Multiscale analysis through the use of biomass residues and CO_2 towards energetic security at country scale via methane production

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Abstract

This work develops a systematic comprehensive comparison for the modelling of the renewable methane from different sources, such as lignocellulosic dry residues via gasification, anaerobic digestion of wet waste, and synthetic methane production from CO_2 captured and renewable electrolytic hydrogen using a multiscale approach. Initially, a techno-economic evaluation determines facility key performance indicators (KPI) and renewable energy sources. Then, a Facility Location Problem (FLP) identifies optimal facility locations. The decentralized use of lignocellulosic and wet waste, along with CO_2 captured from point and dilute sources is analyzed due to the high availability of the raw material and the transportation costs. The problem is formulated as a Mixed-integer linear programming (MILP), optimizing waste and CO_2 utilization, plant locations, PV panel surface areas, and wind turbine numbers across 356 shires in Spain. Lignocellulosic dry waste and point sources CO_2 capture using MEA are preferred. A sensitivity analysis reveals methane price ranges from 3.818 €/MMBTU to 30.229 €/MMBTU to 3.818 €/MMBTU to 13.837 €/MMBTU from 2022 to 2050. Considering carbon taxes, the most competitive price of 3.146 €/MMBTU is projected for 2050, competitive with current natural gas prices.

Keywords: DAC, CO_2 hydrogenation, electrolysis, green hydrogen, synthetic natural gas

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1. Introduction

The development of human activities, mainly related to the industry and transportation sector, has generated a set of intrinsic emissions, generally composed of CO_2, one of the most greenhouse gases (GHG) contributors to global warming. These emissions have experienced a dramatic increase, moving from around 300 ppm in the late 19th century to 425 ppm in 2024 (NASA, 2024), causing an estimated increase in temperatures of 1ºC. To prevent this, in 2015 the Paris Agreement was ratified by 195 countries in the world, where a global warming limit of 1.5°C was stipulated and emissions reductions of 55% were scheduled for 2030 (European Commission, 2020) and net zero emissions for 2050 through decarbonization policies. International cooperation is crucial for the energy and chemicals sectors. Recent European events, such as gas shortages due to the Ukrainian conflict affecting Nord Stream, and disruptions in shipping from the Red Sea conflict impacting Suez Canal routes, have caused significant global energy supply fluctuations. Despite discoveries like shale gas, Europe faces challenges in natural gas availability due to high consumption. Synthetic methane emerges as an important alternative, aligning with circular economy principles to reduce reliance on imported fossil fuels. There are two alternatives to capture CO_2 and produce synthetic natural gas (SNG), biomass growth and its capture from the air and other sources.

On the one hand, biomass has been employed to obtain energy and products, making possible the direct CO_2 capture from the atmosphere through the photosynthesis process to build new structures. Thus, from lignocellulosic dry waste biomass high value-added products have been obtained mainly employing gasification processes, such as bioethanol (Martín and Grossmann, 2011), DME (Peral and Martin, 2015), methanol, and biodiesel (Martín and Grossmann, 2014, 2017a). However, the growth of biomass from cultures presents some major issues, such as the high use of land, water, and fertilizer, and the costs of collection, transportation, and pretreatment. Other wastes generated, such as manure, MSW (municipal solid waste), and sludge present a large amount of moisture content, and they must be processed employing anaerobic digestion. However, manure can cause nutrient pollution, leading to the eutrophication of water bodies and soil deterioration if they are not properly treated (Menzi et al., 2009). Besides, in densely populated areas, where the production of MSW and sludge is an issue, the treatment of this waste is quite inefficient, losing almost 25% of these residues into landfills or incinerated (European Commission, 2021). These technologies offer means to convert organic waste into valuable resources, such as biomethane and biogas, thus promoting energy security and reducing fossil fuel dependence. On the other hand, industrially, carbon capture, utilization, and storage (CCUS) involve capturing CO_2 from different sources, then storing it in geological formations or using it for other uses (Beuttler et al., 2019; IEA, 2022a). Diverse capture methods are required depending on the CO_2 concentration in the source. The physisorption processes like pressure swing adsorption (PSA), temperature swing adsorption (TSA), and vacuum swing adsorption (VSA) use sorbent beds for CO_2 capture (Gupta and Li, 2022), although stability issues and energy-intensive regeneration require multiple beds for continuous operation (Leonzio et al., 2022a, 2022b; Ketabchi et al., 2023). In addition, the use of amine-based chemisorption with concentrated CO_2 sources such as steel and thermal power plants (Hasan, 2017), but struggle with dilute sources like air. Despite efforts, challenges persist with microalgae-based photobioreactors due to low yields (Yan et al., 2016; Zhu et al., 2016). Promising methods to capture CO_2 from diluted include direct air capture (DAC), where fans circulate air through an alkaline solution like KOH to enhance CO_2 dissolution. DAC can also be achieved through bipolar membrane electrodialysis (BPMED) (Sabatino et al., 2020), which separates CO_2 into ions. The captured CO_2, when combined with hydrogen produced via water electrolysis using PV panels and wind turbines (Martín, 2016a, 2017), can be utilized in chemical synthesis, notably methane or methanol.

Spain boasts a robust nationwide network and regasification infrastructure for natural gas distribution. Moreover, there is ample availability of residues and CO_2 from various sources, which can be harnessed to produce biomethane, biogas, and synthetic methane. (Taifouris and Martin, 2023) investigated biomethane and biogas production's potential to reduce Spain's reliance on fossil natural gas. They developed a network to evaluate operating costs, investment, facility scale, site selection, and waste management budget optimization within the circular economy framework (Floudas, 2016). Additionally, utilizing renewable energy for fuel synthesis (Martín and Grossmann, 2018), methanol production from CO_2 and hydrogen via water electrolysis (Martin, 2016a), and capturing CO_2 from the atmosphere with renewable energy for methanol production (Galán et al., 2023), along with methanol and methane production across Spain using CO_2 from various sources within a provincial-scale facility location problem (FLP) (Galán et al., 2024), enables a systematic comparison of technologies regarding waste utilization and CO_2 sources. There is no comprehensive analysis of the use of CO_2 in methane production to reduce the dependence on fossil fuels.

In this work, a comprehensive framework based on an extender FLP over 356 Spanish shires is presented, utilizing as raw materials lignocellulosic dry waste, manure, MSW, sludge, the CO_2 captured from point and diluted sources employing solutions based on monoethanolamine (MEA) and the conventional DAC process to produce methane. Additionally, hydrogen production via water electrolysis powered by renewables is also considered. This framework provides information on the optimal selection of the type, size, waste, location of the treatment and CO_2 capture installations, total operating and investment costs, the percentage of self-sufficiency of methane, and the production cost of the methane produced. The rest of the work is organized as follows: **Section 2** presents the methodology that covers the description of processes, waste utilization, CO_2 capture, renewable hydrogen, and energy production via PV panels and wind turbines, and methane production. The FLP is also presented. **Section 3** delves into the case study of Spain, spotlighting its constraints, limitations, and restrictions, and the characteristics of the dataset. **Section 4** reports the case study and summarizes findings on waste treatment plant locations, CO_2 capture, chemical production, and power generation technologies. It examines decentralized raw material usage for methane production and conducts a sensitivity analysis on technology prices over time. Finally, **Section 5**, discusses and summarizes the conclusions of this work.

2. Methodology

A comprehensive approach is proposed to evaluate the use of renewable resources and wastes such as lignocellulosic dry waste, manure, MSW, and sludge, alongside the capture of CO_2 from various sources to reduce the dependency on fossil fuels. This approach aims to identify optimal locations for these activities. Processes such as waste treatment, CO_2 capture, hydrogen electrolysis, and methane production are analyzed to gauge their efficiency and economic viability. The formulation of the FLP hinges on the decentralized utilization of waste and CO_2, leveraging energy from PV panels and wind turbines used in previous work (Galán et al., 2024)

The following sections describe the process-level analysis for utilizing lignocellulosic dry wastes, manure, MSW, and sludge, as well as capturing and utilizing CO_2. Subsequently, surrogate models are developed, and finally, an FLP is formulated considering decentralized CO_2 utilization on a national scale. The main text presents a general description of the processes used. The complete models, along with certain parameters and data sets, are found in the present work, and the Supplementary Material, based on the previous work by the authors (Galán et al., 2024), and (Taifouris and Martín, 2023).

2.1. Process level analysis

This section delineates the various technologies for waste management, along with the capture of CO_2 for subsequent conversion into methane via the power-to-x process. It describes waste treatment, CO_2 capture, energy generation, renewable electrolytic hydrogen, and methane synthesis.

2.1.1 Wastes treatment technologies

Gasification: A gasification process treats lignocellulosic dry waste, involving syngas upgrading, methane production, and biomethane refinement. It utilizes an indirect gasification system comprising a gasifier and a combustor (Sánchez et al., 2019) so that the combustion of char provides the gasification heat aided by olivine. Syngas composition is estimated via temperature correlations (Phillips et al., 2007), with upgrading employing cyclones, steam reforming, and a ZnO bed for hydrogen sulfide removal (León and Martín, 2016). Steam reforming follows equilibrium conditions (Roh et al., 2010), except for methane conversion. The water gas shift reactor (WGSR) adjusts the H_2/CO ratio for methane production, employing equilibrium models for temperature and syngas composition. Methanation is isothermal, capped at 773 K (Appl, 1999). Lastly, a PSA system reduces CO_2 to 2% and removes NH_3 and water (León and Martín, 2016), yielding pipeline-ready biomethane. Further details are summarized in the Supplementary Material and the work developed by Taifouris and Martín (2023).

Anaerobic digestion: An anaerobic digestion system is proposed for processing high-water-content waste, incorporating waste-specific information derived from León and Martín (2016) and Taifouris and Martin (2018). The model integrates physicochemical parameters of the waste to adjust gas distribution between gaseous (biogas) and liquid phases (digestate). The biogas undergoes purification utilizing an iron bed to remove H_2S and a PSA system to reduce CO_2, NH_3, and H_2O content, ensuring an acceptable biomethane composition. This purification process is detailed in the Supplementary Material and the work developed by Taifouris and Martín (2023).

2.1.2 CO_2 capture

Point sources: CO_2 capture from flue gas utilizes aqueous amine solutions, MEA, at low temperatures, 25-30°C and CO_2 pressures aver 0.05 bar, achieving 90-95% removal efficiency at 0.1 bar (Sánchez and Martín, 2021). A scrubber contacts amine solution with CO_2 gas, with flow rates dependent on solution concentration: 20% for MEA (GPSA 2012). Amine regeneration via distillation incurs solution losses, while refrigeration maintains operating efficiency. Exhaust gases typically contain 1-33% CO_2 (Wang and Song, 2020), affecting efficiency and cost; analysis assumes average MEA yield due to clustering. Further details can be found in the Supplementary Material and the work developed by Galán et al. (2024).

Diluted sources: CO_2 is captured from the air using conventional DAC technology (Holmes and Keith, 2012; Keith et al., 2018; Galán et al., 2023; Galán et al., 2024). The process involves fans and alkaline solutions based on KOH (Keith et al., 2018). KOH converts CO_2 into HCO_3^- and CO_3^{2-} ions through pH-influenced reactions, and it is recycled for CO_2 capture. $Ca(OH)_2$ regenerates the solution, forming $CaCO_3$, which is separated and dried with released heat. Steam generated can be used for heating or power. The process requires temperatures of around 900ºC and can utilize biogas as fuel to maximize efficiency. Further details are summarized in the Supplementary Material and the work developed by Galán et al. (2024)

2.1.3. Electrolytic hydrogen production

Green hydrogen is produced via water electrolysis with solar or wind power. The hydrogen stream, including water vapor and trace oxygen, undergoes purification through condensation and deoxygenation. Zeolite is used for hydrogen dehydration. An oxygen byproduct stream, with water vapor and residual hydrogen, is treated with condensation and zeolites. The oxygen is mainly for commercial sale.

2.1.4. Energy collection

Wind energy: Utilizing a GE 1.5sle turbine model (SAM) with a nominal power of 1,500 kW. The estimated installation and operating costs in 2022 are 1,600 €/kW (Stehly et al., 2018), accompanied by a cost of 0.026 €/kWh (Tovar-Facio et al., 2021). Turbine performance data are derived from (Davis and Martín, 2014a). It considers a number of wind turbines per shire in the range of 20 to 50 per shire, since they constitute the average values of the size of wind farms in Spain (see Supplementary Material). The CO2 emissions associated are accounting as 0.012 kgCO_2/kWhe (Martín and Grossmann, 2018).

Solar energy: PV panels, which harness solar energy, generate power. Each panel, covering an area of 4 m^2, produces 1 kWp. The panel efficiency is assumed to be 25% (η_{PV}). Expanding to 7 m^2 accommodates spacing requirements. The installation and operating costs in 2022 amount to 1,050 €/kWp (Fu et al., 2018) and 0.042 €/kWh (Tovar-Facio et al., 2021), with a measured CO_2 emission value of 0.048 kgCO_2/kWhe (Martín and Grossmann, 2018).

2.1.5. Methane synthesis

Methane production*:* Methane production consists of CO_2 methanation using renewable hydrogen with captured CO_2, similar to methanol synthesis. Operating within a pressure range of 1 to 30 bar and temperatures of 140ºC to 350ºC (Gassner and Marechal, 2009), developing an isothermal control of the H_2/CO/CO_2 mixture. Post-water removal, methane with a composition exceeding 95% is obtained, and for higher purity, additional purification steps may be necessary (Davis and Martín, 2014a, 2014b). Alternatively, syngas from biomass gasification is used as a raw material, performing the methanation of CO. Through anaerobic digestion biogas is generated, which just upgraded by CO_2 removal as described above (Taifouirs and Martín, 2023).

2.2. Process technologies surrogate modeling and process scale-up

Wastes treatment*:* Tailored processes are developed for each waste type, along with their corresponding CAPEX and OPEX. Up to 50 designs with varying treatment capacities are modeled, spanning the minimum and maximum sizes for manure and MSW of 63-368 kt/year and 20-53 kt/year, with yields waste to biomethane of 0.012 $kg_{Biomethane}$/kg, and 0.070 $kg_{Biomethane}$/kg, respectively. For lignocellulosic dry biomass, these capacities correspond to 10-820 kt/year, presenting a yield to biomethane of 0.285 $kg_{Biomethane}$/kg (Taifouris and Martín, 2023). Optimization aims at minimizing biomethane production costs.

CO_2 capture: To include the technologies at the country scale, yields, energy requirements, CO_2 availability from sources in every location, as well as linearized total capital expenditures or investment costs (CAPEX) and operating expenditures or operating costs (OPEX), represent the surrogate models MEA and conventional DAC capture (see Tables SP-9-SP-11 in the Supplementary Material).

To estimate OPEX, fixed and variable costs follow Sinnott's methodology (2005), excluding waste disposal expenses. Auxiliary costs (steam, water, energy) derive from process balances, while fixed costs (labor, maintenance, labs, depreciation, insurance) align with Sinnott. CAPEX estimation entails analyzing equipment sizing and costing initially (Tables SP-9-SP-11). Reactor economic assessment refers to Sánchez et al. (2019), while design correlations for compressors, heat exchangers, and fire heaters come from Cooper et al. (2005). Almena and Martín (2016) provide data for electrostatic precipitators, filters, and cyclones, and the digester design aligns with Taifouris and Martin (2018). Additional capital costs use a factorial method (Sinnott, 2005). Supplementary Material and Taifouris and Martin (2023) provide further details.

2.3. CO_2 utilization at country scale: Facility Location Problem (FLP)

2.3.1. Description of resources

An extended FLP is formulated to include the features of the different CO_2 capture technologies and utilization. The problem relies on the data for the availability of CO_2, wastes, and energy resources across the shires.

Wastes availability: The available quantities of waste are expressed in kt/year, differentiating between dry, lignocellulosic waste, from agricultural activities and subjected to gasification, and wet waste such as manure, MSW, and sludge, which come from livestock and human activities, which are subjected to anaerobic digestion.

CO_2 Availability: The CO_2 emissions are expressed in kt_{CO2}/year, coming from pointed and diluted from point and diluted sources. An issue with CO_2 data availability in certain shires has arisen during this work. Ideally, emissions data should include other chemicals like NO_2, SO_2, and organic volatiles, contributing to the greenhouse effect. Currently, only captured CO_2 is considered. The MEA solutions are mainly used over flue gas in point sources like thermal and steel industries, as well as refineries, while conventional DAC processes capture CO_2 directly from diluted sources such as atmospheric air, including emissions from agriculture, livestock, and transportation.

Sun and wind availability: The availability of sunlight is determined by the annual average value of direct solar irradiance in kW per m^2 (Global Solar Atlas). Therefore, a full year is used to calculate the energy generated by PV panels. Wind availability is measured using the annual average value of wind velocity in m per s (Global Wind Atlas).

Shire distribution: The available surface of each shire is limited to 2% of the total available area expressed in km^2. The approach considers 356 Spanish shires as possible locations for the facilities and the renewable power generation technologies, mainly focused on PV panels. The modelling allows for calculation and determinates the spatial distribution of the required facilities, PV panels, and wind turbine farms (Galán et al, 2024)

2.3.2. Formulation of the FLP problem

Each shire is determined by a representative point, using the latitude and longitude of the most representative city, typically the capital. Moreover, the availability of CO_2 captured from point and diluted sources is considered. The annual sun and wind availability, and the shire available area to locate the space for the PV panels.

The aim consists of maximizing CO_2 capture within spatial and energy constraints while minimizing costs. Key decisions include CO_2 source, utilization, and facility location. Sections cover CO_2 capture from point and diluted sources, hydrogen production, PV panels, and wind turbines, with synthesis product modeling. Surrogate models are developed for each process based on experimental and first-principles data. Complete modeling details are in the Supplementary Material.

3. Case study

3.1. General settings, restrictions, and assumptions of the case study

The FLP's implementation covers the 356 Spanish shires, including Ceuta, Melilla, Balearic, Canary, and Chafarinas Islands. Some areas like Alborán Island, Alhucemas, and Vélez de la Gomera rocks are excluded due to data scarcity or small size. It assesses the availability of raw materials like lignocellulosic dry waste, manure, MSW, and sludge, and CO_2 emissions from point sources such as thermal plants, refineries, and cement plants, among others, and diluted sources, primarily from the transport sector and livestock. The decentralized option is considered for raw material utilization, waste, and CO_2 usage. Based on the studies developed by Galan et al. (2024) and Taifouris and Martin (2023)

The problem is as a MILP structure, the first case consisting of 23,911 equations. 91,192 continuous variables and a total of 73,692 binary variables written in GAMS® (GAMS, 2021). The solver used has been CPLEX (IBM, 2021). The methodology and structure of the mathematical model can be used and adapted to the conditions of other areas and locations by updating and changing the data sets, waste availability, and CO_2.

The problem's constraints, primarily stem from data availability. Firstly, for waste, assumptions have been made about average compositions due to the significant variability inherent in different types of lignocellulosic dry wastes from crops, livestock activities generating manure, and the diverse sources contributing to MSW and sludge. From these residues biomethane and biogas can be obtained. To facilitate the analysis is considered that biomethane can be produced from all of them but takes into account that biogas is upgraded to biomethane. The methane produced from CO_2 captured will be considered synthetic methane. Secondly, data related to emissions from both point and diluted sources is limited to the year 2022 and early 2023 due to the lack of updated information. The flue gas concentration is not considered, since they can present values near 20% in the majority of the cases, they could vary. This aspect directly affects the absorption kinetic and the process. Moreover, the concentration of the flue gas is often considered as confidential information. Additionally, solvent losses are not accounted for in the modeling. Renewable energy generation from PV panels and wind turbines faces limitations in available parameters such as performance, CO_2 emissions, water and mineral consumption, and their evolution over the specified time horizon. The data available typically represents average values for each location, and more detailed data could lead to more precise results. Carbon taxes are considered to improve the profitability of the process due to the obtention of carbon credits.

3.2. Working data, parameters, variables, and evolution over the time horizon

The study's time horizon spans from 2022, 2030, and 2050, drawing upon the latest available data on lignocellulosic dry waste from various sources such as crops, manure, MSW, and sludge across the shires. These biomass resources are intricately linked with demographic shifts and population dynamics. Over a 30-year horizon, Spain's population is projected to remain within the range of 50 to 60 million people (MITECO, 2023), closely resembling current figures. Consequently, the annual supply of these raw materials for biomethane and biogas production is considered stable.

With the availability of CO_2 from point sources, data were considered directly from operational facilities in each shire of Spain. Despite the closure of the thermal plants due to the decarbonization policies, the specific closure sequence of the rest of the facilities is not known. Therefore, invariant emissions are considered for the analysis of the study (see specifications in section 4). The analysis accounts for the aggregate CO_2 emissions from point sources, allowing flexibility in the model to consider simultaneously CO_2 utilization with biomass, thus not exhausting the entire available supply. For CO_2 emissions from diluted sources, information on population and associated emissions from Spanish Autonomous Communities were collected and distributed based on population density. Challenges emerged concerning population data granularity, especially in regions like Catalonia, Valencia, and the Basque Country. Population Figures were derived at the municipal level and subsequently aggregated into shires. The National Institute of Statistics of Spain (INE) provided population and area data for municipalities, while the National Geographic Institute of Spain (IGN) furnished geographical coordinates. Data on direct solar irradiance and wind patterns at 100-meter altitudes were taken from the Global

Solar Atlas (Global Solar Atlas) and Wind Global Atlas (Wind Global Atlas), respectively. Utilizing average values and accounting for consistent climatic conditions over time, the analysis adopts a seasonal periodicity to facilitate comprehensive evaluation.

The data of the proportions required of CO_2, water for electrolysis, produced H_2 and O_2 in the methane production, and the energetic requirements needed for develop CO_2 capture though MEA and DAC, water electrolysis, and methane synthesis are provided (Tables SP-12, SP-14, and SP-8). The gasification and anaerobic digestion are energetically self-sufficient. Additionally, carbon taxes used for evaluating the techno-economic aspects and enhancing profitability are summarized in Section 4 of the Supplementary Material. Carbon taxes applied in Spain experiment a variation over the time horizon, taking the values of 80.87 €/t_{CO2} in the year 2022 (SENDECO2, 2022), 150 €/t_{CO2} in the year 2030 (Blazquez et al., 2021; GMK Center, 2023) and 180 €/t_{CO2} in the year 2050 (ETUI, 2022) (Table SP-15), due to the tightening of environmental policies.

Parameter values and data are provided in both the main text and Supplementary Material through Figures and Tables. Additionally, an Excel file with the sources and processed data is available for reference and further analysis.

4. Results and discussion

The section is organized following the decentralized utilization of lignocellulosic dry waste, manure, and MSW, along with captured CO_2 for methane production. As it was mentioned in the sections above, the decentralized utilization of raw materials constitutes the most profitable option, selecting it as the optimal solution. Moreover, it comprises subsections dedicated to analyzing production based on two factors: the variation in annual cost as a percentage of the budget allocated by MITECO (MITECO, 2020), which amounts to 10,195 M€, and the variation in the available quantity of lignocellulosic material. Thus, for this study, the total amounts of CO_2 considered are 93,327 ktCO_2/year and 102,250 ktCO_2/year, from point and diluted sources respectively. These values differ from the data provided by Galan et al. (2024) due to a reduction in the emission policies, the closure of several industries, and the reduction of emissions in the transport sector. Concerning the lignocellulosic dry wastes, manure, MSW, and sludge, the available amounts reach up to 29,900 kt/year, 121,400 kt/year, 9,500 kt/year, and 3,400 kt/year, respectively.

It considers that these available amounts of residues will remain constant in the analysis for the time horizon described below. The main reason is based on the growth of the population, which will experience a small increase over the next 30 years (MITECO, 2023). It will maintain similar values of the manure, MSW, and sludge in the year 2022. The CO_2 availability remains constant over a time horizon taking the data from the year 2022, since the subsequent data and plant closure due to the environmental policies are not well-known. However, due to the projected CO_2 emissions reduction, the availability of CO_2 in the year 2022 would be higher than the year 2030, and 2050, guaranteeing enough availability of CO_2 for the processes.

4.1 Study of the variation of annual cost (% budget)

This section presents a case study involving the utilization of 100% of the available budget as the maximum annual cost (Figures 1 and 2, SP-2- SP-5 in the Supplementary Material). This annual cost is composed of the total OPEX and a fraction of the total CAPEX, 0.05, considering the amortization time of 20 years. Additionally, a sensitivity analysis is conducted across various percentages of annual cost availability, ranging from 5% to 300% (Tables SP-18-SP-23 in the Supplementary Material). In addition, the effect of the carbon tax as a credit on the final price of SNG is also evaluated.

4.1.1 Analysis of current state: Year 2022

Use of the different wastes, CO_2 capture, and use: From the results without carbon tax are presented. Biomethane is produced using lignocellulosic dry biomass, manure, and MSW, 26,702 kt/year, 83,070 kt/year, and 7,775 kt/year, respectively. The investment and operating costs for these waste treatment methods to produce biomethane amount to up to 11,174 M€, with operating costs totaling 5,341 M€/year (Table SP-16 in the Supplementary Material).

The utilization of the most representative case using various waste streams (Figures 1 a, 1 b, and 1 c) is described. Lignocellulosic dry waste is selected due to its dry nature, presenting higher yields during processing (Taifouris and Martín, 2023). This waste primarily originates from shires surrounding major Spanish rivers such as the Duero in the west, the Ebro River in the east, and notably the Guadalquivir in the southern region (Figure 1 a), using around 800 kt/year per shire. Manure, primarily sourced from livestock farms scattered across the peninsula, yields lower biomethane outputs, around 2,750 kt/year per shire, significantly less than lignocellulosic dry waste (Taifouris and Martín, 2023). MSW from urban centers, particularly in the central region including Madrid, contributes substantially to biomethane production, approximately 1,790 ktCH_4/year per shire. Notably, shires with higher concentrations of manure, such as the northwest and bordering areas with Portugal, notably Extremadura, are depicted in Figure 1 b. Sludge residue, due to its high moisture content, is unsuitable for biomethane production (Taifouris and Martín, 2023). The source of CO_2 is mainly from point sources, capturing adds up to 5,858 ktCO_2/year (Table SP-16 in the Supplementary Material) notably lower than the values of 10,000 ktCO_2/year and 12,500 ktCO_2/year summarized in (Galán et al., 2024). This discrepancy is mainly attributed to a finer granularity in the study areas and the incorporation of biomass also as a raw material for methane production. The investment cost associated with CO_2 capture is 62 M€, resulting in an operating cost of 256 M€/year (Table SP-16 in the Supplementary Material). The use of MEA solutions is favored over conventional DAC (see Figure 1 d) due to their lower investment cost (see Table SP-9 in the Supplementary Material). They are distributed heterogeneously, predominantly in regions like the Northern Third, targeting areas in the north of Cantabria and the Balearic Islands in the Mediterranean Sea, with less prevalence in inland shires of the peninsula. These selections are primarily influenced by the presence of steel and petrochemical industries.

If carbon taxes are applicable, the solution changes, resulting in a different waste. The total amount of lignocellulosic dry waste adds up to 26,700 kt/year, only composed from lignocellulosic dry waste (Table SP-16). This behavior is shown in Figures 2 a, 2 b, and 2 c. The use of lignocellulosic dry waste remains consistent, while manure and MSW are phased out for biogas production due to their high annual cost, equivalent to 100% of the budget. Focusing solely on lignocellulosic dry waste reduces both investment and operating costs significantly to 6,471 M€ and 2,336 M€/year respectively (Table SP-16 in the Supplementary Material). This translates to a significant decrease of 42.09% and 56.25% in costs compared to using this waste type without considering the carbon tax. MEA solutions allow capturing from 5,858 ktCO_2/year to 9,899 ktCO_2/year, marking a 1.69-fold increase. However, the higher CO_2 capture results in greater investment and operating costs, escalating to 106 M€ and 433 M€/year, respectively, representing a 68.98% increase compared to scenarios without a carbon tax (Table SP-16 in the Supplementary Material). MEA solutions' usage has risen due to strict EU environmental regulations, which hold companies accountable for CO_2 emissions, subjecting them to taxation. The carbon taxes vary by country (Table SP-15). This process effectively prevents CO_2 emissions, making it economically viable and lowering synthetic methane production costs. MEA solutions (Figure 2 d) are favored over conventional DAC due to lower costs and carbon tax incentives. CO_2 capture via MEA is prevalent in industrial shires, shifting from the Balearic Islands to southern Spain near the Strait of Gibraltar, home to petrochemical and biofuel industries.

Production of renewable energy: To run the system, energy is required. Initially, an assessment is conducted on the surface area required for the installation of PV panels, the number of wind turbines, as well as the corresponding investment and operating costs. The total surface area dedicated to PV panels is 147 km^2 (Table SP-17 in the Supplementary Material), which represents around 1.46% of the total surface area available for the installation fo PV panels in Spain (refer to section 2.3.2 of the Supplementary Material). In addition, the total number of wind turbines installed reaches 2,465 (Table SP-17 in the Supplementary Material). In scenarios with moderate energy demands, wind turbines are preferred over PV panels due to lower operating costs (see Table SP-11). However, as energy needs increase PV panels become the technology of choice due to lower investment costs (see Table SP-11). This trend strengthens with decreasing renewable energy costs, limited only by available surface areas. The installation of PV panels and wind turbines represents an investment and operating cost of 28,020 M€, and 2,430 M€/year, respectively (Table SP-17 in the Supplementary Material), generating 7.3 GW, which approximately represents the power generated in Spain by the 7 nuclear reactors, corresponding to more than 6% of the electricity production, 119.2 GW at the end of 2022 (Red Eléctrica, 2022).

On the one hand, the PV panel distribution predominantly spans across the southeastern region of the peninsula, encompassing the shires of the Almería province, and extends to the Canary Islands, where direct solar irradiance values peak, achieving a maximum of 37 km^2 per shire. On the other hand, wind turbine energy generation highly depends on wind speeds across shires. Consequently, their location is preferred in coastal shires, such as the Strait of Gibraltar, the Northern Third, selected areas of the Mediterranean Sea, and mountainous interiors of the peninsula (Figures 1 e and 1 f, Table SP-17 in the Supplementary Material)

However, the use of carbon tax allows the introduction of certain variations in energy production. As described above, the use of carbon taxes increased the capture of CO_2 using MEA solutions. impacting on the energy production. The number of wind turbines installed is 2,465 (Table SP-17 in the Supplementary Material). On the contrary, the surface of PV panels increases from 147 km^2 to 290 km^2 (Table SP-17 in the Supplementary Material), which represents an increase of 96.52% of the surface, evidencing the selection of PV panels preferably for power generation. In addition, the increase in energy production represents an increase in the investment and operating cost, 49,355 M€, and 4,282 M€/year (Table SP-17 in the Supplementary Material), respectively. This corresponds with an increase of 76.14% and 76.21% to the previous case, respectively, also generating 12.4 GW, a 68.98% compared to the previous case, accounting for 10.37% of the total power production in Spain (Red Eléctrica, 2022). PV panels and wind turbine distribution are maintained (Figure 2 e and f). Only PV panels experiment with some modification, with a surface area increase in the southeastern regions of the peninsula.

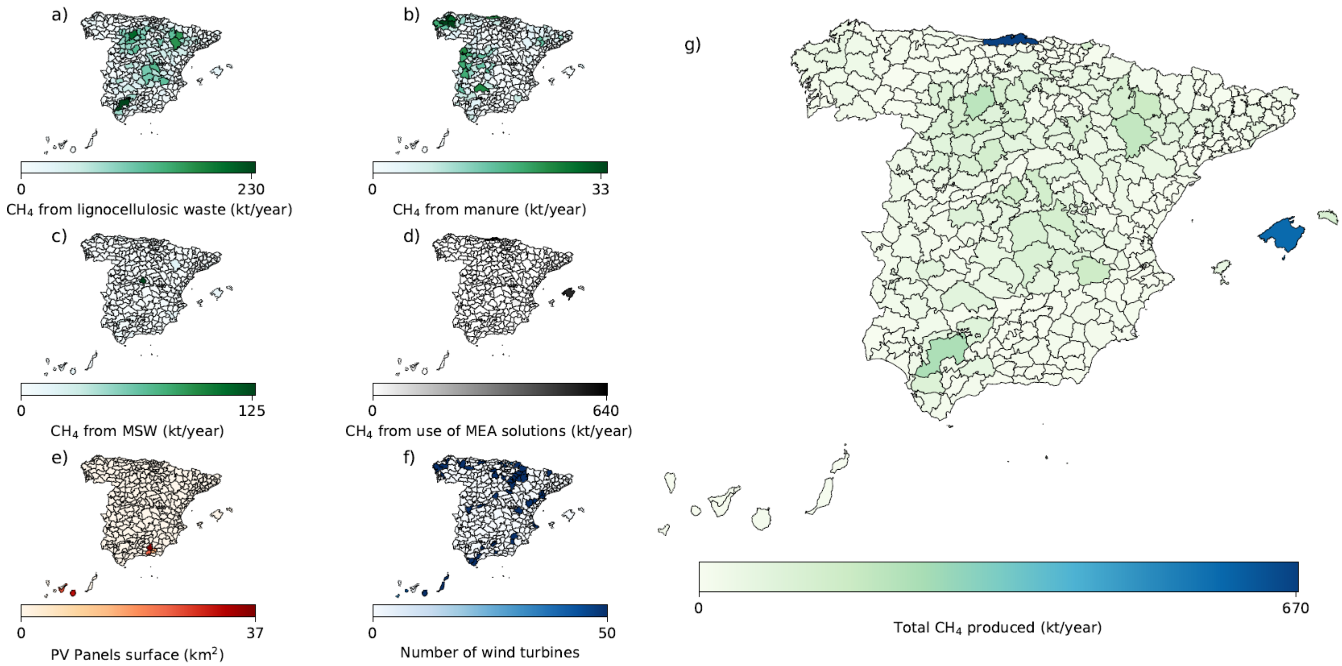

Fig 1. Distribution of the use of lignocellulosic dry waste (a), manure (b), MSW (c), sludge (d), renewable energy generation (e, f), and methane production (g) in Spain by shires in the year 2022 without carbon tax.

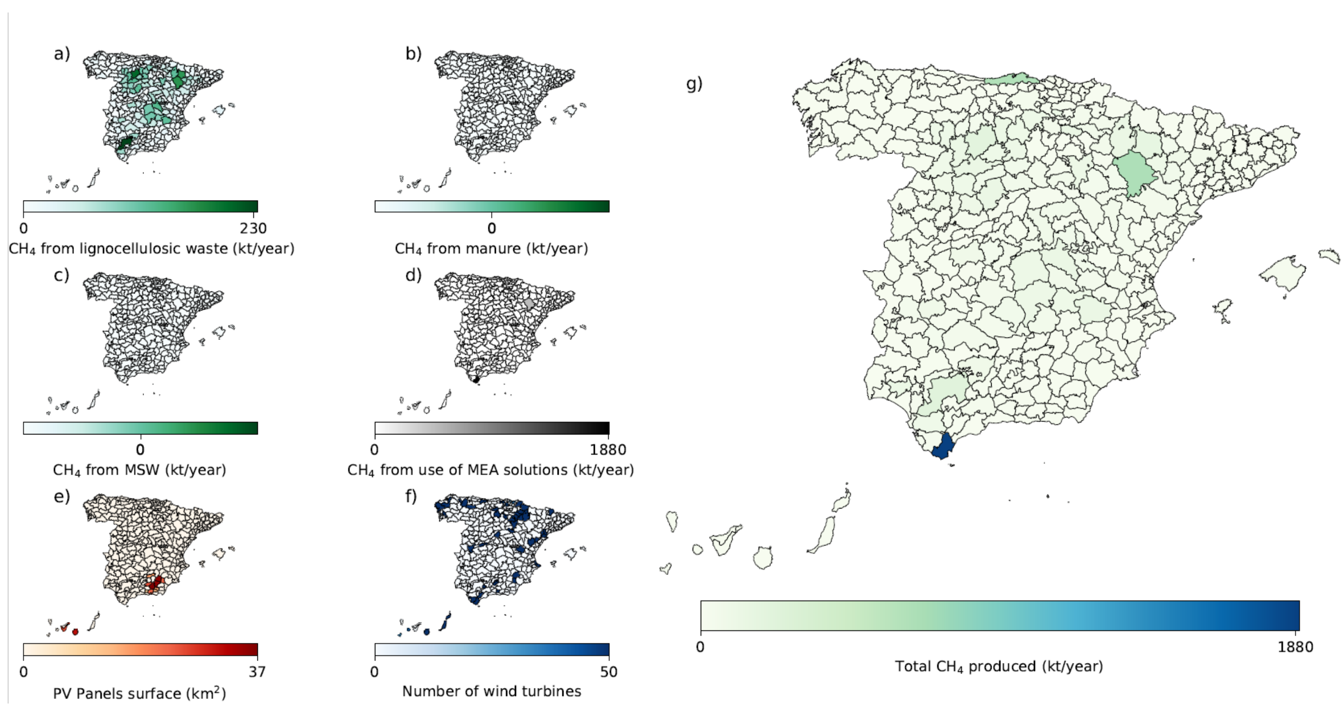

Fig 2. Distribution of the use of lignocellulosic dry waste (a), manure (b), MSW (c), sludge (d), renewable energy generation (e, f), and methane production (g) in Spain by shires in the year 2022 with carbon tax.

Methane production: As described in (Galán et al., 2024), methane production consumes 3 kg_{CO2}/kg_{CH4}, and 0.41 kg_{H2}/kg_{CH4} (see Table SP-12 in the Supplementary Material), requiring substantial carbon capture and production of H_2. First, the total production of biomethane without considering carbon taxes reaches up to 9,151 kt/year, corresponding with 7,610 kt/year from lignocellulosic dry waste, and 997 kt/year and 544 kt/year from manure and MSW, representing 31.5%, 4.1%, and 2.3% (Table SP-16) of the total methane produced to the total methane consumed in Spain for the year 2022 (Taifouris and Martín, 2023). Their costs are 11,174 M€, and 5,341 M€/year (Table SP-16). On the contrary, the generation of synthetic methane adds up to 1,953 kt/year, consuming 805 ktH_2/year and producing 8.1% of the methane to the total methane consumed, showing an investment and operating cost of 3,202 M€ and 304 M€/year (Table SP-16 in the Supplementary Material). Considering that their productions are combined, the total renewable-based methane production reaches 11,104 kt/year, requiring 206 treatment plants for lignocellulosic dry waste. From manure and MSW, the plants installed are 235 and 152, respectively (Table SP-16). Accounting for both methane production pathways and CO_2 capture, the investment and operating costs are 14,440 M€ and 5,900 M€/year. Lignocellulosic dry waste is the best choice as an initial residue due to its biomethane yield (Table SP-18). Following this, CO_2 capture with MEA solutions is the next preferred due to cost factors and CO_2 availability, prioritizing it over manure and MSW (Table SP-9 in the Supplementary Material). The total methane production (Figure 1 g) follows the resource consumption pattern observed in Figures 1 a, 1 b, 1c, and 1 d. Biomethane production, mainly from lignocellulosic dry waste, manure, and MSW is concentrated in the peninsula's interior and the Northern Third regions. Meanwhile, CO_2 captured from MEA solutions disperses across the Northern Third, particularly in areas like the north of Cantabria and the Balearic Islands in the Mediterranean Sea. Maximum methane production values reach 670 $ktCH_4$/year per shire.

As discussed above, implementing the carbon tax affects waste utilization, electricity generation, and methane production. The maximum biomethane production peaks at 7,610 kt/year, exclusively from lignocellulosic dry waste, maintaining the same methane production as the methane consumed in Spain, 31.5%, needing 206 treatment plants, and 6,471 M€ and 2,336 M€/year of investment and operating costs, respectively (Table SP-

16). Furthermore, synthetic methane production amounts to 3,300 kt/year, representing a 68.98% increase compared to the previous scenario, increasing its methane production the produced from 8.1% to 13.7%, consuming 1,361 ktH_2/year, presenting 5,412 M€ for investment and 515 M€/year for operating costs (Table SP-16 in the Supplementary Material), representing a 68.98% increase compared to the previous scenario. The total methane production reaches 10,910 kt/year, presenting 11,989 M€ and 3,284 M€/year for investment and operating costs considering both methane production pathways and CO_2 capture, as described above. This cost experimenting with a reduction of 17%, and 44.34%, respectively. Figures 2 a, 2 b, 2 c, 2 d, and 2 g summarize the results with carbon taxes. Biomethane production is sustained through biomass gasification, maintaining consistent maximum output at 230 kt/year per shire, discarding manure and MSW as an effect of carbon taxes, and employing an annual cost equivalent to 100% of the budget. MEA solutions improve CO_2 capture, driven by carbon taxes, yielding peak values of 1,880 ktCH_4/year per shire. As in the previous scenario, lignocellulosic dry waste is mainly selected for biomethane production, followed by CO_2 capture using MEA solutions. In addition, manure no longer serves as a raw material, and MSW is utilized instead, prioritizing CO_2 capture using MEA solutions (Table SP-19 in the Supplementary Material).

Evolution of self-sufficiency methane percentage, and methane prices: The self-sufficiency of renewable-based methane percentage is defined as the percentage of methane produced with respect to the methane consumed in the absence of the carbon tax, the progression of self-sufficiency methane percentage (blue line) for the year 2022 is illustrated in Figures 3 a, and 5 (Table SP-18 in the Supplementary Material). Initially, up to an annual cost equivalent to 20% of the budget, lignocellulosic dry waste emerges as the primary raw material due to its superior performance in biomethane production (Taifouris and Martín, 2023). The evolution of self-sufficiency methane percentage demonstrates a curve-like behavior during this phase. However, from an annual cost of 25% of the budget onwards, alternative technologies are adopted to complement biomethane production. MEA solutions for CO_2 capture are introduced, resulting in a 0.6% self-sufficiency methane percentage at this point, while lignocellulosic dry waste's contribution increases from 11.7% to 30.7% (Figure 3 a, 5, and Table SP-18 in the Supplementary Material). Progressing further, from an annual cost of 25% of the budget to 300%, the use of manure and MSW becomes evident, keeping their self-sufficiency methane percentage values of 4.21% and 2.26%, respectively. Lignocellulosic dry biomass maintains its contribution at 31.5% up to 300% of the budget, while MEA solutions continue increasing, achieving 42.9% self-sufficiency methane percentage (Figure 3 a). This trend modification is attributed to the availability of CO_2 from point sources and the more competitive costs. Consequently, the combined contribution of all technologies reaches 80.9% of the self-sufficiency methane percentage, changing the trend to a straight line. However, although the maximum annual cost rises 300% of the budget, it is possible to determinate it for a self-sufficiency methane percentage of 100%. Figure 6 shows that budget, 410.4 %. The contribution of the wastes remains the last behavior but varying the value of self-sufficiency methane percentage from 42.9% to 61.9%, 19%. The methane price evolution (black line) is shown in Figures 3 a, and 5 (see Table SP-18 in the Supplementary Material). A double price axis is plotted, one of them in €/MMBTU and another for its equivalence, in €/MWh. When methane production only relies on lignocellulosic dry biomass, the cost ranges from 3.818 €/MMBTU to 6.047 €/MMBTU (13.028 €/MWh to 20.616 €/MWh), displaying nearly linear growth. From the annual cost of 25% to 50% of the budget the introduction of MEA solutions results in a methane price evolution from 7.108 €/MMBTU to 12.088 €/MMBTU (24.254 €/MWh to 41.246 €/MWh) (Table SP-18), increasing a 70% in this range. From 50% to 75% of the budget, the prices increase from 12.088 €/MMBTU to 16.082 €/MMBTU (41.246 €/MWh to 54.874 €/MWh) experimenting with an increasing of 33%. A further increasing of 20.45% is obtained when the annual cost is 100% of the budget, showing a methane price of 19.371 €/MMBTU (66.103 €/MWh) at this stage. The reduction in the methane price follows a convex curvilinear trend, resulting in progressively smaller increases within each interval. Thus, at an annual cost of 300% of the budget, the methane price reaches 33.054 €/MMBTU (112.785 €/MWh), experiencing a 7.14% price increase. Finally, for a self-sufficiency percentage of 100%, this price reaches 36.543 €/MMBTU (124.69 €/MWh) (Figure 6 a)

The impact of implementing carbon tax for the year 2022 is presented in Figures 4 a, and 5 (see Table SP-19 in the Supplementary Material). The observed patterns are similar to the previous case, but the implementation of a carbon tax expedites CO_2 capture, particularly through MEA solutions. It is due to the obtention of carbon credits, an extra income makes that process more profitable and competitive. In this case, for an annual cost of 15% of the budget, lignocellulosic dry waste remains the primary raw material due to its superior biomethane production yield (Taifouris and Martín, 2023), maintaining a methane self-sufficiency percentage of 31.5%. Manure is discarded, employing instead MSW, which starts at an annual cost of 125% of the budget (Figure 4 a and Table SP-19 in the Supplementary Material), resulting in a methane self-sufficiency percentage of approximately 2.1%, marking a 7% reduction compared to the previous scenario. Meanwhile, the production of synthetic methane via MEA solutions growths owing to the application of carbon taxes, peaking itself at 46.8% of methane self-sufficiency percentage for an annual cost of 300% of the budget, representing a 9.09% increase with respect to the case above, showing an overall methane self-sufficiency percentage of 80.4%, reducing only a 0.62% reduction with respect the case above. To achieve that it is necessary to employ 3 times the budget provided by MITECO, 10,195 M€. This use becomes 4 times to achieve a 100% methane self-sufficiency percentage (Figure 6 b), moving from 46.8% to 62.4%, increasing 0.81% concerning the case above. Moreover, the methane price evolution (Figures 4 a, 5, and Table SP-19 in the Supplementary Material) follows a similar behavior to the previous scenario, with a notable reduction in prices attributed to the use of carbon taxes. Specifically, when CO_2 capture is conducted via MEA solutions (Table SP-18-SP-19 in the Supplementary Material). Comparing both cases, consistent prices ranging from 3.818 €/MMBTU to 5.141 €/MMBTU (13.028 €/MWh to 17.542 €/MWh) for annual costs between 5% and 15% of the budget. However, at an annual cost representing 20% of the budget, the price decreases from 6.047 €/MMBTU (20.633 €/MWh) without carbon taxes to 6.042 €/MMBTU (20.616 €/MWh) when they are applied (Tables SP-18- SP-19 in the Supplementary Material), representing a decrease of 0.08%. These prices are comprised into the feasible range from 3 €/MMBTU to 7 €/MMBTU (10.236 €/MWh to 10.236 €/MWh) with a target price of 5 €/MMBTU (17.06 €/MMBTU) (Figure 5), considered the feasible region to produce methane with a competitive price. The reduction between both situations continues until reaching 8.55% when the annual cost accounts for 300% of the budget, resulting in a methane price of 30.229 €/MMBTU (103.146 €/MWh) (Figure 5 and Table SP-19 in the Supplementary Material), reducing it an 8.55%. This reduction becomes 10.35% when the cost of 33.359

€/MMBTU (113.826 €/MWh) is obtained from the use of an annual cost of 410.5% of the budget achieving 100% of methane self-sufficiency,

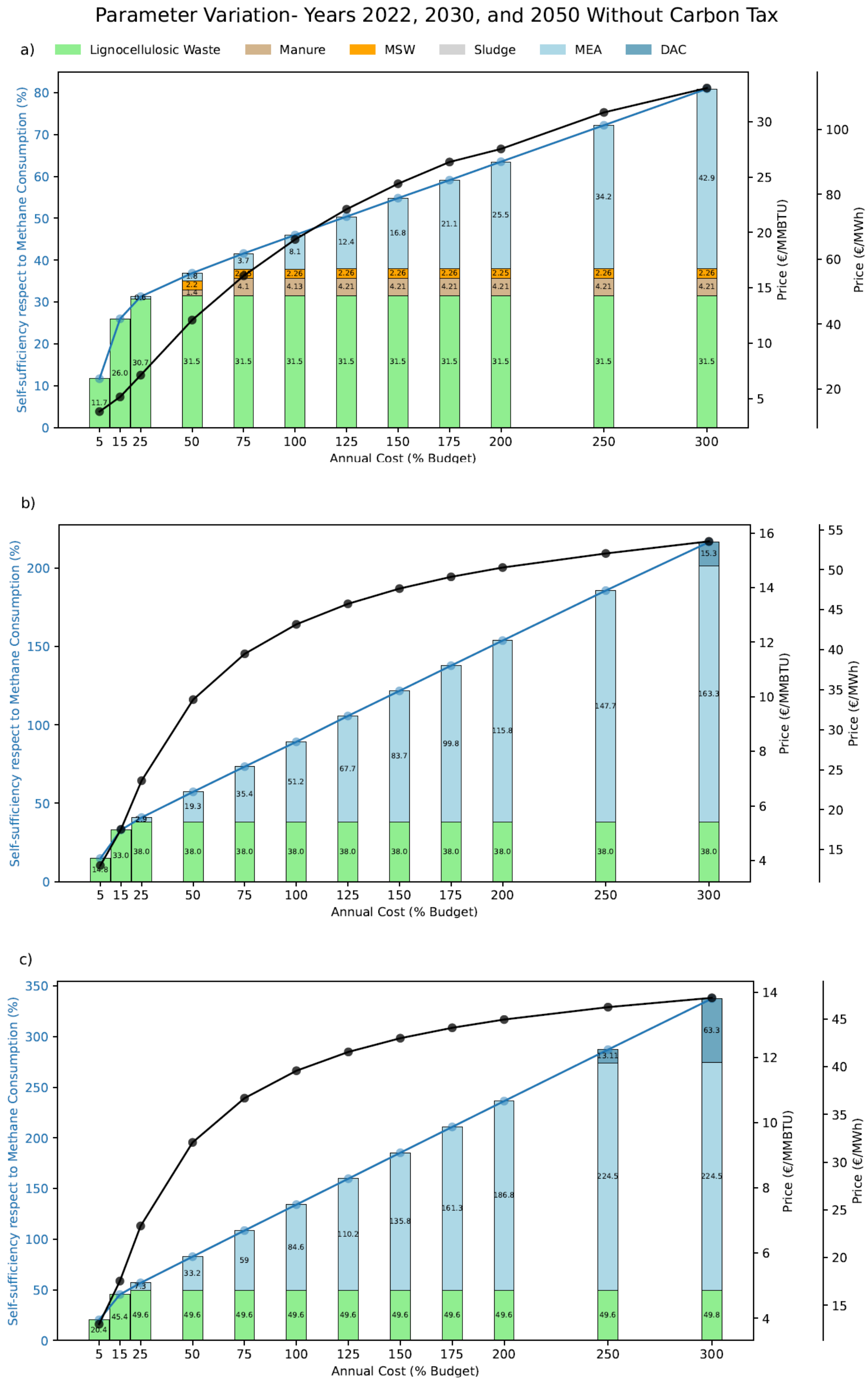

Fig 3. Evolution of the self-sufficiency percentage, and the methane price with respect to the annual cost for the years 2022 (a), 2030 (b), and 2050 (c) without carbon tax.

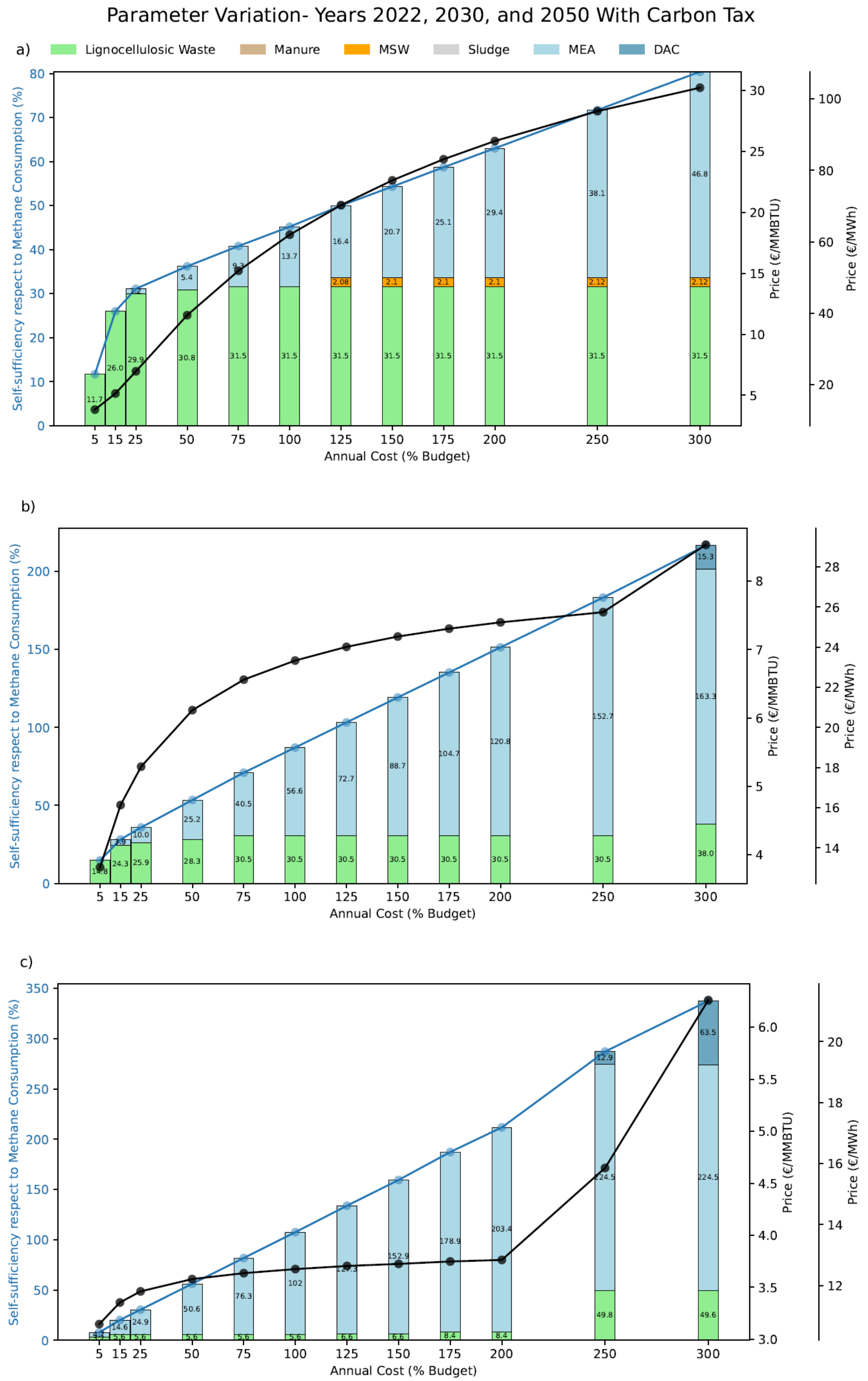

Fig 4. Evolution of the self-sufficiency percentage, and the methane price with respect to the annual cost for the years 2022 (a), 2030 (b), and 2050 (c), with carbon tax.

Based on these results, there is a reduction in manure and MSW use, favoring the production of synthetic methane using MEA solutions with a carbon tax. Thus, experimenting with a reduction in methane price with a reduction in the methane self-sufficiency percentage represents a reduction in methane production. Less methane is produced, but cheaper, due to carbon tax incentives (Table SP-19 in the Supplementary Material). This effect will be more pronounced as costs decrease due to improvements in technologies, in the time horizon of the years 2030 and 2050.

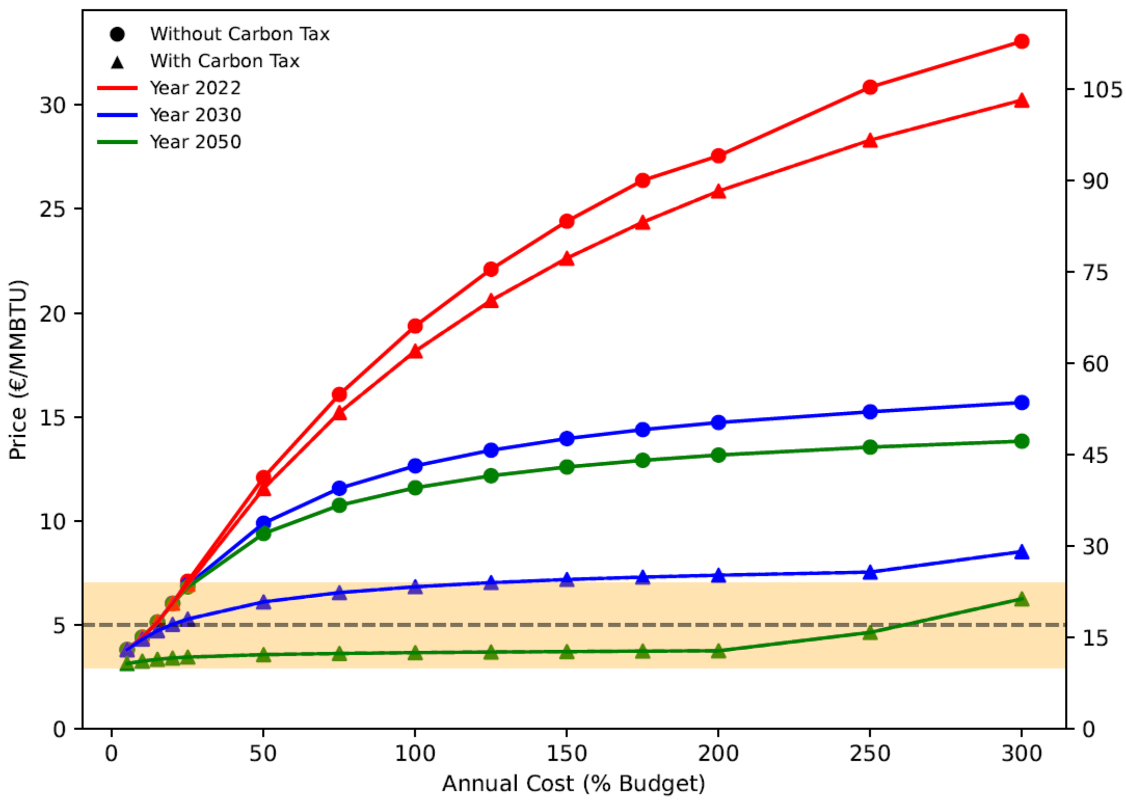

Fig 5. Comparative evolution of the methane price with respect to the annual cost for the years 2022, 2030, and 2050 without and with carbon tax.

Environmental impact of energy generation: CO_2 emissions, minerals required, and water use. Renewable energy sources, PV panels, and wind turbines, present significant environmental impacts throughout their lifecycle, including production, maintenance, and associated emissions (Table SP-17 in the Supplementary Material).

Emissions linked to PV panels add up to 2,325 ktCO_2/year, while those from wind turbines amount to 188 ktCO_2/year, both representing 1.19% and 0.10% of the total CO_2 emissions in Spain. Despite this, PV panels are favored for power generation mainly due to the costs, thus contributing significantly to CO_2 emissions from renewable sources. In addition to operational and maintenance emissions, those from mineral extraction for PV panels and wind turbine production must be factored in. The primary minerals required in their manufacture, include Cu, Ni, Mn, Cr, Mo, Zn, Rare Earth, and Si along with associated CO_2 emissions from extraction (IEA, 2022b; KU Leuven, 2022; Worldsteel, 2023). The total mineral requirements comprise 20,783 tCu, 726 tNi, 1,392 tMn, 839 tCr, 177 tMo, 9,981 tZn, 25 tRare Earth, and 21,836 tSi, resulting in CO_2 emissions of 321 kt_{CO2} and 86 kt_{CO2} for PV panels and wind turbines, respectively Their combined emissions amount to 2,646 ktCO_2 and 273 ktCO_2, representing 45.17% and 4.67%, respectively, both nearly representing around 50% of the CO_2 emissions captured using MEA solutions, 5,858 ktCO_2/year (Tables SP-16-SP-17 in the Supplementary Material). Furthermore, water requirements for maintaining and cleaning PV panels and wind turbines are determined by ratios of 0.33 L/kWh$_e$ and 0.043 L/kWh$_e$, respectively (Jin et al., 2019), as well as water usage in renewable electrolytic hydrogen production. Total water consumption for operating and maintaining PV panels and wind turbines, accounting for $1.6\cdot10^7$ m^3/year and $6.7\cdot10^5$ m^3/year, representing 0.028% and 0.001%, respectively, of Spain's total water storage capacity, 56,07 hm^3 (Embalses.net, 2023). Water dedicated to renewable electrolytic hydrogen production is $7.3\cdot10^6$ m^3/year (Figure SP-1 and Table SP-17 in the Supplementary Material), equivalent to 0.013% of Spain's total water reserves.

The carbon taxes drive CO_2 capture via MEA solutions, incentivizing expanded PV panel deployment for increased power production. However, this expansion escalates CO_2 emissions from the operation, cleaning, mineral consumption, and water usage. Total mineral requirements amount to 35,022 tCu, 731 tNi, 1,392 tMn, 839 tCr, 177 tMo, 10,133 tZn, 25 tRare Earth, and 41,758 tSi, with associated CO_2 emissions of 604 ktCO_2 and 86 ktCO_2 for PV panels and wind turbines, respectively. However, Mn, Cr, Mo, and Rare Earths show consistent usage exclusive to wind turbine production, with identical quantities. Combined emissions from PV panels and wind turbines total 5,051 ktCO_2 and 273 ktCO_2, representing 2.58% and 0.14% of the total CO_2 emissions in Spain. These values change to 51.025% and 2.762%, respectively, employing the CO_2 emissions captured by

MEA solutions, 9,899 kt_{CO2}/year (Figure SP-1 and Table SP-17). Water consumption increases with PV panel utilization, while usage by wind turbines remains steady. This results in $3.06 \cdot 10^7$ m^3/year and $6.7 \cdot 10^5$ m^3/year, equivalent to 0.055% and 0.001% of Spain's total water storage capacity (Embalses.net, 2023). Water employed for renewable electrolytic hydrogen production amounts to $1.23 \cdot 10^7$ m^3/year (Table SP-17), representing 0.022% of the water reserves. Water consumption by PV panels rises by 91.18%, while that for hydrogen production increases by 68.98%. with the case considered above.

4.1.2 Analysis of the future status: Years 2030 and 2050

The results for subsequent decades follow a consistent structure, incorporating variations in technology costs due to the improvements applied to the technology, increasing the value of carbon taxes applied in Spain due to the environmental policy, and a diminution in methane consumption over time (Tables SP-9, SP-10, SP-11, SP-14, and SP-15).

Use of the different wastes, CO_2 capture, and use: In 2030, only lignocellulosic dry waste will be used for biomethane production, using 25,292 kt/year. This value represents a decrease of 5.28% with respect to the lignocellulosic dry waste employed in 2022. The costs related are 5,604 M€ and 1,826 M€/year, representing reductions of 49.84% and 65.81% compared to 2022. The distribution of waste use is the same as in the year 2022, achieving a maximum use value of 800 kt/year per shire (Figure SP-2 a, and Table SP-16 in the Supplementary Material).

The source of CO_2 continues to be the point source, capturing 29,378 $ktCO_2$/year, a fivefold increase compared to 2022 due to the decrease of the technology cost and the increment of its efficiency in the time horizon (Tables SP-9–SP-11 and SP-15). The corresponding investment and operating costs are 315 M€ and 1,286 M€/year (Table SP-16 in the Supplementary Material), experimenting with a parallel increase with the rise in CO_2 capture by MEA solutions. MEA solutions extend across northern Asturias, coastal zones of the northwest and south, Balearic Islands, and select inland shires (Figure SP-2 d).

The introduction of a carbon tax for the year 2030 presents more impact than the year 2022 in Spain since it moves from 80.87 €/t_{CO2} (SENDECO2, 2022) to 150 €/t_{CO2} (Blázquez et al., 2021; GMK Center, 2023) due to the more restrictive emissions policy. Thus, it prompts the use of 20,328 kt/year of lignocellulosic dry waste, a 19.62% lower concerning the use of carbon taxes for the year 2022. (Table SP-16 in the Supplementary Material). The investment and operating costs are 3,688 M€ and 1,130 M€/year, respectively, declining 43% and 51.63% compared to 2022 applying the carbon taxes.

MEA solution captures 32,231 ktCO_2/year a threefold increase compared to 2022. (Table SP-16 in the Supplementary Material). The cost presents the same increase, presenting 346 M€ and 1,411 M/€year, Investment, and operating costs, respectively. The deployment is established across the peninsula, focusing on southern and eastern industrial zones (Figure SP-3 d)

The trends observed for the year 2050 mirror those of 2022, with lignocellulosic dry waste distribution remaining largely unchanged, aside from minor variations in interior shires. It employs 24,052 kt/year of lignocellulosic dry waste, discarding manure and MSW as raw material, and showing a decreasing of 9.93% to the year 2022. Thus, the investment and operating costs reach up to 5,044 M€ and 1,591 M€/year, respectively, marking declines of 54.86% and 70.22% compared to 2022. The distribution maintains stable behavior as discussed above. (Figure SP-4 a)

The capture of CO_2 using MEA solutions represents 35,069 ktCO_2/year (Table SP-16 in the Supplementary Material), six times more CO_2 than in 2022, with investment and operating costs of 376 M€ and 1,535 M€/year, respectively, increasing proportionally with CO_2 capture by MEA solutions. They are extensively deployed across the peninsula, the Balearic Islands, northern Asturias, and northwest coastal areas (Figure SP-4 d),

The carbon tax in the year 2050 is projected to evolve to 180 €/t_{CO2} in Spain (ETUI, 2022). It allows using 2,719 kt/year of lignocellulosic dry waste, representing a reduction of 89.82% compared with the year 2022 applying the carbon taxes. The investment and operating costs reduced by 94.22% and 95.82% by 2022, representing 374 M€ and 98 M€/year. (Table SP-16 in the Supplementary Material). The distribution is performed in selected interior areas and the Guadalquivir River basin (Figure SP-5 a)

The total capture of CO_2 is yielded also with MEA solutions, reaching 42,258 ktCO_2/year, 4 times more than the CO_2 captured in the year 2022 using the carbon taxes due to their increase (Table SP-16 in the Supplementary Material). Investment and operating costs amount to 453 M€ and 1,850 M€/year, respectively, also increasing proportionally with CO_2 capture. MEA solutions (Figure SP-5 d) are mainly deployed across the peninsula, especially in industrial zones in northern Asturias, the northwest coast, and southern and eastern shires (Figure SP-5 d).

Production of renewable energy: In 2030, energy production relies solely on PV panels, with no contribution from wind turbines, primarily due to lower installation and operating costs (Figure SP-2 f, Table SP-11, Table SP-17). The distribution of PV panels is concentrated in the southeast of the peninsula, particularly in Almería province, central shires, and to a lesser extent, the Canary Islands, covering a maximum installed area of 95 km^2 in a single shire (Figure SP-2 e). The installed surface area increases significantly from 147 km^2 in the year 2022 to 1,042 km^2, representing 10.29% of 10,120 km^2, which is calculated as 2% of the total Spain surface in 2022. The investment and operating costs rise to 59,956 M€ and 2,892 M€/year, respectively, representing increases of 113.97% and 19.02% with respect to the year 2022. The power generation capacity is 36.7 GW, five times more than in 2022. The implementation of a carbon tax enables increased synthetic methane production capacity through CO_2 capture from MEA solutions, leading to augmented electrical energy production using PV panels (Figure SP-3 d, and Tables SP-16-SP-17 in the Supplementary Material). The distribution of PV panels remains largely unchanged, with a slight expansion in the central peninsula (Figure SP-3 e). The total area utilized increases to 1,145 km^2, representing 11.32% of the surface area available, stipulated as 2% of the total surface in Spain in 2022. Investment and operating costs reach 65,910 M€ and 3,173 M€/year, increasing 33.54% and a decreasing of 25.90% with respect to the year 2022 respectively, generating 40.2 GW, representing 3.27 times more than the year 2022 applying the carbon taxes This production is equivalent to 33.76% of Spain's electricity production at the close of 2022.

In 2050, the distribution of PV panels remains consistent with previous scenarios, with an expanded surface area in the central peninsula. The installed surface area increases to 1,248 km^2, representing 11.32% with respect to the 2% available surface area in Spain in 2022, and marking an 8.5-fold rise compared to 2022 (Figure SP-4 e and Table SP-17 in the Supplementary Material). The installation entails an investment and operating cost of 53,105 M€ and 3,452 M€/year, respectively, representing increases of 89.53% and 42.07% with respect to 2022. Reduced investment costs from 2030 stem from declining technology expenses over time. Power generation will reach 43.8 GW, six times higher than in 2022, equivalent to 30 nuclear reactors and 36.74% of Spain's electricity production by the end of 2022. Implementation of a carbon tax by 2050 further boosts synthetic methane production, leading to increased electricity production from PV panels, mirroring the distribution, with a slight expansion in the peninsula's center. (Figures SP-5 d, SP-5 e, and Tables SP-16-SP-17 in the Supplementary Material) The total area increases to 1,511 km^2, representing 14.83% with respect to the 2% of the surface area available in Spain in 2022, and being 5 times higher than the surface area in 2022 employing the carbon taxes. Investment and operating costs rise to 64,280 M€ and 4,160 M€/year, increasing 30.23% and decreasing 2.85% with respect to 2022 considering carbon taxes, respectively. Power generation is 52.8 GW, 4.27 times more than the year 2022 applying carbon taxes and representing 44.27% of Spain's electricity production by 2022's close.

Methane production: In 2030, total biomethane production is influenced by technological advancements, focusing on gasification of lignocellulosic dry waste and methane synthesis using CO_2 captured with MEA solutions (Figures SP-2 a, SP-2 d, SP-2 g, and Table SP-16, and Table SP-20), while anaerobic digestion of manure and MSW is disregarded due to its lower yield (Taifouris and Martín, 2023). Methane production is concentrated in the interior peninsula river basins, notably in northern Asturias and the southern region. Total biomethane output reaches 7,208 ktBiomethane/year, a 5.29% reduction from 2022, with the number of required treatment plants decreasing by 30% to 144. However, the self-production of methane rises by 38% due to the estimated reduction of methane consumption in Spain, declining from 24,160 ktCH_4/year to 18,990 ktCH_4/year (Taifouris and Martín, 2023; Table SP-14 in the Supplementary Material). Synthetic methane production amounts to 9,793 ktCH_4/year, fivefold compared to 2022, with self-production covering 51.2% of methane consumption. Additionally, 4,040 ktH_2/year of renewable electrolytic hydrogen is generated, with investment and operating costs considering all of the production paths and the CO_2 capture, 19,968 M€, and 4,609 M€/year, respectively (Tables SP-16-SP-17 in the Supplementary Material). The carbon tax in 2030 will maintain the distribution similar to the previous case. Biomethane production decreases by 23.87% with respect to the year 2022, from 7,610 ktBiomethane/year to 5,793 ktBiomethane/year, with 62.62% fewer treatment plants (206 to 77). In contrast, synthetic methane production increases by 3.56 times, from 3,300 ktCH_4/year to 10,744 ktCH_4/year with respect to the year 2022 considering carbon taxes, with investment and operating costs of 19,446 M€ and 4,182 M€/year, following the procedure described above.

In 2050, advancements in technology would lead to a continued trend of decreasing lignocellulosic dry waste utilization, resulting in 6,855 ktBiomethane/year production, a 9.93% decrease from 2022, with treatment plants reduced by 42.23% to 119. Domestic methane consumption estimation in Spain declines from 24,160 ktCH_4/year to 13,815 ktCH_4/year (Table SP-14 in the Supplementary Material), driving self-production up to 49.6%. Synthetic methane production surges to 11,690 ktCH_4/year, sixfold compared to 2022, with investment and operating costs of 16,359 M€ and 4,824 M€/year, respectively. By implementing a carbon tax in 2050, incentives for CO_2 capture through MEA solutions increase. Biomethane production drastically drops to 775 ktBiomethane/year, an 89.82% reduction to the year 2022 using the carbon taxes, and treatment plants decrease to 4, 98% fewer. Synthetic methane production rises to 14,086 ktCH_4/year, 4.27 times more than the year 2022. It presents investment and operating costs totaling 14,008 M€ and 3,993 M€/year, respectively (Taifouris and Martín, 2023; Table SP-16 in the Supplementary Material).

Evolution of self-sufficiency methane percentage, and methane prices: In 2030, due to reduced technology costs, lignocellulosic dry waste and CO_2 remain key raw materials. The methane price trend mirrors that of 2022 but with less slope due to MEA solutions' increased profitability. Sole reliance on lignocellulosic dry waste raises costs from 3.818 €/MMBTU to 5.142 €/MMBTU (13.028 €/MWh to 17.545 €/MWh) for annual costs from 5% to 15%. CO_2 capture with MEA solutions further escalates methane prices from 6.044 €/MMBTU to 15.693 €/MMBTU (20.623 €/MWh to 53.547 €/MWh) for annual costs from 20% to 300% of the budget. At 100% budget utilization, methane costs become 12.652 €/MMBTU (43.170 €/MWh), 34.69% lower than in 2022. Lignocellulosic dry waste represents 38% of methane demand due to reduced Spanish methane consumption (18,990 ktCH_4/year). The use of MEA solutions rises from the cost of 2.9% to 163.3% of the budget, reaching 300% annual value. At this point, conventional DAC processes, contributing 15.3%, are employed (Figure 3 b).

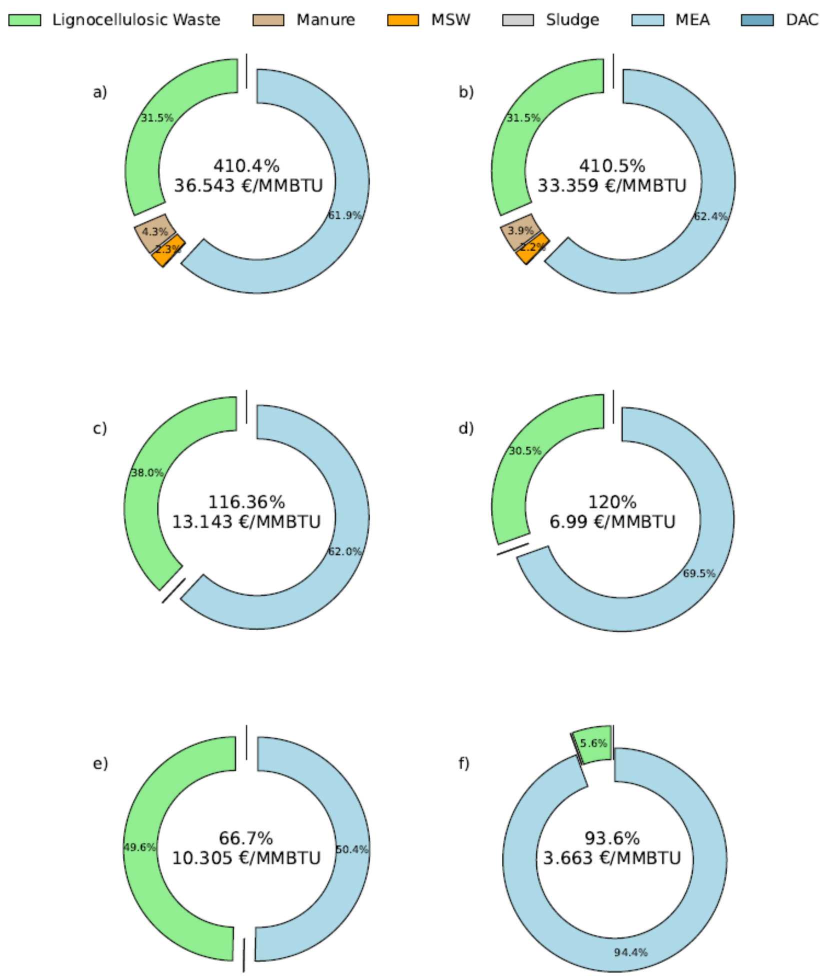

Fig. 6. Distribution of annual cost, and methane prices for a self-sufficiency methane percentage without considering carbon taxes for the years 2022 (a), 2030 (c), and 2050 (e), and considering carbon taxes for the year 2022 (b), 2030 (d), and 2050 (f)

100% of the methane self-sufficiency percentage employs an annual of 116% of the budget (Figure 6 c), decreasing 64% with respect to 2022. The use of carbon taxes in 2030 (Figure 4 b, 5, and Table SP-21 in the Supplementary Material) amplifies this trend, promoting MEA solutions over biomethane from waste gasification. The sole use of lignocellulosic dry biomass costs 3.818 €/MMBTU (13.028 €/MWh), with MEA solutions kicking in at 4.324 €/MMBTU to 8.532 €/MMBTU (14.754 €/MWh to 29.112 €/MWh) for annual cost from 10% to 300%. At 100% budget utilization, methane costs become 6.839 €/MMBTU (23.336 €/MWh). This represents a 62.35% decrease, similar to the variation in methane prices with and without carbon taxes in 2030 (Figure 5). In all of the cases, only the methane prices are comprised in the feasible region highlighted from 3 €/MMBTU to 7 €/MMBTU (10.236 €/MWh to 10.236 €/MWh) when the annual cost is between 5%-25% of the budget, an using carbon taxes for the years 2030 and 2050 (Figure 6). Figure 4 b shows a decline in the price curve, with a sudden slope increase between 250% and 300% annual cost. This is mainly due to the limited benefits of DAC from carbon tax incentives, owing to challenges in accounting for emissions from diluted sources and their regulation. Thus, 100% of methane self-sufficiency percentage is achieved for an annual cost of 120% of the budget, reducing its methane price by 79.05% with respect to the year 2022 considering carbon taxes to 6.99 €/MMBTU (20.480 €/MMBTU)

2050, a continuation of the trend observed in 2030 is evident, driven by advancements in technology that further promote the utilization of MEA solutions for lignocellulosic dry waste (Figure 3 c, 5, and Table SP-22). Similar to 2030, a slight curvilinear trend persists in biomass prices, increasing from 3.818 €/MMBTU to 5.142 €/MMBTU (13.028 €/MWh to 17.545 €/MWh). MEA solutions come into play, elevating prices from 6.024 €/MMBTU to 13.837 €/MMBTU (20.555 €/MWh to 47.214 €/MWh). At 100% of the budget, methane costs 11.599 €/MMBTU (39.577 €/MWh), 40.12% lower than in 2022 (Table SP-22 in the Supplementary Material). Lignocellulosic dry waste maintains 49.6% methane self-sufficiency due to reduced Spanish methane consumption in 2050, 13,815 $ktCH_4$/year (Table SP-14 in the Supplementary Material). Figure 3 c illustrates MEA solutions usage exceeding 200% at annual costs of 250% and 300% of the budget, with conventional DAC processes reaching 13.11% and 63.3%, respectively, surpassing the 300% methane self-sufficiency threshold.

Finally, for a 100% methane self-sufficiency percentage, the price decreased by 71.25% to 2022, presenting a methane price of 10.305 €/MMBTU (35.171 €/MWh) and an annual cost of 66.7% (Figure 6 e).

The implementation of carbon taxes by 2050 will further drive the decline in lignocellulosic dry waste usage and boost carbon capture for synthetic methane production. Referring to Figures 4 c, and 5 (Table SP-16 in the Supplementary Material), both lignocellulosic dry waste and MEA solutions are simultaneously utilized, with prices evolving from 3.146 €/MMBTU to 6.260 €/MMBTU (10.735 €/MWh to 21.360 €/MWh) for an annual cost from 5% to 20% of the budget. As related above, the first region from 5%-20% and the application of carbon taxes for the years 2030 and 2050 allow reducing the price until competitive values (Figure 5). At 100% of the budget, methane costs 3.675 €/MMBTU (12.540 €/MWh), marking an 81.08% decrease compared to 2022 without carbon taxes (Table SP-23 in the Supplementary Material). A notable decrease in methane prices is observed, closely matching the variation with and without carbon taxes in 2050 (Figure 5). Figure 4 c depicts a consistent methane price curve until approximately 3.8 €/MMBTU (12.97 €/MWh), where MEA solutions usage exceeds 200%, lignocellulosic dry waste utilization reaches 49.6%, and conventional DAC processes contribute 12.9% and 62.5%, respectively, achieving methane self-sufficiency close to 350%. The change in curvature, similar to 2030 with carbon taxes, is attributed to their application solely to MEA solutions and the challenges in applying them to dilute sources. In addition, achieving a 100% methane self-sufficiency percentage presents the same behavior, with a methane price of 3.663 €/MMBTU (12.501 €/MWh), 89% lower than 2022, for an annual cost of 93.6% of the budget (Figure 5 f)

Environmental impact of energy generation: CO_2 emissions, minerals required, and water use. In 2030, technological advancements drive down costs, establishing PV panels as the primary energy source. Emissions from PV panels use a total of 15,425 ktCO_2/year, with an additional 2,095 ktCO_2 emitted from mineral extraction. Combined, these account for 59.635% of CO_2 captured by MEA solutions, totaling 29,378 ktCO_2/year. Excluding wind turbines, mineral requirements include $1.04 \cdot 10^5$ tCu, 38 tNi, 1,101 tZn, and $1.45 \cdot 10^5$ tSi. PV panel operation consumes $1.06 \cdot 10^8$ m^3/year of water, equivalent to 0.19% of Spain's total water storage capacity (Embalses.net, 2023). Additionally, renewable electrolytic hydrogen production uses $3.64 \cdot 10^7$ m^3/year, representing 0.065% of total capacity. Carbon tax implementation in 2030 spurs CO_2 capture from point sources, increasing PV panel-related emissions to 16,923 $ktCO_2$/year, with mineral extraction emissions rising to 2,298 $ktCO_2$. Together, they contribute 59.635% of CO_2 captured by MEA solutions, totaling 32,231 $ktCO_2$/year (Table SP-17 in the Supplementary Material). Mineral requirements grow by 9.7%, with water consumption reaching $1.16 \cdot 10^8$ m^3/year (0.21% of Spain's total capacity) and hydrogen production using $4 \cdot 10^7$ m^3/year (0.071% of total capacity) (Embalses.net, 2023).

In 2050, the upward trajectory of expanding the surface area of PV panels for energy production persists, leading to increased CO_2 emissions from their operation, as well as heightened mineral demand and water usage. PV panel usage alone contributes 18,413 $ktCO_2$/year to emissions, with an additional 2,501 $ktCO_2$/year attributed to mineral extraction. Combined, these emissions constitute 59.635% of CO_2 emissions captured by MEA solutions, totaling 35,069 $ktCO_2$/year. Mineral requirements include $1.24 \cdot 10^5$ tCu, 44 tNi, 1,314 tZn, and $1.73 \cdot 10^5$ tSi Operating water consumption for PV panels amounts to $1.266 \cdot 10^8$ m^3/year, representing 0.23% of Spain's total water storage capacity (Embalses.net, 2023), while renewable electrolytic hydrogen production accounts for 0.077%, equivalent to $4.34 \cdot 10^7$ m^3/year. Furthermore, the introduction of a carbon tax in 2050 amplifies CO_2 emissions from PV panel use to 22,188 $ktCO_2$/year and mineral extraction to 3,013 $ktCO_2$, together comprising 59.635% of captured CO_2 emissions at 42,258 $ktCO_2$/year. Mineral demands increase by 20.5%, with requirements of $1.50 \cdot 10^5$ tCu, 53 tNi, 1,583 tZn, and $2.08 \cdot 10^5$ tSi (Table SP-17 in the Supplementary Material). Water consumption rises to $1.53 \cdot 10^8$ m^3/year for PV panel maintenance, constituting 0.27% of total water storage, while renewable electrolytic hydrogen production consumes 0.093% (Embalses.net, 2023), equivalent to $5.23 \cdot 10^7$ m^3/year.

5. Conclusions

This work has developed a systematic analysis to produce renewable methane using lignocellulosic dry waste via gasification and the anaerobic digestion experienced by manure, MSW, and sludge due to their wet nature. The biogas obtained is upgraded for biomethane production. Additionally, hydrogenation of captured CO_2 using MEA solutions and the DAC process to produce synthetic methane is developed, leveraging renewable hydrogen from water electrolysis powered by PV panels and wind turbines. The problem is formulated as an FLP following a decentralized use of wastes and CO_2 across the 356 Spanish shires comprising the case study. The study considers the variation of the budget available and the application of carbon taxes over the years 2022, 2030, and 2050. The maximum available surface is assumed to be 2% of the shires' surfaces, representing 10,120 km^2. The number of wind turbines to be installed per shire falls within the range of 20-50.

The energy required is based on the use of CO_2 capture technologies, and green hydrogen production, while waste treatment is self-sufficient. PV panels are mainly selected due to their competitive cost, choosing wind turbines to produce additional power. The use of PV panels reaches from 147 km^2 to 1,511 km^2, meanwhile, wind turbines reach up to 2,465, generating power from 7.3 GW to 52.8 GW. The maximum use of lignocellulosic dry waste to biomethane adds up to 7,610 kt/year, capturing 42,258 ktCO_2/year from point sources with associated production of synthetic methane of 14,086 kt/year. The use of lignocellulosic dry waste Is mainly selected for biomethane production, discarding manure, MSW, and sludge mainly due to their mainly due to their moisture content. The CO_2 capture from point sources becomes the second selected technology, avoiding the DAC process for the elevated costs. The shires first selected correspond to the southeastern region of the peninsula, and the coastline, employing the 1.46% of the surface area available, selecting point sources to capture 5,858 ktCO_2/year, and producing 11,104 kt/year of methane. The investment of 20,338 M€ for waste treatments, CO_2 capture from point sources, and methane synthesis in the year 2022.

The reduction in the cost of the technology is expected until the year 2050. Here, the PV panels and wind turbines show a cost of around 64 M€. The use of power generated is employed for CO_2 capture using MEA solutions. Up to 42,260 ktCO_2/year are captured, representing 45.28% of the total point sources emissions in Spain from the industrial zones in northern Asturias, the northwest coast, and southern and eastern shires, producing 14,861 kt/year of methane, needing an investment of 13,300 M€. Therefore, part of CO_2 emissions comes from power generation, moving in the ranges from 49.8% to 59.6%.

Methane production from waste treatment and CO_2 can be economically viable, mainly employing the gasification of lignocellulosic dry wastes as the main technology, and the hydrogenation of CO_2 captured from MEA solutions. The maximum methane production moves from 11,000 kt/year to 18,500 kt/year from 2022 to

2050 completely using the available budget, being able to increase the production up to 46,600 kt/year when the annual cost represents 3 times the budget. The prices move between 3 €/MMBTU -7 €/MMBTU as a competitive range by using gasification up to an annual cost of 20%, moving the prices in the range from 3.818 €/MMBTU to 30.229 €/MMBTU in 2022 resulting of the use of MEA solutions. The methane production range of 2,820-19,430 kt/year. These prices are projected to decrease to 13.837 €/MMBTU by 2050. The inclusion of carbon taxes can further reduce the price to 3.146 €/MMBTU, aligning it with the natural gas prices currently.

CRediT authorship contribution statement

Guillermo Galán: Writing - original draft, Software, Investigation, Methodology, Formal analysis. **Manuel Taifouris Silva**: Software, reviewing, Investigation. **Mariano Martín**: Conceptualization, Writing - reviewing & editing, Resources, Supervision, Funding Acquisition, Project administration. **Ignacio E. Grossmann**: Supervision, Conceptualization - reviewing & editing.

Nomenclature

BPMED	Bipolar Membrane Electrodialysis
BTU	British Thermal Unit
CAPEX	Capital expenditures, investment cost (€/kW-€/t)
CCUS	Carbon capture, utilization, and storage
Cr	Chromium
Cu	Copper
DAC	Direct Air Capture
DEA	Diethanolamine
DME	Dimethyl ether
EU	European Union
EOR	Enhanced oil recovery
FLP	Facility Location Problem
GAMS	General Algebraic Modelling System
GHG	Greenhouse gas
IEA	International Energy Agency
IGN	Instituto Geográfico Nacional/ National Geographic Institute of Spain
INE	Instituto Nacional de Estadística/ National Institute of Statistics
kWp	kW peak
kWh_e	kWh of electrical energy
KPI	Key performance indicators
MEA	Monoethanolamine
MILP	Mixed-integer linear programming
Mn	Manganese
Mo	Molybdenum
MSW	Municipal solid waste

Ni	Nickel
OPEX	Operating expenditures, operating costs(€/kW-€/t)
PSA	Pressure swing adsorption
PV	Photovoltaic
Si	Silicon
SI	International System of units
SNG	Synthetic natural gas,
SP	Supplementary Material
TEA	Triethanolamine
TSA	Temperature swing adsorption
VSA	Vacuum swing adsorption
WGSR	Water gas shift reactor
Zn	Zinc

Subindexes

e	Electrical

Symbols

η_{PV}	Efficiency of PV panel

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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