methane

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Methane Production from Sugarcane Vinasse Biodigestion: An Efficient Bioenergy and Environmental Solution for the State of São Paulo, Brazil

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Abstract: This study mapped the bioenergy production from sugarcane vinasse according to the mesoregions of the State of São Paulo (SP), Brazil, assessing the magnitude of biogas-derived electricity and biomethane production and estimating the greenhouse gas (GHG) emissions. SP holds 45% of the Brazilian ethanol-producing plants, in which 1.4 million m^3 of carbon-rich vinasse are generated daily. The electricity generated from vinasse has the potential to fully supply the residential consumption (ca. 6.5 million inhabitants) in the main sugarcane-producing mesoregions of the state (Ribeirão Preto, São José do Rio Preto, Bauru, Araçatuba and Presidente Prudente). In another approach, biomethane could displace almost 3.5 billion liters of diesel, which represents a 26% abatement in the annual state diesel consumption. Energetically exploiting biogas is mandatory to prevent GHG-related drawbacks, as the eventual emission of methane produced under controlled conditions (261.2×10^6 kg-CO_2eq d^{-1}) is ca. 7-fold higher than the total emissions estimated for the entire ethanol production chain. Meanwhile, replacing diesel with biomethane can avoid the emission of 45.4×10^6 kg-CO_2eq d^{-1}. Implementing an efficient model of energy recovery from vinasse in SP has great potential to serve as a basis for expanding the utilization of this wastewater in Brazil.

Keywords: bioenergy production mapping; diesel displacement; greenhouse gas abatement; sugarcane biorefinery; vinasse management

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1. Introduction

The Brazilian sucro-alcohol industry is a highly consolidated biorefinery model, providing sugar, ethanol and bagasse-derived thermoelectricity as major products [1]. Brazil is the second largest ethanol producer in the world, leading the rank when specifically considering sugarcane as the feedstock [2,3]. Ethanol production from sugarcane in Brazil reached 26.5 million m^3 in the 2022/2023 harvest, with the potential to reach 27.7 million m^3 in 2023/2024 [4]. Despite the continent-equivalent area of Brazil, sugarcane cultivation and processing are unevenly distributed and highly concentrated in the center-south region, which accounts for more than 90% of the total Brazilian ethanol production [4]. A closer spatial distribution analysis shows an additional concentration of ethanol plants in the State of São Paulo (SP), where 45% of the Brazilian biorefineries are installed [5] providing 45% of the Brazilian ethanol production [4].

The renewable character and the lower carbon content relative to gasoline are major environmental advantages of ethanol in relation to fossil fuels [2]. However, limitations in wastewater management still impose challenges on the environmental suitability of sugarcane processing towards ethanol [1,6]. Focus is given to vinasse, the primary wastewater from distillation, which concentrates high amounts of organic and inorganic constituents [7,8]. The potassium-rich character of vinasse stimulates its use in the fertirrigation of sugarcane fields as a strategy to recycle water and nutrients as well as to minimize costs of mineral fertilization [7]. Short-term analyses of fertirrigated areas show some improvements in nutrient availability [9]; however, the long-term soil application of vinasse has potential to trigger numerous adverse environmental impacts on soil, water and air [10–12]. The emission of greenhouse gases (GHGs) is of particular interest, as methane and nitrous oxide can be produced by the soil microflora [12–15]. In addition, the high organic load of vinasse concentrates ca. 10% of sugarcane's energy content, characterizing the uncontrolled conversion of organic matter during fertirrigation as a relevant waste of bioenergy [6].

Anaerobic digestion (AD) is considered by far the best technological approach to manage sugarcane vinasse, considering a series of benefits: (i) minimization of the polluting organic load, (ii) opportunity for recovering bioenergy in the methane-rich biogas stream and (iii) maintenance of the nutrient-rich character of fresh vinasse in the digestate [1,6,8]. Despite all these advantages, real-scale experiences with vinasse AD are very restricted in Brazil, with the recent implementation of only some lagoon-based bioenergy-producing plants [16,17]. In practical terms, numerous aspects of sugarcane vinasse AD at industrial scale are still unknown, including the energy potential, the real fertilizer character of digestate and the reduction in carbon emissions in sugarcane fields.

Knowing the real-scale energy potential of vinasse AD is imperative to understand potential applications, which stimulated numerous scenarization-based investigations in the last decade [18–22]. However, these studies are based on micro- or macro-scale analyses, in which the energy production is assessed within the boundaries of a single biorefinery (micro) or considering the whole volume of vinasse produced in Brazil (macro) without considering the aforementioned uneven spatial distribution of biorefineries. Identifying regions in which vinasse-derived energy can be promptly utilized is of utmost importance to define hotspots for the implementation of AD plants, leading to a regional characterization of vinasse's energy potential. This analysis is of great importance within the concept of decentralized energy supply, in which losses are minimized and more efficient energy utilization is achieved [23].

This study utilizes SP, the largest ethanol-producing and most populous area in Brazil, as the reference to innovatively assess the spatial distribution of the energy potential of vinasse according to the mesoregions of the state. The energy production and the opportunities for its local utilization were assessed considering the production of electricity and biomethane. Avoided GHG emissions were also calculated, providing a basis for understanding the environmental gains of the process. This study is fully integrated into the Plano Estadual de Energia 2050 or State Energy Plan 2050 in free translation, which aims to plan the energy sector in SP with focus on achieving CO_2 neutrality [24]. In practical aspects, spatially locating areas with low or great potential to produce bioenergy is extremely important to optimize the production and prompt utilization of this energy (electricity or biomethane in this case) in order to minimize costs incurred with transmission (electricity) or distribution (biomethane) and directly supply local demands. In other words, the approach presented in this study is an excellent tool for planning bioenergy production and use.

2. Methods

2.1. Description of the State of São Paulo

SP is the most populous state in Brazil, with a total of 44.4 million inhabitants (almost 22% of the Brazilian population) according to the most recent census data from the Brazilian Institute of Geography and Statistics [25]. SP is divided into 15 mesoregions, each one characterized by uneven population distribution patterns (Table 1). The distribution of sugarcane cultivation areas is also uneven in SP (Table 1), with a great concentration (over 42%) in the northeast region of the state (mesoregions of Ribeirão Preto and São José do Rio Preto; Table 1). Currently, 149 sugarcane processing plants are installed in SP [5].

Table 1. Population and sugarcane harvesting area in the mesoregions of SP.

Mesoregion (Code)	Sugarcane Cultivation Area (ha) [a]	Population (inhab) [b]
Araçatuba (ATB)	642,856	752,643
Araraquara (AQR)	440,205	862,355
Assis (ASS)	338,426	610,527
Bauru (BAU)	687,991	1,605,543
Campinas (CPS)	236,016	4,042,278
Itapetininga (ITP)	52,649	902,208
Litoral Sul Paulista (LSP)	Zero	501,794
Macro Metropolitana Paulista (MMP)	24,246	2,772,114
Marília (MAR)	87,279	486,204
Metropolitana de São Paulo (MSP)	Zero	22,833,820
Piracicaba (PCB)	298,680	1,479,941
Presidente Prudente (PPT)	569,335	927,930
Ribeirão Preto (RPO)	1,402,877	2,525,050
São José do Rio Preto (SRP)	1,127,087	1,696,313
Vale do Paraíba Paulista (VPP)	Zero	2,421,738
Total	5,907,647	44,420,459

[a] Referring to the 2020/2021 season period [26]; [b] Referring to the 2022 Census [25].

2.2. Input Data for Calculations

In addition to the sugarcane cultivation areas listed in Table 1, input data used in the calculations included sugarcane productivity and ethanol yield, as well as specific vinasse generation (Table 2). It is important to stress that the ethanol yield considered in this study refers to the one observed in annexed sugarcane biorefineries, in which both juice and molasses (the latter remaining from sugar production) are used as substrates in yeast fermentation. Annexed biorefineries account for over 80% of the sugarcane plants installed in SP [27]. Performance data related to biodigestion are also listed in Table 2, including substrate conversion and methane production results observed for the processing of sugarcane vinasse derived from annexed plants [28].

2.3. Energy Assessment Methodology

2.3.1. Calculation Procedure

The calculation protocol was initially based on the calculation of the vinasse production rate (VPR; $m^3\ d^{-1}$) and biogas production rate (BPR; $Nm^3\ d^{-1}$) according to Equations (1) and (2). SCA is the sugarcane cultivation area (ha), whilst the terms SCP, EY, SVG, HP, COD, ER_{COD}, MY and C_{CH4} are described in Table 2. The total energy potential of biogas (TEP; $MJ\ d^{-1}$) was calculated using Equation (3), considering a lower heating value (LHV_{biogas}) of 29.0 MJ Nm^{-3} for the biogas with $C_{CH4,biogas}$ = 81.2% (value estimated assuming the LHV of pure methane as 35.72 MJ Nm^{-3}; [29]). VPR, BPR and TEP, as well as the energy production approaches described in the sequence, were calculated for each mesoregion.

$$VPR = \frac{SCA \times SCP \times EY \times SVG}{HP} \quad (1)$$

$$BPR = \frac{VPR \times COD \times ER_{COD} \times MY}{C_{CH4,biogas}} \quad (2)$$

$$TEP = BPR \times LHV_{biogas} \quad (3)$$

Table 2. Input data used in the calculations.

Parameter (Symbol)		Value	Unit	Reference
Sugarcane production and processing	Sugarcane productivity (SCP)	81.529	$TC\ ha^{-1}$	[4]
	Ethanol yield [a] (EY)	53.4	$L_{ethanol}\ TC^{-1}$	[18]

Table 2. *Cont.*

Parameter (Symbol)		Value	Unit	Reference
Sugarcane production and processing	Specific vinasse generation (SVG)	13.0	$L_{vinasse}\ L^{-1}_{ethanol}$	[30]
	Harvesting period (HP)	240	d	[27]
Vinasse biodigestion	Chemical oxygen demand (COD) of vinasse [a]	35.4	$g\ L^{-1}$ or $kg\ m^{-3}$	[30]
	COD removal efficiency (ER_{COD}) [b]	86.5	%	[28]
	Methane yield (MY) [b]	0.343	$Nm^3CH_4\ kg^{-1}COD_{removed}$	[28]
	Methane concentration in biogas ($C_{CH4,biogas}$) [b]	81.2	%	[28]

[a] Refers to annexed biorefineries; [b] Considering the application of an organic loading rate of 10.0 kg COD m^{-3} d^{-1} and hydraulic retention time of 24.0 h in an anaerobic structured-bed reactor. TC: tons of sugarcane.

The energy recovery from biogas was assessed from two perspectives, considering the generation of electricity and the production of biomethane ($bioCH_4$), i.e., purified biogas with energy content similar or equivalent to that of natural gas (NG). Electricity generation was assessed assuming conservative and optimized scenarios, each one characterized by different electric conversion efficiency (η) levels: 0.38 (conservative approach referring to an internal combustion engine-based power plant) [18] and 0.60 (optimized approach referring to a combined-cycle-based power plant) [31]. Scenarization-based studies addressing potential uses for vinasse-derived biogas traditionally consider engines as the prime movers [18,32,33]; however, some recent investigations indicated significant energy gains with the use of the combined cycle [19,22]. The electric energy production (EEP; MJ or MWh, considering 1 MWh = 3600 MJ) was calculated according to Equation (4).

$$EEP = TEP \times \eta \times HP \quad (4)$$

The $bioCH_4$ production rate (BmPR; $Nm^3\ d^{-1}$) was calculated using Equation (5) assuming a methane concentration ($C_{CH4,bioCH4}$) of 95%, which exceeds the minimum concentration (90%) required by the Brazilian legislation [34]. The term IPL represents the intrinsic performance losses associated with biogas upgrading, with a fixed value of 2% [35]. The $bioCH_4$ energy potential (BmEP; MJ or MWh) was finally calculated according to Equation (6), considering a lower heating value (LHV_{bioCH4}) of 33.93 MJ Nm^{-3} for $bioCH_4$.

$$BmPR = \left(\frac{VPR \times COD \times ER_{COD} \times MY}{C_{CH4,bioCH4}}\right)\left(1 - \frac{IPL}{100}\right) \quad (5)$$

$$BmEP = BmPR \times LHV_{bioCH4} \times HP \quad (6)$$

2.3.2. Comparative Analyses: Electricity Production

The magnitude of vinasse-derived electricity was assessed by comparing EEP with both the electric residential consumption in each mesoregion and the thermoelectricity production from bagasse by calculating the energy replacement potential (ERP; %). In the first case, the ERP was calculated according to Equation (7), in which the terms Pop and pcEC are the population of each mesoregion (according to Table 1; inhab) and the annual per capita electricity consumption in SP (2926 kWh $inhab^{-1}$) [36]. The numerical term 8/12 corrects the pcEP to eight months, i.e., equivalent to the HP (240 d). Equation (8) describes the ERP relative to the thermoelectricity production from bagasse, in which the term EYB is the electricity yield from bagasse (58.98 kWh TC^{-1} or 58.98×10^{-3} MWh TC^{-1}) [37].

$$ERP = \left[\frac{Pop \times pcEC \times (8/12)}{EEP}\right]100 \quad (7)$$

$$ERP = \left(\frac{SCA \times SCP \times EYB}{EEP}\right)100 \quad (8)$$

2.3.3. Comparative Analyses: $bioCH_4$ Production

The magnitude of vinasse-derived $bioCH_4$ was initially assessed by calculating the potential to replace diesel in heavy-duty machinery and trucks in mills. The diesel-to-$bioCH_4$ equivalence (DB_{eq}; L), which represents the amount of diesel saved per harvest, was calculated according to Equation (9), in which the term LHV_{diesel} is the lower heating value of diesel oil (35.50 MJ L^{-1}) [38]. In addition, the sugarcane harvesting equivalent (SCH_{eq}; TC) was calculated using Equation (10), in which the term SDC is the specific diesel consumption (4 L TC^{-1}) [38] and represents the amount of diesel consumed during sugarcane harvesting, transportation and processing. The number of heavy-duty trucks (HDT_{eq}) potentially fed with $bioCH_4$ per season was also estimated (Equation (11)), considering an average truck efficiency of 1.15 km L^{-1} and that each truck covers 200 km daily [18].

$$DB_{eq} = \frac{BmEP}{LHV_{diesel}} \tag{9}$$

$$SCH_{eq} = \frac{DB_{eq}}{SDC} \tag{10}$$

$$HDT_{eq} = \frac{DB_{eq}}{[(200/1.15) \times HP]} \tag{11}$$

2.4. Environmental Assessment Methodology

The environmental assessment was based on the methodology proposed elsewhere [9], in which the uncontrolled methane emission (UME; kg-CO_2eq d^{-1}) resulting from the degradation of vinasse's organic content in sugarcane fields and the amount of non-emitted methane (NEM; kg-CO_2eq d^{-1}) resulting from the production and use of biogas in biodigestion systems were calculated. UME and NEM calculation protocols were modified, as detailed in the sequence. UME was obtained from Equation (12), in which the terms SME and GWP are the specific methane emission from vinasse (0.062 kg-CH_4 m^{-3} vinasse) [15] and the global warming potential of methane (25 kg-CO_2eq $kg^{-1}CH_4$) [39]. NEM was calculated using Equation (13), in which the numerical term 0.714 is used to convert the methane production from Nm^3 to kg.

$$UME = VPR \times SME \times GWP \tag{12}$$

$$NEM = 0.714 \times VPR \times COD \times (ER_{COD}/100) \times MY \times GWP \tag{13}$$

The amount of non-emitted GHG (NEGHG; kg-CO_2eq d^{-1}) resulting from diesel oil replacement by $bioCH_4$ was finally calculated according to Equation (14). In this case, the direct (DGHGE) and indirect (IGHGE) GHG emissions associated with diesel use were assumed as 74.1 and 14.5 g-CO_2eq MJ^{-1} [40], respectively.

$$NEGHG = \frac{BmEP \times (DGHGE + IGHGE) \times 10^{-3}}{HP} \tag{14}$$

2.5. Mapping of Key Results

Selected results from the calculation procedures described in Sections 2.3 and 2.4, namely, VPR, BPR, EEP, ERP, BmEP, DB_{eq}, SCH_{eq}, UME, NEM and NEGHG, were mapped following the mesoregions of SP. The software QGIS version 3.30.3 by OSGeo was used to build the maps. The shapefile of SP's mesoregions was obtained from the Brazilian Institute of Geography and Statistics [41].

3. Results and Discussion

3.1. Spatial Distribution of Vinasse-Derived Biogas in SP

The mapping of the biogas production rate from sugarcane vinasse (BPR) in the SP mesoregions is depicted in Figure 1b and clearly matches the production of vinasse throughout the state (Figure 1a). This distribution pattern is intrinsic to the areas covered by sugarcane crops in the SP mesoregions, which are remarkable in Ribeirão Preto (1,402,877 ha; Table 1) and São José do Rio Preto (1,127,087 ha; Table 1), with both accounting for ca. 43% of the total harvested area in the state (5,907,647 ha). Within this scenario, both the VPR and BPR presented high generation/production potentials in the mesoregions covering the largest sugarcane crop areas, with Ribeirão Preto (VPR = 330,830 m^3 d^{-1} and BPR = 4,279,200 Nm^3 d^{-1}), São José do Rio Preto (VPR = 265,793 m^3 d^{-1} and BPR = 3,437,957 Nm^3 d^{-1}), Bauru (VPR = 162,244 m^3 d^{-1} and BPR = 2,098,581 Nm^3 d^{-1}), Araçatuba (VPR = 151,600 m^3 d^{-1} and BPR = 1,960,905 Nm^3 d^{-1}) and Presidente Prudente (VPR = 134,262 m^3 d^{-1} and BPR = 1,736,644 Nm^3 d^{-1}) standing out and together representing ca. 75% of the total vinasse-derived biogas potential in the state.

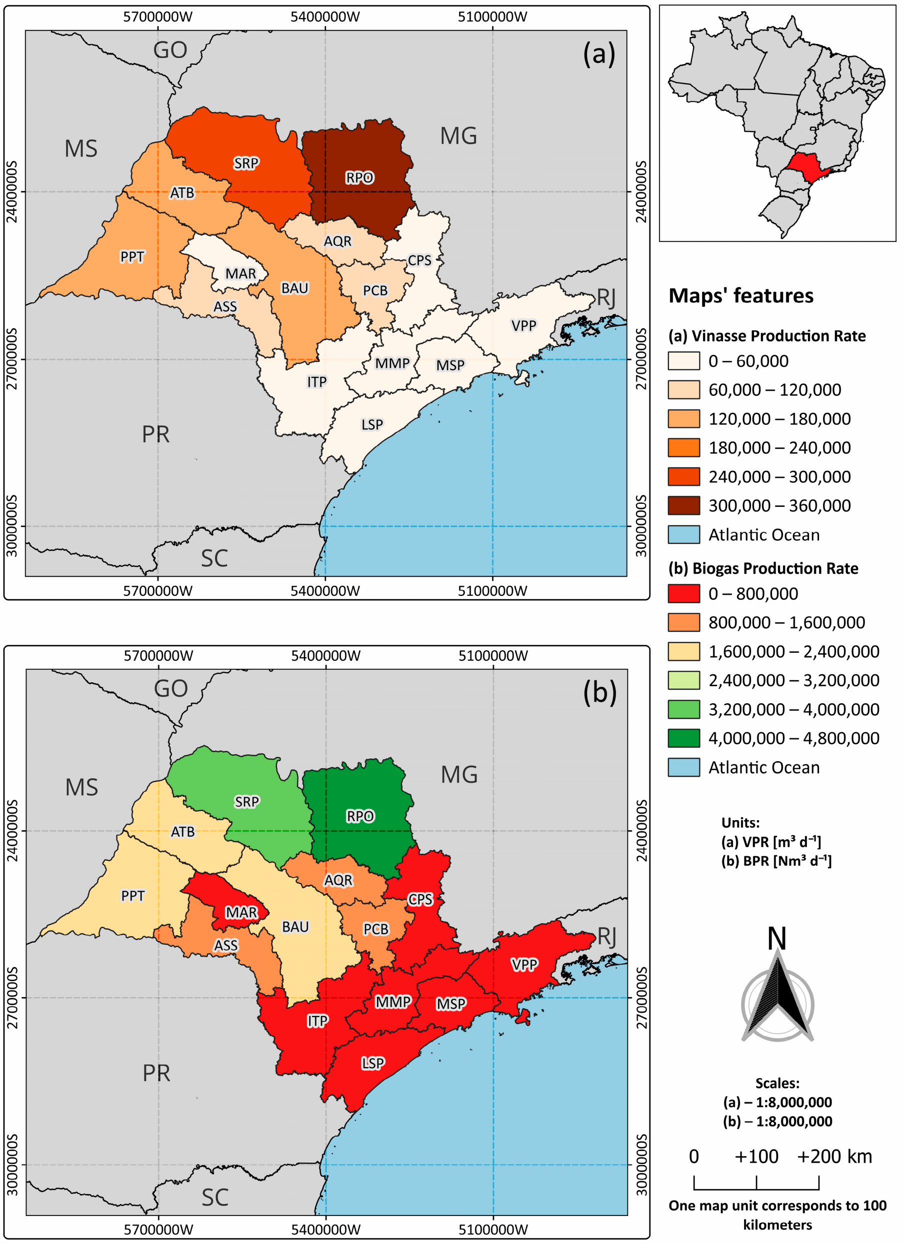

Figure 1. Overall characterization: mapping of the (**a**) vinasse production rate (VPR) and (**b**) biogas production rate (BPR) according to the mesoregions of the State of São Paulo. Mesoregions: ATB, Araçatuba; AQR, Araraquara; ASS, Assis; BAU, Bauru; CPS, Campinas; ITP, Itapetininga; LSP, Litoral Sul Paulista; MMP, Macro Metropolitana Paulista; MAR, Marília; MSP, Metropolitana de São Paulo; PCB, Piracicaba; PPT, Presidente Prudente; RPO, Ribeirão Preto; SRP, São José do Rio Preto; VPP, Vale do Paraíba Paulista.

These results reveal a spatial sectoring of the resource recovery potential from sugarcane vinasse in the mesoregions located in the northern (Ribeirão Preto, São José do Rio Preto and Araçatuba), northwestern (Presidente Prudente, Assis and Marília) and central (Bauru, Araraquara and Piracicaba) portions of SP. On the other hand, the southern (Itapetininga, Macro Metropolitana de São Paulo and Litoral Sul Paulista) and southeastern (Campinas, Metropolitana de São Paulo and Vale do Paraíba Paulista) regions offer the lowest energy recovery potential from vinasse. These six mesoregions concentrate more than 75% of the population of the state, and some are characterized by densely populated urban areas, reflecting the different patterns of land use and occupation in the state. It is worth highlighting that these areas offer different opportunities for recovering energy from biogas, such as by applying the anaerobic technology in the treatment of sewage or in the stabilization of the biological sludge generated in activated sludge systems.

In practical aspects, the sectorization pattern of vinasse-derived biogas production potential is a cornerstone for decision-makers involved in the ethanol production industry who aim to minimize the environmental impacts of fertirrigation and add energy value to the sugarcane biorefinery supply chain [9].

3.2. Electric Potential of Vinasse-Derived Biogas

The comparative assessment of the electric energy production (EEP) for the conservative (Figure 2a,b) and optimized (Figure 2c,d) scenarios reveals a different spatial sectoring pattern for the energy recovery potential (ERP; Figure 2b,d) in the mesoregions. Overall, the EEP from vinasse-derived biogas for the conservative (Figure 2a) and optimized scenarios (Figure 2c) is directly related to the generation of this wastewater in each mesoregion, and therefore its spatial sectoring is depicted in a pattern very similar to the mapping of the VPR (Figure 1a) and BPR (Figure 1b). As a result, the mesoregions of Ribeirão Preto (EEP-conservative = 3144 GWh and EEP-optimized = 4965 GWh), São José do Rio Preto (EEP-conservative = 2526 GWh and EEP-optimized = 3988 GWh), Bauru (EEP-conservative = 1542 GWh and EEP-optimized = 2434 GWh), Araçatuba (EEP-conservative = 1441 GWh and EEP-optimized = 2275 GWh) and Presidente Prudente (EEP-conservative = 1276 GWh and EEP-optimized = 2014 GWh) stood out compared to the other mesoregions, accounting for 75% of the total electric potential resulting from vinasse-derived biogas in conservative (9928 GWh) and optimized (15,675 GWh) scenarios.

According to the 2023 Statistical Yearbook of Electricity prepared by the Brazilian Energy Research Company [36], which provides key information on the Brazilian energy supply chain using data referring to the year 2022, SP holds 11.4% (ca. 23.54 GW or 206,351,640 GWh) of the country's installed generation potential (206.5 GW or 1,810,179,000 GWh) and ranks first in the national scenario. Within this context of adding vinasse-derived biogas as an alternative energy source in SP, its representativeness corresponds to 0.0064% and 0.0101% for the conservative and optimized scenarios, respectively, which can be considered to have little impact compared to the overall SP energy supply chain. However, analyzing the per capita electricity consumption in SP, the total potential for recovering electricity from vinasse-derived biogas to supply household demand is 15.28% (13,239 GWh) and 24.14% (20,903 GWh) for the conservative and optimized scenarios, respectively. The leading regions of Ribeirão Preto, São José do Rio Preto, Bauru, Araçatuba and Presidente Prudente account for an electricity recovery capacity of 11.5% (EEP-conservative = 9928 GWh) and 18.1% (EEP-optimized = 15,675 GWh) of all household electricity consumed in SP (86,664 GWh) considering the conservative and optimized scenarios, respectively.

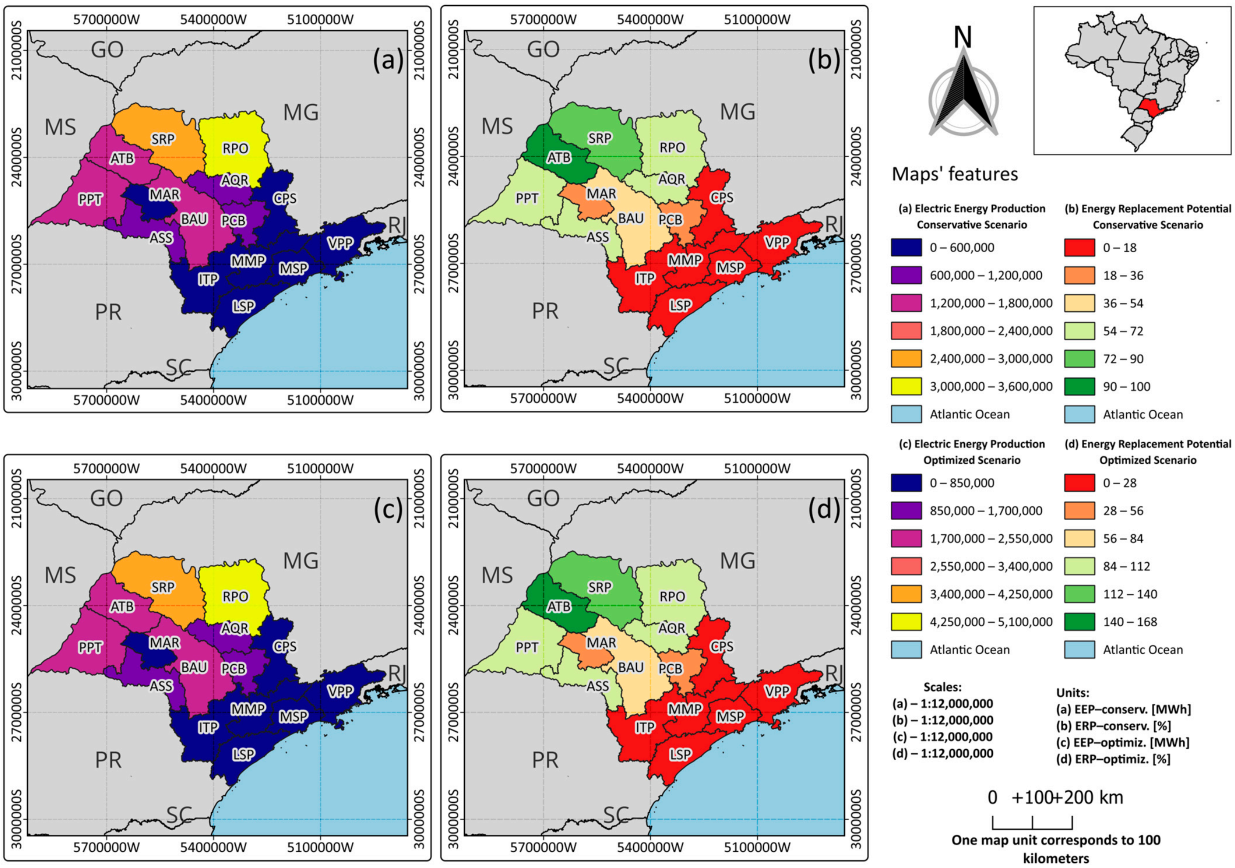

Figure 2. Energy assessment: mapping of the (**a**) electric energy production (EEP) and (**b**) energy replacement potential (ERP) for the conservative approach; and (**c**) EEP and (**d**) ERP for the optimized approach according to the mesoregions of the State of São Paulo. Mesoregions: ATB, Araçatuba; AQR, Araraquara; ASS, Assis; BAU, Bauru; CPS, Campinas; ITP, Itapetininga; LSP, Litoral Sul Paulista; MMP, Macro Metropolitana Paulista; MAR, Marília; MSP, Metropolitana de São Paulo; PCB, Piracicaba; PPT, Presidente Prudente; RPO, Ribeirão Preto; SRP, São José do Rio Preto; VPP, Vale do Paraíba Paulista.

Although these results are remarkable, assessing the local ERP, i.e., within the boundaries of a given mesoregion, provides a better understanding of the magnitude of the EEP. Overall, the mesoregions that showed the greatest potential for recovering electricity via vinasse-derived biogas also had prominent positions in terms of ERP, but in different rankings, including Araçatuba (ERP-conservative = 98.1% and ERP-optimized = 154.9%), São José do Rio Preto (ERP-conservative = 76.3% and ERP-optimized = 120.5%), Presidente Prudente (ERP-conservative = 70.5% and ERP-optimized = 111.3%) and Ribeirão Preto (ERP-conservative = 63.8% and ERP-optimized = 100.8%). The potential of vinasse-derived electricity to fully supply the residential consumption in these mesoregions when considering the optimized approach is noteworthy. Surprisingly, regions with relatively low EEP (less than 1,000,000 MWh) showed very attractive ERP values. For instance, Assis and Araraquara indicated ERP values of 63.7–100.5% and 58.6–92.6% under the conservative and optimized scenarios, respectively. The use of electricity in areas close to the generation site is of utmost importance for minimizing losses in transmission lines. In addition, the sugarcane harvesting period coincides with the dry season in Brazil (about from May/June to September/October), so the electricity from vinasse has the potential to supply the electric consumption mainly during events of low availability of hydroelectricity, also decreasing the dependency on oil-fueled thermoelectricity plants.

The electricity generated from vinasse has the potential to increase the installed capacity of sugarcane biorefineries by 46.6% (conservative approach) and 73.6% (optimized approach), using the bagasse-derived thermoelectricity as a comparative reference. The EEP relative to the amount of processed sugarcane corresponded to 27.49 kWh TC^{-1} (conservative approach) and 43.40 kWh TC^{-1} (optimized approach), comprising values that can fully supply the electricity consumption in ethanol production (12.47 kWh TC^{-1}) [42]. Currently, the sucro-energy sector provides ca. 7% of the total electric power granted by the Brazilian Electricity Regulatory Agency (ANEEL) [37], so using vinasse-derived electricity can markedly increase the participation of sugarcane biorefineries in the electric sector.

EEP values were also compared with the energy sources that make up the Brazilian electricity matrix. Except for the case of hydropower, with an annual contribution of 427,114 GWh or 427.1 TWh [36], the vinasse-derived electricity produced in SP would correspond to large fractions (in some cases exceeding the entire installed capacity) of consolidated energy sources. For instance, the EEP calculated for the conservative approach (13,293 GWh) could individually replace 90% of the nuclear power, as well as the entire electricity generated from coal and oil derivatives (Figure 3). Meanwhile, the EEP derived from the optimized approach (20,903 GWh) accounted for 49.7%, 40.4% and 69.4% of the electricity generated from NG, biomass and the Sun, respectively (Figure 3). The contribution of vinasse-derived electricity has the potential to approximately double the values depicted in Figure 3, as more than 90% of the Brazilian ethanol production is concentrated in the center-south region [4]. Nevertheless, the size of the biorefinery, which impacts the amount of vinasse available for processing and, consequently, the biogas production, directly impacts the economics of recovering bioenergy from vinasse. In practical terms, converting vinasse into electricity (or $bioCH_4$) may not be economically feasible at small-scale sugarcane facilities, so considering the implementation of biogas hubs capable of receiving organic substrates from different industrial plants may offset these scale limitations [43].

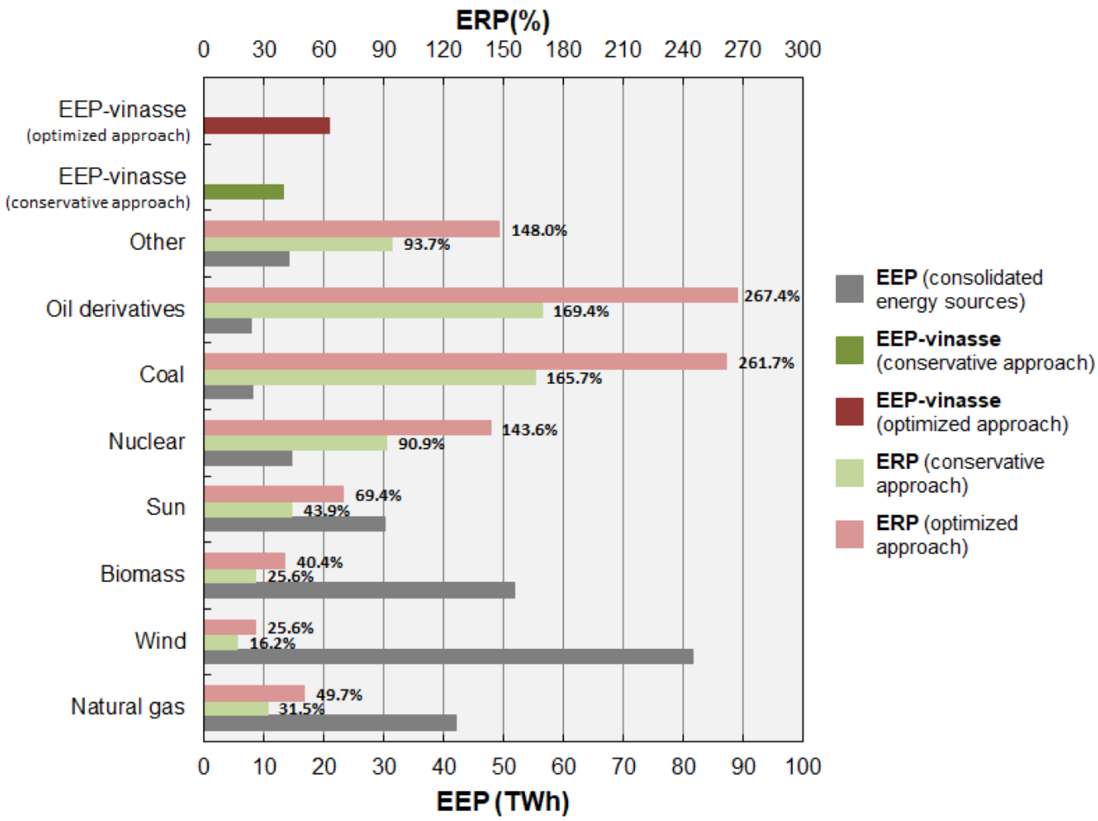

Figure 3. Electric energy production (EEP) from vinasse compared with consolidated energy sources in the Brazilian electricity matrix and resulting energy replacement potential (ERP). Percentage values refer to the ERP. Comparative EEP data (natural gas, wind, biomass, Sun, nuclear, coal, oil derivatives and other sources) obtained elsewhere [36].

3.3. Biomethane Production from Sugarcane Vinasse

The biogas evolved from sugarcane vinasse biodigestion can be subjected to different upgrading processes for removing CO_2 and other impurities, mainly hydrogen sulfide (H_2S), to produce bioCH_4 (methane concentration $\geq$ 90% in the Brazilian case) [34]. BioCH_4 is a versatile energy source and is also known as the "renewable natural gas" and can be applied in different sectors, such as in transportation (from light- to heavy-duty vehicles) and industry (engines, heating systems and gas turbines), as well as a direct alternative to NG in distribution systems [44,45]. The total bioCH_4 energy potential of SP was estimated as 34.1 $\times$ 10^6 MWh (Figure 4a), considering a total bioCH_4 volume of 3.6 $\times$ 10^9 Nm^3 per harvest. The spatial distribution of bioCH_4 followed the same pattern of electricity production, based on the availability of vinasse per mesoregion. Consequently, the mesoregions of Ribeirão Preto (8.1 $\times$ 10^6 MWh), São José do Rio Preto (6.5 $\times$ 10^6 MWh), Bauru (4.0 $\times$ 10^6 MWh), Araçatuba (3.7 $\times$ 10^6 MWh) and Presidente Prudente (3.3 $\times$ 10^6 MWh) (Figure 4a) were characterized as the main bioCH_4 producers.

The total bioCH_4 vinasse-derived production potential of SP is equivalent to 3.46 $\times$ 10^9 L of diesel oil (Figure 4b), which means that 26% of the annual diesel oil consumption in the state (12.6 $\times$ 10^9 L) [46] could be replaced. BioCH_4 also has the potential to replace 69% of the overall natural gas consumption in SP considering all types of consumers, i.e., 5.2 $\times$ 10^9 Nm^3 $year^{-1}$ [47]. It is important to stress that the distribution of bioCH_4 depends directly on the availability of gas grids close to sugarcane biorefineries because process profitability also takes into consideration the costs incurred with pipeline installation [19,32,42]. Hence, investing in electricity production from biogas may be a more rational strategy on a short- to medium-term basis [42], as this option does not depend on significant infrastructural modifications.

Focusing on the sugarcane biorefinery context, producing bioCH_4 could represent a huge gain both from economic and environmental perspectives. The diesel-oil-fueled heavy-duty machines and trucks used for harvesting and transportation, respectively, represent a considerable operational expense and one of the main environmental drawbacks for the sucro-alcohol industry [32,38,48]. The calculated diesel oil equivalent (Figure 4b) could supply almost 83,000 heavy trucks, maintaining the machinery used in the harvesting and transportation of 865 $\times$ 10^6 tons of sugarcane per season (Figure 4c). Given that a total of 484 $\times$ 10^6 tons of sugarcane is annually processed in SP, as estimated in the calculations considered herein, no more than 56% of the total bioCH_4 produced from sugarcane vinasse would be required to replace the overall diesel consumption in sugarcane distilleries per season.

The remaining amount of bioCH_4, i.e., 15 $\times$ 10^6 MWh or 1.6 $\times$ 10^9 Nm^3 per season, would still be available for sale, diversifying the bioenergy recovery within the sugarcane biorefinery concept. This energy would be able to offset 30.4% of the overall NG consumption in SP or could supply the whole NG demand for residential, commercial, transportation, cogeneration and thermogeneration in the state (1.1 $\times$ 10^9 Nm^3 $year^{-1}$) [47]. An overview of biomethane's specifications, sources and uses was released in August 2015 by the Brazilian National Agency of Oil, Natural Gas and Biofuels (ANP). The regulation allows for the injection of any proportion of bioCH_4 into NG grids, once the specifications defined by ANP are met [49].

Joppert et al. [50] carried out an energetic evaluation regarding the production and injection of bioCH_4 in the gas pipelines available in SP. The authors found that only 33.5% (66/197) of the operating biorefineries at that time were located in a radius of 20 km from a gas pipeline, which would be economically feasible for grid injection and gas distribution by utilizing an already existing infrastructure. Therefore, injecting surplus bioCH_4 into the gas grid would not be economically feasible in most of the sugarcane biorefineries of the state. Nevertheless, considering the high population density of SP, surplus bioCH_4 could be commercialized with local NG distribution companies. Moreover, as the substrate (i.e., sugarcane vinasse) comes from a biofuel production sector, the regulatory agency (ANP) and the potential customers (gas station distributors) would be the same, which could, in theory, facilitate the regulation and distribution. Finally, each biorefinery will naturally define the most suitable approaches to utilize biogas according to the surrounding demands, so that the sugarcane-processing plants located close to the gas grid can invest in $bioCH_4$ production, while the remaining facilities can implement electricity production.

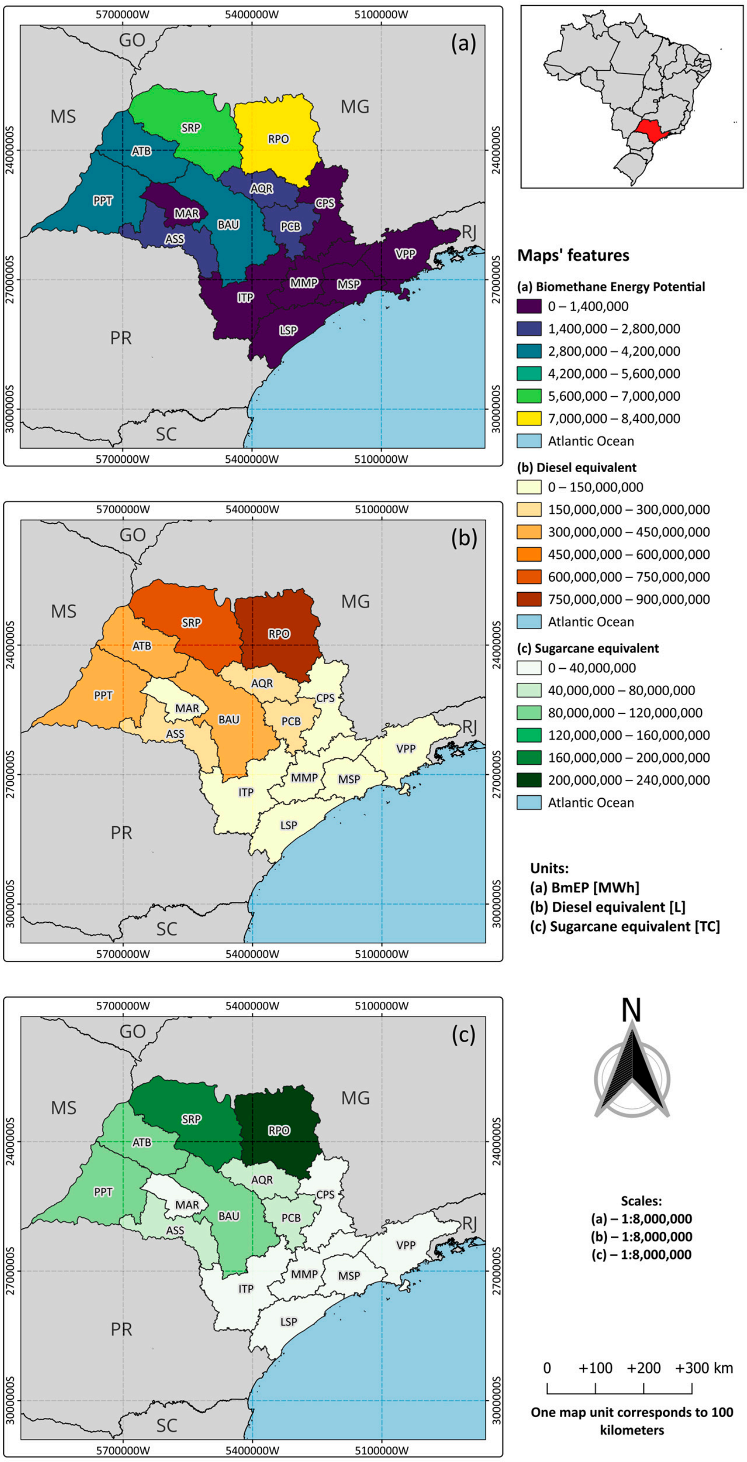

Figure 4. Energy assessment: mapping of the (**a**) $bioCH_4$ energy potential (BmEP), (**b**) diesel-to-$bioCH_4$ equivalence (DB_{eq}) and (**c**) sugarcane harvesting equivalent (SCH_{eq}) according to the mesoregions of the State of São Paulo. Mesoregions: ATB, Araçatuba; AQR, Araraquara; ASS, Assis; BAU, Bauru; CPS, Campinas; ITP, Itapetininga; LSP, Litoral Sul Paulista; MMP, Macro Metropolitana Paulista; MAR, Marília; MSP, Metropolitana de São Paulo; PCB, Piracicaba; PPT, Presidente Prudente; RPO, Ribeirão Preto; SRP, São José do Rio Preto; VPP, Vale do Paraíba Paulista.

3.4. Assessment of GHG Emissions

Figure 5 presents the estimates for uncontrolled methane emission (UME), non-emitted methane (NEM) and non-emitted greenhouse gas (NEGHG) in the 15 mesoregions of SP. All values varied markedly according to the spatial distribution of the sugarcane cultivation areas, following the same patterns observed for the production of energy (Sections 3.2 and 3.3). UME values ranging from 300,000–600,000 kg-CO_2eq d^{-1} were estimated for the mesoregions in which sugarcane cultivation is concentrated, namely, Ribeirão Preto, São José do Rio Preto, Bauru, Araçatuba and Presidente Prudente (Figure 5a). The total UME calculated for SP reached 2.2×10^6 kg-CO_2eq d^{-1}, corresponding to less than 2% of the GHG emissions associated with electricity production from all sources in Brazil (121.4×10^6 kg-CO_2eq d^{-1}) [36]. Considering all steps of ethanol production, the total amount of GHG emissions was estimated as 37.0×10^6 kg-CO_2eq d^{-1}, based on the specific emission value of 345 kg-CO_2eq m^{-3} ethanol reported elsewhere [51]. Hence, UME corresponds to ca. 6% of the total emissions in the sucro-energy sector in SP, so the contribution of other steps, such as the production of ethanol itself (which releases CO_2) and the use of diesel in agricultural operations and transportation, accounts for the remaining share of the total emissions. Results obtained for the UME might have been underestimated in this work because the emission of nitrous oxide (N_2O), which has also been measured in areas subjected to vinasse application [14,52], was not included. The GWP of N_2O exceeds 300 kg-CO_2eq $kg^{-1}N_2O$ [14], characterizing an additional source of GHG mainly when nitrogen concentrations are unbalanced. Equally, direct CO_2 emissions were not considered in this calculation, as focus was given to methane emanation.

Once decision-makers in the sucro-energy sector define biodigestion as the primary management approach for vinasse, the exploitation of biogas will be mandatory, considering NEM values (Figure 5b) about 120-fold higher than those obtained for UME. The total NEM calculated for SP reached 261.2×10^6 kg-CO_2eq d^{-1}, a value ca. 7-fold higher than the total emissions estimated for the entire ethanol production chain (37.0×10^6 kg-CO_2eq d^{-1}), as mentioned above. Under controlled conditions (anaerobic reactors), the activity of methanogenic microbes is greatly enhanced in comparison to that of "natural" environments, such as soils subjected to vinasse application, which demands efficient management of biogas. In practical aspects, designing an anaerobic processing plant for vinasse management cannot be justified only by the bias of reducing the polluting organic load of the wastewater. While the emission of high volumes of methane is a significant environmental burden, using flares as an alternative to attenuate such emissions is economically unfeasible [19].

In addition to the issues related to methane, Kabeyi and Olanrewaju [53] highlighted the potential of CO_2 sequestration via biogas utilization, specifically when considering bioCH_4 production. Biogas contains approximately 30% CO_2, which is extracted during the upgrading process and can be utilized in various applications inside or outside the biorefinery's borders. CO_2 can be used in juice clarification as a strategy to replace sulfitation considering an on-site utilization, whilst applications in the food industry (carbonation processes) and value-added chemical production are alternatives based on CO_2 commercialization [54]. The same strategies can also be used to manage the high-purity CO_2 (>99%) from yeast fermentation.

It is important to stress that not using the methane produced in biodigestion, i.e., assuming the release of NEM-related emissions, is more harmful to the environment than not replacing diesel with bioCH_4. The NEGHG (Figure 5c) considering the sum of all mesoregions corresponded to 45.4×10^6 kg-CO_2eq d^{-1}, a value almost 6-fold lower than the total NEM (261.2×10^6 kg-CO_2eq d^{-1}). Interestingly, NEGHG exceeded the total GHG emissions associated with ethanol production (37.0×10^6 kg-CO_2eq d^{-1}), showing a positive balance in sugarcane biorefineries. This scenario was much more favorable than the one reported elsewhere [38], in which the authors demonstrated that 27.5% of GHG emissions can be avoided by replacing diesel in mill operations. Nevertheless, these differences should be analyzed with caution because of the different calculation methodologies used in each study.

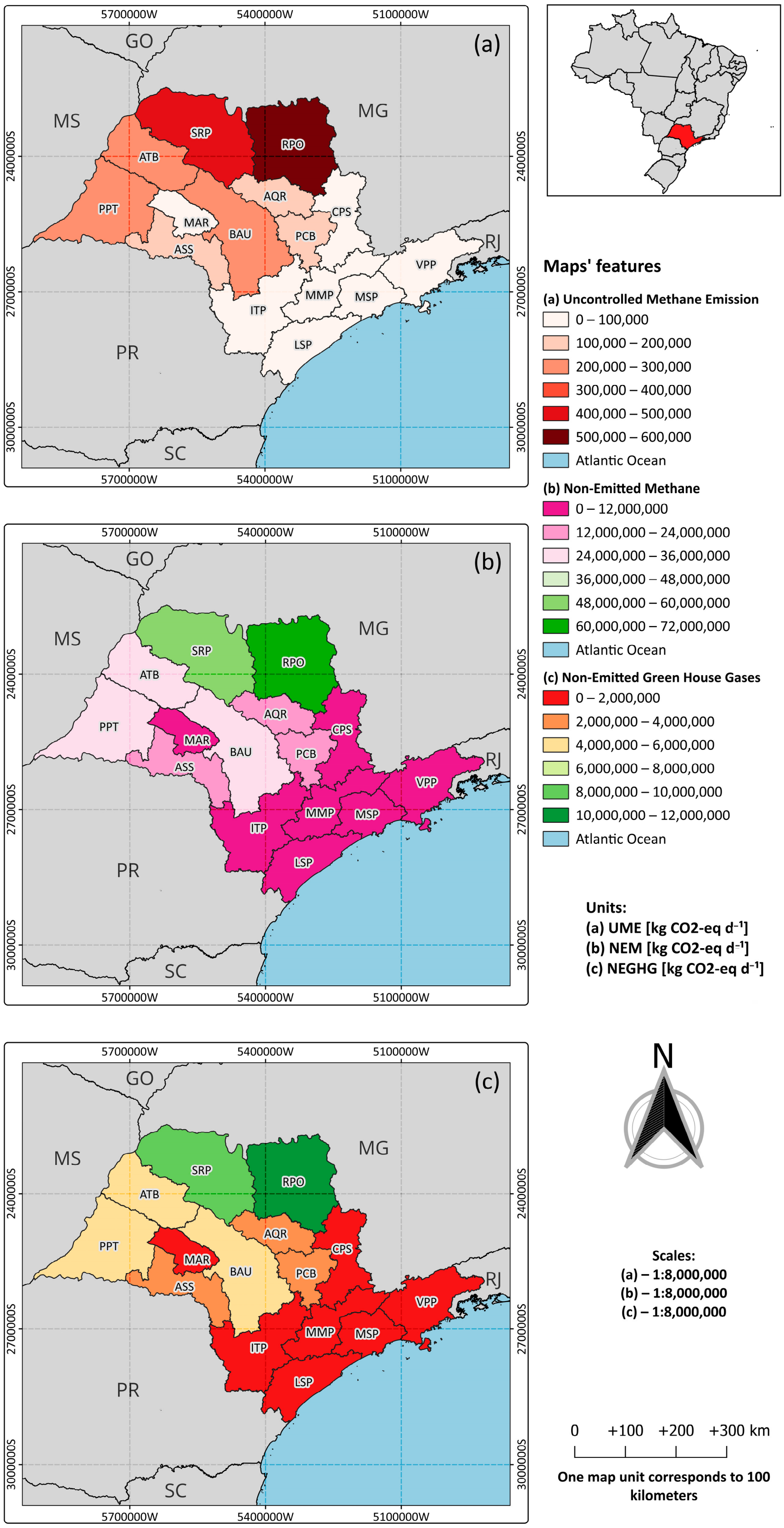

Figure 5. Environmental assessment: mapping of the (**a**) uncontrolled methane emission (UME), (**b**) non-emitted methane (NEM) and (**c**) non-emitted greenhouse gas (NEGHG) according to the mesoregions of the State of São Paulo. Mesoregions: ATB, Araçatuba; AQR, Araraquara; ASS, Assis; BAU, Bauru; CPS, Campinas; ITP, Itapetininga; LSP, Litoral Sul Paulista; MMP, Macro Metropolitana Paulista; MAR, Marília; MSP, Metropolitana de São Paulo; PCB, Piracicaba; PPT, Presidente Prudente; RPO, Ribeirão Preto; SRP, São José do Rio Preto; VPP, Vale do Paraíba Paulista.

Brazil is one of the main global sources of methane emissions related to organic residues from both urban and agricultural activities [55]. Considering this scenario, the country has been undergoing a transition to a carbon-neutral economy. The Zero Methane Plan stands out in this transition effort, representing the commitment of the Brazilian Federal Government, together with more than 100 countries, to globally reduce methane emissions by 30% by 2030 compared to 2020 levels. In a practical way, this plan will provide residue producers with an opportunity to transform residues into renewable energy and biofertilizer in order to generate income and operational savings while improving the environmental quality. In this way, the anaerobic digestion of vinasse emerges as a key strategy for reducing GHG emissions.

Finally, although not directly related to GHG emissions (NEM), sulfur-related air pollution may also become an important feature when dealing with vinasse-derived biogas. The high sulfate concentrations usually found in vinasse buildup in the form of highly corrosive and toxic sulfide in biogas after biological conversion [22] demand a removal step prior to the energetic exploitation of biogas. If not removed, sulfide is oxidized into sulfur dioxide (SO_2) in the prime movers (or flares), a precursor of acid rain events [56]. Considering a simple calculation, assuming the total BPR calculated for SP (18.0×10^6 Nm^3 d^{-1}) and a sulfide concentration of 2% in biogas [57,58], the release of 5.5×10^5 kg-H_2S d^{-1} would result in a total sulfur dioxide emission of 1.0×10^6 kg-SO_2 d^{-1}, which is of great interest to measure environmental impacts related to terrestrial acidification [20,59].

4. Conclusions

Investing in sugarcane vinasse biodigestion was demonstrated to be a key strategy to improve the offer of bioenergy and reduce GHG emissions within the context of the State of São Paulo. From an energetic perspective, the electricity generated from vinasse in combined-cycle plants has the potential to fully supply the residential consumption in the main sugarcane-producing mesoregions of the state, i.e., Ribeirão Preto, São José do Rio Preto, Bauru, Araçatuba and Presidente Prudente, corresponding to a population of more than 6.5 million inhabitants. In overall terms, about 25% of the total electric residential demand could be supplied by vinasse-derived electricity in São Paulo. When targeting biogas upgrading, almost 3.5 billion liters of diesel could be displaced by biomethane, representing a 26% abatement in the annual diesel consumption in the state, or 69% of the overall natural gas consumption could be replaced. The total amount of biomethane produced has the potential to roughly double the amount of sugarcane harvested in the state. Regarding environmental aspects, once produced, biogas has to be properly exploited, because potential GHG emissions related to methane production (261.2×10^6 kg-CO_2eq d^{-1}) in anaerobic reactors were estimated to be ca. 7-fold higher than the total emissions estimated for the entire ethanol production chain, as well as almost 6-fold higher than the emissions avoided with the replacement of diesel by biomethane. The State of São Paulo, or more precisely its northern and northwestern regions, is an extremely fertile ground for recovering energy from sugarcane vinasse. The adopted model could be the basis for approximately doubling the share of energy derived from vinasse in the Brazilian energy matrix, as the results presented here refer to just under half (45%) of the vinasse available in Brazil. Hence, exploiting sugarcane vinasse-derived bioenergy is a great opportunity to increase the share of renewable energy in the Brazilian energy matrix within the short to medium term. Economic assessments will be welcome to define the most suitable products, i.e., electricity or biomethane, for each region, taking into account local demands and the size of the biogas plant.

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