2018 41.98 41.98 1 2018 1 4 50.15 50.15 46.85 2018 2 1 45.64 45.64 42.30 2018 2 2 46.56 46.56 43.74 2 3 43.91 43.91 42.41 2018 2018 2 4 54.27 54.27 52.13 2018 3 1 52.01 52.01 49.96 2018 2 57.25 57.25 55.60 3 2018 3 3 60.67 60.67 58.56 2018 3 4 56.31 56.31 54.65 2018 3 5 59.08 59.08 56.61 2018 4 10.19 10.19 10.19 1 2018 2 22.24 22.24 20.29 4 2018 3 31.81 31.81 29.99 4 2018 4 4 35.21 35.21 33.26 2018 56.51 56.47 53.17 5 1 5 2 74.60 2018 79.40 79.40 2018 5 3 84.72 84.72 79.60 2018 5 4 88.43 88.43 83.10 2018 1 106.34 106.34 96.60 6 2018 2 116.13 116.13 104.55 6 3 2018 6 122.41 121.83 116.21 2018 4 123.31 123.31 117.66 6 2018 5 128.22 128.22 128.22 6 7 2018 128.22 128.22 128.22 1

**Table 2 -** Brazilian price for Electric Energy.

2018	7	2	128.22	128.22	128.22
2018	7	3	128.22	128.22	128.22
2018	7	4	128.22	128.22	128.22
2018	8	1	128.22	128.22	128.22
2018	8	2	128.22	128.22	128.22
2018	8	3	128.22	128.22	128.22
2018	8	4	128.22	128.22	128.22
2018	8	5	128.22	128.22	128.22
2018	9	1	125.11	125.11	121.21
2018	9	2	127.95	127.95	122.46
2018	9	3	127.02	127.02	120.91
2018	9	4	113.43	113.43	109.91
2018	10	1	98.90	98.90	96.66
2018	10	2	83.25	83.25	81.80
2018	10	3	69.79	69.79	66.52
2018	10	4	59.08	59.08	56.94
2018	11	1	36.46	36.46	35.72
2018	11	2	38.46	38.46	37.35
2018	11	3	30.94	30.94	30.28
2018	11	4	30.64	30.64	29.57
2018	11	5	25.81	25.81	25.12
2018	12	1	15.08	15.08	14.81
2018	12	2	16.86	16.86	16.66
2018	12	3	20.35	20.35	19.63
2018	12	4	22.37	22.37	21.82

Source: Brazilian Electrical Energy Trade Chamber (CCEE, 2018).

Pr	ices for Industrial Sector	Prices (without commercial national tax) <sup>A</sup>		
Classes	Volume m <sup>3</sup> /month	Variable – USD/m <sup>3</sup>		
1	Until 50.000,00 m <sup>3</sup>	0.540		
2	50.000,01 to 300.000,00 m <sup>3</sup>	0.380		
3	300.000,01 to 500.000,00 m <sup>3</sup>	0.362		
4	500.000,01 to 1.000.000,00 m <sup>3</sup>	0.358		
5	1.000.000,01 to 2.000.000,00 m <sup>3</sup>	0.352		
6	> 2.000.000,00 m <sup>3</sup>	0.346		

**Table 3-** Brazilian Natural Gas prices for the leader supply in the location of the study case paper mill

<sup>A</sup> This Brazilian commercial tax acronym is *ICMS*, which is charged for all goods and services that circulates. It is accounted for in separate and for price definitions it is excluded. Source: COMGÁS, 2019.

## Financial analysis support information

## **Financial Resources**

Full paper mill equipment park = USD 50,155,200.00 UASB acquisition cost = USD 3,400,000.00 Financing time = 10 years Payment = Main stream + Interest rate Interest rate = Financing cost + BNDES Spread + Financial Agent Spread Interest rate = 9.25%a.a. Financing cost = LTIR = Long Term Interest Rate = 6.85%a.a. BNDES Spread = 1.40%a.a. Financial Agent Spread = 1.00%a.a.

Payment (main stream) = USD 340,000.00/year Payment (interest) = USD 31,450.00/year

#### **Depreciation fund - on Permanent Assets**

Rate = 3%a.a (Brazilian usual rate. Authors assumption: This rate is only an approximation once in the Financial Balance the depreciation feeds a discount account for the asset value.) Permanent Assets = USD 3,400,000.00 Depreciation horizon for UASB =25years Depreciation fund = USD 4,080.00/year

#### Payment in installments fund - on Intangible Assets

Rate = 3%a.a (Authors assumption) Intangible Assets = 10% of acquisition cost(Authors assumption) Intangible Assets = USD 340,000.00 Payment in installments fund = USD 10,200.00/year

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COMGÁS, 2019. Tarifas do Gás Natural Canalizado. https://www.comgas.com.br/tarifas/industrial/. In Portuguese.

# Capítulo 3 - Usos finais do biogás na forma de biometano: Análise de Impacto Ambiental

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(Impact Factor: 7.2)

# Environmental impact assessment of end-uses of biomethane

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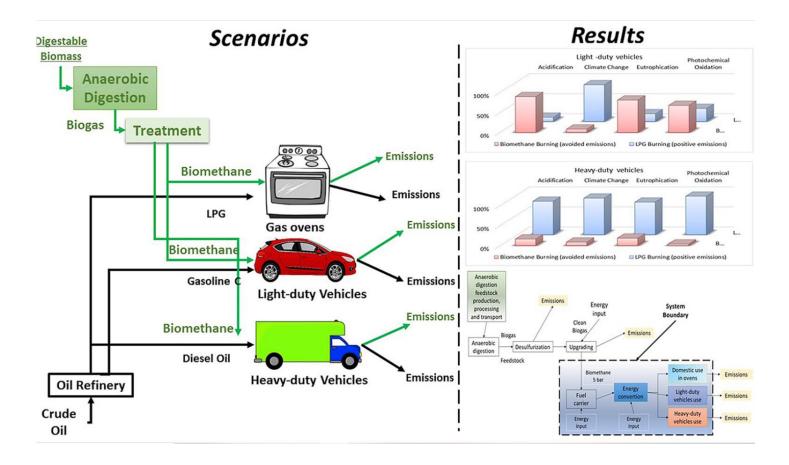
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## Abstract

The growing global population, hunger for energy and worried about climate change, demands the development of new sources of energy. In this scenario, biomass stands out due its renewability and availability. Biogas, one type of energy that results from the anaerobic digestion of organic materials, is composed of about 60% of methane and 35% of carbon dioxide, and can be converted to biomethane, a fuel with high energy content. Biomethane can be used in ovens for cooking, light-duty vehicles for transportation and heavy-duty vehicles for work. This study compared the impacts of the use of biomethane in all of these three end-uses applying the life cycle assessment methodology. Four impact categories were evaluated: acidification, climate change, eutrophication and photochemical oxidation, when replacing one traditional fossil fuel use for those biomethane end uses. The results showed that the replacement of Diesel Oil in heavy-duty vehicles was beneficial in all impact categories, the replacement of liquefied petroleum gas in gas ovens impacted positively only climate change and the replacement of gasoline-C in light-duty vehicles was disadvantageous (except for climate change). For all the uses, the replacement of the traditional fossil fuel by biomethane for climate change impact was beneficial. The contribution analysis showed that the burning of the fuel was the most relevant process for all four impact categories. This study aims to supply data for further analysis of the full life cycle of biomethane, considering the source of biomass, which can support a whole well-to-wheel approach.

**Keywords:** Bioenergy, Anaerobic Digestion, Environmental Sustainability, Biogas, Life Cycle Assessment

# **Graphical Abstract**



#### 1. Introduction

The global phenomenon of urbanization and growing population requires a rise in food production in an increasingly intense manner and, consequently, greater production of residual biomass and wastewater. Organic wastes from agricultural residues (crops and animal) amounting to one-third of global food production (Gustavsson et al., 2011) have severe environmental implications and generate large upstream and downstream emissions to the environment, potentially affecting future food and energy security (Tonini et al., 2018).

Currently, three crisis are underway, namely: a) food security crisis, especially in poor countries; b) the energy crisis, due to a global energy matrix strongly based on fossil fuels with low renewability in nature and with high polluting potential - intense use and burning promote large emissions of greenhouse gases whose effect is deleterious to the environment), and c) climate crisis, especially global warming (Bley Jr. et al., 2009). The imperative transition en route to a low-carbon economy requires the optimization of production chains including the energy conversion of wastes and the application of coproducts (biomaterials or bioenergy) in the chains themselves as an strategic planning focused on socioeconomic and socio-environmental sustainability, especially for vulnerable populations (Fortier et al., 2019). The three crisis mentioned are interlinked and are strongly associated with the existing environmental deterioration. In addition to the climatic issue, the inappropriate disposal of waste, depending on its nature, e.g. animal waste and untreated industrial wastewater, can cause nitrogen (N), phosphorus (P), potassium (K) saturation, heavy metals and other chemical agents in soil and water bodies as well as climate changes related to greenhouse gas (GHG) impact associated to residual biomass management (Coimbra-Araújo et al., 2014).

According to the Brazilian Ministry of Environment, organic waste (agricultural and industrial) accounted for more than 50% of all of the municipal solid waste in 2010, reaching 800 million of tons per year. A previous assessment of the potential use of Brazilian's main

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crops residues (sugarcane, soybean and maize among others) for bioenergy purposes showed that the country has enough raw material to support different biomass conversion technologies, to aggregate value to the food industry supply chain and to cope with the development of a biobased economy (Forster-Carneiro et al., 2013).

In addition, domestic and industrial wastewater treatment and biogas recovery through anaerobic digestion, a low cost, economically viable and environmentally friendly technology, could be easily and widely adopted to maximize energy recycling and, furthermore, available technologies are prone to create great economic advantages (Berni et al., 2014). The sustainability assessment of biogas, electric energy generation and organic fertilizer production, as well as, GHG mitigation in high intensive Brazilian agricultural systems reinforced the environmental and economic benefits related to the adoption of anaerobic digestion for energy cycling (Buller et al., 2015). The introduction of biodigesters into industrial and rural systems has spread throughout the world, including Brazil, where the introduction of the Federal Program for Low Carbon Agriculture (ABC Program) set up in 2010 after the 2009 United Nations Framework Convention on Climate Change (Unfccc) meeting in Copenhagen, induced the adoption of Clean Development Mechanisms. The estimated potential energy source from biogas for Brazilian main agricultural residues and for domestic wastewater treatment, could reach 6.9 MW (Silva dos Santos et al., 2018).

Anaerobic digestion makes possible to produce clean energy, once biogas, the gas mixture resultant of the organic biodegradation, can be burnt as a fuel (Ghodrat et al., 2018; Náthia-Neves et al., 2018). Biogas contains from 50-75% in volume of methane, 20-50% of carbon dioxide and traces of other gases, such as hydrogen sulfide and nitrogen. This mixture has a high calorific content (approximately 5,200 kcal/Nm<sup>3</sup>) due the high concentration of methane (Wellinger et al., 2013). However, due to the presence of hydrogen sulfide, the oxidation can generate corrosive gases that damage the equipment, so before combustion, the biogas has to go through a desulphurization process.

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Biogas can be enriched to biomethane, by concentrating the methane in the mixture there are several methods to do it (Leme and Seabra, 2017). Biomethane, similar to other fuels, is regulated in Brazil by the Oil, Gas and Biofuels National Agency, must contain at least 96.5 % of methane, about 3.0 % of carbon dioxide and traces of other gases(Anp, 2015).

Besides being in its early stages, there are some options for biomethane applications in Brazil, such as automotive fuel (both light and heavy-duty vehicles), mainly by the Itaipu Technological Park (Coimbra-Araújo et al., 2014) and its insertion in gas pipelines for selling to domestic use or heat source (ANP, 2019). The possible uses of biogas and / or biomethane in power grids (by converting to such efficient engines to sustain the required level of investment) and domestic use (in ovens instead of liquefied petroleum gas) represent an important advance for the whole society (Aziz et al., 2019). However, for developing countries the transition is still a challenge to overcome (Leontopoulos et al., 2015; Muniz Kubota et al., 2017).

It is noticeable that after the 21st Conference of the Parties (COP 21) to the UNFCCC<sup>1</sup>, Brazilian efforts to increase the production and use of renewable energy were strongly accelerated. A new national policy called RenovaBio<sup>2</sup> aims to draw up a joint strategy to recognize the strategic role of all types of bioenergy in the Brazilian energy matrix, both for energy security and for mitigation of greenhouse gas emissions (GHG) (MME, 2018).

RenovaBio approach to establish the guidelines for the energy transition is based on Life Cycle Assessment (LCA) methodology (ISO, 2006a; 2006b) which is internationally recognized for evaluating environmental impacts of products and technologies. The term 'life cycle' refers to all of the stages of a process, from the extraction of raw material to the final disposal, including all intermediary processes (Guinée and Heijungs, 1993). LCA generates

<sup>&</sup>lt;sup>1</sup><u>https://www.un.org/sustainabledevelopment/blog/category/climate-change/cop21/</u>

<sup>&</sup>lt;sup>2</sup><u>http://www.planalto.gov.br/ccivil\_03/\_ato2015-2018/2017/lei/L13576.htm</u> (In Portuguese)

quantitative indicators related to several categories of environmental impact in three protection areas: Human Health, Natural / Artificial Environment and Natural Resources; such as global warming (impacts on human health and the environment), eutrophication of water and soil (impacts on the environment), acidification (impacts on the environment) and photochemical oxidation (impacts on human health and the environment), among others.

Several works and regulations concerning environmental impacts of biofuels focus on GHG emissions, however LCA studies have shown that a reducing GHG emission may not reduce other environmental impacts, and may even increase them (Bolin et al., 2009; Caponio et al., 2013; Czyrnek-Delêtre et al., 2017).

Hakawati et al. (2017) assessed 49 biogas-to-energy routesusing LCA and the results showed that the higher efficiency route is the direct use of biogas in the surroundings of the anaerobic digestion facility, however energy efficiency is not the only relevant factor for decision making; others such as economic or environmental should be considered as well. Ravina and Genon (2015) assessment of global and local emissions of a biogas plant considering two scenarios, combustion of biogas in a combined heat and power unit and the upgrading of biogas to biomethane and subsequent injection to the gas grid or use in transportation, indicated that biomethane both end-uses could partly or totally avoid local impacts.

The use of biogas energy as a substitute for liquefied petroleum gas (LPG) in households was assessed by Nhu et al. (2015) in Vietnamese farms, while Alexander et al. (2019) proposed a home-style biogas systems to produce energy and biofertilizer as well as the domestic end-use of biogas from food waste anaerobic digestion in urban areas. Jury et al. (2010) assessed the injection of biogas into the natural gas grid for different possible uses in Luxemburg. For the case of end-use of biogas in light and heavy-duty vehicles, scientific findings are far well developed in Brazil (Coimbra-Araújo et al., 2014) but a broad assessment of environmental impacts is still necessary to support the technology dissemination on a large scale. According to Patterson et al. (2011a), for biogas production and end-use systems the LCA should be done at regional level to properly assist the infrastructure development and to back up decision making processes. End-uses of biogas were evaluated in Mexico (Chan Gutiérrez et al., 2018), in Argentina (Morero et al., 2015), United Kingdom (Patterson et al., 2011b, 2013) and for several scenarios of biogas production from landfills in Europe and USA (Beylot et al., 2013).

So far, as the authors knowledge, there are no previous works focusing on biomethane end-uses and its environmental impacts for the Brazilian scenario. The present work aims to provide an environmental impact analysis of four impact categories for domestic use (ovens) and engines (only vehicles) of biomethane and to aid future researches that will consider a full well-to-wheel LCA approach. These further studies will demand data from the production of biomass, generation of biogas, purification and enrichment, which vary strongly with the processes and types of biomass, thus becoming very case-specific.

#### 2. Materials and methods

#### 2.1. Life Cycle Assessment (LCA)

LCA, a robust methodology for environmental assessment internationally recognized, is based on the collection of all the materials and energy resources necessary to a product system from the raw material extraction until the product final disposal (Guinée et al., 2002). This way LCA captures the full life cycle in a systemic way.

According to ISO Norms 14040/44 (ISO, 2006a; 2006b) the methodology contains 4 steps: 1) objective and scope; 2) inventory analysis; 3) impact assessment and 4) interpretation.

The objective is to compare the environmental impacts of three alternatives (domestic use in ovens, use in light-duty vehicles and use in heavy-duty vehicles) to replace fossil fuels by biomethane by means of an attributional LCA, i.e. only direct emissions from life cycle were accounted for considering MJ of energy (work or heat) by each alternative as the functional unit.

This work applied a well-to-wheel (WtW) approach when analyzing the fossil fuels life cycle. This approach considers the energy use from the production of the fuel to its final use (Alamia et al., 2016). WtW and LCA methodologies can have the same system boundaries (Alamia et al., 2016; Czyrnek-Delêtre et al., 2017), which is the case of this work. For fossil fuel use in domestic ovens, the WtW approach was maintained, because of the assumption related to the energy use of a fuel from production to end use.

For the biomethane scenario, however, a gate-to-wheel approach was done, because of the assumption that biomethane comes from the biorefinery, after the desulphurization step, to the end use. This approach was chosen to avoid a generalization related to the organic feedstock production chain which influences the impacts magnitude. The impacts related to production, enrichment and desulphurization were not accounted for (as a full WtW approach would require). This was done because those processes are very case-specific and the applicability of our results would be reduced.

In summary, this LCA focused in the comparison of environmental performance of three alternatives of biomethane destination until its final use: biomethane used for the production of heat in gas ovens, as fuel for light-duty vehicles and for heavy-duty vehicles, measured in MJ, considering the burning efficiency of each process. Biomethane avoids the use of a specific fossil fuel for each scenario. The environmental performance for biomethane use for each scenario was compared to fossil fuel use considering the avoided emissions. For each alternative, biomethane produced from the desulphurisation and enrichment of the biogas from a wastewater treatment station was considered, as shown in **Figure 1**. Therefore, systems' boundaries for the fossil fuels embrace the environmental impact and assessment of extraction and transportation (both included in the upstream flows).

The impact categories chosen were acidification, eutrophication, climate change and photochemical oxidation. These impact categories were selected because of their direct relationship with effects related to wastewater and/or sludge inappropriate disposal in the environment (Menichetti and Otto, 2009).

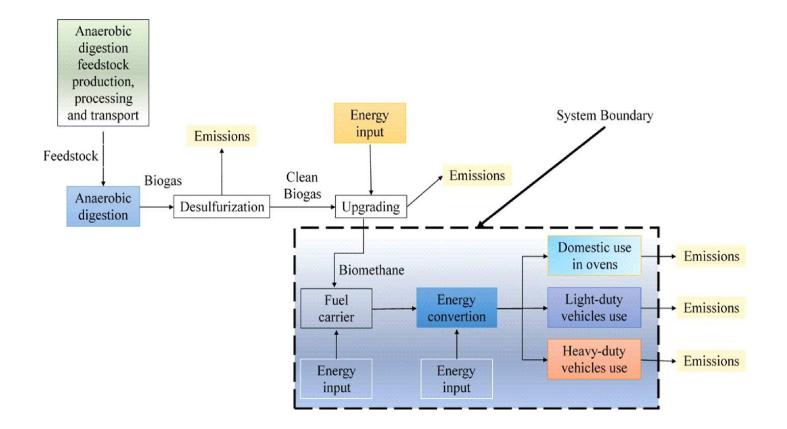


Figure 1. Alternatives and control volume for each scenario of biomethane produced from

the desulphurization and biogas enrichment from a wastewater treatment plant.

Data Inventory is presented in the **Supplementary material**. The impact assessment was done using the free software OpenLCA®, according to **Equation 1** described in the Handbook of Life Cycle Assessment (Guinée et al., 2002).

Environmental impact = 
$$\sum_i Factor_i * m_i$$
 (Eq. 1)

Where *Factor*<sub>i</sub> is the environmental impact factor of molecule *i* and  $m_i$  is the mass of molecule *i* emitted. Firstly to assess all the upstream processes related to biomethane production (using default methods on the software) and in the sequence to the assessment of biomethane use in substitution of fossil fuels, namely: natural gas, gasoline-C and Diesel oil; according to the detailed methodology presented in the following items. In addition, other input data (emission factors, etc.) and the followed steps and additional information obtained from OpenLCA® are presented in the

# Supplementary material.

The summary hypotheses and assumptions done (further detailed in sections 2.1.1 and 2.1.2) are:

✓ Biomethane use as a substitute for liquefied petroleum gas in gas ovens:

- a. Local generation of biomethane
- b. LPG generated at REPLAN (Brazilian Oil Refinery), estimation of 250 km of distance
- c. Cylinders of 15kg (empty)
- d. Cylinders of 28kg (full)

✓ Biomethane as a substitute for gasoline-C:

a. Local generation of biomethane

- b. Gasoline-C as a mixture of gasoline-A and anhydrous ethanol
- c. Pressure at the injection of 200 bar

✓ Biomethane as a substitute Diesel oil in vehicles:

- a. Local generation of biomethane
- b. Diesel as a mixture of 5% biodiesel and 95% pure Diesel oil
- c. Pressure at the injection of 250 bar

# 2.1.1. Biomethane use as a substitute for liquefied petroleum gas in gas ovens

We didn't consider the losses in the tubes, because the proposed scenario was considering a residential facility close to the industrial one, so making the eventual losses negligible.

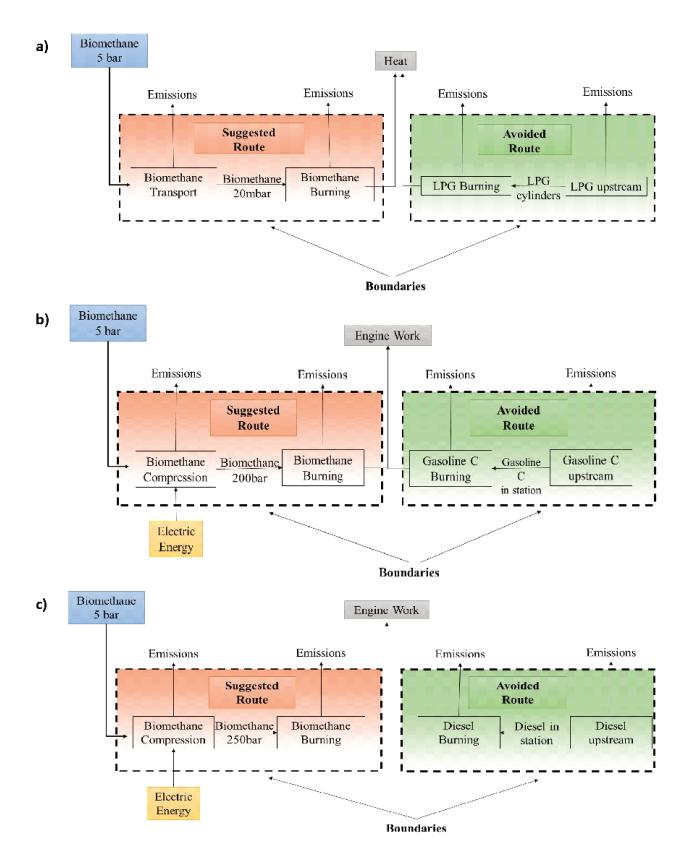
Systems' boundaries include environmental impact assessment of extraction, transportation and so on according to **Figure 2**, which presents each alternative. Biomethane at 5 bar, obtained from the desulphurization process, is piped to fuel carriers and energy conversion process for each alternative (domestic use in ovens, light-duty vehicles use and heavy-duty vehicle use).

The system boundary for the case of environmental performance of the use of biomethane in substitution for liquefied petroleum gas (LPG) in ovens was established considering that the biomethane comes out from a desulphurisation and enrichment system. In addition, it is transported by pipes to a residence, avoiding the use of LPG in a P-13 canister, as shown in **Figure 2a**.

In addition, the assumption is that LPG was produced at the Replan-Paulínia refinery (Brazil). It was also considered that it is transported in trucks in 28 kg cylinders (15 kg of the empty cylinder and 13 kg of LPG), and the gas cylinders run 250 km full for the delivery and more 250 km (empty) on the return route to the distribution center, according to the Brazilian National Agency of Petroleum, Natural Gas and Biofuels (ANP, 2017).

The transportation and production data at the refinery were obtained from EcoInvent Database v. 3.4.Gas leakages in the transportation of biomethane were neglected (Alamia et al., 2016) and it was assumed that the pressure drop was enough to reach the pressure required for use in stoves.

The inventory data was obtained from literature (Jungbluth, 1997), considering that biomethane has the same emissions as natural gas, except for the NMVOC (non-methane volatile organic carbons), which was considered zero for biomethane. It was also considered that 60% of energy was useful heat, and the rest was lost to the environment.



**Figure 2.** System boundaries for biomethane: a) gas ovens; b) light-duty vehicles; c) heavyduty vehicles.

#### 2.1.2. Biomethane as a substitute for gasoline-C and Diesel oil in vehicles

For the environmental performance of biomethane as a substitute for gasoline-C and Diesel oil, as before for domestic use, the system boundary also considered biomethane coming out from a desulphurization and enrichment system and being transported by pipes to light-duty vehicles (Otto-cycle engine) and heavy-duty vehicles (Diesel-cycle engine), as shown in **Figures 2b and 2c**, in substitution for gasoline-C and Diesel oil.

Due to Brazilian regulatory demands, gasoline-C is a mixture composed of 78% of gasoline-A (pure gasoline) and 22% of anhydrous ethanol from sugarcane. Similarly, Diesel oil is a mixture composed of 95% of pure Diesel oil and 5% of biodiesel. The upstream processes for anhydrous ethanol, gasoline-A and Diesel oil were considered background data and were obtained from EcoInvent Database v. 3.4. The upstream processes for biodiesel were not considered.

The emissions were obtained from the National Inventory of Atmospheric Emissions by Automotive Vehicles (MMA, 2014) and the energy and physical densities from the National Energy Balance, 2016 (EPE, 2017). It was assumed that the light-duty vehicle would perform 12 km per liter of gasoline-C and 12 Km per Nm<sup>3</sup> of biomethane and the heavy-duty vehicle would require 208 g of Diesel oil per kWh and would perform 3.4 km per liter of Diesel.

For both biomethane in light-duty vehicles and in heavy-duty vehicles, the burning efficiency remained the same as for the original fossil fuels, respectively, 30% and 41% (MMA, 2014; Alamia et al., 2016).

It was assumed that biomethane has the same emissions of Vehicular Natural Gas (VNG) disregarding non-methane hydrocarbons and considering carbon dioxide as biogenic, according to the limits of the P-7 phase of the Brazilian regulation PROCONVE (Conama, 2008).

The emission of carbon dioxide for biomethane was estimated considering 99% of oxidation and nitrous oxide ( $N_2O$ ) emission was estimated according to the Intergovernmental Panel on Climate Change Guidelines (IPCC, 2006).

The injection of biomethane into Otto cycle motors for light-duty vehicles was carried out at a pressure of 200 bar according to Brazilian regulation of National Agency of Petroleum, Natural Gas and Biofuels for liquefied petroleum gas (LPG) (ANP, 2001). In this way, a compression of 5 to 200 bar of biomethane was considered to calculate the electricity demand, performed through a gas energy balance, considering pure methane, assuming 90% of efficiency in the compressor. It was considered that 22% of carbon dioxide emissions from gasoline-C are from renewable sources due to the addition of anhydrous ethanol coming from Brazilian sugarcane. The impacts due to the use of electricity were also obtained by the EcoInvent v. 3.4 and considered background data; refer to **Table 1** to check database information.

**Table 1.** Background data for light and heavy-duty vehicles.

Process	Name in Database
Gasoline-A Upstream	Market for petrol., low-sulphur, RoW
Anhydrous Ethanol Upstream	Dewatering of ethanol from biomass, from 95% to 99.7% solution state, BR
Electricity Use	Market for electricity, medium voltage, production mix, BR
Diesel Oil Upstream	Market for diesel, low-sulphur, RoW

In addition, in the case of injection of biomethane in Diesel cycle engines for heavy vehicles, according to the national laws (BRASIL, 2016), 8% of the volume of diesel oil must be composed of biodiesel, therefore upstream processes were not considered, it was assumed that 8% of the carbon dioxide (CO<sub>2</sub>) emitted was biogenic.

Another assumption was that biomethane injection was carried out at a pressure of 250 bar (Alamia et al., 2016) a compression of 5 to 250 bar was considered and to calculate its electricity demand an energy balance was done considering pure methane, and assuming an efficiency of 90% in the compressor. It was also considered as if all of the upstream processes were for pure fossil diesel oil (a given background, refer to **Table 1**).

The final emissions and burning efficiency comparing biomethane and fossil fuels for the three alternatives are shown in **Table 2**.

#### 3. Results and Discussion

### **3.1 Environmental Impacts**

For all scenarios, LCA results are presented considering only the impacts of the effectively final energy usage for biomethane replacing LPG, gasoline-C and Diesel Oil in the end-use (**Table 3**). Refer to section 2.1, where the calculation steps are presented.

In the replacement of LPG for biomethane in domestic use, the impacts related to LPG raw material obtaining, processing and transportation showed to be minimal compared to the emissions related to biomethane (refer to **Supplementary Material** to observe all the assumptions).

In addition, for biomethane burning in domestic ovens, all of the emissions, except for CH<sub>4</sub>, are similar; this is due to the regulation of LPG, which must not contain methane (ANP, 2004). Regarding global warming, although the emissions are similar, fuel sources differ, consequently,  $CO_2$  generated from the biomethane burning does not cause impacts in this category once it is accounted for as a biogenic source (IPCC, 2006). **Table 2.** Emissions and efficiencies of biomethane and LPG for Diesel-Cycle Engines and Otto-Cycle Engines.

	Oven	S	Engines				
			Diesel-Cycl	e Engines	Otto-Cycle Engines		
Emissions (kg/MJ)	Biomethane	LPG	Biomethane	Diesel	Biomethane	Gasoline-C	
СО	2.50.10 <sup>-5</sup>	2.50.10 <sup>-5</sup>	4.55.10 <sup>-4</sup>	1.14.10-2	1.82.10 <sup>-4</sup>	9.31.10 <sup>-5</sup>	
CO <sub>2</sub>	5.55.10 <sup>-2</sup>	6.36.10 <sup>-2</sup>	3.40.10-2	7.33.10 <sup>-2</sup>	5.43.10 <sup>-2</sup>	5.35.10 <sup>-2</sup>	
NO <sub>X</sub>	2.60.10 <sup>-5</sup>	2.60.10 <sup>-5</sup>	2.27.10 <sup>-4</sup>	1.14.10-3	9.45.10 <sup>-5</sup>	1.12.10 <sup>-5</sup>	
N <sub>2</sub> O	5.00.10 <sup>-7</sup>	5.00.10 <sup>-7</sup>	1.77.10 <sup>-5</sup>	2.87.10 <sup>-6</sup>	1.02.10 <sup>-5</sup>	7.82.10 <sup>-6</sup>	
CH <sub>4</sub>	5.00.10 <sup>-7</sup>	0.00	1.25.10 <sup>-4</sup>	0.00	7.17.10 <sup>-5</sup>	9.68.10 <sup>-6</sup>	
NMVOC	0.00	4.00.10 <sup>-6</sup>	0.00	0.00	0.00	0.00	
NMHC	0.00	0.00	0.00	1.58.10 <sup>-1</sup>	0.00	5.21.10 <sup>-6</sup>	
PM	1.00.10 <sup>-7</sup>	1.00.10-7	0.00	1.39.10 <sup>-3</sup>	0.00	4.09.10 <sup>-7</sup>	
Aldehydes	0.00	0.00	0.00	0.00	1.24.10 <sup>-6</sup>	6.33.10 <sup>-7</sup>	
Formaldehyde	2.00.10 <sup>-7</sup>	2.00.10 <sup>-7</sup>	0.00	0.00	0.00	0.00	

# **Table 3.** Impacts Assessments of biomethane and LPG for Diesel-Cycle Engines and Otto-Cycle Engines.

Impact Assessment Category	Unit	Over	ns	Engines			
				Diesel-Cycle Engines		Otto-Cycle Engines	
		Biomethane	LPG	Biomethane	Diesel	Biomethane	Gasoline-C
Acidification potential – average Europe	kg SO <sub>2eq</sub>	1.30.10 <sup>-5</sup>	1.30.10 <sup>-5</sup>	1.14.10 <sup>-4</sup>	5.68.10 <sup>-4</sup>	4.72.10 <sup>-5</sup>	5.58.10 <sup>-6</sup>
Climate change - GWP100	kg CO <sub>2eq</sub>	2.09.10 <sup>-4</sup>	6.38.10 <sup>-2</sup>	9.27.10 <sup>-3</sup>	9.57.10 <sup>-2</sup>	5.18.10 <sup>-3</sup>	5.63.10 <sup>-2</sup>
Eutrophication - generic	kg PO4 <sup>3-</sup> eq	3.52.10 <sup>-6</sup>	3.52.10 <sup>-6</sup>	3.43.10 <sup>-5</sup>	1.49.10-4	1.50.10 <sup>-5</sup>	3.56.10 <sup>-6</sup>
Photochemical oxidation - high Nox	kg ethylene <sub>eq</sub>	7.82.10 <sup>-7</sup>	7.79.10 <sup>-7</sup>	1.30.10 <sup>-5</sup>	3.07.10 <sup>-4</sup>	5.35.10 <sup>-6</sup>	2.57.10 <sup>-6</sup>

Finally, for Photochemical Oxidation category, the small difference is due to the differences in emissions of NMVOC and CH<sub>4</sub>. Biomethane does not emit NMVOC, because methane is the only hydrocarbon produced in anaerobic digestion and the methane emissions is because of the unburnt methane. The opposite is true for LPG, which does not contain methane and the NMVOC emitted is due the unburnt ones. Although the emissions are different, the overall impact of Photochemical Oxidation is similar.

The burning of biomethane as replacement for Diesel oil and gasoline-C, in heavyduty and light-duty vehicles, respectively, was evaluated considering only the burning efficiency once the upstream flows not showed to be highly impacting on its impact assessment magnitude order (See **Supplementary Material**).

Evaluating emissions for biomethane burning in Diesel-cycle engines, lower emissions, are found for CO, NOx, and CH<sub>4</sub>. This explains the lower value for Climate Change impact in heavy-duty vehicles. Especially for PM and NMVOC, no emissions were observed for biomethane in replacement of diesel, which strongly contributes to its better assessment for this route. Considering the alternative of biomethane replacing gasoline-C, still PM and NMVOC strongly contributes to its better assessment for this route. These results follow Ravina and Genon (2015) findings that biomethane end-use as fuel in transport result in lower local GHG emissions and in the reduction of NOx and PM.

Comparing all the alternatives for biomethane use and the selected impact categories, the best use for biomethane is the replacement of Diesel oil in heavy-duty vehicles followed by the substitution of LPG in domestic gas ovens **Figure 3**.

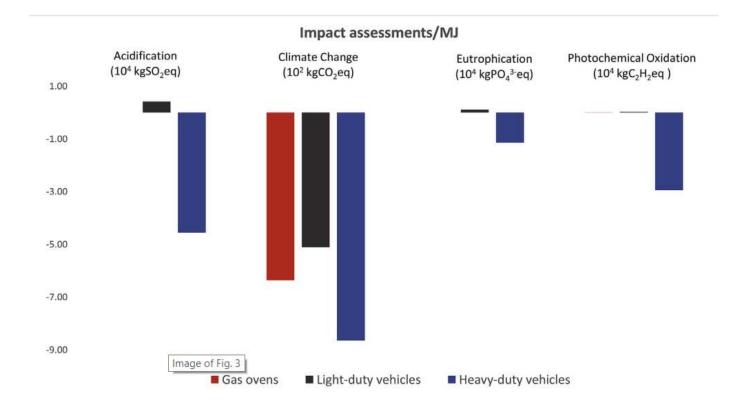


Figure 3. Environmental impacts for each use of biomethane.

**Figure 3** shows the environmental impacts for each use of biomethane for all of the impact categories. The negative results show that the substitution for biomethane reduced the impacts, and the positive results show increasing impacts. The higher decrease in impact assessment for the heavy-duty vehicles is noticeable. In all of the impact categories evaluated, the replacement of Diesel Oil by biomethane in heavy-duty vehicles presented superior environmental performance, even considering the low-sulfur content of Brazilian Diesel Oil and a small percentage of biodiesel.

Because of the large variety of methodological assumptions in bioenergy LCA assessments, the existence of many variables involved in specific-cases and their strongly dependence on regional factors (Cherubini and Strømman, 2011), such as system boundaries, statistical methods, scarceness of updated and accurate local data, and local environmental and political conditions (Jin et al., 2018), a direct comparison of the results here presented would be debatable.

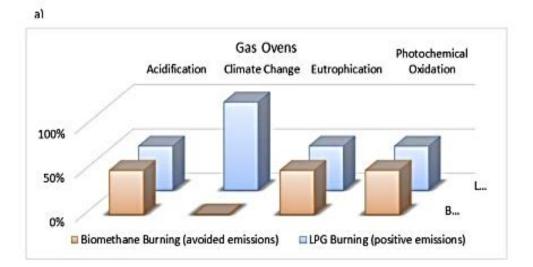
Notwithstanding the aforementioned, Ardolino et al. (2018) quantified the potential impacts of biomethane end-uses using the burning of biogas for combined heat and power as base scenario and emphasized that the better environmental results in the LCA approach is the substitution of fossil fuels in transport. The results of the present study showed that the substitution of Diesel oil by biomethane in Brazil has a meaningful potential to keep up with the reduction of emissions in the transport sector, which amounted to 48 % of the total country's emissions in 2018 (IEA, 2018). Considering the anaerobic digestion of the main wastes in Brazil, a fleet of more than 180,000 buses could be supplied with biomethane in the replacement of Diesel oil (Silva dos Santos et al., 2018).

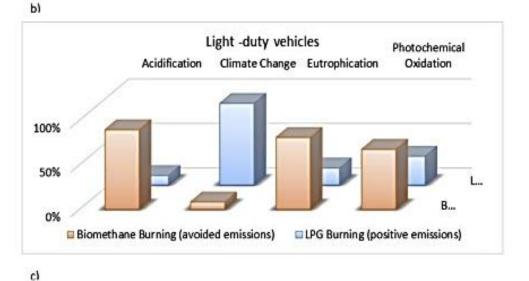
## **3.2** Contribution Analysis

**Figure 4** presents the relative contribution for each process in the final impact. All columns have a length of 100%. Positive values indicate that the specific process causes impact in the environment and the negative ones indicate that the process avoids impacts. The length of each process indicates the relative contribution to the result presented in **Figure** 

3.

All the processes that had at least 1% of contribution in each impact category are presented in **Figure 4**. In all of the studied scenarios, the process that contributes the most is the fuel burning processes, making the other processes irrelevant to the analyzed impact categories.





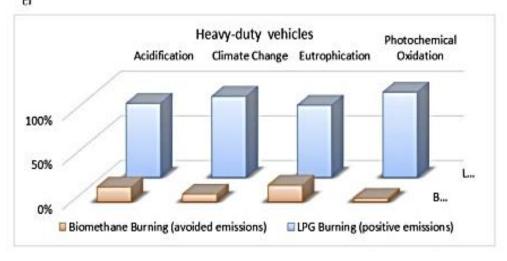


Figure 4. Contribution analysis in each impact category for each use of biomethane.

For domestic use, aside the climate change impact, where the use of biomethane almost totally eliminates the impacts of LPG, in all other impact categories the substitution of LPG by biomethane is indifferent.

For light-duty vehicles, the replacement of gasoline-C by biomethane is strongly beneficial for the climate change impact category, however in all of the other categories the substitution is disadvantageous, due to the emissions of CO, NO<sub>x</sub>, N<sub>2</sub>O and aldehydes during biomethane burning.

In the heavy-duty scenario, the substitution of Diesel Oil by biomethane is strongly beneficial, being the most relevant for the photochemical oxidation impact category.

#### 4. Conclusions

The replacement of LPG for biomethane in domestic use presents a special contribution to cope with climate change mitigation. However, for better inferences for this route and to get more robust recommendations, biomethane supplied in pipelines for domestic use, which is in its first stages in Brazil, should be considered in a further assessment. Biomethane in the substitution of gasoline-C could present lower emissions using engines and exhaustion systems more suitable to biomethane (or VNG, which is chemically similar), since in Brazil the vehicles are adapted after manufacturing to use VNG.

In all of the impact categories evaluated, the replacement of Diesel Oil by biomethane in heavy-duty vehicles presented superior environmental performance, even considering the low-sulfur content of Brazilian Diesel Oil and a small percentage of biodiesel. This points out to an opportunity to reinforce biomethane technologies for automotive use focusing on the establishment of a cleaner energy matrix in Brazil, a country of continental dimensions, that until nowadays depends on road transportation to flow its agricultural and industrial production. Likewise, the substitution of Diesel for biomethane in public transport, which is heavily dependent on buses in Brazil, since rail and subway services exist only in some capitals with a low coverage network, would bring propitious benefits. The results suggest that the introduction of biomethane in heavy-duty vehicles can diminish the carbon footprint of several agricultural and urban systems.

This study took into account the biomethane generated from a wastewater treatment station after the enrichment of biogas to its final use. The outcomes revealed that LCA approach offers relevant environmental impact indices supporting decision-making on the development of a new energy matrix in Brazil and it helps the selection of the optimal biomethane end-uses. A further assessment to complete this first approach should be carried out to evaluate other sources of residual biomass, which are plenty in Brazilian agricultural and industrial systems, considering a virtuous integration between rural and urban areas, including industrial processing, raw material and finished goods transportation, favoring strategies and public policies to give shape for the development of a circular economy.

# 5. Acknowledgments

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# **Supplementary material**

In this annex we are presenting an additional material that was used to prepare this manuscript. For biomethane in ovens the results in **Table 1** refer to 60% of burning efficiency.

Table 1: Emissions from biomethane and LPG for burning in gas ovens in kg/MJ burned

	Biomethane	LPG
NOx	2.60.10-5	2.60.10 <sup>-5</sup>
PM	1.00.10 <sup>-7</sup>	1.00.10-7
CO	2.50.10-5	2.50.10-5
NMVOC	0	4.00.10 <sup>-6</sup>
N <sub>2</sub> O	5.00.10-7	5.00.10-7
CO <sub>2</sub>	5.55.10-2	6.36.10 <sup>-2</sup>
Aldehydes	2.00.10-7	2.00.10-7
CH4	5.00.10-7	0

The inventory, for the calculation of the environmental performance of replacing biomethane for gasoline-C, considered that Brazil's automotive gasoline-C is composed of 22% anhydrous ethanol and 78% gasoline-A. The upstream activities of anhydrous ethanol and gasoline as data background were obtained from EcoInvent Database v. 3.4. The emissions were obtained from the National Inventory of Atmospheric Emissions by Automotive Vehicles (Mma, 2014) and the energy densities of the National Energy Balance, 2016 (Moraes *et al.*, 2017). It was assumed that biomethane has the same emissions of Natural Gas (NGV) disregarding non-methane hydrocarbons and considering carbon dioxide as biogenic. The emissions and burning efficiency in Otto-cycle engines for biomethane and

gasoline-C are shown in **Table 2**. In order to calculate the environmental performance of the replacement of biomethane for Diesel oil, it was considered biomethane and Diesel oil burned in a Diesel-cycle engine. In addition, emissions similar to the NGV were considered, according to the limits of the P-7 phase of the Brazilian regulation PROCONVE (Conama, 2008) for the use of fuel in Diesel-cycle engines, considering carbon dioxide as a renewable source and excluding emissions of non-methane hydrocarbons. The emission of carbon dioxide for biomethane was estimated considering 99% of oxidation and that of nitrous oxide (N<sub>2</sub>O) estimated by the Intergovernmental Panel on Climate Change (IPCC, 2006).

	Diesel-	CycleEngines	Otto-Cycle	Engines	
	Diesel Biomethane		Biomethane	Gasoline-C	
	(kg/MJ)	(kg/MJ)	(kg/MJ)	(kg/MJ)	
CO	$1.14.10^{-2}$	4.55.10-4	1.82.10-4	9.31.10 <sup>-5</sup>	
NOx	$1.14.10^{-3}$	2.27.10-4	9.45.10-5	1.12.10 <sup>-5</sup>	
Aldehyde	0	0	1.24.10-6	6.33.10 <sup>-7</sup>	
S					
NMHC	$1.58.10^{-1}$	0	8.47.10-6	5.21.10-6	
CH4	0	1.25.10-4	7.17.10 <sup>-5</sup>	9.68.10 <sup>-6</sup>	
PM	1.39.10-3	0	0	4.09.10 <sup>-7</sup>	
N <sub>2</sub> O	2.87.10-6	1.77.10 <sup>-5</sup>	1.02.10-5	7.82.10-6	
CO <sub>2</sub>	7.33.10 <sup>-2</sup>	3.40.10-2	5.43.10-2	6.86.10 <sup>-2</sup>	
Efficiency	41%	41%	30%	30%	

Table 2. Data from emissions and efficiency of the burning in Diesel and Otto Cycle engines

The injection of biomethane into Otto-cycle motors for light vehicles was carried out at a pressure of 200 bar according to Brazilian regulations of National Agency of Petroleum, Natural Gas and Biofuels for liquefied petroleum gas (Anp, 2001). In this way, a compression of 5 to 200 bar of biomethane was considered by a compressor to calculate the electricity demand, performed through a gas energy balance, considered as pure methane, assuming 90% efficiency in the compressor. It was considered that 22% of carbon dioxide emissions from gasoline-A are from renewable sources due to the addition of anhydrous ethanol coming from Brazil's sugar cane. The impacts due to the use of electricity were also obtained by the EcoInvent v. 3.4 and considered background data. In addition, in the case of injection of biomethane in diesel cycle engines for heavy vehicles, according to the legislation 8% of the volume of diesel oil must be composed of biodiesel in Brazil, so the upstream processes of this fuel were not considered, it was assumed that 8% of the carbon dioxide (CO<sub>2</sub>) emitted was biogenic. The biomethane injection was carried out at a pressure of 250 bar (Alamia et al., 2016). For this it was considered the compression of 5 to 250 bar of the biomethane by a compressor, to calculate its electricity demand was realized a balance of energy of the gas, considered like pure methane, and assuming an efficiency of 90% in the compressor. It was also considered as if all processes upstream of the diesel were pure fossil diesel oil and considered as a given background.

Tables 3 to 5 present all the process contribution in the LCA.

Process	BiomethaneBurning	LPG atrefinery	LPG Burning	LPG Transport		
Location		GLO		GLO		
Impactcategory	Referenceunit	ProcessContribution				
Acidificationpotential – averageEurope	kg SO <sub>2 eq</sub>	1.30.10-5	0	1.30.10-5	0	
Climatechange - GWP100	kg CO <sub>2 eq</sub>	2.09.10-4	0	6.38.10-2	0	
Eutrophication - generic	kg PO4 <sup>3</sup> eq	3.52.10-6	0	3.52.10-6	0	
Photochemicaloxidation - high NOx	kg C <sub>2</sub> H <sub>2 eq</sub>	7.82.10-7	0	7.82.10-7	0	

# **Table 3:** Process Contribution for Ovens

 Table 4: Process Contribution for Light-Duty Vehicles

Process		Biomethane Burning	Biomethane Compression	Electricity voltage transformation from medium to low voltage	Electricity, high voltage, production mix	Market for ethanol	Market for electricity, low voltage	
Locati	ion			BR	BR		BR	
Impactcategory	Reference unit		ProcessContribution					
Acidificationpotenti al- averageEurope	kg SO <sub>2eq</sub>	4.72.10-5	0	0	0	0	0	
Climatechange - GWP100	kg CO <sub>2eq</sub>	5.18.10-3	0	0	0	0	1.32.10 <sup>-12</sup>	
Eutrophication - generic	kg PO4 <sup>3-</sup> eq	1.50.10-5	0	0	0	0	0	
Photochemicaloxida tion - high NOx	kg C <sub>2</sub> H <sub>2 eq</sub>	5.35.10-6	0	0	0	0	0	

**Table 5:** Process Contribution for Heavy-duty Vehicles

Proces	6S	Biomethane Burning	network electricity medium		Diesel Burning	Transmission network construction, electricity, medium voltage		
Locatio	on				BR	GLO		RoW
Impact category	Reference unit				Pro	ocess Contribution		
Acidification potential – average Europe	kg SO <sub>2 eq</sub>	1.14.10-4	0	0	0	0	5.68.10-4	0
Climate change - GWP100	kg CO <sub>2 eq</sub>	9.27.10-3	0	0	6.20.10 <sup>-8</sup>	0	9.57.10-2	1.46.10 <sup>-8</sup>
Eutrophication - generic	kg PO <sub>4</sub> <sup>3</sup> <sub>eq</sub>	3.43.10-5	0	0	0	0	1.49.10-4	0
Photochemical oxidation - high NOx	kg C <sub>2</sub> H <sub>2 eq</sub>	1.30.10 <sup>-5</sup>	0	0	0	0	3.07.10-'4	0

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# Capítulo 4 – Gerenciamento de resíduos e recuperação energética do processamento de açaí na região amazônica brasileira: Uma perspectiva para uma economia circular

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Running Head: Bioenergy recovery from açaí agroindustrial waste in the Brazilian Amazonian region

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#### Abstract

This study aims to evaluate the Brazilian production of açaí, focusing on its waste generation and addressing mass and energy balances set from the cultivation, extraction, processing, and waste disposal. Besides, a new technological route for açaí's waste management was introduced for bioenergy recovery within the circular economy concept. In 2018, Brazil produced 1.7 million tons of açaí fruit for an income of 1.07 billion USD, and for this production, the waste generation (seeds) was estimated at 85%. Due to the high production of waste, an innovative approach was developed for a system boundary, including the waste management of solid and liquid wastes through anaerobic digestion (AD). The results showed that from 1 ton of açaí fruit fed into the facility for processing, it was generated 1.2 ton of solid waste and wastewater. This waste submitted to AD can produce 2.77 m<sup>3</sup> of biogas, with a methane composition of 50%. The completely industrial process demands 25 kWh per ton of frozen pulp, the local energy produced by the biogas burning could be selfconsumed, establishing a circular energy economy for this sector. With the adoption of AD waste management, about 61% of the external electricity requirement for the açaí fruits processing can be replaced from the biogas produced. The adoption of this technology can be a positive outcome as a contributor to the energy matrix decarbonization. Furthermore, the implementation of AD could support the transition for a circular economy, with environmental, social, and economic benefits for the local and regional sustainable development.

Keywords: Wastewater; Solid Waste; Anaerobic technology; Circular economy

### **1. Introduction**

Brazilian biodiversity is worldwide notable, especially in the Amazon rainforest, the most significant native forest of the world.<sup>1</sup> However, the Amazonian region lacks infrastructure and basic sanitation, and most cities from the northen region do not have appropriate waste collection and management systems. From an environmental perspective, agro-industrial waste can be potential feedstocks for bioenergy production, since they are renewable, abundant, and non-food sources, able to contain the drastic climate change attributed to the excessive dependence on fossil fuels.<sup>2</sup> The production of renewable fuels from biomass can be carried out in different technological routes, such as converting lignocellulosic material to produce fermentable sugars, bioethanol, organic acids, bioactive compounds, and biogas.<sup>3-6</sup> A previous study demonstrated the high potential of açaí waste to produce methane in a laboratory-scale anaerobic reactor,<sup>7</sup> which can be more environmentally friendly waste disposal, while also generating bioenergy.

Açaí (*Euterpe oleracea*), one of the essential cultivars from the Amazonian region, is mostly extracted from the river basins natural vegetation, and it's a vital source of food and economic resource for the local population.<sup>8,9</sup> The main product extracted from açaí is the pulp, which is consumed in the whole country due to its nutraceutical properties, as well as it is exported.<sup>10</sup> Beyond that, açaí presents various applications described in the literature for the food, pharmaceutical, and cosmetic industries.<sup>11</sup> In 2018, the Brazilian production of açaí reached 1.7 million tons, 1.5 million from cultivated or managed production,<sup>12</sup> and 221.6 thousand tons from the extractive production,<sup>13</sup> representing a robust economic sector to the north region of the country. Notwithstanding, 85% of the fruit's mass is not edible, and just 15% is processed to produce fresh and frozen pulp.<sup>14-17</sup> In Brazil, no treatment or appropriate disposal is convenient for açaí agroindustrial waste. The part of these waste is sold for masonry or compost production, and the large part is a challenge. A recent Brazilian law

established the National Biofuel Policy, known as RenovaBio, as an integral part of the country's energy policy, aiming to increase the use of bioenergy in the energy matrix while contributing to a proper trade-off of energy efficiency and greenhouse gases mitigation.<sup>18</sup> Besides, the European Union approved the Directive EU 2018/2001, establishing a mandatory renewable energy consumption of at least 32% of total energy until 2030.<sup>19</sup> With the approval of national and international law, there is attracting attention in the production of renewable fuels from biomass, as an alternative technological route for bioenergy production.<sup>20</sup> The agro-industrial waste generated in high quantity with greater availability throughout the year could be repurposed for bioenergy production, in a circular economy concept (**Figure 1**). The fruit waste is promising lignocellulosic biomass, which conversion allows the production of reducing sugars, bioethanol, organic acids, bioactive compounds, and also biogas (derived from the AD).<sup>21</sup> A technological route for the destination of solid and liquid industrial wastes could support the decrease of açaí's industry carbon footprint in Brazil.

Based on those mentioned above, this study aims to evaluate the current system boundary established in the Brazilian production of frozen açaí pulp, focusing on the waste generated during the agro-industrial processing and its environmental impacts. In the meantime, a new technological route for açaí's waste management energy recovery was proposed – in a circular economy concept.

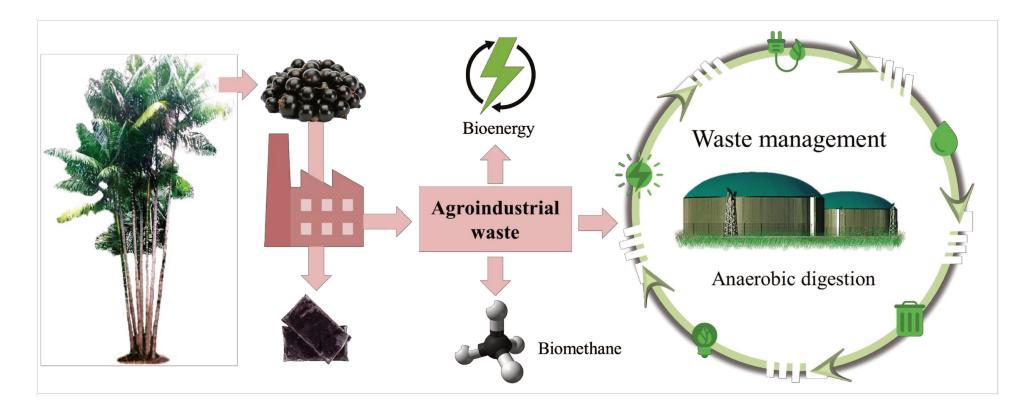


Figure 1. Schematic diagram of bioenergy production from açaí agroindustrial waste in a

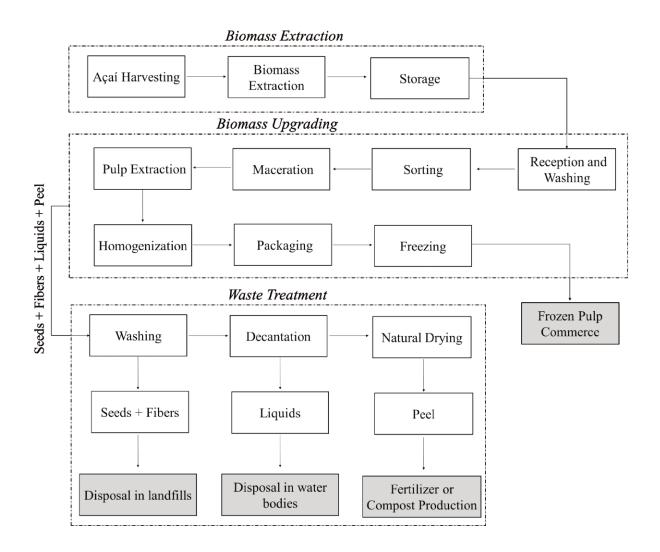
circular economy concept.

### 2. Current system boundary for açaí production and processing

The current system boundary for the açaí production chain was set from the production of fruits, then extraction, processing, and ending at the waste disposal (**Figure 2**). From the fruit, a drink called "açaí pulp" is extracted, which has been winning the national and international markets as an energetic and functional food, in addition to being a raw material in the pharmaceutical and cosmetics industry.<sup>10</sup> This growing demand is due to the large number of bioactive compounds contained in açaí, which present antioxidant, anti-inflammatory, anticarcinogenic, antimicrobial, analgesic, and vasodilatory properties.<sup>9,22</sup> This places açaí as one of the new "superfruits".<sup>22</sup>

The production of açaí fruits can be done using two different management techniques: extraction and cultivation. According to data from the Brazilian Institute of Statistics and Geography<sup>13</sup> related to Vegetable Extraction and Silviculture, for 2018, 221.6 thousand tons of açaí fruit were produced, generating a revenue of 162 million USD – average exchange rate for 2018 of 3.65 USD/R\$.<sup>23</sup> Regarding the data of production in temporary and permanent crops,<sup>12</sup> Brazil produced, in 2018, the amount of 1.5 million tons of açaí fruit, generating revenue of 910 million USD. Adding the two types of production, Brazil presented in 2018 a total of 1.7 million tons of açaí fruit for an income of 1.07 billion USD.

Extractive production occurs in all of the seven states in the northern region and in the State of Maranhão in the Northeast region of Brazil (the latter represents only 8% of the national output). The State of Pará is the leading producer of açai fruit (67% of the total) following of the state of Amazonas (21%). Production by cultivation technique does not occur in the states of Acre and Amapá in the Northern region. Nevertheless, it occurs in the states of Alagoas, Bahia, and Espirito Santo. Pará has an even greater hegemony, accounting for 95% of cultivated production. This production generates an income equivalent to 3% of the State's Gross Domestic Product (GDP), according to the Fruit and Derivatives Industries Syndicate report.<sup>24</sup>

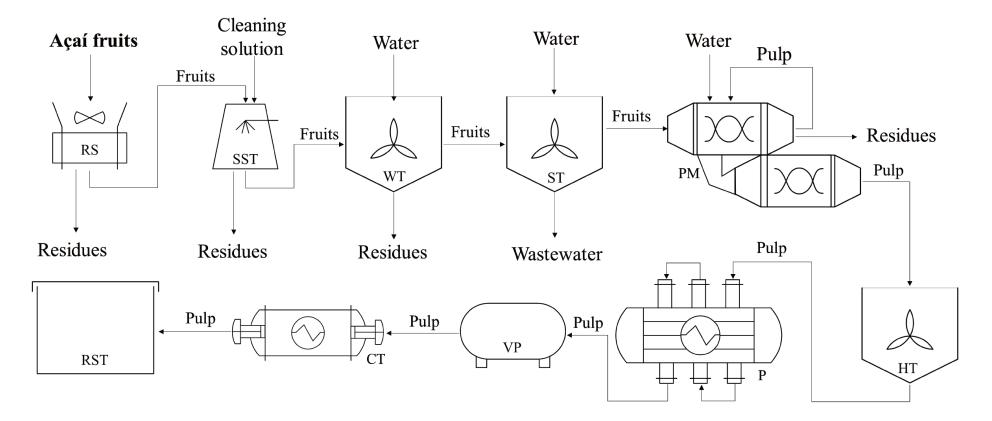


**Figure 2.** Current system boundary for açaí extraction, pulp upgrading, and waste treatment.

The açaí fruits have a globular, rounded shape, with a diameter of 1 to 1.6 cm and an average weight of 0.8 to 2.3 g, presenting a very thin epicarp of violet-purple color, almost black, when mature. After harvesting, the fruits are destined for industrial processing, according to **Figure 3**. Initially, the solids are separated from the fruits in a rotary separator. Fruits are sanitized with sodium hypochlorite solution and washed. In a softening tank, fruits are submitted to a heating process to facilitate pulp separation from the seeds. After pulp separation and homogenization, pasteurization follows to eliminate microbiological agents. The pasteurized pulp is packaged, refrigerated in a cooling tube, and then stored in a refrigerated chamber until the commercialization.

From **Figure 3**, a large amount of residual biomass and wastewater are generated during the açaí processing. Therefore, açaí seeds are the primary waste from the açaí processing industry, accounting for 85% of the fruit volume.<sup>9-16</sup> Although being used as a fertilizer by composting naturally handcrafted or burnt for thermal energy generation, these applications are not sufficient to absorb the amount of waste generated. Açaí seeds are considered urban waste and are currently a considerable inconvenience to the well-being and health hygiene of cities in northern Brazil.<sup>17</sup>

The conversion of this waste into bioenergy or biofuels would be attractive from commercial and environmental approaches. The açaí seed is lignocellulosic biomass, containing 43.81% of cellulose, followed by hemicellulose (25.89%) and lignin (24.56%), the latter composed by 22.99% of insoluble lignin and only 1.57% of soluble lignin.<sup>7</sup> Açaí seed has a high content of carbohydrates (cellulose and hemicellulose), approximately 70%, which justifies its use for biofuels. Besides, there are few studies in the scientific literature on its energy use through biotechnological platforms,<sup>25</sup> and AD can be a promising technology for this residue repurposing.



**Figure 3.** Industrial scheme to produce frozen açaí pulp. Legend: RS, rotary separator with grid; SST, sanitization and separation tank; WT, washing tank; ST, softening tank; PM, pulping machine; HT, homogenization tank; P, pasteurizer; VP, vacuum packer; CT, cooling tube; RST, refrigerated storage chamber.

AD is a natural process that occurs in the absence of oxygen, resulting in the production of a gas mixture known as biogas, mainly composed of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>).<sup>26,27</sup> AD is employed for wastewater treatment, as well as for the treatment of organic waste with high moisture content, solids, and organic materials. Since AD is a complex dynamic system involving microbiological, biochemical, and physicochemical processes, it is necessary to establish operational parameters to produce biogas with a high content of methane.<sup>28-29</sup>

Solids, pH, alkalinity, ammonium nitrogen, chemical oxygen demand, and volatile fatty acids are the most critical variables to control along with AD, once they directly affect the biogas yield.<sup>30-31</sup> Setting the better operational parameters for the AD, biogas with high methane contents can be produced, and further applied to generate electric and thermal energies. Both could be used for self-consumption, while any eventual electrical energy surplus could be sold to the grid. The biogas can undergo a purification process to the elimination of sulphur compounds and carbon dioxide.<sup>32,33</sup> This process is necessary to avoid the corrosion in the motor-generator and to reduce the density of the gas.<sup>32,33</sup> Therefore, after the purification, more than 95% of methane is obtained, and the biomethane can be injected directly into existing natural gas networks.<sup>34</sup> From an environmental perspective, beyond reducing the content of organic matter in a waste, AD produces clean energy, a value-added product to energy matrix decarbonization.

Due to the issues related to the high production of açaí waste during the processing, an innovative approach focusing on waste management was developed. **Figure 4** shows the suggested system boundary compared to the waste management of solid and liquid waste through AD.

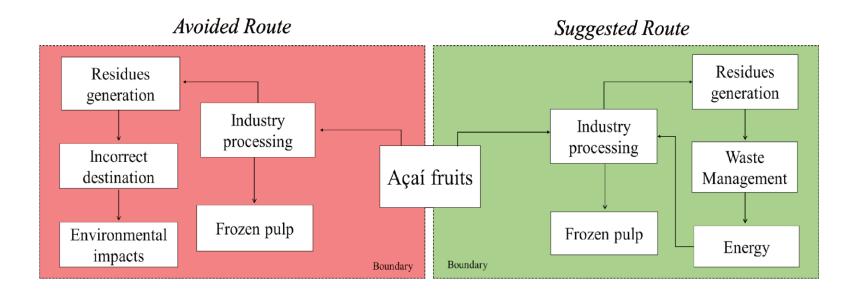


Figure 4. Suggested system boundary for açaí production, focusing on waste management.

For açaí processing industries, AD applied under mesophilic conditions produces energy (methane). According to Maciel-Silva et al.,<sup>7</sup> an anaerobic reactor of 4.3 L fed by 25% of açaí solid waste (high content of solids), 30% of mesophilic sludge, and 45% of açaí processing wastewater, with a hydraulic retention time of 35 days, the accumulated volume of biogas was around 4 L, composed by 50% of methane, with a yield of 7.79 L biogas kg<sup>-1</sup> of total volatile solids. This yield can eventually be considered low, and dry AD turns the waste in a more easily digestible waste, so taking less time to decompose. Futures studies of co-digestion and pre-treatment steps could increase the biogas yield for industrial applications. These require extensive research and possibly the combination of several processes to improve the returns and economic balance to solve the problem.

The conventional AD is a worldwide spread well-known technology and, for a real application to the industry, requires a mass and energy balance to assess the technological transfer. **Figure 5** presents a case study based on one (01) ton of açaí fruits daily processing. The industrial balance aims to demonstrate a possible route to the conversion of agro-industrial waste into new products. An initial base of 1,000 kg of açaí fruits was established for daily processing. The açaí yield is estimated in only 15%, and to each ton of fresh fruits processed, 0.92 L of water is necessary to all the process (*in loco* estimative) (**Table 1**). Beyond, for the frozen pulp production, 564.26 L of water is added in the pulp to produce the commercialized frozen pulp.

Parameters	Value Unit		Reference	
Yield of açaí edible part	0.15	kg açaí pulp kg <sup>-1</sup> of fresh fruit	9	
Ratio of açaí fruits waste	0.85	kg açaí seed kg <sup>-1</sup> of fresh fruit	9	
Frozen pulp production	1.4	kg of fresh fruit kg <sup>-1</sup> of frozen pulp	In loco estimative	
Total water demand	0.92	L of water ton <sup>-1</sup> of fresh fruit	In loco estimative	
Electric energy demand	25	kWh ton <sup>-1</sup> of frozen pulp	In loco estimative	
Wastewater generation	0.51	L kg <sup>-1</sup> of frozen açaí pulp	35	
Biogas production from AD	2.28	m <sup>3</sup> of biogas ton <sup>-1</sup> of waste	7	
Methane content	50	%	7	
Electricity production	3.95    kWh m-3 of biogas		Estimative*	

Table 1: Assumptions adopted to calculate the mass balance of the açaí industry.

\* Estimation based on the data obtained Maciel-Silva et al.<sup>7</sup> with the theoretical calculation

based on Jiménez-Castro et al.<sup>29</sup>

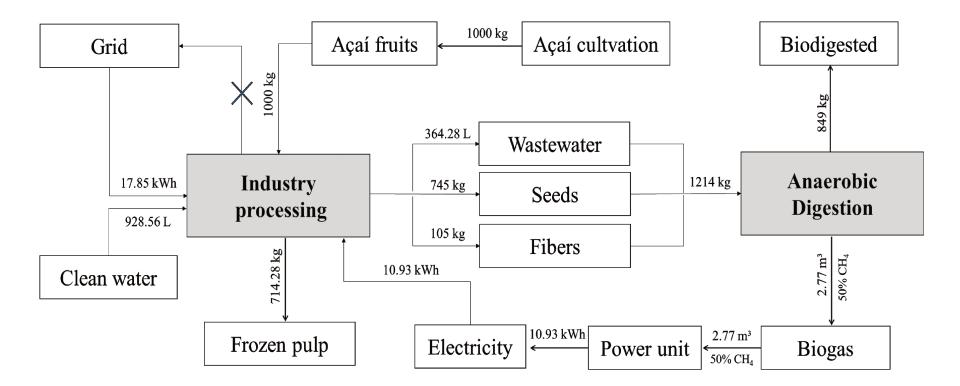


Figure 5. Waste management of açaí through anaerobic digestion: mass and energy balance.

Consequently, from 1000 kg of fresh fruits, 150 kilograms of fresh pulp are produced, with total water consumption of 928.56 L, and 364.28 L are used utility and finally discarded as wastewater. An outstanding outcome is that 78% of the frozen açaí pulp commercialized is composed of the water added in the processing. In contrast, 22% is the fruit itself. Furthermore, for this daily basis adopted for this simulation, the wastewater generated is estimated in 0.51 L kg<sup>-1</sup> of frozen açaí pulp produced.<sup>35</sup> Then, 364.28 L of açaí wastewater is generated, and this value represents 36% of the initial açaí mass fed in the industry.

Around 85% of the açaí solid waste is composed of seeds,<sup>9</sup> and the other fraction is represented by fibers, which constitute 745 kg and 105 kg, respectively. This agro-industrial waste is a potential feedstock to produce biogas because of its high chemical oxygen demand.<sup>7</sup> After AD, around 850 kilograms of biodigested are generated, and this organic material could return to the field to be used as a natural fertilizer, avoiding the use of mineral fertilizers. On the other hand, the biodigested can proceed to stabilization ponds and return to water bodies, contributing positively to the açaí industrial chain, reducing environmental impacts.

A scenario for the production of electricity from methane was proposed. The 1000 kg of waste fed in a continuous reactor produced 2.28 Nm<sup>3</sup> of biogas, operating with a hydraulic retention time of 35 days.<sup>7</sup> Then, 1214 kilograms of açaí waste (including solid and liquid fraction) could produce up to 2.77 Nm<sup>3</sup> of biogas with a methane content around 50%. This biogas can be burned in a motor-generator to generate electricity, as described by Jiménez-Castro et al.,<sup>29</sup> taking into account the volume of biogas produced, the lower calorific value of methane, percentage of methane in biogas, and engine efficiency.

Considering that the whole process to produce frozen açaí pulp demands 25 kWh per ton of frozen pulp (data obtained *in loco*) – equivalent to 17.85 kWh per 714.28 kg of frozen pulp – the local energy produced by the biogas burning could be self-consumed, establishing

a circular energy economy for the industry. Based on the condition described and using theoretical calculations to quantify the potential electric energy generation, around 10.93 kWh could be produced from the burning of 2.77 Nm<sup>3</sup> of biogas rich in methane. This energy could supply the facility's demand, while, as in the current case, there is no energy surplus to sold back to the grid. In summary, the açaí industry here simulated needs to buy only 6.92 kWh from the grid, 61% lower than the conventional process without energy recovery, which can be seen as a positive outcome and a contributor to the energy matrix decarbonization.

### 4. Circular economy for sustainable industrial development

Despite the benefits previously demonstrated with bioenergy production from AD, the recovery of industrial waste can enhance the cost-effective of the agro-industrial supply chain.<sup>36</sup> Stable and straightforward AD technology can facilitate its implementation in small and medium-size factories, which is a benefit to the transition to a circular economy. The main advantages of AD technology to a circular economy include cost savings, closing production-to-waste loops, and increasing the reuse and recycling of bioenergy and biomaterial.<sup>37,38</sup> The circular economy is also a conceptual model widely used for appropriate waste management,<sup>39</sup> and agroindustrial waste is a feedstock well defined to play sustainable development based on bioeconomy.<sup>37</sup> Notwithstanding, with the crescent demand for energy, the biogas produced can be a substitute for fossil fuels, reducing the environmental impacts and contributing to the energy matrix decarbonization.<sup>40</sup> In addition, Brazil is one of the largest producers of agricultural commodities, which generated large amounts of solid and liquid waste. The circular economy implies waste prevention, reuse, and recycling. It is a powerful concept to foster resource use, waste generation, and GHG emissions, which contribute to the environmental and economic policymakers' decisions.<sup>41,42</sup> Figure 6 presents an illustration related to the transition of linear to the circular economy, focusing on sustainable growth. For instance, it is necessary to balance the industrial development with environmental conservation, focusing on strategies for economic gain and valorization of agro-industrial waste.<sup>38</sup> Beyond, a circular economy aims the creation of economic, social, and environmental value. In the definition of bioeconomy, it is necessary to prevent unhealthy working conditions, uncorrected raw materials extraction, and even improper destination of natural resources.<sup>43</sup>

### 5. Concluding remarks: açaí waste management for a circular economy

Currently, climate change related to the fossil-based economy has been an international concern. The research on renewable energy production aims to introduce innovative sources of cleaner energy. AD is one of the most promising low-cost technologies for this purpose. Biogas is a renewable energy that could replace fossil fuels in electric and thermal energy generation and even used as a vehicular fuel. Moreover, the circular economy paradigm involves the wide adoption of technologies to reduce natural resource scarcity like freshwater and environmental side-effects caused by fossil fuel. Developing countries, commonly, do not present effective waste management policies to avoid improper disposal of agroindustrial waste, especially for small and medium plants. In the case of açaí, *in loco* observations, evidenced that the pulp production was centered in low productive and smaller companies. On the other hand, even in low processing capacity, high amounts of waste are generated. The adoption of viable and practical solutions to waste management is a necessary local strategy to reduce environmental impacts. Besides, technologies such as AD should be encouraged to produce clean energy, likewise biogas. A schematic illustration with the alternative proposed route can be observed in **Figure 7.** 

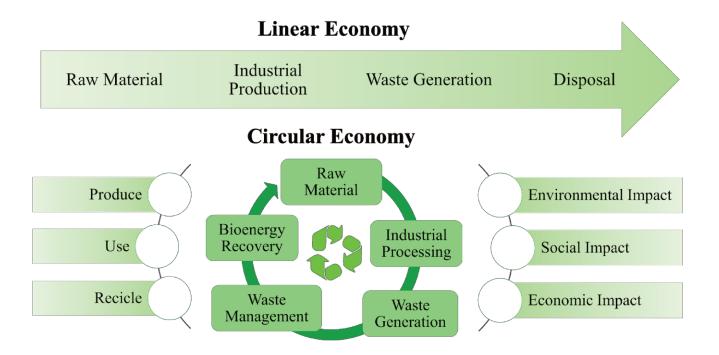


Figure 6. Illustration between linear and circular economy focusing on sustainability.

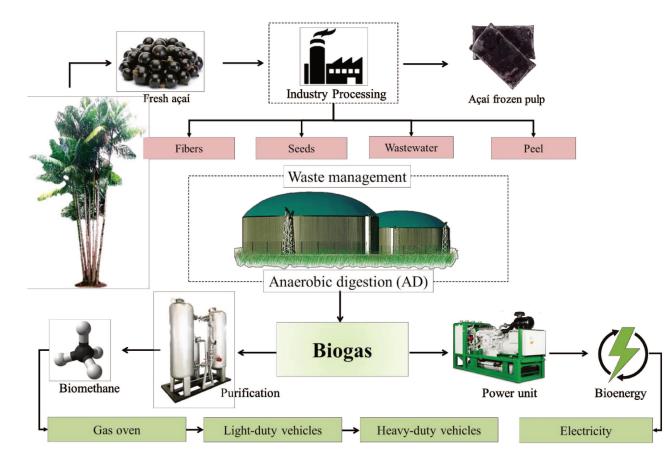


Figure 7. Waste management of açaí waste through anaerobic digestion technology.

In the açaí industry, most of the energy employed is destined to produce a global yield of 37%, considering the raw materials input flows (water and açaí fruits) and the final production of frozen açaí pulp (**Figure 5**). In the industrial processing of açaí (**Figure 3**), most of the physical operations demand electric energy, especially the pulping machine, pasteurizer, and cooling tube. The global yield calculated in the present study is directly related to the fraction of waste generated in the physical operations. For instance, rotary separator, sanitization and separation tank, washing tank, softening tank, and pulping machine make a significant amount of solid and liquid waste. With the AD adoption for açaí waste management, biogas can be produced and further converted into electric and thermal energy. In addition, it can be purified into biomethane to the use of a gas oven, light, and heavy-duty vehicles.<sup>34</sup> In the case of biomethane, the several possible uses aforementioned are additional routes to mitigate greenhouse gas emissions.<sup>34</sup> Such routes are strategies for the development of a circular economy that mitigate environmental side-effects while reinforcing productive chain sustainability.

This study addressed a well-consolidated technology for the treatment of açaí agroindustrial waste, which could be applied as an immediate solution for a real and local problem. From an environmental perspective, the results demonstrated a potential solution to the proper disposal of solid waste generated by this strong agro-industrial sector in the Brazilian Amazonian region (which faces limited access to utilities in its remote areas). Moreover, the operational performance of the anaerobic digester fed with açaí seeds and wastewater can be considered favorable for a real implementation. Besides, the dry AD converts the lignocellulosic waste into a more easily and quickly digestible substrate. The application of AD is feasible for bioenergy recovery, in contrast to landfills, composting, and burning, contributing to the energy matrix decarbonization. In a circular economy, the adoption of new waste management routes can be a practical solution to produce bioenergy, contributing to beneficial environmental impacts of the açaí industries, towards local and regional sustainable development.

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## Capítulo 5 – Discussão Geral

Dentro do conceito de bioeconomia, que é a implementação de insumos orgânicos dentro de cadeias de valor com o objetivo de oferecer soluções para sustentabilidade. Este trabalho estuda a digestão anaeróbia que é uma técnica ambientalmente favorável para tratamento de resíduos orgânicos e geração do biogás, com isso, agregando valor ao que antes seria apenas um resíduo, gerando eletricidade, reduzindo a emissão de gás carbônico, também permitindo a criação de empregos e renda. A seguir será apresentada uma discussão mais detalhada de cada capítulo.

## 5.1. Discussão sobre o Capítulo 2 - Estudo de caso: Utilização de um reator UASB em uma planta de celulose

Neste capítulo foi analisado a implantação de um reator UASB no sistema de tratamento de uma planta integrada de papel e celulose. Foi considerada a utilização do biogás gerado tanto para a geração de calor, quanto para a geração de eletricidade, verificando custos e impactos ambientais.

Os resultados mostraram que tanto o uso de biogás para geração de calor e eletricidade reduzem os impactos ambientais de aquecimento global, sendo o impacto evitado na ordem de 10 vezes maior para a geração de calor, pois nesse caso substitui um combustível fóssil, o gás natural e no caso da geração de eletricidade, substitui energia gerada por matriz energética relativamente renovável, com uma grande participação de energia hidrelétrica.

No caso dos custos evitados, o resultado é inverso, sendo o biogás usado para geração de energia elétrica a alternativa que permite maior custo evitado, isso ocorre devido a uma boa parte (aproximadamente 40%) da energia gerada no Brasil vir de termelétricas o que encarece a energia devido aos custos do combustível.

Os resultados financeiros mostraram que a implementação do sistema UASB na usina em estudo teve pouco impacto no resultado financeiro total da usina, mostrando assim sua implementação não afeta a competitividade da usina.

## 5.2. Discussão sobre o Capítulo 3- Usos finais do biogás na forma de biometano: Análise de Impacto Ambiental

Neste capítulo foram estudados 3 usos de biometano: Em fogões domésticos, em veículos leves e em veículos pesados. Para cada uso foi considerado que o biometano substituía um combustível fóssil padrão. Foram analisadas quatro categorias de impacto (mudanças climáticas, eutrofização, oxidação fotoquímica e acidificação) usando a Avaliação de Ciclo de Vida (ACV).

Os resultados mostraram que o uso de biometano em substituição de óleo Diesel, em veículos pesados, foi vantajosa em todas as categorias, sendo mais vantajosa na categoria de mudanças climáticas. Mesmo com características peculiares do óleo Diesel brasileiro, como 5% em volume de biodiesel, e baixo teor de enxofre (comparado a padrões mundiais), o biometano ainda se mostra vantajoso nesse uso.

O caso da substituição de GLP (gás liquefeito de petróleo) por biometano em uso doméstico, a substituição é praticamente indiferente, com exceção da categoria de mudanças climáticas, isso se deve ao biometano ter praticamente as mesmas emissões do GLP, porém no caso do CO<sub>2</sub>, o do biometano não tem impacta nessa categoria, visto que o biometano é um combustível renovável. Não foram consideradas as perdas nas conexões das tubulações, pois no cenário proposto o gás seria utilizado em uma residência próxima de onde ele é gerado. As perdas por vazamentos na tubulação podem impactar consideravelmente o resultado da análise ambiental final.

No caso dos veículos leves, a substituição da gasolina C por biometano é desvantajosa em todas as categorias exceto mudanças climáticas. Isso se deve principalmente aos motores de veículos a gás natural (quimicamente semelhante ao biometano, porém de origem fóssil) são veículos a gasolina adaptados, portanto produzem muitas emissões de particulados e óxidos de nitrogênio, o que contribui para as categorias de impacto de eutrofização.

# 5.3. Discussão sobre o Capítulo 4- Gerenciamento de resíduos e recuperação energética do processamento de açaí na região amazônica brasileira: Uma perspectiva para uma economia circular

Neste este estudo foi feita uma análise da produção brasileira de açaí, com foco na geração de resíduos e abordando os balanços de massa e energia definidos a partir do cultivo, extração, processamento e destinação do resíduo. Além disso, foi introduzida uma nova rota tecnológica para a gestão dos resíduos do açaí para valorização da bioenergia dentro do conceito de economia circular.

Em 2018, o Brasil produziu 1,7 milhão de toneladas de açaí para uma receita de 1,07 bilhão de dólares, e para essa produção, a geração de resíduos (sementes) foi estimada em 85%. A produção extrativa ocorre em todos os sete estados da região Norte e no Estado do Maranhão na região Nordeste do Brasil (este último representa apenas 8% da produção nacional). O Estado do Pará é o maior produtor da fruta açaí (67% do total), seguido do estado do Amazonas (21%).

Devido à alta produção de resíduos, uma abordagem inovadora foi desenvolvida para um limite de sistema, incluindo o gerenciamento de resíduos sólidos e líquidos por meio da digestão anaeróbia (AD).

Os resultados mostraram que a partir de 1 tonelada de açaí alimentada na unidade para processamento, foi gerada 1,2 tonelada de resíduos sólidos e efluentes. Esse resíduo submetido à AD pode produzir 2,77 Nm<sup>3</sup> de biogás, com composição de metano de 50%, podendo repor 61% da necessidade de eletricidade externa para o processamento do açaí.

A adoção dessa tecnologia pode ser um resultado positivo como contribuinte para a descarbonização da matriz energética. Além disso, a implementação do AD poderia apoiar a transição para uma economia circular, com benefícios ambientais, sociais e econômicos para o desenvolvimento sustentável local e regional.

# Capítulo 6 – Conclusão Geral

Neste trabalho, foi feito avaliações do potencial de aproveitamento energético do biogás resultante da digestão anaeróbia de resíduos orgânicos através de estudos de potencial energético em diversos cenários e avaliações do impacto ambiental causado pela adoção do biogás como fonte de energia.

Uma abordagem integrada foi feita para explorar um reator UASB para reciclagem de energia na indústria de celulose e papel no capítulo 2. Este estudo de caso em um país em desenvolvimento permitiu as seguintes conclusões pontuais:

• A adoção de reatores UASB na indústria de papel e celulose auxilia nas questões ambientais (qualidade do ar e da água) e contribui para a transição para uma matriz energética com maior participação de fontes renováveis;

• A análise financeira mostrou que o investimento em um reator UASB apresenta resultados atrativos. O aporte inicial não afeta o balanço financeiro da indústria, tem um retorno sobre o investimento (> 50%) com um curto tempo de reembolso. Além disso, considerando a produção total brasileira de papel, se poderia evitar a emissão de 1.06 x  $10^5$  ton CO<sub>2eq</sub> por ano, efetivamente contribuindo para a mitigação das emissões;

• Considerando os crescentes custos energéticos e preocupações relativas as mudanças ambientais, o biogás se torna uma fonte renovável de energia economicamente atrativa. Pode ser usado para substituir fontes convencionais de energia, tais como gás natural ou óleo Diesel, ao mesmo tempo que reduz a pegada de carbono.

A avaliação do impacto ambiental das utilizações finais do biometano mostrado no capítulo 3 permitiu as seguintes conclusões pontuais:

• A substituição do GLP por biometano em usos domésticos apresenta uma contribuição especial para contribuir para a mitigação de mudança climática;

• Biometano substituindo gasolina-C pode apresentar menores emissões usando motores mais apropriados para o biometano;

• Em todas as categorias de impacto analisadas, a substituição de óleo Diesel por biometano em veículos pesados, apresentou desempenho ambiental superior, mesmo considerando o baixo conteúdo de enxofre no óleo Diesel brasileiro e uma fração de biodiesel na mistura;

• O uso de tecnologia de biometano em automóveis poderia estabelecer uma matriz energética mais limpa no Brasil, e auxiliar no transporte de produtos agrícolas e industriais;

• A substituição de óleo Diesel por biometano no transporte público, que depende fortemente de ônibus, pode trazer impacto significativo e favorável;

• A introdução de biometano em veículos pesados poderia diminuir a pegada de carbono de vários sistemas agrícolas e urbanos;

• O uso de biomassa que gera o biometano poderá permitir uma integração benéfica dentre os setores industriais e agrícolas, favorecendo estratégias que dão lugar a uma economia circular.

Uma abordagem integrada foi feita quanto ao gerenciamento de resíduos e recuperação energética do processamento de açaí na região amazônica brasileira no capítulo 4. Esta perspectiva para uma economia circular permitiu pontuar as seguintes conclusões:

 Digestão anaeróbica é uma das tecnologias de baixo custo e o biogás poderá promover energia renovável para substituir os combustíveis fósseis na geração de energia elétrica e térmica e até mesmo utilizada como combustível veicular;

• O processamento de açaí evidencia que a produção de polpa está centrada em empresas de pequeno porte que focam no mercado local, entretanto estas são capazes de gerar grande quantidade de resíduos;

• Na indústria do açaí, a maior parte da energia empregada (37%) estão nas etapas de fluxos de entrada de matérias-primas (água e frutos do açaí) e a produção final da polpa de açaí congelada, principalmente, máquina de polpação, pasteurizador e tubo de resfriamento;

• A fração de resíduos que são gerados nas operações físicas, por exemplo, separador rotativo, tanque de sanitização e separação, tanque de lavagem, tanque de amaciamento e máquina de polpação geram uma quantidade significativa de resíduos sólidos e líquidos;

• O gerenciamento de resíduos do açaí pode ser feito através da digestão anaeróbia e produção de biogás, que pode ser convertido em energia elétrica e térmica, ou purificado em biometano para uso em forno a gás, veículos leves e pesados;

• O biometano pode estabelecer rotas ou estratégias para o desenvolvimento de uma economia circular que mitiguem os efeitos colaterais ambientais e reforcem a sustentabilidade da cadeia produtiva;

• Do ponto de vista ambiental, os resultados demonstraram uma solução potencial para o descarte adequado dos resíduos sólidos gerados por esse forte setor agroindustrial na região amazônica brasileira (que enfrenta dificuldades de acesso a utilidades em suas áreas remotas). Finalmente, conclui-se que a digestão anaeróbia é uma técnica ambientalmente favorável para tratamento de resíduos orgânicos e geração do biogás. A implementação de reatores anaeróbios para o tratamento de resíduos orgânicos poderia apoiar a transição para uma economia de baixo carbono, com benefícios ambientais, sociais e econômicos para o desenvolvimento sustentável.

O aluno Samuel Fontenelle Ferreira ingressou como doutorando na UNICAMP em 2017 através de processo seletivo do Departamento de Engenharia de Alimentos (DEA). Com aprovação de bolsa de doutorado do CNPq (processo nº 142091/2017-0) durante 42 meses. Além das disciplinas cursadas no mestrado em 2015 e 2016 na mesma universidade, foram cursadas em 2017, 2018 e 2019: BI003 Sustentabilidade Social, Econômica e Ambiental; IC770 Tópicos em Saneamento Ambiental V; PE162 Política Energética, Planejamento e Regulação; PE172 Avaliação do Ciclo de Vida Aplicada à Energia; TP199 Seminários e participação no Programa de Estágio Docente grupo C (PED C) com atividades de apoio parcial à docência da disciplina de: Laboratório de Operações Unitárias (TA 035) no 1° semestre de 2017; 1° semestre de 2018 e 1° semestre de 2019.

A aluno participou em 2017 no evento 12º SLACA (Simpósio Latino-Americano de Ciência dos Alimentos), realizado em Campinas. Em 2018 participou do Fórum Sul-Brasileiro de Biogás e Biometano, realizado em Foz do Iguaçu.

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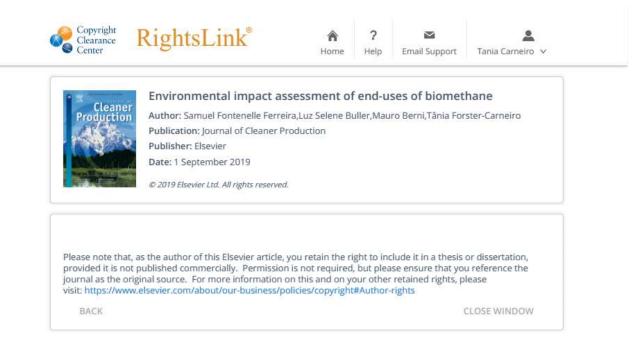
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