# Liquefied biomethane from sugarcane vinasse and municipal solid waste: sustainable fuel for a green-gas heavy duty road freight transport corridor in Sao Paulo state

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**Abstract** – Diversifying the energy components of a country's transport sector is essential to guarantee the fuel supply to consumers and increase the market dynamics and competitiveness. Among the known alternative fuels, biogas is a renewable source and after upgrading to biomethane, it presents a similar composition to natural gas (> 90% of $CH_4$; 35-40 $MJ.m^{-3}$). In addition, it can be produced from a wide variety of biological resources and at different scales In this study, two scenarios have been developed that evaluate the use of liquefied biomethane (LBM) as a diesel replacement option in the freight sector of an area of 248,223 $km^2$ (equivalent to the area of the UK). Sugarcane vinasse (SVC) and Municipal Solid Waste (MSW) were the sole feedstocks for biogas production. The first scenario, non-restricted scenario (NRS), covered the entire territory while, the second scenario, restricted scenario (RS), includes only the area where gas pipelines are available. An economic assessment of the entire biogas value chain including, biogas production units, purification, transport and end-use was performed. The minimum selling price (MSP) of biomethane throughout the biogas chain was then estimated. LBM is estimated to be a cost-effective and affordable fuel choice compared to diesel. The technical potential of biogas production by the sugarcane mills and landfills of Sao Paulo state can replace up to half of the diesel consumed in the territory. The minimum distances and optimal locations methodology indicated the need for 120 liquefaction plants in the NRS, 35 injection points in the RS, and 7 refueling stations to supply LBM throughout the state of Sao Paulo. The units for $CO_2$ removal had the greatest influence on capital costs (~60%) in both scenarios. Expenditure associated with the gas injection operation and its transport comprised more than 90% of the operating costs of the RS. Electricity purchasing represented the highest share of the operating costs at biogas purification (20%-30%) and biomethane liquefaction (65%-91%) units. Personnel costs are observed along the entire biogas chain, especially, in the biomethane transport step (40%), indicating an opportunity to generate wealth, jobs, and income. Despite our projections for the cost-effective and competitive supplies of LBM as a diesel replacement fuel, policy support measures such as a feed-in tariff, are likely to be necessary in order to overcome non-technical barriers and gain wider acceptability.

**Keywords:** sugarcane vinasse, municipal solid waste, liquefied biomethane, transport sector, economic assessment, diesel replacement.

**Abbreviations:**

AD – Anaerobic digestion
BC – Blue Corridor
BDS – Biological desulfurization systems
BRA – Brazilian Real
CBM – Compressed biomethane
CNG – Compressed-natural gas
COD – Chemical Demand of Oxygen
DER – Department of Highways of Sao Paulo
EPE – Brazilian Energy Research Company
EU – European Union
GHG – Greenhouse gases
GPD – Gross Domestic Product
HDV – Heavy-duty vehicles
HRT – Hydraulic Retention Time
IPCC – Intergovernmental Panel on Climate Change
LBM – Liquefied biomethane
LFBG – Biogas from landfills
LNG – Liquefied-natural gas
MDR – Brazilian Ministry of Regional Development
MRC – Mixed refrigerant cycle
MSP – Minimum selling price
MSW – Municipal solid waste
NRS – Non-Restricted Scenario
OFMSW – Organic fraction of municipal solid waste
OLR – Organic Loading Rate
RS – Restricted Scenario
SCV – Sugarcane Vinasse
SNIS – Brazilian Sanitation Information System
UASB – Upflow Anaerobic Sludge Blanket reactor
USD – United States Dollar

# 1 INTRODUCTION

In the 2000s, structural changes in eastern European countries and the integration of some of them into the European Union (EU) have increased the volume of heavy-duty vehicles (HDV) transiting through the continental territory, which lead to high levels of atmospheric pollution such as nitric and sulphur oxides and hydrocarbons [1,2]. The replacement of petrol and diesel could play an important role in cutting such emissions. Hence, the idea of corridors for the HDV using compressed-natural gas (CNG) as fuel was launched and named as Blue Corridor (BC).

Technical and economic assessment of three pilot CNG-BC (1. Moscow – Berlin (International road E30); 2. Berlin – Rome (Road E55 and E45); 3. Helsinki – Moscow (road E18-E105)) was performed based on traffic volumes (16,000 lorries), savings in fuel costs, reductions in emissions and number of existing CNG fuelling stations. For instance, the fuel savings on Moscow – Berlin corridor would amount to more than 300 million Euros per year and harmful exhaust emissions would be reduced up to 60%. For optimal fuelling potential, 25 new CNG stations would be required at an average cost of 250,000 Euros each [1].

Gas liquefaction provides a reduction in volume (up to 1:600 at standard conditions for temperature) and, consequently, the increment of its energy density [3,4]. Therefore, it might represent an alternative to minimise the number of fuelling stations from a greater autonomy of HDV. Mouette et al. [5] evaluated the adoption of liquefied-natural gas (LNG) on inter-cities-BC with a traffic rate between 145-200 trips per day within an equivalent area of the United Kingdom. For the mentioned scenario, LNG-BC could, indeed, reduce the cost of fuel by 40%. However, the greenhouse gases (GHG) emissions would be reduced by 5.2% only.

A greener approach towards sustainable fuels in the transport sector is imperative. Furthermore, it is in agreement with the regular scientific assessments on climate change provided by the Intergovernmental Panel on Climate Change (IPCC) [6]. In this sense, biogas is one of the greatest examples of renewable biofuel.

Biogas is formed during the decomposition of residues by microorganisms. This process is also known as anaerobic digestion (AD) and can occur both naturally under lacking dissolved oxygen environments but, also at biogas plants with controlled conditions. It can be produced from a large variety of feedstock (substrate) including food industry wastewater [7], lignocellulosic biomass [8], and municipal solid waste [9]. This feature might also be allied to different plant-scales and mobility (i.e., centralised- or decentralised AD-plants) [10], which makes AD a low-cost multipurpose technology.

One of the biogas components is methane, a versatile energy carrier that can be used to supply a local thermal demand, electricity generation or, upgraded to fuel vehicles and injected into the gas grid as biomethane [11]. However, it is important to bear in mind that both feedstock and biogas end-uses are determined by the socioeconomic context of a given territory.

Sugarcane Vinasse (SCV) is a residual liquid stream from the ethanol production process, generated in the distillation stage at an average ratio of 12 L per litter of ethanol produced [12]. It has been extensively studied to produce biogas [13–16] and reported as an excellent feedstock in biogas end-use scenarios [17–20] due to its large generation volume (490 billion litters - 2019-2020 Brazilian season) [21] and high organic matter and nutrients content [15,22]. Economic assessment of diesel replacement in agricultural operations of sugarcane mills has been pointed out as the best scenario for biogas end-uses in detriment to the energy electricity generation via Combined Heat and Power (CHP), a biogas-default application [17,18,23].

Broader scenarios have also been investigated aiming to identify business opportunities for the sugarcane plants. Poveda [18] evaluated the integration of compressed biomethane (CBM) into the transport sector of Metropolitan Region of Ribeirão Preto, Brazil (equivalent area of Montenegro) and its environmental benefits. According to the author, a medium sized mill (600 $m^3_{ethanol}.d^{-1}$) has the potential to fuel about 740 HDV during the sugarcane harvest and, then, supplemented with CNG. Furthermore, negative GHG emissions ($-29.0$ kg $CO_2 .m^{-3}{}_{SCV}$) were computed, regarding the scenario with SCV fertigation: a common practice at the mills.

Biogas from landfills (LFBG) can be addressed to surplus the lack of biogas/biomethane during the sugarcane off-season besides attending the continuous demands outside the economic radius of the mills (20 km) [18,24–26], considering that municipal solid waste (MSW) is one of many anthropogenic biomass residues widely available.

Similarly, MSW has been studied as a feedstock to produce biogas [9,27,28] being the electricity generation, in this case, the most common end-use [9,28,29]. Di Maria et al. [30] estimated a value for energy recovery from LFBG ranged from approximately 11 to 90 kWh per ton of disposed mechanically sorted organic fraction of MSW. At a macro level, the LFBG would benefit the Brazilian energy system by supplying an additional 79.4 MW of electricity each month [31]. The viability in the use of liquefied or compress biogas to replace diesel in urban bus fleet was also determined by Nadaletti et al. [29]. In the mentioned study, Brazil has the potential to generate about 16,131,857 $Nm^3_{biogas}.h^{-1}$ which could supply the actual bus fleet, estimated in 107,000 vehicles; and prevent emissions of GHG (4.3 $tonCO_2. d^{-1}$).

Most of the aforementioned technologies are established and reliable today. However, their techno-economic feasibility must be analysed to assess the net benefit. Therefore, the aim of this study is to evaluate the emerging market for both sugarcane mills and landfills to produce biogas; and the corresponding market for liquefying biomethane to fuel the freight transport system of Sao Paulo state.

## 2 MATERIALS AND METHODS

In this section, the scenarios for biogas production, upgrade and use as Liquified Biomethane (LBM) in the freight transport system of Sao Paulo state will be contextualised. The input data and methodologies for the technical-economic assessment will be presented in detail, providing the basis for replicating to other areas.

### 2.1 Overview of the Sao Paulo state

Sao Paulo state (248,222.8 $km^2$ - equivalent area of the UK) has a major industrial complex of the Federative Republic of Brazil, being responsible for 33.9% of the country's Gross Domestic Product (GPD) [32]. With more than 46 million inhabitants in 2019, Sao Palo state centralises a significant infrastructure network, for instance, more than 200,000 km of highways (including roads and municipal highways) [32] and 3,500 km of gas pipelines [33] which makes this territory a likely area for introducing emerging technologies.

Geographical regions of the State of Sao Paulo were obtained from the Brazilian Institute of Geography and Statistics [34]. The main roads in the state were obtained from the Sao Paulo State Data Analysis System Foundation [35]; Gas pipelines shapefiles were obtained from the Energy Research Company [36]. The information on the location of ethanol mills and their annual ethanol production (in $m^3$) were taken from the Geographic Information System of the Brazilian Energy Sector, created by the Brazilian Energy Research Company (EPE) [36]. Data on landfills and ethanol mills were taken from two primary sources. Landfill location and daily municipal waste received (in ton) information was obtained from the National Sanitation Information System (SNIS), maintained by the Ministry of Regional Development (MDR) [37]. Out of 208 landfills registered on the SNIS, 148 did not present any location data. Further 83 landfill locations were collected manually online, leaving 65 landfills out of the study due to lack of location information. Figure 1 shows an overview of the Sao Paulo State and its infrastructure corresponding to the biogas-chain in the freight transport system.

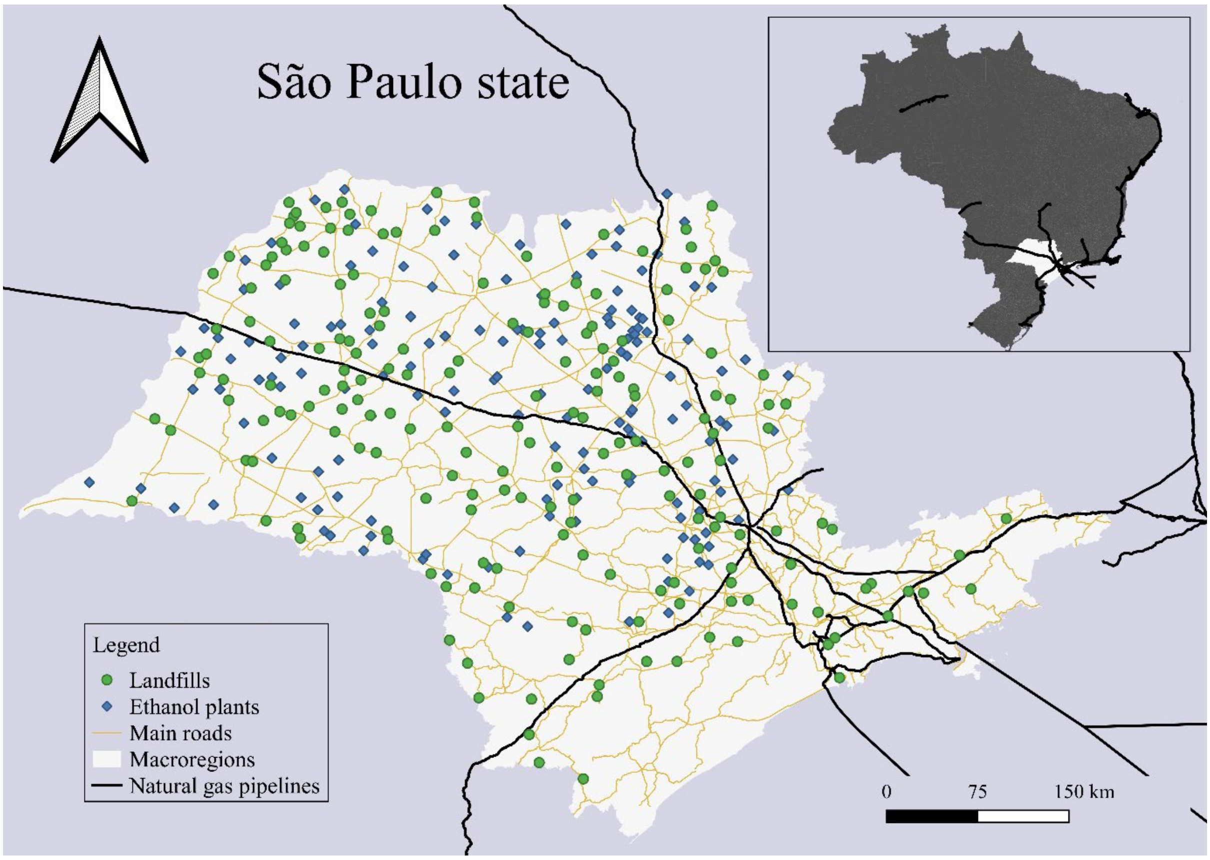

Figure 1. Overview of Sao Paulo State and its infrastructure corresponding to the biogas-chain in the freight transport system. **Reference:** Author.

## 2.2 Sugarcane mills, vinasse generation and use for biogas production

The cultivation of sugarcane (*Saccharum officinarum L.*) in Brazil dates to its colonial times, but it was with the oil crisis, in the 1970s, that ethanol produced from this grass became a strategic national product. Today, Brazil produces $3.57 \times 10^{7}$ m³ of ethanol (sugarcane season 2019/2020) consolidating as a global reference in production and use of biofuels [38].

Typically, the flowchart of a sugarcane mill has few variations, being divided mainly in the following steps: 1. Reception, preparation and grinding, 2. Sieve and broth treatment, 3. Sugar factory, 4. Ethanol distillery, 5. Utilities (power generation), 6. Disposal of effluents, 7. Product storage [14]. The mills operate 24 h per day, with an uptime of around 85%, during 167-218 days per year due to the sugarcane seasonality [17,20]. In terms of ethanol production, the literature indicates a range of 75-90 litters of ethanol per ton of cane milled in autonomous-type plant whilst, in annexed-type plant, ethanol production is considerably less (33-39 litters) given that part of the sugarcane-juice is used to produce sugar [39]. In any case, the SCV production, residual liquid stream generated in the distillery unit, ranges from 7 L to 18 L per litter of ethanol produced being 12 $L_{SCV}$. $L^{-1}_{ethanol}$ considered as an average value.

SCV has a high organic content (15-150 $gO_2.L^{-1}$) and considerable concentrations of potassium (1.2-10.0 g $K^+.L^{-1}$) and sulphate (0.76-9.50 $gSO_4^{2-}.L^{-1}$), acidic and corrosive characteristics (pH 3.5-5.0) [22]. The sum of these features makes SCV the main residual stream from sugarcane mills with potential for energy recovery.

Thermophilic AD-plant (Upflow Anaerobic Sludge Blanket reactor (UASB), 75 m³) has demonstrated capability of producing biogas from SGV since the 1980s at a high-rate (25 $kgCOD.m^{-3}.d^{-1}$) [16]. The methane yield reported is approximately of 0.23 N $m^{-3}_{CH4}$. $kg^{-1}_{CODremoved}$ with a pollutant load decrement of 60%, indicating that the AD of SCV is a reliable technology and fully available to the sugarcane mills. It should be recalled that thermophilic AD of SCV was considered, in this study, as operating condition (55 °C) due to its superior stability [15,40]. Furthermore, SCV leaves the distillation operation in the temperature range of 85–90 °C, thus, representing no additional costs to the process. The main input data used for characterising the sugarcane mill, vinasse generation and biogas yield are compiled in Table 1. The daily biogas production from SCV was estimated according to the Equation 1.

$$V_{SCV-Biogas} = Q_{SCV}.C_{COD}.\mathcal{E}_{COD}.Y_{Biogas} \qquad \text{Equation 1}$$

Where $Q_{SCV}$ is the flow of SCV in $m^3$ SCV. $d^{-1}$; $C_{COD}$is the initial concentration of Chemical Oxygen Demand (COD) in $kgO_2.m^{-3}_{SCV}$; $\mathcal{E}_{COD}$ is the COD removal in % and $Y_{Biogas}$ is the biogas yield in N-$m^3_{biogas}$. $kg^{-1}COD_{removed}$.

Table 1. Main input data for the sugarcane mills, vinasse generation and biogas/biomethane yield.

| **Input data (ratios, variables and parameters)** | **Value** | **Unit** | **(Range) or Remarks** | **References** |
|---|---|---|---|---|
| Plant operation | 210 | d | (167-218) | [17,20] |
| SCV generation | 12 | $m^3$. $m^{-3}$ ethanol | (7-18) | [15,16] |
| Temperature | 55 | °C | - | [15,16] |
| COD [a] | 35.20 | kg $O_2$. $m^{-3}$ | (15-150 $kgO_2.m^{-3}$) | [15,16] |
| Sulfate | 1.4 | kg $SO_4^{2-}$. $m^{-3}$ | - | [15,16] |
| Buffer | 12.50 | kg $NaHCO_3$. $m^{-3}_{SCV}$ | Reactor start-up (30 days) | [15,16] |
| UASB [b] - OLR [c] | 25 | kg COD. $m^{-3}$. $d^{-1}$ | (15 – 30) | [15,16] |
| HRT [d] | 1.40 | d | | [15,16] |
| COD removal | 60.70 | % | - | [15,16] |
| Biogas yield [e] | 0.38 | N $m^3_{biogas}$. $kg^{-1}_{CODremoved}$ | - | [15,16] |
| Biogas yield [e] | 83.5 | N $m^3_{CH4}$. $m^{-3}_{Ethanol}$ | - | Author |

a. Chemical Demand of Oxygen (COD). b. Upflow Anaerobic Sludge Blanket (UASB) reactor-type. c. Organic Loading Rate (OLR). d. Hydraulic Retention Time (HRT). e. The normal standard condition is adopted in order to facilitate the comparison between biogas flow. Hence, the volumetric flow and/or yield is presented in this “N” condition that means 1.013 bar pressure (1 atm), 0 (zero) degrees centigrade and 0% relative humidity.

## 2.3 Municipal Solid Waste, landfills, and biogas production.

Around 2 billion tonnes per year of MSW are generated globally, of which 34–53% is comprised of organic biodegradable waste also known as organic fraction of MSW (OFMSW) [41]. Landfilling consists of an inevitable final stage in waste management and is the most common method of MSW disposal around the world [28]. In Brazil, 79 million tons of MSW were produced in 2018. Of this, 92% (72.7 million) were collected and 58% (41.9 million) was transported to a suitable destination. Regular collection of MSW is routinely carried out throughout the state of Sao Paulo. Recent data (2018) from the Brazilian Institute of Geography and Statistics (IBGE) show that this service is practically universal and serves 98.8% of the state's households [42].

When MSW is disposed in landfills, most of the organic fraction will be degraded over a longer (> 100 years) or shorter period (< 1 year). Most of this process will occur via biological pathways depending on conditions in situ [41]. The main biodegradation products are carbon dioxide ($CO_2$), water and heat for the aerobic process (early stage – up to 2 years) and methane ($CH_4$) and $CO_2$ for the anaerobic process (stationary stage reached in mid-term). An interesting fact is the landfills useful life, that is about 15–20 years [29]. The daily biogas production from OFMSW was estimated according to the Tier 1 methodology recommended by the Intergovernmental Panel on Climate Change – IPCC (Equation 2) [41]. The method assumes that the full theoretical potential to produce biogas is exploited during the same year the waste is disposed.

$$V_{OFMSW-Biogas} = \left(\frac{(MSW_T \,.MSW_F \,.MCF.DOC.DOC_F.F.\frac{16}{12}R)\,.(1-OX)}{\rho_{CH4}\,.\,8760}\right).10^9 \qquad \text{Equation 2}$$

Where, $MSW_T$ is the total MSW generated in Gg. year$^{-1}$; $MSW_F$ is the fraction of MSW disposed to solid waste disposal sites; $MCF$ methane correction factor (fraction) (IPCC default value of 0.6); $DOC$ is the degradable organic carbon (fraction) in kg-C.kg$^{-1}$ SW; $DOC_F$ is the fraction DOC dissimilated (IPCC default value of 0.77); $F$ is fraction of CH4 in landfill gas (IPCC default value of 0.5); 16/12 is the conversion rate of carbon into methane [Dimensionless fraction]; $R$ is the amount of recovered methane in Gg$CH_4$.year$^{-1}$ (not applied - not emitted, burned in the flare); OX is the oxidation factor [Dimensionless fraction] (not applied - there is no formation of $CO_2$ before combustion of methane in landfill); $\rho_{CH4}$ is the density of methane (716 g. N $m^3$). 8760 is the value for the conversion from years to hours. Note: Gg is Gigagram (*i.e.,* 1 Gigagram or Gg is equivalent to $10^9$ grams).

## 2.4 Biogas upgrading units in sugarcane mills and landfills

Biogas comprises a mixture of gases that varies according to the substrate and the AD conditions. In general, the following composition is assumed on a dry-basis: 60%-70% of methane ($CH_4$) and 30%-40% of carbon dioxide ($CO_2$). However, other components such as hydrogen ($H_2$), ammonia ($NH_3$), hydrogen sulphide ($H_2S$), nitrogen ($N_2$) and oxygen ($O_2$) might also be detected. Resolution 685/2017 [43] of the Brazilian oil and gas agency establishes the quality standards of biomethane so that there is perfect interchangeability between biomethane and natural gas (Table 2).

Table 2. Biomethane quality standards and SCV and landfills biogas compositions.

| **Parameter** | **Units** [a] | **Biomethane (ANP 685/2017)** | **SCV Biogas** [b] | **Landfills Biogas** [c] |
|---|---|---|---|---|
| Higher Heating Value (HHV) | kJ.$m^{-3}$ | 35.0 - 43.0 | - | - |
| $CH_4$ | % mol | 90.0 | 55-65 | 45-60 |
| $O_2$ | % mol | 0.8 | ~ 0 | 0.1-1 |
| $CO_2$ | % mol | 3.0 | 35-45 | 40-60 |
| $CO_2 + O_2 + N_2$ | % mol | 10 | 35-45 | 40-60 |
| Total Sulfur [d] | mg.$m^{-3}$ | 70 | 9500 – 42,500 | 0-14,000 |
| $H_2S$ | mg.$m^{-3}$ | 10 | 10,000 – 45,000 | - |
| $H_2O$ (dew point at 1 atm) | °C | 39 | Saturated | Saturated |
| Others | % mol | - | Traces | Traces |

a. Composition in dry basis. b. According to Ferraz Junior et al. [20] Koyama et al. [22] and Leme et al. [23] b. According to the United States Environmental Protection Agency [44]. d. It is assumed that all Sulphur in biogas is in $H_2S$.

The process from “raw” biogas to liquefied biomethane has several major steps that can be solved in an integrated upgrading/liquefaction or in separate steps combining different technologies followed by transport and storage facilities. Briefly, $H_2S$ must be removed for its corrosive and toxic nature. In turn, the water vapor and $CO_2$ removal are required to prevent hydrate formation and, the deposition of solid that would block passages and soon make the process inoperative, respectively [45].

### 2.4.1 $H_2S$ removal

The volume of hydrogen sulphide ($H_2S$) produced during the anaerobic digestion of SCV depends on a number of factors and is hard to be predicted. However, it was assumed that all Sulphur (mostly sulphates) in the SCV was reduced to sulphide which ends up as $H_2S$. Based on that, each mol of $SO_4^{2-}$ (96 g) produced one mol of $H_2S$ (22.4 L). The volume of $H_2S$ was estimated at standard temperature and pressure using the Equation 3:

$$V_{H2S} = \frac{\frac{[SO_4^{2-}]}{64}.R.T}{P} \qquad \text{Equation 3}$$

Where, $V_{H2S}$ is the volume of $H_2S$ in $m^3$, $\frac{[SO_4^{2-}]}{64}$ is the number of mols given by the ratio of concentration of sulfate of SVC in $g.m^{-3}$ and its molecular weight in $g.mol^{-1}$, $R$ is the gas constant in $J.K^{-1}.mol^{-1}$, $T$ is the temperature in K and $P$ is the pressure in pascal. Given the $H_2S$ concentration in SCV (Table 2) and biogas production (Equation 1), the Sulphur load of the example project is 1.74-ton $SO_4^{2-}$. $d^{-1}$. Furthermore, the concentration of $H_2S$ in the biogas resulted in 4.7% in volume (dry basis) or 47000 ppmv based on Equation 3. Hence, the Sulphur removal technology would be recommended.

Biological desulfurization systems (BDS) are used on a large scale and operate with a limit of 20,000 ppmv of $H_2S$ at a flow of 322,000 $Nm^3.d^{-1}$ or arranged in series for higher concentrations [46]. Basically, BDS are placed in two stages: 1) continuous $H_2S$ absorption in sodium hydroxide solution (NaOH). 2) regeneration of the adsorbent solution in aerobic condition. In the latter condition, sulphide is converted into elemental Sulphur or sulphates. This arrangement tends to have affordable investment costs, associated with relatively lower operational costs [20].

$H_2S$ concentrations in landfill gas samples often range from under detection limits to thousands part per million [47]. The UK Environment Agency (2002) reviewed trace landfill gas data from 79 sites [48]. In the reviewed data, $H_2S$ concentration was reported as high as about

70,000 ppm. This magnitude of $H_2S$ concentration is not often observed in MSW landfill gases. The median concentration and average concentration of $H_2S$ was 2 ppmv and 960 ppmv, respectively. Due to these relative low concentrations and the following units process for biogas purification, a desulfurization system unit in the landfills were not considered.

### 2.4.2 $CO_2$ removal and dehydration

There are a number of different technologies that can remove $CO_2$ and water from biogas. In this study, water scrubber (WS) was chosen for being usually adopted [49–51]. WS method is used as a solvent to remove $CO_2$ and $H_2S$, considering the $CH_4$ solubility in water is much less than the target components, in this case, $CO_2$ mainly. Operating pressure of 8 bar(g) was chosen and a conventional electrical glycol chiller was selected to ensure that process water was cooled down to 6-7 °C. The water content in the biogas, downstream of $CO_2$ removal unit, was estimated assuming its water saturation at 35°C and 1 atm which means that the water content in biogas was 5.0%, regardless the feedstock (Table 2). In this context, dehydration is required and it can be accomplished using dehydrators [52]. From this point on, high purity of methane is addressed (90%-99%).

### 2.4.3 Compression, odorization, and liquefaction

For all biogas-upgrading units, some level of biogas compression was required. First biogas compression involved the use of centrifugal blowers to pull it from the biodigester through the BDS. The second compression was determined by the WS unit (8 bar(g)). For biomethane compression, an electrical reciprocating compressor was chosen. Odourisation was performed using conventional equipment that injects controlled amounts of odouriser (e.g., mercaptans) into the gaseous stream based on its flow rate. The mixed refrigerant cycle (MRC) was chosen for the liquefaction unit due to its simpler process, less equipment and investment; and easy operation [45]. MRC process uses a mixture of C1 to C5 hydrocarbons and nitrogen as refrigerant (up to -150 °C) at a liquefaction rate of 0.88 [45]. Table 3 lists the process parameters for biogas purification including, biomethane compression and its liquefaction.

Table 3. Process parameters for biogas purification and biomethane compression.

| Input data (ratios, variables and parameters) | Technology | Value | Unit | (Range) or Remarks | References |
|---|---|---|---|---|---|
| $H_2S$ removal | Biological | 10000 | ppmv | 0.44 kg-NaOH. $Kg^{-1}S_{removed}$ | [46] |
| $CO_2$ removal | Water scrubber | Up to 2000 | $Nm^3.h^{-1}$ | 129 kg-$H_2O$.$ton^{-1}CO_{2removed}$ | [53] |
| Gas dehydration | Chemical | Up to 1400 | $Nm^3.h^{-1}$ | Desiccant | [52] |

| | | | | | |
|---|---|---|---|---|---|
| Compressor | - | Up to 3000 | $Nm^3.h^{-1}$ | (6 - 250 bar) | [54] |
| Liquefaction | Mixed refrigerant cycle | Up to 833 | $Nm^3.h^{-1}$ | - | [45] |

## 2.5 LBM in the freight transport system: transport sector, gas grid and scenarios

The spatial distribution of transport logistics in the Brazilian territory has a predominance of highways, concentrated mainly in the Centre-South of the country, especially in the state of Sao Paulo. It counts with nearly 200,000 km of roads and highways that are distributed as federal (1,055 km), state (15,402 km) and municipal (175,821 km) highways, while other 6,716 km of highways are in charge of concessionaires (Department of Highways of Sao Paulo – DER-SP [55]). Complementarily, the National fleet has 110 million vehicles, being 28.5% of this value registered in the state of Sao Paulo and more than 4.6 million vehicles classified as heavy-duty vehicles (i.e., able to be fuelled with liquefied biomethane) (National Traffic Department – [56]). The gas pipelines in the Sao Paulo state are depicted in Supplementary Table 1.

Supplementary Table 1. Gas grid classified as transport pipelines in the Sao Paulo state.

| **Name** | **Origin** | **Destiny** | **Diameter (pole)** | **Length (km)** | **Maximum operation pressure (bar)[a]** |
|---|---|---|---|---|---|
| GASBOL (North stretch) | Corumbá (MS) | Guararema (SP) | 32-24 | 1,417.00 | 100 |
| GASBOL (South stretch) | Paulínia (SP) | Canoas (RS) | 24-16 | 1,176.00 | 100/75 |
| Campinas - Rio (GASCAR) | Paulínia (SP) | Japeri (RJ) | 28 | 450.00 | 100 |
| GASPAL (ESVOL/MAUÁ) | Volta Redonda (RJ) | Mauá (SP) | 22 | 325.70 | 65/51 |
| Caraguatatuba - Taubaté | Caraguatatuba (SP) | Taubaté (SP) | 28 | 98.00 | 100 |
| Paulínia - Jacutinga | Paulínia (SP) | Jacutinga (MG) | 14 | 93.00 | 100 |
| GASAN (RPBC/RECAP) | Cubatão (SP) | Mauá (SP) | 12 | 37.00 | 51 |
| GASAN II - 22" | Sao Bernardo do Campo (SP) | Sao Paulo (SP) | 22 | 38.00 | 74 |

*a.* Gas pipelines operating license issued by ANP [33].

Based on this information (*subheads 2.1-2.5*), two possible scenarios for using LBM in the freight transport system of Sao Paulo state were determined (Figure 2). The first one, called Non-Restricted Scenario (NRS), encompasses the entire state of Sao Paulo regardless of the gas grid, while the second scenario, called Restricted Scenario (RS) includes only the geographic regions served by gas pipelines.

1. The NRS considers that biogas produced from SCV and MSW will be compressed and transported by truck to gas liquefaction plants located on the Sao Paulo’s state highways. The gas is liquefied and further transported by truck/trailers to supply a number of refuelling stations (Figure 2A and B).
2. The RS considers that biogas produced from SCV and MSW is compressed and transported by truck to injection points located in intersections between gas pipelines and Sao Paulo’s state highways. Here, the biogas is injected into the pipeline and mixed with natural gas. A number of refuelling stations are then installed in intersections between Sao Paulo’s highways and gas pipelines, so that a volume of the biogas/natural gas mixture is extracted and liquefied, serving the refuelling stations (Figure 2A and C).

The assumptions, constraints, and the methods used to address both scenarios are explained in the supplementary data.

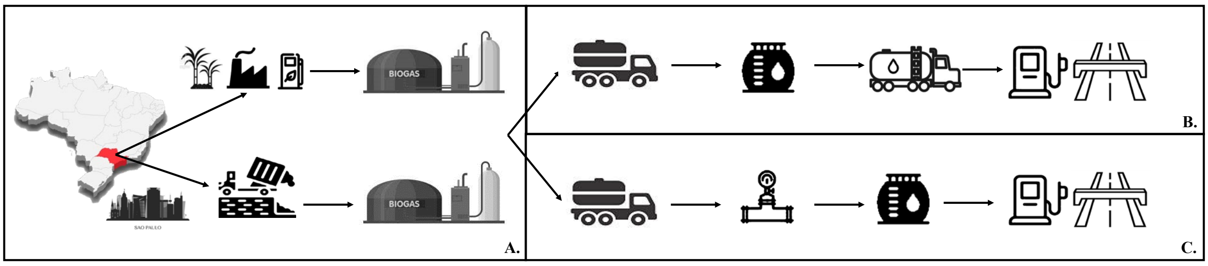

Figure 2. A. Sugarcane vinasse and municipal solid waste as feedstock for biogas production. B. Schematic representation of non-restricted scenario (NRS). C Schematic representation of restricted scenario (RS).

## 2.6 Cost Analysis

This study analyses the minimum selling price of LBM for each refuelling station, liquefaction plant, and corresponding biogas plants, in each scenario. The costs include capital costs and operational costs for biogas production and purification, distribution, liquefaction, and refuelling. Overall, these investment and operating costs, raw materials, chemicals and electricity costs were based on a conversion rate of USD 0.17 per Brazilian Real (April 2021).

The cost of LBM per energy unit was calculated using the minimum selling price method. In the NRS, the biogas is transported to local liquefaction plants. In the liquefaction plants, LBM is transported by trucks to the refuelling stations. In NRS, the total cost is spread among the refuelling stations based on their volume shares of total output. Equation 4 through 7 show the calculations for scenario 1:

$$TCB_k = \sum_i \left(O_{Bio_{i,k}} + O_{Dist_{i,k}}\right) + \sum_i \left(C_{Bio_{i,k}} + C_{Dist_{i,k}}\right) \cdot \frac{r \cdot (1+r)^n}{(1+r)^n - 1}$$

Equation 4

$$TCL_{k,s} \ = TCB_k + O_{Liq_k} + O_{Tran_{k,s}} + C_{Liq_k} + C_{Tran_{k,s}} \cdot \frac{r \cdot (1+r)^n}{(1+r)^n - 1}$$

Equation 5

$$TCR_s = \sum_k \ TCL_{k,s} + O_{Ref_s} + NG_p \cdot NGQ + C_{Ref_s} \cdot \frac{r \cdot (1+r)^n}{(1+r)^n - 1}$$

Equation 6

$$LBM_{p,s} = \frac{TCB_k + TCL_k + TCR_s}{Sup_{y,s}}$$

Equation 7

Where, $TCB_k$ is the total annual cost of biogas plants delivering to liquefaction plant *k*, $TCL_{k,s}$ is the total annual cost of liquefaction plant *k*, $TCR_s$ is the total cost of refuelling in refuelling station *s*, $LBM_{p,s}$ is the price in USD/GJ of liquefied biogas in refuelling station *s*, $O_{Bio_{i,k}}$ and $O_{Dist_{i,k}}$ are the annual operational costs of biomethane production in biogas plant *i* and biomethane transportation from biogas plant *i* to liquefaction plant *k*, $O_{Liq_k}$ is the annual operational cost of liquefaction, $O_{Tran_{k,s}}$ is the annual operational cost of transporting LBM from liquefaction plant *k* to refuelling station *s*, $O_{Ref_s}$ is the annual operational cost of refuelling station *s*, $NGQ$ is the quantity of natural gas used to maintain the same supply all year round, $NG_p$ ·is the price for natural gas molecule, $C_{Bio_{i,k}}$ is the capital cost of biogas plant *i*, $C_{Dist_{i,k}}$ is the distribution capital cost from biogas plant *i* to liquefaction plant *k*, $C_{\mathrm{Liq}_k}$ and $C_{Tran_{k,s}}$ are the total capital cost of liquefaction plant *k* and LBM transport from liquefaction plant *k* to refuelling station *s*, respectively, $C_{Ref_s}$ is the total capital cost of refueling station *s*, *r* is the expected rate of return of investments, *n* is the duration of the project, and, $Sup_{y,s}$ is the total LBM supply (in GJ) in year *y* in refuelling station *s*.

In RS, biogas is transported to the injection sites of the natural gas pipelines. The liquefaction in RS is done on-site in the refuelling stations. The costs of biogas plants and of each injection point are attributed to the closest refuelling station. In the same way as the NRS, RS includes natural gas from the grid to maintain a constant supply throughout the year. Equation 8 shows the calculations of total annual cost for biogas plants, and Equation 9 shows the cost for injection. Finally, the minimum selling price for LBM is calculated in Equation 10.

$$TCB_j \ = \sum_i (O_{Bio_{i,j}} + O_{Dist_{i,j}}) + \ \sum_i \left( C_{Bio_{i,j}} + C_{Dist_{i,j}} \right) \cdot \frac{r \cdot (1+r)^n}{(1+r)^n - 1}$$

Equation 8

$$TCI_{j,s} = TCB_j + O_{Inj_j} + C_{Inj_j} \cdot \frac{r\cdot(1+r)^n}{(1+r)^n-1} \qquad \text{Equation 9}$$

$$LBM_{p,s} = \frac{\sum_j TCI_{j,s}+O_{Liq_s}+O_{Ref_s}+NG_p\cdot NGQ+(C_{Liq_s}+C_{Ref_s})\cdot\frac{r\cdot(1+r)^n}{(1+r)^n-1}}{Sup_{y,s}} \qquad \text{Equation 10}$$

Where, TCIj,s is the total annual cost of injection attributed to refuelling station, $NG_p$ is the price for the natural gas molecule, NGQ is the quantity of natural gas used to maintain supply all year round, $O_{Bio_{i,j}}$ and $O_{Dist_{i,j}}$ are the annual operational costs of biomethane production in biogas plant *i* and biomethane transportation from biogas plant *i* to injection site $j$, $O_{BInj_{j,s}}$ is the annual operational costs of injection site $j$, $LBM_{p,s}$ is the price in USD/GJ of liquefied biogas in refuelling station $s$, $O_{Liq_s}$ and $O_{Ref_s}$ are the annual operational costs of liquefaction and refuelling in refuelling station $s$, $C_{Bio_{i,a}}$ is the capital cost of biogas plant *I*, $C_{Dist_{i,j}}$ is the distribution capital cost from biogas plant *i* to injection site $j$, $C_{BInj_j}$ is the total capital cost of injection site $j$,$C_{\mathrm{Liq}_s}$ and $C_{R\mathrm{ef}_s}$ are the total capital cost of liquefaction and refuelling in station $s$, *r* is the expected rate of return of investments, *n* is the duration of the project, $Sup_{y,s}$ is the total supply of LBM plus natural gas in year *y* in refuelling station *s* in GJ.

### 2.6.1 AD plant costs (Biogas production and purification)

The capital cost of the AD plants in the sugarcane mills was estimated based on the construction of equalisation tanks, UASB-type reactor and the acquisition of equipment, for instance, pumps, water seal and gas meters (Table 4). The number of reactors was determined considering the processing of the total volume of SCV generated daily in the mills, and the reactor operating conditions listed in Table 1. Biogas capture infrastructure in landfills was assumed to be pre-existing. On the other hand, the units for upgrading biogas and compressing biomethane were computed for both sugarcane mills and landfills. The latter did not count with the $H_2O$ removal system due to the composition of biogas previously described in *subhead 2.4.1.* Installation costs (i.e., mechanical, hydraulic, electric and automation systems) were set to 20% of the investments with civil engineering works and equipment. The operating costs included personnel (operations, maintenance, and administration), the maintenance of the AD-plants itself and annual chemical and electricity costs (Table 5).

Table 4. Capital costs of an AD plants: biogas production, and its upgrading and biomethane compression.

| Civil work/equipment | Cost (USD) | (Range) Remarks | References |
|---|---|---|---|
| Equalisation tank [a] | 404,729.50 | (USD 350 per $m^3_{constructed}$)<br>Reinforced Concrete | [57] |
| Online pH | 3,900.00 | - | [57] |
| Centrifugal pump | 4,500.00 | 64.00 $m^3.h^{-1}$ | [57] |
| UASB reactor [b] | 686,000.00 | (USD 350 per $m^3_{constructed}$)<br>Reinforced Concrete | [57] |
| Water seal | 2,080.00 | Stainless steel | [57] |
| Gas meter | 5,200.00 | - | [57] |
| $H_2S$ removal | 20,797.40 | - | [46] |
| $CO_2$ removal | 4,700,000.00 | - | [53] |
| Gas dehydration | 31,299 | - | [52] |
| Compressor | 535,649.97 | - | [54] |

a. Dimensions (W x L x H (m)) of 14.5x14.5x5.5 resulting in a total and working volume of 1156.37 $m^3$ and 1051.25 $m^3$, respectively. b. Dimensions (W x L x H (m)) of 14.0x20.0x7.0 resulting in a Total and working volume of 1960 $m^3$ and 1747.2 $m^3$, respectively.

Table 5. Operating cost of an AD plant: biogas production, and its upgrading and biomethane compression.

| Operating cost | Value | Unit/Cost | (Range) Remarks | References |
|---|---|---|---|---|
| Personnel – operations | 14,918.85 | $USD.(employee.year)^{-1}$ | 3 workers per day | [5] |
| Personnel - supervision | 24,470.11 | $USD.(employee.year)^{-1}$ | 3 workers per day | [5] |
| Electricity [a] | 0.352 | $kWh.m^{-3}$ SCV | UASB reactor and $H_2S$ removal unit | [18] |
| Bicarbonate | 0.49 | $USD.kg^{-1}$ | Start-up of UASB reactor | [8] |
| NaOH | 0.61 | $USD.m^{-3}$ | Adsorbent - $H_2S$ removal | [8] |
| Electricity [a] | 400 | kWh | $CO_2$ removal | [53] |
| Water [b] | 129 | kg $H_2O.ton^{-1}_{CO2\ rem.}$ | $CO_2$ removal unit – Water of process | [58] |
| Electricity [a] | 19.6 | kWh | Gas dehydration unit | [52] |
| Electricity [a] | 350 | kWh | Compressor | [54] |

a. The electricity cost was 0.09163 $USD.kWh^{-1}$ according to [59] b. The consumption of water was not accounted, considering the use of industrial water.

### 2.6.2 Liquefaction

Liquefaction occurs at different stages in each scenario. While NRS liquefaction occurs after biomethane production to later be transported to the refuelling station, liquefaction in RS occurs after biomethane is transported via pipelines, mixing with natural gas.

For both scenarios, capital costs for the liquefaction stage was based on [5] and [60] for scales between 0.05 and 1 mtpa, with a cost 1036.56USD (2020) per ton of liquefied biomethane. In the case of liquefaction with scale outside this range, the capital cost was adjusted using Equation 11 [61]:

$$C_2 = C_1 \cdot \left(\frac{S_1}{S_2}\right)^n \qquad \text{Equation 11}$$

Where, the capital cost $C_2$, associated with scale $S_2$, is calculated using a known cost $C_1$, associated with scale $S_1$. An average exponent *n* of 0.6 was used [62].

The liquefaction stage encompasses a gas treatment unit, a liquefaction train, one LNG storage tank of 150,000 $m^3$ (single containment), and LNG lorry loading facilities [63]. The operational costs of liquefaction include personnel (operations, maintenance and administration), electricity consumption, general maintenance, insurance and the consumption of refrigerants, and data is shown in Table 6.

Table 6. Operating costs of a liquefaction plant.

| **Item** | **Value** | **Unit/Cost** | **(Range) Remarks** | **Reference** |
|---|---|---|---|---|
| Operating personnel | 27,631 | USD. (employee. year)$^{-1}$ | 3 workers per day | [64] |
| Maintenance personnel | 25,542 | USD. (employee. year)$^{-1}$ | | [64] |
| Administration personnel | 41,893 | USD. (employee. year)$^{-1}$ | 6 workers per day | [64] |
| Consumables - Refrigerants | 1,530.87 | Tonne. Tonne per annum$^{-1}$ | Ethane | [5] |
| Consumables - Refrigerants | 677.96 | Tonne. Tonne per annum$^{-1}$ | Propane | [5] |
| Electricity consumption | -0.0331*D+0.9984 | kWh.kg$^{-1}$ | 1-15 MMSCFD $^{a}$ | [65] |
| Electricity consumption | 0.7 | kWh.kg$^{-1}$ | > 15 MMSCFD | [65] |
| Electricity price | 0.09163 | USD.kWh$^{-1}$ | | [59] |
| Maintenance | 2 | % of capital. year $^{-1}$ | | [5] |
| General Administration | 20 | % from (Personnel + Maintenance). year $^{-1}$ | | [5] |

|  |  |  |  |
|---|---|---|---|
| Insurance and Duties | 0.75 | % of capital. year $^{-1}$ | [5] |

[a] Total electricity consumption is a function of scale, with D being the processed natural gas in MMSCFD (Million standard cubic feet per day).

### 2.6.3 Transportation costs

There are three basic components of transportation in this study: in the NRS scenario, the transportation of biomethane from biogas plants to liquefaction plants and the transportation of LBM from liquefaction plants to refuelling stations. In the RS, the transportation of biogas to injection points, which inject biomethane into the gas pipeline to be transported to the refuelling stations.

The number of trucks necessary for each route was calculated based on the distance and an average speed of 40 km/h. The number of hours travelled was divided by 24, which yields the number of trucks necessary to make all daily trips. Similarly, the number of drivers was calculated based on the total hours travelled divided by 6, the maximum number of hours travelled per driver. The values for capital and operational costs in the transportation and distribution of biogas and LBM are shown in Table 7 and Table 8, respectively.

Table 7. Capital costs of transportation

| Item | Value | Unit/cost | (Range) Remarks | Reference |
|---|---|---|---|---|
| **Biomethane transport (NRS and RS)** |  |  |  |  |
| Pressurised tank | 31,338 | USD.tank$^{-1}$ | 45m³ | Online research |
| Truck | 30,000 | USD.truck$^{-1}$ | 6 axles | [5] |
| **LBM transport (NRS)** |  |  |  |  |
| Cryogenic Tank | 175,749 | USD.tank$^{-1}$ | 30m$^3$ | [5] |
| Truck | 30,000 | USD.truck$^{-1}$ | 6 axles | [5] |
| **Biomethane injection (RS)** |  |  |  |  |
| Grid injection capital costs | 0.02 | USD.m$^{-3}$CH$_4$ |  | [66] |

Table 8. Operational costs of transportation

| Item | Value | Unit/Cost | (Range) Remarks | Reference |
|---|---|---|---|---|
| **Biomethane transport (NRS and RS)** |  |  |  |  |
| Fuel cost | 0.91 | USD.liter$^{-1}$ | Lorry yield: 2.2 km/litter | [5] |
| Tires | Variable |  | 11% from the total logistic cost | [5] |

| | | | | |
|---|---|---|---|---|
| Maintenance | Variable | | 14% from the total logistic cost | [5] |
| Labour | 13,724 | USD.(driver.year)$^{-1}$ | The number of drivers was estimated based on a maximum of 6 hours per day per driver | [5] |
| | | **Biomethane injection (NRS)** | | |
| Grid injection operational costs | 0.08 | USD.m$^{-3}$CH$_4$ | | [66] |
| Gas transport tariff | 1.316 | USD.MMBTU$^{-1}$ | Regulated tariff charged for transport infrastructure use | [67] |

### 2.6.4 Refuelling infrastructure costs

The capital costs of the refuelling infrastructure include civil and electrical work, LBM storage for one day based on supply of biomethane, LBM pumping, and dispensers. This analysis was estimated based on Mariani [68] . Each refuelling was estimated to last an average of 5 minutes and the estimation of the number of dispensers was based on this time for the functional flow of daily supply. Table 9. shows the input data for the estimation of the capital costs of the refuelling infrastructure.

Table 9. Capital costs of refuelling stations

| Item | Value | Unit | (Range) Remarks | Reference |
|---|---|---|---|---|
| Engineering | 4,210.23 | USD.t$^{-1}$ | | [68] |
| Electrics | 11,770.91 | USD.t$^{-1}$ | | |
| Auxiliary facilities | 2,925.00 | USD.t$^{-1}$ | | |
| Civil work | 7,941.26 | USD.t$^{-1}$ | | |
| LNG Storage (m$^3$) | | | | |
| 20 | 111,150.00 | USD.tank$^{-1}$ | Tank size and quantity were estimated based on a one-day storage | |
| 30 | 134,550.00 | USD.tank$^{-1}$ | | |
| 60 | 157,950.00 | USD.tank$^{-1}$ | | |
| 500 | 1,008,000.00 | USD.tank$^{-1}$ | | |
| LBM conditioner | 941.76 | USD.t$^{-1}$ | | |
| LBM dispenser [a] | 56,912.31 | USD.dispenser$^{-1}$ | | |

[a] Includes flow meter, pocket nozzle heating, automated nozzle cleaning system and return hose

The operational cost for the refuelling infrastructure is based on the information in Table 10. It includes personnel, (attendants and administration), certification and testing, general expenses, and electricity for LBM pumping.

Table 10. Operational costs for refuelling stations

| Item | Value | Unit | (Range) Remarks | Source |
|---|---|---|---|---|
| Personnel - attendant | 10,900.7 | USD.(employee.year)$^{-1}$ | 1 employee for every 3 dispensers | [64] |
| Personnel – administration | 40,640 | USD.(employee.year)$^{-1}$ | 6 per day | [64] |
| Maintenance | variable | % of capital | 1% of capital | [68] |
| Technical cost | 1.05 | USD.kg$^{-1}$ | certify, testing | [68] |
| General | 0.47 | USD.kg$^{-1}$ | phone, cleaning, illumination | [68] |
| LBM pumping | 0.01 | kWh.kg$^{-1}$ | | [68] |
| Natural gas molecule | 0.4267 | USD.m$^{-3}$ | Regulated natural gas for use in transport | [67] |

## 3 RESULTS AND DISCUSSION

Two scenarios based on the use of liquefied biomethane (LBM) were designed for replacing diesel in the freight sector of Sao Paulo state. Sugarcane vinasse (SVC) and Municipal Solid Waste (MSW) were set as feedstock. The first scenario, non-restricted scenario (NRS), covers the entire territory of Sao Paulo state, regardless of the gas grid availability. The second scenario, restricted scenario (RS), includes only the areas where gas pipelines are available. In this section, the results for each scenario jointly with their economic analysis are presented.

### 3.1 Biogas potential

In the NRS, 154 sugarcane mills and 200 landfills were computed while, in the RS, a set of 42 sugarcane mills and 57 landfills were observed. Sugarcane mills were classified according to their processing capacity (Table 11). The plants are evenly distributed among the ranges and more than 80% of them presented processing capacities up to 30 thousand tons of sugarcane per day for both scenarios. With regards to the landfills, all the MSW plants were classified as landfill plants indicating the absence of hazardous waste and the presence of infrastructure to drain the biogas generated [69]. Based on Equations 1 and 2, the biogas potential for the state of Sao Paulo was calculated for each sugarcane mill and landfill considered, respectively. The potential for biogas production from SCV was estimated at 14.7 million Nm$^3$ per day in NRS, while in the RS, the correspondent production was four times lower (3.6 million Nm$^3$ per day), which is coherent to the dimension of the given scenario. On the other hand, the potential for producing biogas from landfills was estimated at 4.8 and 4.2 million Nm$^3$ per day for NRS and RS, respectively. The small difference between these values is explained by the gas pipelines availability covering 12 metropolitan areas of Sao Paulo state, that means 86% of the state's population (~ 34.9 million inhabitants).

Table 11. Capacity of processing sugarcane and municipal solid waste in this study.

| **Sugarcane mill (ton. d$^{-1}$)** | **Scenarios (%)** | | **Landfill (ton. year$^{-1}$)** | **Scenarios (%)** | |
|---|---|---|---|---|---|
| - | NRS | RS | - | NRS | RS |
| Up to 9,999 | 27.3 | 38.1 | Up to 999 | 22.4 | 6.9 |
| 10,000 – 19,999 | 35.1 | 33.3 | 1,000 – 9,999 | 46.8 | 39.7 |
| 20,000 – 29,999 | 16.9 | 9.5 | 10,000 – 24,999 | 11.4 | 6.9 |
| 30,000 – 39,999 | 13.6 | 9.5 | 25,000 – 49,999 | 6.5 | 13.8 |
| > 40,000 | 7.1 | 9.5 | > 50,000 | 12.9 | 32.8 |

As mentioned in Section 2, each AD-plant is connected by truck to a liquefaction plant in the NRS, and to an injection point to feed the natural gas pipelines in the RS. Refuelling stations are optimally placed on top of gas pipelines to allow for natural gas consumption on the days sugarcane mills are inoperative. A total of 7 refuelling stations were designed to allow for the maximum coverage of trucks using liquefied gas. Figure 3A-B show the resulting infrastructure for the NRS and RS, respectively.

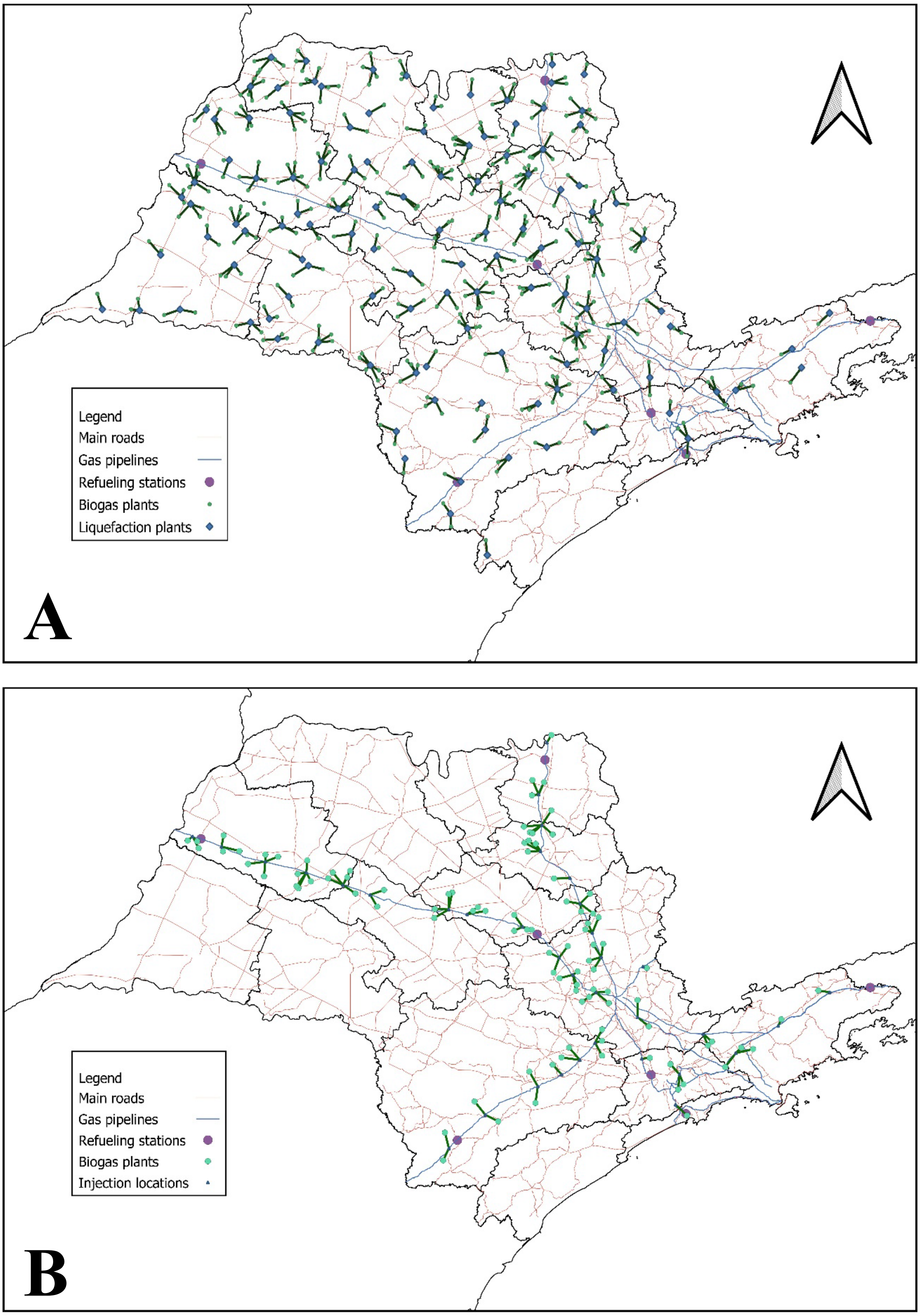

Figure 3. A. AD-plants, liquefaction plants and refuelling stations in the NRS and, B. AD-plants, Injection points and refuelling stations in the RS.

Given the difference of the potential of biogas/biomethane production between NRS and RS, a surplus of natural gas is necessary in order to obtain the relative costs of LBM costs. More details will be presented (*subhead 3.4.*).

### 3.2 Investment costs of AD-plants (Biogas production and purification)

To evaluate the cost of AD plants, including biogas purification units, an economic analysis was performed to both scenarios. Within the data used to obtain the minimum price of biogas, the capital costs were approximately USD 480 million per year in NRS, and USD 165 million per year in RS. These costs refer to the decrease in the value of the equipment, considering the interest rate of 0.08 and the project lifetime of 30 years. In turn, operating costs represented 1.7 times over the respective values of capital costs. These high values are associated with the study coverage area and the complexity of operations at the AD plants (production and purification of biogas), each has its own set of needs to function properly. Figure 4 shows the aggregated costs of the biogas plants in each scenario.

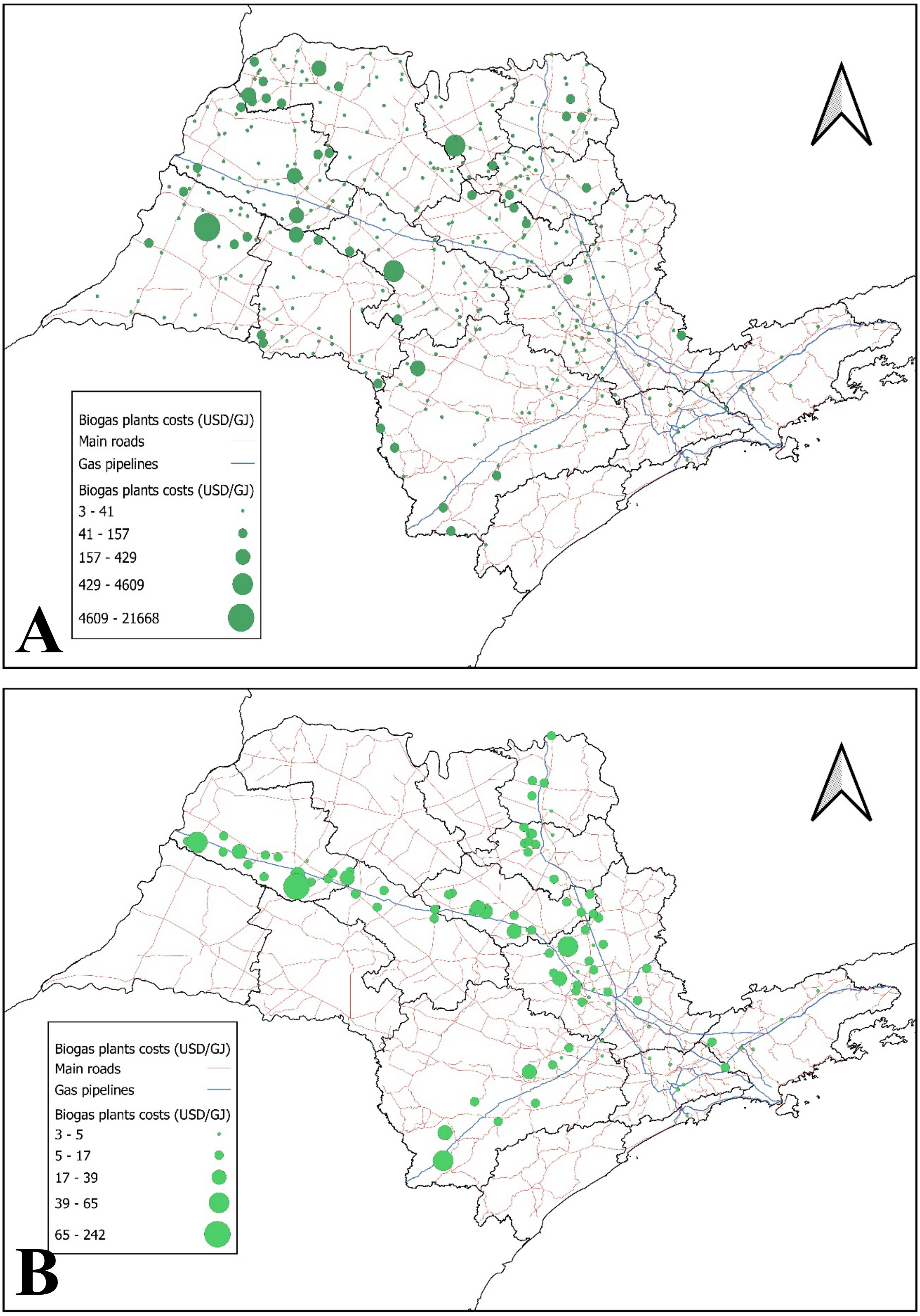

Figure 4. A. AD-plants costs (USD/GJ) in the NRS and B. AD-plants costs (USD/GJ) in the RS.

To break the costs down further, Figure 5A-B depicts the approximate expenditure of AD plants per scenario and feedstock-based plant. Interestingly, the units for $CO_2$ removal had the greatest influence on capital costs (~60%) in both scenarios, indicating the need for a technology of greater economic attractiveness in the biogas purification facilities. Construction and installation costs with mechanical, hydraulic, electrical, and automation systems of UASB reactor and equalisation tank represented 21%-30% and 6%-9% of capital cost, respectively, in both scenarios. Finally, the compression units represented approximately 6% of the total capital cost. The same behaviour (distribution of capital costs) is observed when the plants are evaluated based on the feedstock. However, the $CO_2$ removal units reached more than 80% of total costs in landfills plants since the infrastructure to drain the biogas generated is pre-existing (*subhead 3.1*). As also shown in Figure 5C-D, alkalising agent (chemical – UASB) represented 50%-65% of the annual operating costs in both scenarios. Phase separation could be considered as a strategy to reduce the amount of alkalising used, as demonstrated in Ferraz et al. [15]. However, it would require additional tank and equipment increasing the capital costs. The feasibility of Sodium bicarbonate and soda in the anaerobic digestion of SCV is discussed in Fuess et al. [57]. Alternatively, alkalising agents could be recovered from the ashes generated at the mill's bagasse cogeneration system as described in Ferraz et al.[8]. Next, electricity costs in the $CO_2$ removal and compression units represented 20%-30% and 7%-10%, respectively, followed by the expenditure on personnel (5%) which includes supervisor and operating staffs. As regards the expenditure with electricity on the AD-plants, there is probably little that can be done as far as technological improvements are concerned. Furthermore, electricity expenses would not represent a direct cost for most of modern sugarcane mills, considering that the surplus capacity would come from the mill's bagasse cogeneration system. As for personnel expenditure, it can be reasonably assumed as an opportunity to generate wealth, jobs, and income.

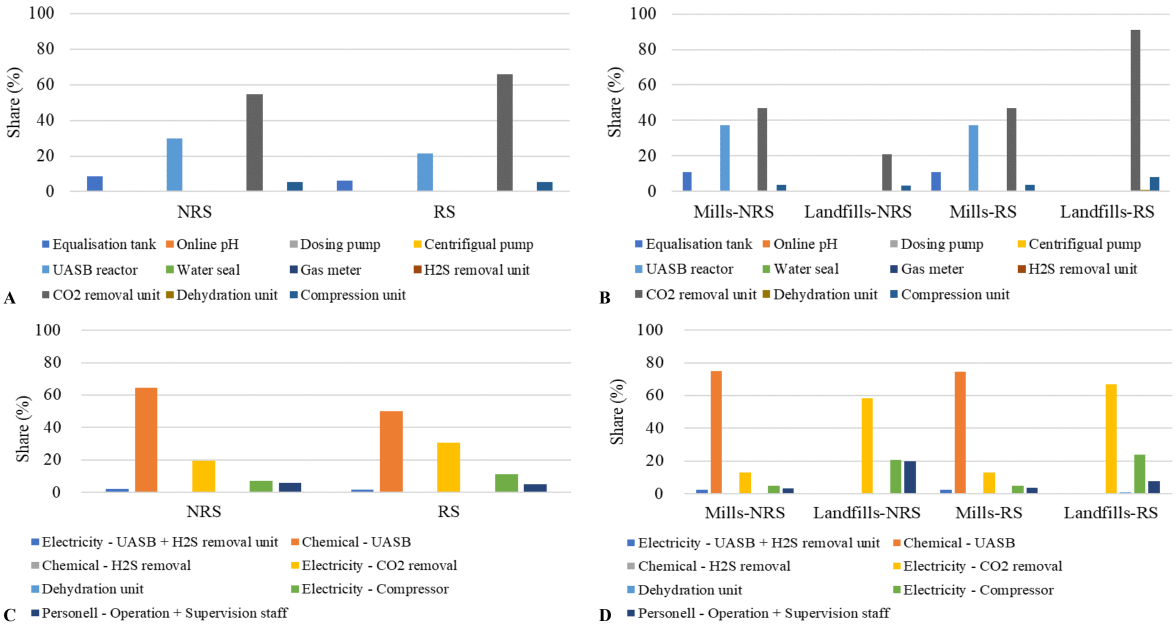

Figure 5. Cost distribution of AD-plants, including the units for biogas purification. A. Capital costs of NRS and RS. B. Capital cost of sugarcane mills and landfills of the NRS and RS. C. Operating cost of NRS and RS. D. Operating cost of sugarcane mills and landfills of the NRS and RS.

As previously mentioned, in the NRS, biomethane is transported to local liquefaction plants whilst, in the RS, biomethane is transported to pipelines to be injected. The capital costs of compressed biomethane transporting from AD-plants to liquefaction station is nearly USD 21.7 million per year while transporting it to the injection points of gas grid would cost USD 6.1 million per year. This high difference is mostly related to the amount of biomethane produced in each scenario. The compression level here assumed (250 bar(g)) represents one of the highest available in the market and it is the optimum of the current compressor's technology [54]. However, the transport of compressed biomethane can be further optimised by increasing the number of axles and carts which will reduce expenditure on the number of trucks, personnel and fuel. The latter is beyond the scope of this paper (Figure 6A and B).

### 3.3 Liquefaction of biomethane and its transport (NRS), and injection into the gas grid (RS)

Regarding the optimisation for locating the liquefaction plants and refuelling stations, 120 liquefaction plants in the NRS were found, along with 35 injection points in RS, within a radius of 20 km from the sugarcane mills/landfills. The capital cost of the liquefaction units was estimated in approximately USD 374 million per year in the NRS. From this value, 10% corresponds to the LBM transport to the refuelling stations. Regarding the RS, USD 200 million per year would be necessary to do the same investment (i.e., capital costs) , excluding capital expenditure of transport (Figure 6C and D). The annual operating costs was equivalent to the annual capital costs in the NRS, and 25% higher than the RS, based on its ratio equal to 1.00. Expenditure on electricity and personnel in the liquefaction units of both scenarios represented the greatest share, summing 65%-91% and 3%-18%, respectively, of the total operating costs. These data suggest the need of innovative electricity-saving technologies, but also perspectives of opportunities for professionals in sustainability and economic development throughout the biogas chain: production, transport and final use. Conversely, when the capital costs of injection are included in the RS, the value increases to USD 467 million per year. The total operating costs/total capital costs ratio were 3.0 and 0.8 in the NRS and RS, respectively. Personnel (40%), fuel (32%) and tires (17%) represented the main expenditure in the NRS, while the gas injection operation and its transport tariff represented more than 90% within operating costs share in the RS. Expenditures with biomethane injection into the gad grid and its transport tariff could be reduced via Feed-in tariffs which is a mechanism used as public policies aimed at accelerating investment in renewable energy technologies, widely adhered by countries such as Australia [70], Austria [6], Canada [71], Denmark [3] and United kingdom [72]. Figure 7 shows the aggregated costs of the intermediary steps of biomethane production in each scenario.

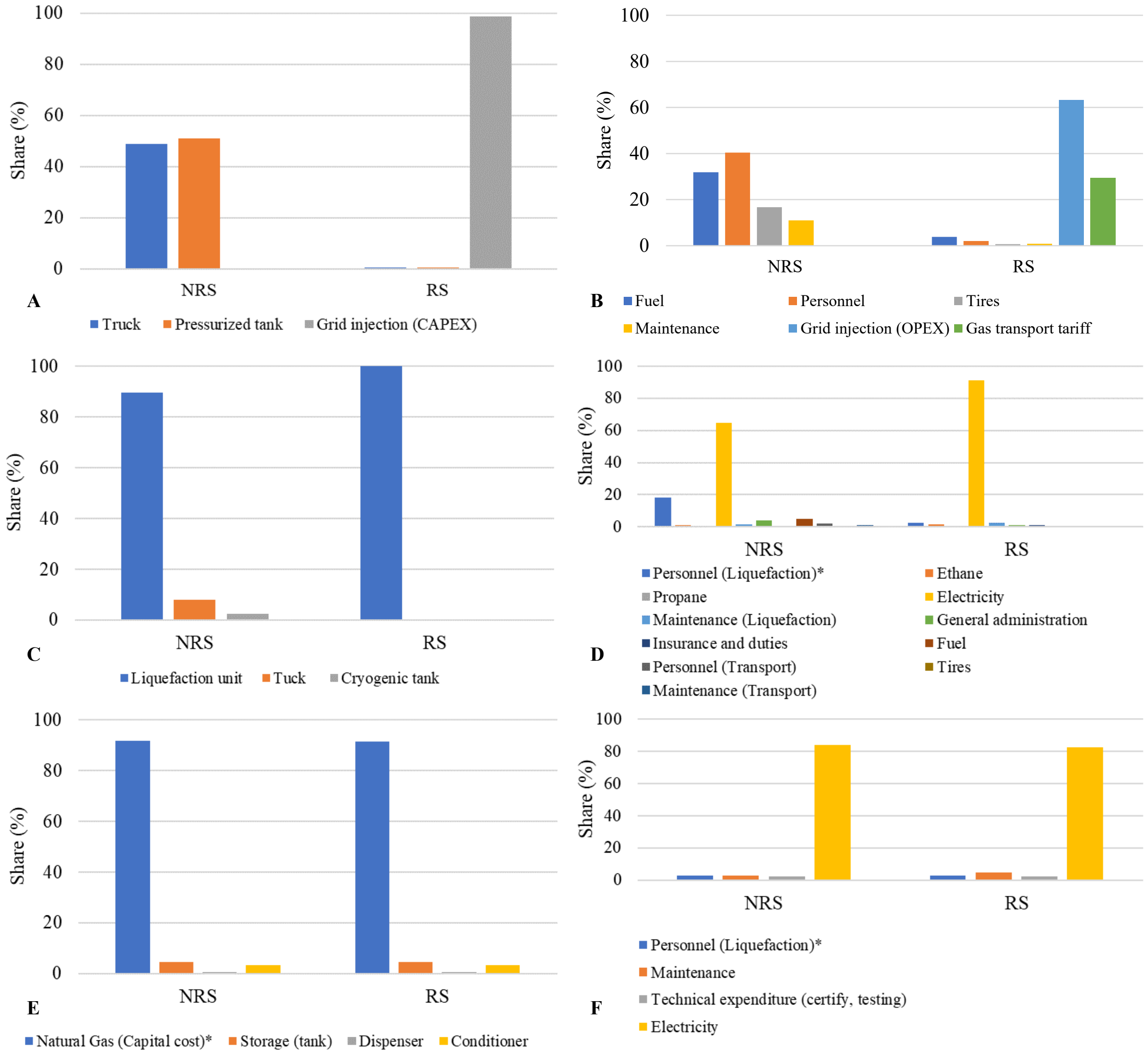

Figure 6. A. Capital cost distribution of transport and injection of biomethane in the NRS and RS. B. Operating costs distribution of transport and injection of biomethane in the NRS and RS. C. Capital cost distribution of liquefaction and transport of LBM in the NRS and RS. D. Operating cost distribution of liquefaction and transport of LBM in the NRS and RS. Personnel (liquefaction) includes personnel per se and its administration and maintenance. E. Capital cost distribution of refuelling stations in the NRS and RS. F. Operating cost distribution of refuelling stations in the NRS and RS. Capital costs of natural gas include engineering, civil work, electrics, and auxiliary utilities.

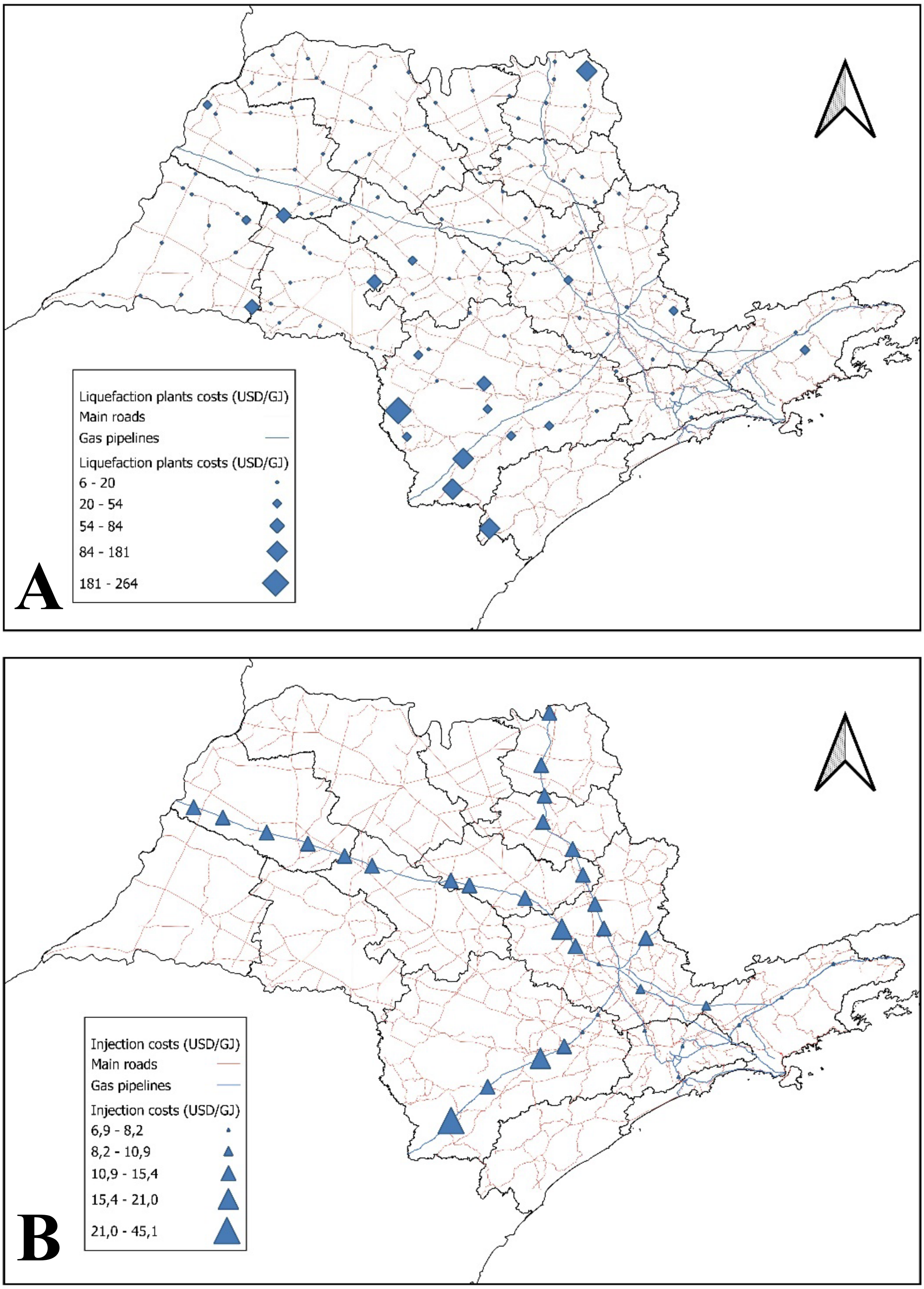

Figure 7. Costs (USD/GJ) of intermediary steps. A. Biomethane liquefaction plants in the NRS and B. Biomethane injection into the grid.

### 3.4 Refuelling stations

Downstream of AD-plants, it was observed roughly 4.9 billion $Nm^3$ (1.76 x 10^8 GJ) and 2.3 billion $Nm^3$ (8,35x10^7 GJ) of biomethane per year in the NRS and RS, respectively. These corresponded values are able to replace 50.3% and 23.9% of the total diesel consumed in the Sao Paulo state (3.58 x 10^8 GJ) [73]. Further 2.3 billion $Nm^3$ and 564 million $Nm^3$ per year of natural gas was needed to maintain the annual supply in the NRS and RS, respectively, at a price of 0.43 $USD.m^{-3}$ [67], due to the difference in working days between sugarcane mills (210 days per year) and the landfills (365 days per year), i.e., in 155 days of the year, the refuelling stations consume natural gas to maintain a steady supply throughout the year. Furthermore, 7 refuelling stations are required to supply of LBM throughout the state of Sao Paulo and ensuring the trucks' autonomy (400 km), as previously shown in Figure 3. Finally, the capital costs at the refuelling stations were estimated in USD 94.7 million per year and USD 56.2 million per year in the NRS and RS, respectively. The greatest expenditure share was represented by the engineering, civil work, electrics, and auxiliary utilities of natural gas liquefaction on site (subsection 3.3.) which could be reduced by increasing biomethane production, in particular as the production of the first-generation of ethanol (1G Ethanol) has been endorsed by the government through the Federal Law No. 13,576/2017 [74]. The National Biofuel Policy (RenovaBio) foresees expansion of biofuels by 2030. Briefly, the program's guidelines aim to fund new industrial units and technologies, including the second-generation of ethanol (2G Ethanol) which may result in an increment of up to 50%-60% of its current production. Interestingly, the biodigestion of residues from the 1G2G sugarcane ethanol biorefinery has been demonstrated in batch experiments with useful results as preliminary drivers to scale up [40]. Once more, expenditure with electricity had the highest impact on the operating costs, representing more than 80% of shares at the refuelling stations (Figure 6E and F).

### 3.5 Minimum selling price throughout the biogas chain

The minimum selling price (MSP – tax not included) of biomethane was estimated as a function of the total costs of the full biogas value chain including: (i) AD-plants – biogas production and purification, (ii) liquefaction units, (iii) injection points and (iv) refuelling stations. The first MSP of biomethane was estimated based on the capital and operating costs of each AD-plant and the respective costs of biomethane delivery prior its liquefaction, considering the designed scenarios. Figure 4 depicts five different clusters of biomethane price, being the lowest value of 3.0 USD.GJ$^{-1}$ and the highest of 21,688 USD.GJ$^{-1}$. More than 76.3% of the values in the NRS were lower than 23.15 USD.GJ$^{-1}$ which is referred to the current diesel price [75], while 97.1% of values in the NRS were lower than the threshold (diesel price). The wide range in the MSP of biomethane downstream of AD-plants is associated with the scale factor of the plants, therefore, further investigation must be performed to optimise capital and operating investment. Next, the MSP of biomethane downstream of liquefaction units and injection points were between 6-264 USD.GJ$^{-1}$ and 6.9-45.1 USD.GJ$^{-1}$, respectively. More than 85% of the values were above the threshold in both scenarios, indicating a better distribution of capital and operating costs and, therefore, a normalisation of different AD-plants' scales (Figure 7). Finally, the average MSP of biomethane (i.e., LBM) at the refuelling stations was estimated at 12.9 USD.GJ$^{-1}$ in the NRS and 17.4 USD.GJ$^{-1}$ in the RS, suggesting a better market attractiveness for biomethane compared to diesel, considering the examples outlined (Figure 8 – Table 12). It is worth mentioning that the values estimated above are very specific to the examples proposed. Regardless, this study has meticulously addressed the cost of unit process of AD-plant, biogas purification, biomethane transport, injection, liquefaction, and refuelling plants.

Table 12. Summary of minimum selling price of BM throughout the biogas chain vs. Diesel price.

| **Sugarcane mill (ton. d$^{-1}$)** | **USD.GJ$^{-1}$** | | |
|---|---|---|---|
| | **BM-NRS** [a] | **BM-RS** [a] | **Diesel** [b] |
| **AD-plants** | 3.8-27543.8 | 3.8-307.3 | 23.15 |
| Liquefaction units | 7.6-335.3 | - | |
| Injection | - | 8.8-57.3 | |
| Refuelling station | 9.3-21.3 | 11.9-44.5 | |

a. Tax included. Federal taxes (Cide-Fuels), PIS/Pasep and Cofins of 9% and State tax of 18% (ICMS). b. Diesel price in June 2021.

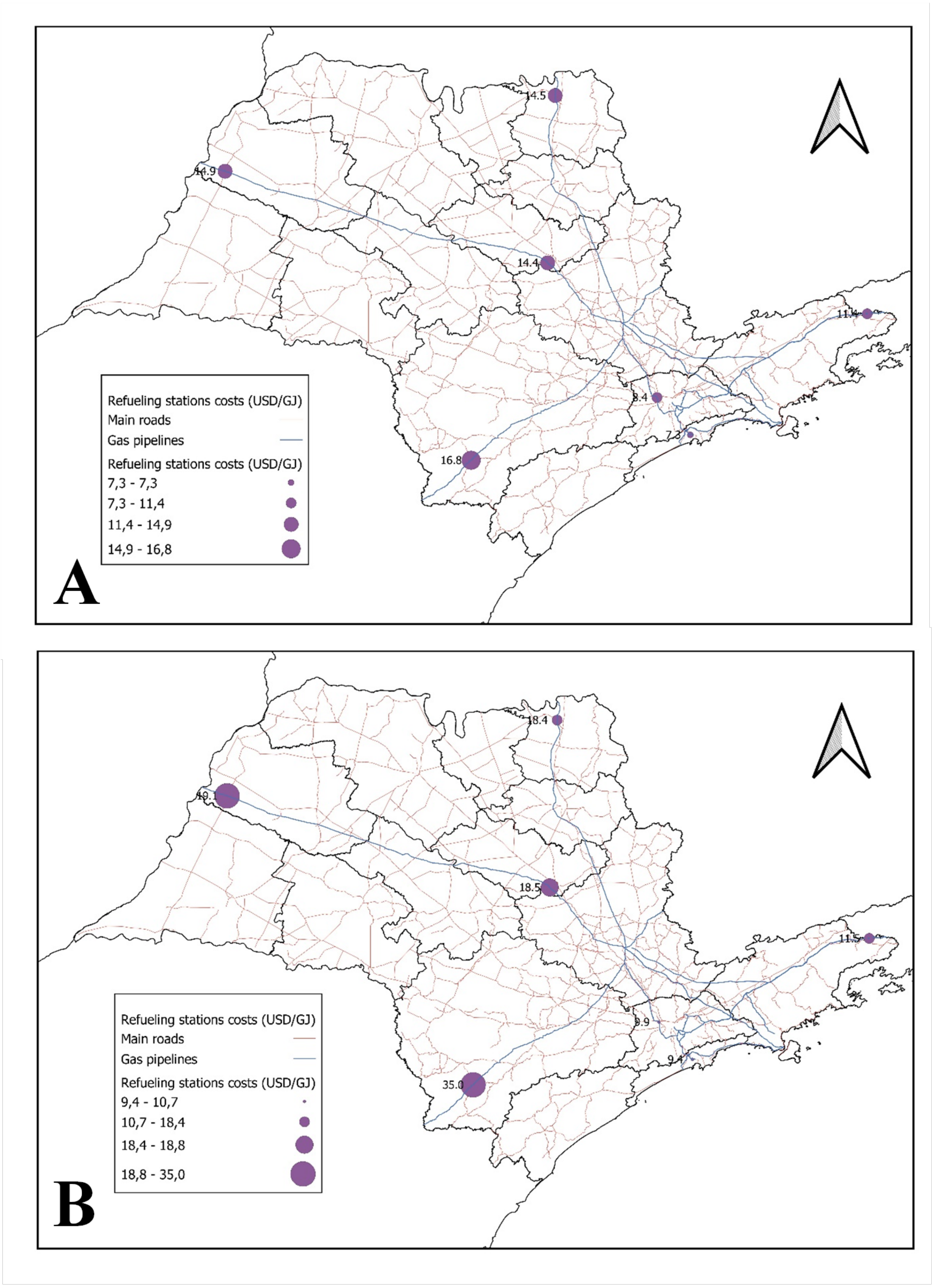

Figure 8. Minimum selling price of biomethane in the delivery point, i.e., the refuelling stations. A. NRS and B. RS.

# 4 REMARKS, PERSPECTIVES AND LIMITATIONS

We provide a few remarks and clarify some of the limitations of this study. First, the lack of geographic location information for some of the landfill sites in the state of Sao Paulo hinders the calculation of the full potential of biogas production. These landfills, however, most probably have low participation in overall potential. Another limitation that increases the uncertainty in the estimation of biogas potential is the lack of meta-information on sugarcane mills provided by EPE. The geographic information made available by the Brazilian Energy Research Company does not disclose the annual data on volumes of ethanol production in each mill; therefore, some of the information on ethanol production capacities could be outdated.

Limitations to the costs estimations. The estimates for the number of employees, level of employment and associated wage rates in biogas and liquefaction plants and refuelling stations could be further detailed in order to reduce the uncertainty in the labour cost calculations. Estimating water consumption biogas purification / upgrading. We assumed that all the water requirements for the purification would be provided by the ethanol and/or sugar processing units in sugarcane mills, which might not be the case in sugarcane mills which are efficient users of process water.

Finally, the size and number of refuelling stations will need careful evaluation and consideration within the context of State and National transport infrastructure development. Concentrating 25 dispensers in one refuelling station could bring logistical issues and hidden costs that have not been evaluated in this study. With regards of perspectives on renewable fuels in Brazil, biogas/biomethane is still a new energy vector in the national energy matrix and must be considered strategically in national energy planning. According to the results of this study, up to half of diesel could be replace in the freight system of Sao Paulo state using SCV and MSW as feedstock. The two designed scenarios could be implemented in two steps. 1. The RS through the existing natural gas infrastructure followed by a progressive extension of RS via new infrastructure (dedicated pipelines) until full coverage of the given area (NRS - Step 2). Methodologies that include the temporal costs and income dynamics, perhaps via a Net Present Value calculation, could assist in such decision. However, the use of biogas/biomethane in Brazil is still one step back, considering that less than 2% of its potential is used today. Regulations are necessary to make biogas/biomethane chemically, economically and politically “visible’’ in the country. Therefore, it is always important to highlight the main advantages of biogas/biomethane: 1. The chain of biogas production, purification, transport, liquefaction, and final use generates economic results through planning, implementation, operation, and maintenance services. 2. Biogas/Biomethane is projected to be self-financing and not affected by the exchange rate fluctuations and international price changes. 3. Global and local GHG and air pollutant emissions can be reduced especially when biogas/biomethane replaces diesel (not the scope of this study). 4. Biofertilizer is an inseparable product of AD-plants and can represent an important supply of plant nutrition and play a role in a transition towards closed loop, circular economies in the agricultural sector.

# 5 CONCLUSIONS

Under the assumptions in our study, LBM is estimated to be a cost-effective option when compared to diesel. The potential of biogas production by the sugarcane mills and landfill sites of Sao Paulo State can replace up to half of the diesel consumed within the State. The minimum distances and optimal locations methodology indicate a requirement for 120 liquefaction plants in the NRS, 35 injection points in the RS, and 7 refueling stations to supply LBM throughout the state of Sao Paulo. The $CO_2$ removal units had the greatest influence on capital costs at the AD-plants in both scenarios. Expenditure on the gas injection operation and the transport tariff took up a significant share of the total operating costs of the RS. Expenditure on electricity represented the highest share of the operating costs at biogas purification and biomethane liquefaction units. Labour costs are observed along the entire biogas value chain, especially, in the biomethane transport step, indicating an opportunity to generate wealth, jobs, and income. The feed-in tariff is recommended to accelerate investment in renewable energy technologies.

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