Sveučilište u Zagrebu

FACULTY OF MECHANICAL ENGINEERING AND NAVAL ARCHITECTURE

ANA KODBA

ECONOMICAL AND SUSTAINABLE UTILIZATION OF AGRICULTURAL RESIDUES AND INDUSTRIAL BY-PRODUCTS FOR BIOGAS PRODUCTION

DOCTORAL THESIS

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DOKTORSKI RAD

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PREFACE

Between stimulus and response there is a space. In that space is our power to choose our response. In our response lies our growth and our freedom.

Viktor E. Frankl

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The work presented in this thesis was carried out at the Department of Energy, Power and Environmental Engineering of the Faculty of Mechanical Engineering and Naval Architecture, University of Zagreb. First and foremost, I would like to express my sincere gratitude to my supervisor who supported me through this work and proved to be the best supervisor I could envies, Professor Tomislav Pukšec. I feel incredibly fortunate to have had the opportunity to complete my doctoral studies under his supervision and mentorship, and I am particularly grateful for all the discussions that helped to shape my research ideas and especially the patience, support and understanding he always had for me during this challenging process. Most importantly, I am thankful for the belief he had in me, especially in the most challenging times. I would also like to thank to Professor Neven Duić and Boris Ćosić for their support in the earlier phase of my PhD research.

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SUMMARY

The European Commission is aiming for a resource-efficient, competitive, and low-carbon economy by 2050. Core of this vision is the circular bioeconomy, integrating both bioeconomy and circular economy principles. It seeks to achieve sustainability by minimizing waste, maximizing resource efficiency, and promoting the use of renewable biological resources throughout their entire lifecycle. Biogas production via anaerobic digestion technology provides numerous benefits as it not only recovers a portion of the energy stored in biomass but also aligns with the goals of the circular bioeconomy. However, concerns about biogas production from feed and food crops raise the need for utilisation of novel biogas feedstocks that does not compete with other uses, such as agricultural residues and industrial by-products.

The main objective of this doctoral thesis is to prove the value of the use of a graph theory approach in modelling a residual biomass supply network, for different types of case study areas, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria. For this purpose, the novel approach for the assessment of seasonality of technical potential of agricultural residues and industrial by-products for biogas production was developed, constraints for which different types of industrial by-products and agricultural residues meet sustainable and greenhouse gas emissions saving criteria were defined and the mathematical model for residual biomass supply network modelling from sustainability, greenhouse gas emissions savings and economic point of view was developed. The hypothesis of this research is that an economically feasible residual biomass supply network for biogas production, that meets sustainability and greenhouse gas emissions saving criteria, could be determined with a graph theory approach. This doctoral thesis draws from five papers published in CC journals, with three in Q1 journals, one in a Q2 journal, and one in a Q3 journal.

The results presented validate the proposed hypothesis. Innovative Geographic Information System (GIS) methodologies were introduced, incorporating the seasonality of residual biomass generation into GIS mapping. These results underline the importance of considering seasonal variations when assessing potential. An analysis was conducted on greenhouse gas emissions, specific emission savings, and sustainability requirements for biogas production using agricultural residues, municipal bio-waste, and industrial by-products as feedstock. This method was grounded in Directive 2018/2001. Based on the calculations, a maximum travel distance for the considered feedstocks was defined, highlighting the substantial influence of transportation emissions on biogas production's ability to achieve necessary emission reductions.

Finally, a novel multi-period P-graph-based model for optimizing biomass supply networks was developed, advancing the integration of environmental constraints into the Process Network Synthesis (PNS) network. The optimization objective embedded within the mathematical model is to minimize the biomass supply network's costs while meeting greenhouse gas emission reduction targets and maximizing the utilization of seasonally available biomass. The model presents a comprehensive overview of GHG emissions at each stage of biogas production, compared to threshold values.

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SAŽETAK

Europska komisija teži ekonomiji koja će do 2050. biti učinkovita u korištenju resursa, konkurentna i niskougljična. U središtu ove vizije je cirkularna bioekonomija koja integrira načela bioekonomije i cirkularne ekonomije. Cilj joj je postići održivost smanjenjem otpada, maksimiziranjem učinkovitosti korištenja resursa i promicanjem korištenja obnovljivih bioloških resursa kroz cijeli njihov životni ciklus. Proizvodnja bioplina putem tehnologije anaerobne digestije nudi brojne prednosti s obzirom na to da ne samo da oporabljuje dio energije pohranjene u biomasi, već je i u skladu s ciljevima cirkularne bioekonomije. Međutim, zabrinutosti zbog proizvodnje bioplina iz stočne hrane i prehrambenih usjeva potiču potrebu za korištenjem novih vrsta sirovina za proizvodnju bioplina koje ne konkuriraju drugim primjenama, kao što su poljoprivredni ostaci i industrijski nusproizvodi.

Glavni cilj ove doktorske disertacije je dokazati vrijednost korištenja pristupa teorije grafova u modeliranju opskrbne mreža otpadnom biomasom za različite tipove područja interesa, koja je ekonomski isplativa, ali također zadovoljava kriterije održivosti i smanjenja emisije stakleničkih plinova. U tu svrhu razvijen je novi pristup za procjenu sezonalnosti tehničkog potencijala poljoprivrednih ostataka i industrijskih nusproizvoda za proizvodnju bioplina, definirana su ograničenja za koja različite vrste industrijskih nusproizvoda i poljoprivrednih ostataka zadovoljavaju kriterije održivosti i smanjenja emisije stakleničkih plinova te je razvijen matematički model za modeliranje mreže opskrbe otpadnom biomasom s aspekta održivosti, ušteda u emisijama stakleničkih plinova i ekonomike. Hipoteza ovog istraživanja je da se ekonomski isplativa mreža opskrbe otpadnom biomasom za proizvodnju bioplina, koja zadovoljava kriterije održivosti i smanjenja emisije stakleničkih plinova, može odrediti pomoću pristupa teorije grafova. Ova doktorska disertacija temelji se na pet radova objavljenih u CC časopisima, od kojih su tri u Q1 časopisima, jedan u Q2 časopisu i jedan u Q3 časopisu.

Rezultati prikazani u ovoj doktorskoj disertaciji potvrđuju predloženu hipotezu. Inovativne metode bazirane na Geografskom informacijskom sustavu (GIS), koje integriraju sezonalnost generiranja otpadne biomase u postupak GIS mapiranja potencijala biomase su predstavljene. Dobiveni rezultati ističu važnost uzimanja u obzir sezonskih varijacija prilikom procjene potencijala. Provedena je analiza emisija stakleničkih plinova, specifičnih ušteda emisija i zahtjeva vezanih uz održivost proizvodnje bioplina temeljene na korištenju poljoprivrednih ostatke, komunalnog biootpada i industrijskih nusproizvode kao sirovina. Ova metoda bazira se na Direktivi 2018/2001. Na temelju izračuna, određena je maksimalna transportna udaljenost

za razmatrane sirovine, pri čemu se ističe velik utjecaj koji emisije generirane transportom sirovina imaju na sposobnost bioplinske proizvodnje da ostvari potrebna smanjenja emisija.

Konačno, razvijen je novi više-periodni P-graf model za optimizaciju mreža opskrbe biomasom, unapređujući integraciju okolišnih ograničenja u mrežu za sintezu procesa (PNS). Optimizacijski cilj integriran u matematički model je minimizirati troškove mreže opskrbe biomasom, pri čemu se ispunjavaju postavljeni ciljevi smanjenja emisija stakleničkih plinova i maksimizira iskorištavanje sezonski dostupne biomase. Model pruža sveobuhvatan pregled emisija stakleničkih emisija generiranih u svakoj fazi proizvodnje te usporedne granične vrijednosti.

PROŠIRENI SAŽETAK

Ključni element u postizanju resursno učinkovite, konkurentne i niskougljične ekonomije je usvajanje načela bioekonomije i njezinog naprednijeg oblika, kružne bioekonomije. Bioekonomija se fokusira na zamjenu neobnovljivih, ekološki štetnih resursa održivim biološkim alternativama. Ova paradigma utječe na razne sektore, uključujući poljoprivredu, šumarstvo i ribarstvo.

Bioplin je integralni dio vizije Europske Unije (EU) za kružnu bioekonomiju, koja integrira načela minimizacije otpada te maksimizacije učinkovitosti korištenja resursa kroz cijeli životni ciklus. Bioplin, koji se uglavnom sastoji od metana i ugljikovog dioksida, proizvodi se iz organskih materijala poput poljoprivrednih ostataka, biootpada i gnojevke procesom anaerobne digestije (AD). Tehnologija anaerobne digestije je tržišno zrela tehnologija koja nudi višestruke prednosti, kao što su oporaba energije sadržane u biomasi, recikliranje hranjivih tvari u obliku organskih gnojiva te smanjenje emisija stakleničkih plinova. EU je prepoznala ove prednosti klasifikacijom AD-a kao operacije recikliranja u sklopu EU zakona i planova o gospodarenju otpadom

Proizvodnja bioplina je u skladu s političkim ciljevima smanjenja ovisnosti o fosilnim gorivima te doprinosi ciljevima proizvodnje energije iz obnovljivih izvora energije i poticanja održive ekonomije. Ovaj sektor je značajno napredovao zbog tehnološke zrelosti i političke podrške, no postoji potreba za navigacijom izazova povezanima s odabirom sirovina i održivosti korištenja istih. Većina trenutnih postrojenja za proizvodnju bioplina u EU uvelike koristi kukuruznu silažu kao sirovinu. To izaziva zabrinutost zbog utjecaja uzrokovanih izravnim i neizravnim promjenama u korištenju zemljišta te potencijalne konkurencije s proizvodnjom hrane i stočne hrane.

Revidirana Direktiva o promicanju uporabe energije iz obnovljivih izvora (D2018/2001) adresira ove zabrinutosti postavljanjem kriterija održivosti, uključujući kriterij uštede stakleničkih plinova od najmanje 70% za postrojenja koja započinju s radom od 2021. godine i 80% za one koji započinju s radom od 2026. Dodatno k tome, RePowerEU plan za cilj postavlja značajno smanjenje ovisnost EU o ruskim fosilnim gorivima te ubrzavajući prijelaz prema čistoj energiji, što uključuje ambiciozni cilj proizvodnje 35 milijardi kubnih metara biometana do 2030. godine.

Ova doktorska disertacija temelji se na pet radova objavljenih u CC časopisima, od kojih su tri u objavljana Q1 časopisima, jedan u Q2 časopisu i jedan u Q3 časopisu. Rad 1 prikazuje pristup za georeferenciranom mapiranju potencijala poljoprivrednih ostataka i komunalnog biootpada za proizvodnju bioplina. U sklopu ovog rada, uz prostornu distribuciju uzeta je u obzir i sezonska varijacija potencijala sirovina. Rezultati pokazuju da postoji snažna potreba za uključivanjem utjecaja sezonalnosti u procjeni potencijala bioplina za lignocelulozne poljoprivredne ostatke. Prednosti ovog pristupa prikazane su u dva primjera koja su rezultirala sa 12 % i 40 % nižim kapacitetom skladišnih prostora, u usporedbi s trenutačno korištenim pristupima koji uzimaju u obzir godišnji potencijal. Slično tome, Rad 2 se također fokusira na procjenu utjecala sezonalnosti na potencijal proizvodnje bioplina, no fokus stavlja na sirovine koje nastaju kao nusproizvodi u industrijskim postrojenjima. Rezultati su pokazali kako i za slučaj ovih sirovina postoji snažna potreba za uključivanjem sezonskog aspekta pri definiranju potencijala biomase pogodnog za proizvodnju bioplina. Za razmatrane sirovine i studije slučaja potencijalnih lokacija za proizvodnju bioplina, rezultati pokazuju kako se godišnji faktor opterećenja kreće od 0,1-0,24 u slučaju kada skladištenje sirovina nije dostupno. U slučaju 6-mjesečnog skladištenja sirovina, godišnji faktori opterećenja se poboljšavaju te variraju od 0,58-0,82, no ocjena ekonomske isplativosti zahtjeva pažljivo razmatranje zbog potencijalnih problema povezanih s nepravilnim skladištenjem te dodatnih investicijskih troškova.

U sklopu Rada 3 je provedena analiza emisija stakleničkih plinova, njihovih ušteda i zahtjeva za održivosti proizvodnje bioplina, za razne sirovine za koje nisu definirani emisijski faktori u Direktivi 2018/2001. Rezultati pokazuju da emisije transporta značajno utječu na sposobnost ispunjavanja zahtjeva za uštedom emisija, koja se postavljaju za bioplinska postrojenja. Isto tako, naglašavaju potrebu za proširenjem broja sirovina za koje su definirane tipične i zadane vrijednosti emisija stakleničkih plinova, za svaku od razmatranih kategorija emisija.

Rad 4 predstavlja novi matematički model temeljen na korištenju teorije grafova za optimizaciju mreže opskrbe otpadnom biomasom. Cilj ove optimizacije je dvostruk: pronaći najisplativiju mrežu opskrbe biomasom koja ima minimalne troškove, istovremeno ispunjavajući potrebne uštede emisija stakleničkih plinova definirane u Direktivi 2018/2001 (uštede od 80% u usporedbi s fosilnim gorivima) za proizvodnu i korištenje bioplina. Dodatno k tome, sezonska varijacija u opskrbi biomasom integrirana je u model korištenjem pristupa s više razdoblja. Rad 5 je također fokusiran na optimizaciju mreže opskrbe biomasom, no fokus stavlja na sirovine kod kojih postoji manji izbor mogućnosti u mreži opskrbe biomasom te su dostupne tijekom cijele godine. Ovaj preduvjet omogućuje integraciju ograničenje emisija stakleničkih plinova u postupak evaluacije podataka implementiranog u GIS alatu, čime je umanjena složenost procesa. Rezultanti podaci dobiveni GIS alatom izvoze se u alat temeljen na teoriji grafova za daljnju ekonomsku optimizaciju. Radovi 4 i 5 dokazuju da se ekonomski isplativa mreža opskrbe otpadnom biomasom za proizvodnju bioplina, koja ispunjava kriterije održivosti i smanjenja emisija stakleničkih plinova, može odrediti pomoću pristupa teorije grafova. Sveobuhvatno gledajući, radovi u sklopu ove doktorske disertacije nude sveobuhvatni pogled na modeliranje i optimizaciju proizvodnje bioplina, od samog mapiranja dostupnosti, do analize emisije te ekonomskih razmatranja i izbora sirovina.

Ključne riječi:

Geografski informacijski sustav; bioplin, emisije stakleničkih plinova, teorija grafova

CILJ I HIPOTEZA

Ciljevi ovog istraživanja uključuju sljedeće tri točke:

1. Odrediti prostornu raspodjelu i izračunati utjecaj sezonalnosti proizvodnje otpadne biomase.
2. Definirati ograničenja za koja različite vrste industrijskih nusproizvoda i poljoprivrednih ostataka ispunjavaju kriterije održivosti i smanjenja emisija stakleničkih plinova.
3. Dokazati vrijednost korištenja pristupa teorije grafova u modeliranju opskrbnih mreža otpadnom biomasom za različite tipove područja interesa, koja zadovoljavaju kriterije održivosti i smanjenja emisija stakleničkih plinova.

Hipoteza ovog istraživanja je da se ekonomski isplativa mreža opskrbe otpadnom biomasom za proizvodnju bioplina, koja ispunjava kriterije održivosti i smanjenja emisija stakleničkih plinova, može odrediti pomoću pristupa teorije grafova.

ZNANSTVENI DOPRINOS

Znanstveni doprinosi ovog istraživanja uključuju sljedeće tri točke:

1. Novi pristup za procjenu sezonalnosti tehničkog potencijala poljoprivrednih ostataka i industrijskih nusproizvoda za proizvodnju bioplina

2. Definirana ograničenja za koja različite vrste industrijskih nusproizvoda i poljoprivrednih ostataka zadovoljavaju kriterije održivosti i uštede stakleničkih plinova
3. Matematički model za modeliranje mreže dobave otpadne biomase sa stajališta održivosti, uštede stakleničkih plinova i ekonomičnosti

METODE I POSTUPCI

Metoda implementirana u sklopu ove doktorske disertacije sastoji se od tri dijela: 1) Mapiranje prostorne raspodjele i sezonske varijacije potencijala proizvodnje bioplina temeljene na Geografskom informacijskom sustavu (GIS); 2) Analiza emisija stakleničkih plinova, ušteda emisija stakleničkih plinova i zahtjeva za održivost u proizvodnji i korištenju bioplina; 3) Optimizacija opskrbnog lanca biomase pomoću P-graf pristupa.

Prvi dio metode pruža sveobuhvatan okvir za ocjenu i optimizaciju proizvodnje bioplina. GIS alat korišten je za analizu prostorne raspodjele i sezonske varijacije potencijala bioplina. Kombinirani prostorno-statistički pristup korišten je za procjenu potencijala različitih sirovina kao što su komunalni biootpad, poljoprivredni i stočarski ostaci te industrijski nusproizvodi. U sklopu metode, definirane su jednadžbe za izračun teorijskog i tehničkog potencijala ovih sirovina. Uz mapiranje potencijala bioplina, GIS alat je korišten za identifikaciju optimalnih lokacija za izgradnju bioplinskih postrojenja te za identifikaciju najkraćih transportnih ruta između pružatelja sirovina i bioplinskih postrojenja, kao i za određivanje troškova prijevoza.

Drugi dio metode opisuje postupak izračuna emisija stakleničkih plinova (GHG) generiranih proizvodnjom i korištenjem bioplina, temeljen na Direktivi 2018/2001 koji je razvio Zajednički istraživački centar. Ovaj postupak koristi Globalni potencijal zatopljenja (GWP) kao klimatsku metriku, koja kvantificira utjecaj različitih stakleničkih plinova poput metana i dušikovih oksida tijekom razdoblja od 100 godina. Ukupne emisije generirane kao rezultat proizvodnje i korištenja bioplina predstavljaju sveobuhvatni zbroj različitih komponenti, uključujući emisije iz ekstrakcije ili uzgoja sirovina, promjene u korištenju zemljišta, obradu i prijevoz, među ostalim. Također uzima u obzir uštede emisija koje nastaju kao rezultat poboljšanih poljoprivrednih praksi te primjenom tehnologija za hvatanje i skladištenje ugljičnog dioksida. Ova metoda također proširuje analizu alokacijom emisija stakleničkih plinova toplinskoj i električnoj energiji generiranoj iz bioplina u kogeneracijskim postrojenjima. Dodatno k tome, definirane su uštede stakleničkih plinova za toplinsku i električnu energiju, u usporedbi sa fosilnim komparatorima. Na temelju postignutih ušteda stakleničkih plinova, definirane su maksimalne transportne udaljenosti za svaku od razmatranih sirovina korištenih za proizvodnju bioplina. Dodatno k tome, definirani su i naglašeni zahtjevi održivosti postavljeni na korištenje sirovina za proizvodnju bioplina.

Treći dio metode opisuje razvoj matematičkog model za ekonomsku optimizaciju mreže opskrbe otpadnom biomasom koji integrira zahtjeve za uštedom emisija stakleničkih plinova. Pri tome su razvijena dva različita pristupa: prvi pristup je prilagođen za složene mreže koje se bave sezonski varijabilnim sirovinama, obično prisutnim u ruralnim područjima. Drugi pristup je primjenjiv za jednostavnije mreže opskrbe otpadnom biomase i namijenjen je sirovinama dostupnim tijekom cijele godine, koje su češće u urbanim područjima. Oba pristupa koriste GIS mapiranje za dobivanje podataka o dostupnosti sirovina i transportnim udaljenostima.

Prvi pristup koristi alat temeljen na teoriji grafova za razvoj matematičkog model usmjerenog na pronalaženje ekonomski optimalnih struktura mreže koje ispunjavaju ciljeve održivosti i smanjenja emisija stakleničkih plinova. Nadalje, kroz proširenje modela na više razdoblja, razvijeni model uzima u obzir sezonalnost opskrbe biomasom tijekom godine. Specijalizirani algoritmi MSG (*engl. Maximal Structure Generation*), SSG (*engl. Solution structure generation*) i ABB (*engl. Accelerated Branch and Bound)* korišteni su za generiranje maksimalne strukture, svih kombinatorno izvedivih struktura te rangiranih n- najboljih struktura.

Drugi pristup također započinje s GIS mapiranjem, ali se usredotočuje isključivo na tok biomase. Ograničavanje ukupnih emisija stakleničkih plinova je implementirano u sklopu evaluacije podataka implementiranih u GIS alatu, čime je umanjena složenost procesa. Rezultanti podaci dobiveni GIS alatom izvoze se u alat temeljen na teoriji grafova za daljnju optimizaciju.

KEYWORDS

Geographic information system; biogas, greenhouse gas emissions, graph theory

KLJUČNE RIJEČI

Geografski informacijski sustav; bioplin, emisije stakleničkih plinova, teorija grafova

LIST OF ABBREVIATIONS

ABB	Accelerated Branch and Bound
AD	Anaerobic digestion
CHP	Combined heat and power
EC	European Commission
EU	European Union
GIS	Geographic Information System
GHG	Greenhouse gas
GWP	Global Warming Potential
IPCC	Intergovernmental Panel on Climate Change
LULUCF	Land Use, Land-Use Change, and Forestry
MILP	Mixed-Integer Linear Programming
MSG	Maximal Structure Generation
PNS	Process-Network Synthesis
SSG	Solution structure generation

NOMENCLATURE

A uilised land for agricultural production (m^2)

A_{fiel} area of a specific field (m^2)

A_k the total agricultural area of the region, in which specific field is located (m^2)

b fuel price (EUR/L)

B_{biogas} biogas potential of transported feedstock (GJ)

C_{el} a fraction of exergy in the electricity (-)

C_h a fraction of exergy in the useful heat, calculated as Carnot efficiency (-)

CS_A the quantity of carbon stored per area corresponding to the actual land use (gC/ha)

CS_R the carbon stock per unit area corresponding to the reference land use (gC/ha)

$COMP$ amount of residues which should be left for the feeding and bedding of animals (kg)

C_{trans} specific transport cost (EUR/GJ)

d transport distance (km)

E total emissions from the use of the fuel (gCO_2eq/MJ)

e_{ec} emissions from the extraction or cultivation of feedstocks (gCO_2eq/MJ)

e_l annualised emissions from carbon stock changes caused by a land-use change (gCO_2eq/MJ)

e_p emissions from processing (gCO_2eq/MJ)

e_{td} emissions from transport and distribution (gCO_2eq/MJ)

e_u emissions from the fuel in use (gCO_2eq/MJ)

e_{sca} emission reduction from soil carbon accumulation due to improved agricultural management (gCO_2eq/MJ)

e_{ccs} emission savings from CO_2 capture and geological storage (gCO_2eq/MJ)

e_{ccr} emission savings from CO_2 capture and replacement (gCO_2eq/MJ)

e_B bonus of 29 gCO_2eq/MJ, applied for the case when biomass is obtained from restored degraded land (gCO_2eq/MJ)

E_{bio} methane potential of the considered feedstock (m_3)

$E_{bio,fiel,m}$ the energy value of the biogas potential for the specific field in the specific month m (MJ)

$E_{bio.feedstock}$ biogas potential of loaded biomass feedstocks (MJ)

EC_{el} total GHG emissions associated to electrical energy (gCO_2eq/MJ)

EC_h total GHG emissions associated to thermal energy (gCO_2eq/MJ)

$EC_{F(h)/F(el)}$ total emissions from the fossil fuel comparator for useful thermal energy/electrical energy (gCO_{2eq}/MJ)

EF_{fuel} fuel's emission factor (gCO_2eq/l)

K_{empty} fuel consumption of empty truck (l/km)

K_{full} fuel consumption of a full truck (l/km)

LHV_{CH_4} methane lower heating value (MJ/m^3).

y average biomass yield of the agricultural production (kg/m^2)

y_{bio} specific biogas yield (m^3/kg)

$M_{p.com}$ amount of processed commodities (kg)

MPH manure per head ratio (kg/head)

N number of heads of livestock (head)

P the crop's productivity, expressed as the amount of biofuel produced per hectare per year (MJ/ha*yr);

$P_{th,ag}$ theoretical potential of residues from the agricultural category (kg)

$P_{tech\,,ag}$ technical potential of residues from agricultural production (kg),

$P_{th,liv}$. theoretical potential of manure (kg)

RPP residue-to-product ratio (kg/kg)

$RPPC$ residue to processed commodity ratio (kg/kg)

$SPA_{(a)}$ amount of residues which should be left for the feeding and bedding of animals (kg).

SRR sustainable removal rate (%)

$s_{(a)}$ share of animals to which $SPA_{(a)}$ refers (%)

s_{CH_4} share of the methane contained in biogas (%)

T transport cost correction factor (-)

T_h temperature of the useful heat at the point of delivery (K)

T_0 environmental temperature, set at 273.15 K (K).

V_{truck} truck load capacity (m^3)

$\rho_{feedstock}$ bulk density of feedstock (t/m^3)

η_{el} electrical efficiency, determined as the annual electrical energy output divided by the energy content of annual fuel input (%)

η_h - heat efficiency, determined as the annual useful thermal energy output divided by the energy content of the annual fuel input (%)

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1 INTRODUCTION

This section provides the contextual background of the topic and motivation for this thesis. This is followed by the literature review which provided a knowledge gap analysis of the topic, with a special emphasis given on biomass potential mapping, sustainability and greenhouse gas emissions saving criteria analysis, as well as a P-graph optimization of biomass supply networks. Finally, the objectives and hypotheses of this research are presented, followed by a scientific contribution of this thesis.

1.1 Background and motivation

1.1.1 Circular bioeconomy

The European Commission (EC) has set a long-term goal to develop a resource-efficient, competitive and low carbon economy by 2050 [1]. Bioeconomy has been recognised as a crucial element towards achieving these long-term objectives. The bioeconomy encompasses the substitution of finite and environmentally damaging fossil resources with renewable biological resources, as well as the adoption of bio-based processes and technologies [2]. It entails a comprehensive restructuring of the economy, taking into account environmental constraints and sustainable practices. In essence, the bioeconomy aims to shift away from non-renewable and unsustainable resources, promoting a more sustainable and environmentally friendly economic model. It encompasses a wide range of sectors, such as agriculture, forestry, fisheries, and bio-based industries [3].

The circular bioeconomy is a concept that combines the principles of bioeconomy and circular economy to create a sustainable and regenerative economic system [2]. It aims to minimize waste generation, maximize resource efficiency, and promote the use of renewable biological resources throughout the entire value chain. In the circular bioeconomy, the focus is on reducing, reusing, recycling, and recovering materials and energy from biological resources, such as agricultural residues, food waste, and bio-based products [4]. This approach not only reduces the dependence on fossil fuels but also contributes to mitigating climate change and preserving biodiversity. By closing the loop and ensuring the continuous flow of resources, the circular bioeconomy fosters innovation, creates new business opportunities, and promotes a more resilient and sustainable society [5]. Bioenergy production is intricately linked to the principles of the circular bioeconomy, as it involves the conversion of organic materials, such as crop residues, forest biomass, and organic waste, into usable energy sources like liquid biofuels, biogas (or biomethane), electricity and heat.

1.1.2 Biogas production through anaerobic digestion

Biogas is a renewable energy source that holds great potential in addressing environmental challenges and providing sustainable energy solutions. It is generated by the anaerobic digestion of organic materials, such as agricultural waste, food waste, and sewage sludge. Throughout the anaerobic digestion process, microorganisms decompose the organic matter, resulting in the production of methane and carbon dioxide gases, along with small quantities of other gases [6]. Biogas can be captured, upgraded to biomethane and used as a renewable fuel for a variety of purposes, including electricity generation, heating, and cooking.

Nowadays, the technology of biogas production by anaerobic digestion is mature and well developed [7]. Anaerobic digestion (AD) technology for biogas production offers numerous advantages, such as:

- recover the energy contained in biomass and produce renewable energy in the form of biogas and biomethane;
- contribute to nutrient and micro-nutrient recycling which can be future used in the form of organic fertiliser, thereby substituting fossil fuel based mineral fertilisers;
- threefold greenhouse gas emissions reduction, due to avoided emissions from landfills or improved manure management; production of renewable energy which replaces fossil fuels; production of organic fertilisers which replace energy intensive mineral fertilisers [8].

Those advantages have been recognised by the European Commission, which updated waste legislation [9] and defined AD as a recycling operation in the waste hierarchy.

1.1.3 The role of biogas in current and future energy systems

The EU policies on renewable energy production introduced various support schemes that encouraged the increase of biogas production [10]. By the end of 2021, there were around 19 000 biogas and 1023 biomethane plants in Europe [11]. Figure 1 shows the number of biogas plants per 1 million capita in European countries.

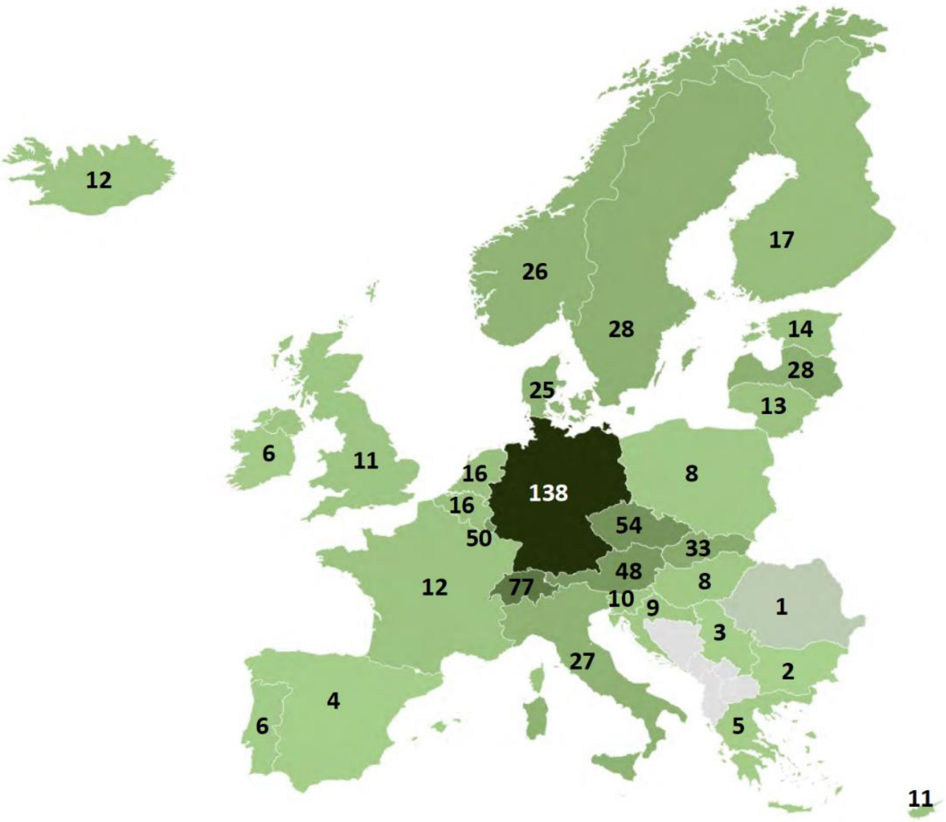


Figure 1 Number of biogas plants per 1 million capita [12]

The biggest biogas and biomethane producer in Europe is Germany, followed by the United Kingdom and France. The majority of Europe's existing biogas production is employed in cogeneration (CHP) facilities to generate both electricity and heat. Subsequently, the electricity is integrated into the existing power grid, while the heat is frequently employed on a local scale, such as in a district heating system or on-site.

The role of the biogas sector has been recognised in numerous EU policies and plans. As the most significant, the following can be highlighted:

- Renewable Energy Directive recast (Renewable Energy Directive 2018/2001/EU) [13]

The recast directive, which extends the legal framework until 2030, introduces a new binding target for renewable energy in the EU of at least 32% by 2030. This target includes a yearly increase of 1.3% for renewable energy in the heating sector and an end target of 14% renewable energy in the transport sector by 2030. The objective of the latter incorporates a sub-target of 3.5% for advanced biofuels and biogas.

The Directive aims to facilitate the integration of biomethane into the natural gas grid, expand the use of guarantees of origin to cover renewable gas, and simplify the cross-border trade of biomethane. Additionally, the Directive establishes specific requirements for greenhouse gas savings and sustainability in the use of biogas for heating, electricity, and transportation. These requirements will be further elaborated upon in the subsequent subsection.

- RePowerEU [14]

RePowerEU aims to rapidly reduce dependence on Russian fossil fuels by fast forwarding the measures to address the consequences of escalating energy costs, enhance the EU's gas sources diversification, and expedite the shift towards clean energy. The European biogas and biomethane industries are dedicated to supplying 35 billion cubic metres (bcm) of biomethane by 2030, thereby aiding the EU in meeting both its climate targets and ensuring energy security. In 2020, the EU generated 18 bcm of biogas and biomethane. By upgrading current biogas facilities to boost biomethane output and expanding production capacity, the EU aim to establish a more robust and environmentally-friendly energy system. Figure 2 presents the projections of combined biogas and biomethane production in EU-27.

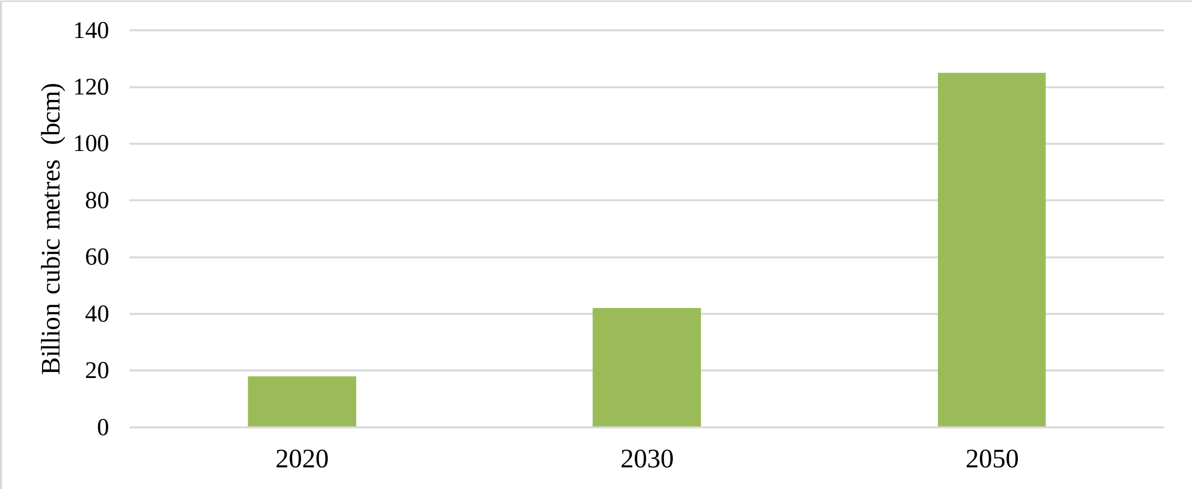


Figure 2 Projections of combined biogas and biomethane production in EU-27 [15]

By 2030, EU-27 should increase biomethane production for 32 bcm, going from 3 bcm in 2022 to 35 bcm in 2030 [15].

1.1.4 Feedstocks used for biogas production

The large span of different biomass sources can be used as a feedstock for bioenergy production. Biogas plants operating in EU use, to the greatest extent (up to 72 % [16]), maize silage as a substrate for biogas production [17], due to high biomass and biogas yields, as well as feedstock storability [18]. However, the utilisation of feedstocks that have been grown on agricultural land (such as maize silage) has caused concerns over the negative environmental impact due to direct and indirect land-use change, as well as socio-economic concerns as biogas production can be in competition with food and feed production [19]. Direct land-use change is defined as the land-use change that occurs when biogas feedstock cultivation displaces a prior crop that was cultivated on that land, for other use (i.e. food or feed production). Thus, a direct connection can be made between biogas production and land-use change [1]. On the other hand, indirect land-use change occurs when the cultivation of crops for biogas (or bioliquids, biomass) production displaces the traditional production of crops for food and feed purposes, which results in additional demand on land. This increasing pressure on land can lead to the extension of agricultural land into areas with high carbon stock such as wetlands, peat land and forests, thus causing additional greenhouse gas emissions [20]. Those concerns are reflected in the revised Renewable Energy Directive [13], which came into effect in December 2018. The Directive established sustainability and the greenhouse gas (GHG) emission-savings criteria that biogas used in transport, electricity, heating and cooling production, must comply with. Additionally, the directive enhances the sustainability requirements for biogas feedstocks and adds new requirements for specific greenhouse gas emission savings from biogas production, which biogas facilities must adhere to in order to contribute to renewable energy goals and qualify for government funding. As one of these criteria, the Directive states that the GHG savings from the use of biomass for heating, cooling and electricity production must be at least 70% for plants that started to work in 2021 and 80% for plants starting operation from 2026. In addition, the Directive states that the share of high indirect land- use change-risk biofuels, bioliquids or biomass fuels produced from food and feed crops for which a significant expansion of the production area into land with high-carbon stock is observed shall gradually decrease to 0 % until December 2030 [13]. In order to diminish this negative environmental impact, residual resources are expected to have increased utilization due to lower environmental impact [21]. A sound knowledge base on the availability of residue potential has therefore been recognized as a necessary prerequisite for the success of the development of a biogas sector [22].

1.2 Knowledge gap analysis

1.2.1 Biomass potential mapping

In recent years, the use of GIS tools for biomass potential mapping has gained prominence. The utilisation of GIS tools offers crucial insights into the spatial distribution of biomass potential, facilitating the optimization of bioenergy production facilities. New directives and concerns about the sustainability of biogas production have resulted in increased interest in underestimated feedstocks for biogas production, such as agricultural residues. Lourinho et al. [29] used a GIS tool for assessing the spatial distribution of agroforestry residues' annual potential, while Haase et al [30] used it for the assessment of the spatial distribution of annual sustainable crop residue potentials. The spatial distribution of annual biogas potential from non-woody biomass of conservation areas and roadsides for biogas was assessed with a GIS tool by Meerbeek et al. [31]. Tańczuk et al. [32] used a GIS tool to assess the annual theoretical and technical potential of chicken manure from various rearing systems in Polish provinces. Chen et al. [33] presented the method for assessing the annual economic potential of biomass supply from crop residues, in which he used a GIS-based approach to identify the areas in China that are likely to produce crop residue. Vukašinović et al. [34], developed a GIS-based combined approach for the determination of the most cost-effective investments in biomass sector. The proposed approach included GIS mapping of annual biomass potential and defining both storage and plant locations. Similar to this, Sharma et al. [35], used a GIS tool to assess annual potential of corn stover, switchgrass and miscanthus in order to assess biofuel production potential and suitable biorefinery locations in the USA, while Comber et al. [36] assessed annual potential of food waste, cattle slurry and wheat straw to locate bio-energy facilities. The annual potential of agricultural waste, co-products and by-products was assessed by Bedoić et al [37], for the member countries of the European Union. Sultana et al. [38] have already shown that the application of GIS tools enables assessment of biomass transportation cost. Valenti et al. [39], used a GIS tool to assess the annual biogas potential of citrus pulp, olive pomace, whey, poultry litter, cattle manure and corn silage. In accordance with the results, the authors conducted an economic assessment which allowed them to determine the size and location of four biogas plants in Sicily. The same feedstocks were considered in the work [40], in which authors developed a GIS-based spatial index of feedstock-mixture availability for anaerobic co-digestion. The developed spatial index describes the availability of the specific feedstock in each municipality, in accordance with the annual production of respective feedstock and enables identification of municipalities which are most suitable for biogas production. Shea et al. [41] used a GIS tool to identify financially viable locations for biomethane injection to a natural gas network, in accordance with the spatial distribution of the annual potential from grass silage and cattle slurry. Natarajan et al. [42] used a GIS tool for annual biomass potential assessment in India, for which authors developed land use maps for the selected pilot regions. Similar to this, Höhn et al. [43] developed a regional GIS-based method to analyse suitable locations and capacities of biogas plants, based on the theoretical annual potential of various biomass resources, as well as transportation distances. Franco et al. [44] developed a model to solve the multi-criteria decision problem of identifying the most suitable location for a biogas plant, taking into consideration the annual potential of slurry, population density, distance to heat plants and transportation-optimal sites.

A seasonality of biomass production affects a requested storage facility capacity and consequently, the cost of the logistic supply chain. Balaman et al. [45] developed a model to maximize the profit of the biomass supply chain. In this research, one of the variables included in the optimization was a unit land cost, which was used to determine capacity and locations (counties) in which a storage facility would be most economically feasible to install. The total storage capacity for all considered regions was determined in accordance with the annual biomass availability (corn silage, layer hen manure, broiler hen manure and cattle manure).

In addition to agricultural residues, industrial by-products have also been recognised as underutilised feedstock for biogas production. This has been proven in various experimental studies, such as the study where Al Afif et al. [46] concluded that the quality of biogas produced from olive mill solid waste was sufficient for all experiments. Furthermore, Duarte et al [47] concluded in their experimental research that industrial residues such as residues from vegetables and fruit industries are promising co-digestion substrates due to the positive synergetic effect demonstrated in increased biogas yield. In this context, the assessment of biogas potential of residues and by-products from industrial production captivates the attention of many researchers. Moreda et al. [48] calculated the yearly methane potential of numerous agricultural residues and by-products from agro-industrial production in Uruguay. In this review work, Moreda et al. detected residues and by-products from a brewery, dairy, fish, malting, poultry, rice, sausage, slaughterhouse, tannery, wine and wool scouring industry as viable for biogas production and assessed the respective annual potential on the national level. Similar to this, Kythreotou et al. [49] assessed the annual biogas potential of several potential sources for biogas production, such as biodegradable fractions of municipal solid waste, residues from food and beverage industries and sewage sludge. Assessment of the biogas potential from manure and slaughterhouse by-products was conducted in the work [50]. In this work, Mahmoud Ali et al. used the GIS tool to present the distribution of annual biogas potential from the above-mentioned feedstocks, between Mauritania's provinces. In another work, Pereira et al. [51] calculated the economic potential for electricity generation from vinasse in accordance with the annual potential of biogas from vinasse, obtained from sugarcane processing. The economic potential was calculated with a GIS tool, for each municipality of the state of São Paulo. In accordance with the annual potential of residues from the palm oil industry, Loong Lam et al. [52] developed an environmental strategy for a sustainable supply chain.

From the literature review, it's evident that there's been significant research focused on developing GIS-based methods to evaluate annual biogas potential. Yet, the production of agricultural and industrial residues and by-product is not consistent throughout the year. Given their low energy density, these feedstocks require substantial storage capacities to bridge the time gap between supply and demand.

Moreover, some industrial by-products can only be stored for a limited duration because they are more prone to degradation and pH changes. This underscores the necessity to address the research gap concerning the integration of both seasonal and spatial variations in biogas potential from agricultural residues and industrial by-products.

1.2.2 Defining constraints for biomass to meet sustainability and greenhouse gas emissions saving criteria

The environmental sustainability of biomass utilisation for energy purposes has raised significant concerns. It has been reported that biomass utilisation may result in unsolved challenges and trade-offs concerning the accounting of GHG and non-GHG emissions [53]. The environmental sustainability of biomass utilisation is a complex problem which depends on various factors such as the feedstock type, feedstock pre-processing and processing technology, transportation and distribution distance, emissions from the fuel in use, etc. Due to its high complexity, the importance of this research problem has increased in the last decades. Hence, a significant number of research papers investigate various types of environmental sustainability performance analysis of biomass utilisation for energy production. Some of them are presented in the following paragraphs.

Hamelin et al. [54] performed the life cycle assessment of biogas production based on manure and the following co-substrates: straw, garden waste, food waste, energy crops and animal urine and faeces. The results, given in $kgCO_2eq$ per functional unit, prioritised source-segregated solid manure as co-substrates, followed by straw and biowastes, while energy crops are identified as co-substrates whose utilization would result in adverse environmental impacts. In their recent work, Meng et al. [55] examined the viability of total or partial replacement for peat by maize straw biogas residues and manure biogas residues. The results show that a biogas plant that produces 10,000 m^3 biogas daily can achieve savings of 439.4 tonnes/year of CO_2 through the proposed replacement. Den Boer et al. [56] calculated that the utilisation of kitchen waste for biogas production could lead to 680,000 tCO_{2eq} savings per year.

The transport distance of the biomass supply and the availability of the biomass throughout the year have a significant impact on the energy conversion efficiency and GHG reductions of anaerobic digestion (AD) technology [57]. Berglun et al. [58] performed the energy life cycle analysis of 8 feedstocks for biogas production. The results showed that the difference between energy output and input is positive in the cases of transport distances less than 700 km for slaughterhouse waste, 580 km for municipal organic waste, 240 km for straw, 220 for pig manure and 200 km for cow manure. In their study, Uusitalo et al. [59] came to the conclusion that using biogas to produce heat and electricity leads to greater GHG reductions than composting feedstock, yet not as high as in the case of its utilisation for transport. In the work of Balcioglu et al. [60], the authors calculated that if 60% of cattle manure and all available chicken manure in Turkey are co-digested with other waste feedstock, this could lead to annual GHG emissions reduction of up to 2.5 %. Wąs et al. [61] assess the GHG mitigation potential in Ukraine for biogas production,which uses agricultural waste and manure as biogas feedstocks. Results indicate that the theoretical potential of GHG savings ranges between 5% to 6.14%, while technical potential varies between 2.3% to 2.8% of total GHG emissions. Tamburni et al. [62] calculated that biogas production from agricultural waste could result in GHG emission savings of up to 3,000,000 $MgCO_{2eq}$ in the region of Emilia-Romagna.

As can be seen from the scientific literature review, environmental sustainability and GHG savings have been studied by many researchers and the results obtained in those studies are calculated by various approaches, thus providing the results in many different ways. However, there are still numerous feedstocks that are recognised as novel feedstocks for biogas production (mostly so-called waste materials), but the constraints for achieving required GHG savings still need to be defined, as there are limited data on GHG-emission from the biogas production chain which can serve as typical and defaults.

To address this research gap, the research object of this thesis is to define the maximal transport distance of various novel biomass feedstock, that complies with the GHG savings of 80%, compared with fossil fuels, as required in Directive 2018/2001 [13].

1.2.3 P-graph optimisation of biomass supply network

It is becoming evident that energy systems modelling is progressively embracing different types of integrative approaches [63]. Murele et al. [64] investigated the influence of the integration of biomass into coal-based energy supply networks. Results of the optimisation aimed to minimise the cost of the energy supply network, obtained through the General Algebraic Modelling System software (GAMS), indicate that a biomass fraction of 7.9% in the mixed solid fuel will provide an optimal solution, as it would result in a balanced cost decrease of the emission cost and increase of the supply network. Simon et al. [65] developed a model that simulates the supply curve of wood biomass from the sustainable management of natural forests. The findings indicate that the maximum admissible distance to the nearest transportation route and the associated transportation expenses are the two factors that exert the greatest impact on both the supply and cost of wood biomass. Rentizelas et al. [66] applied the Data Envelopment Analysis method for assessing the cost, energy and GHG emission efficiency of international biomass supply network pathways. The selection of the most efficient pathway depends on the total cost, energy consumption and emissions, as well as the priorities of the decision maker. Shen et al. [67] developed a novel mathematical optimisation approach that allows the reduction of redundancy of data series to solve the multi-echelon biomass supply problem. This multi-echelon biomass supply problem includes economic, environmental and social indicators, optimised by maximising economic viability and social benefit while minimising environmental emission through a weighted-sum approach and max-min aggregation approach.

The use of P-graphs in energy system modelling has intensified during the past two decades. In their recent paper, Xu et al. [68] implemented the P-graph approach to define optimal energy export strategies of islands, whose objective is to minimise construction, operating and environmental cost (related to greenhouse gas footprint). Results showed that the best operational path and the best economical cost are in the case of export by electricity. Similar to this, the paper published in April 2023, written by Ji et al. [69] presents the implementation of the P-graph approach for the optimisation of multi-period renewable energy systems with hydrogen and battery energy storage. For the developed biomass energy supply scenario, the results indicate that the renewable energy systems with hydrogen storage and battery storage are, respectively, 21.5 % and 5.3 % cheaper than those without energy storage. The developed model investigates CO_2 generation and includes it in the optimisation through the cost of CO_2 emissions.

P-graph application is especially interesting for biomass supply network optimization problems. How et al. [70] developed a decomposition approach for a P-graph application of synthesis of multiple biomass corridors. Stile et al [71] have expanded the use of P-graph-based algorithms to assess the reliability of raw material availability. Malladi et al. [72] have developed a p-graph-based decision support tool for optimizing the short-term logistic of forest-based biomass, by minimizing the biomass logistics cost. Egieya et al [73] used a P-graph framework to optimise the integrated biopower supply network, by maximizing the economic performance. Lo et al. [74] proposed a P-graph based method that considered the incorporation of biomass supply chain uncertainties. Results have shown that a reduction in net present value (NPV) ranges from 1.39% to 12.21% when the biomass shortage scenario was included. Ondruška et al. [75] extended the application of the P-graph approach to perform resource optimization in an aquaponics facility. Aviso et al. [76] implemented a P-graph approach to the development of optimal and sub-optimal biochar-based carbon networks. Here, the objective was to optimise the network in terms of overall carbon sequestered annually, without exceeding constraints on soil contamination. Lam et al. [52] have proposed a model to integrate palm biomass and waste motor oil into the waste-to-energy model. The method to solve the combinatorial of the biomass supply chain in Federal Land Development Authority Jengka was presented by Varbanov et al. [77]. Here, the authors have proposed possible locations for building a new biomass processing facility in the considered region, which should be used for the utilization of waste from oil palm biomass processing. Van Fan et al. [78] applied the P-graph approach to detect cost-optimal and suboptimal pre-and post-treatment pathways for the anaerobic digestion of lignocellulosic waste. The result of the optimisation for the lignocellulosic waste showed that alkali CaO pre-treatment proved to be the cost-optimal pre-treatment option of the lignocellulosic waste, while H2S + membrane separation proved to be the cost-optimal post-treatment (biomethane upgrading) option. Benjamin [79]. developed a P-graph approach to perform a critical analysis of an integrated network of biomass processing industries under scenarios that involve both supply and demand side disturbances. This method enables the reduction of the net product stream output that results from the occurrence of climate change-induced events (supply-side disruptions) and seasonal fluctuations in demand, to be assessed. Vance et al. [80] implemented the P-graph method for the development of economically optimal and suboptimal structures of biomass network that includes corn silage, grass silage, corn straw and wood as feedstock material for combined heat and power (CHP) units. For the obtained results (ranked structures) ecological footprint was assessed, indicating the amount of land required to support and assimilate a given human population's consumption and wastes. The structures whose ecological footprint was lower than the given threshold were considered sustainable structures.

Interest in biomass supply network optimization is evidently increasing. However, to exploit the potential of the waste materials for biogas production, it is crucial to link the availability of biomass and its geographical distribution, with the optimization of the economical performances of a biomass supply network. Furthermore, current models first determine the economically optimal and sub-optimal biomass network and afterwards compare the ecological footprint/environment constraints between optimal and sub-optimal structures. To ensure that the optimal structure is in line with the legislation requirements, it is beneficial to add GHG- related constraints in the optimisation model. Finally, the seasonality of feedstock supply and biogas demand is mostly neglected, although it may have a significant impact on the viability of utilisation of feedstocks with high seasonal fluctuations, such as industrial by-products and agricultural residues.

1.3 Objective and hypotheses of research

Objectives of this research are following. Firstly, to determine the spatial distribution and to calculate the influence of the seasonality of the residual biomass generation. Secondly, to define constraints for which different types of industrial by-products and agricultural residues meet sustainability and greenhouse gas emissions saving criteria. Thirdly, to prove the value of the use of graph theory approach in modelling residual biomass supply network, for different types of case study areas, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria.

The hypothesis of this research is that economically feasible residual biomass supply network for biogas production, that meets sustainability and greenhouse gas emissions saving criteria, could be determined with graph theory approach.

1.4 Scientific contribution

This research has following scientific contributions:

- The novel approach for the assessment of seasonality of technical potential of agricultural residues and industrial by-products for biogas production
- Defined constraints for which different types of industrial by-products and agricultural residues meet sustainable and greenhouse gas emissions saving criteria
- The mathematical model for residual biomass supply network modelling from sustainability, greenhouse gas emissions savings and economic point of view.

2 METHODS

In this section overall modelling approaches and key elements of the conducted research are presented as follows:

1. A Geographical Information System (GIS) based approach for assessing the spatial distribution and seasonal variation of biogas production potential;
2. Analysis of greenhouse gas emissions, greenhouse emission savings and sustainability requirements for the production and use of biogas;
3. Optimisation of biomass supply network with P-graph approach.

2.1 A Geographical Information System (GIS) based approach for assessing the spatial distribution and seasonal variation of biogas production potential

Various methods have been developed for evaluating bioenergy potential. These can be broadly categorized into three groups: statistical evaluation, spatial evaluation, and integrated spatial-statistical evaluation. The statistical evaluation employs data related to land use, agriculture, animal, and industrial production, along with the residue-to-product ratio (RPR). Using this data, the method gauges biomass potential at various statistical units, such as city, county, or country levels. On the other hand, spatial evaluation techniques are rooted in explicitly spatial data, processed through a geographic information system (GIS). The integrated spatial-statistical method merges both techniques, leveraging both statistical and explicitly spatial data. This combined approach is the foundation for this study and will be elucidated in the subsequent subsections. This method is explained in more details in Paper 1 and in Paper 2. The method encompasses the following steps:

2.1.1 Feedstock determination

This work focuses on the assessment of the biogas potential from municipal biowaste, agricultural residues derived from plants (maize stover, wheat straw, barley straw, oat straw, triticale straw, rapeseed straw, soya-beans straw, sugar beet tops, damaged vegetables), livestock production (manure) and industrial residues and by-products. The considered industrial residues and by-products are those which occur in sugar refineries (sugar beet pulp), wineries (grape pressings), tomato (tomato waste), olive oil industry (olive pomace) and breweries (brewer's spent grain).

2.1.2 Assessment of the theoretical potential of considered feedstocks

Theoretical potential of residues from plant production is defined as the annual production of residues generated during agricultural production. As it is shown in equation (1), it is a function of agricultural production and residue to product ratio:

$$P_{th,ag} = y * A * RPP \tag{1}$$

where$P_{th,ag}$ stands for the theoretical potential of residues from the agricultural category (kg), y for the average biomass yield of the agricultural production (kg/m^2), A for utilised land for agricultural production (m^2) and RPP for the residue-to-product ratio for the agricultural category (kg/kg).

In the case of livestock derived residues, the theoretical potential of manure is estimated according to equation (2):

$$P_{th,liv} = N * MPH \tag{2}$$

where $P_{th,liv}$ stands for the theoretical potential of manure (kg), N for the number of heads of livestock (head) and MPH for manure per head ratio: annual manure production per livestock (kg/head).

The theoretical potential of industrial by-products is defined as the annual production of industrial by-products. Similar like for the agricultural production, it is a function of the amount of processed commodities and residue to the processed commodity ratio:

$$P_{th,ind} = M_{p.com} * RPPC \tag{3}$$

where $P_{th,ind}$ stands for the theoretical potential of residues and by-products from industrial production (kg), $M_{p.com}$ for the amount of processed commodities (kg) and $RPPC$ for the residue to processed commodity ratio for a specific commodity (kg/kg).

2.1.3 Assessment of the technical potential of considered feedstocks

Technical potential is defined as the part of the theoretical potential which is available due to competition with other uses (food, feed, land protection etc.). Assessment of this potential is relevant when assessing the potential of agricultural residues, available for the utilisation in bioenergy production. Assessment of this potential is based on the previously calculated theoretical potential, sustainable removal rates and competitive uses (for livestock production), according to equation (4):

$$P_{tech,ag} = P_{th,ag} * SRR - COMP \quad (4)$$

where $P_{tech,ag}$ stands for technical potential of residues from agricultural production (kg), $P_{th,ag}$ for the theoretical potential of residues from agricultural production (kg), SRR for a sustainable removal rate for agricultural category and $COMP$ for the amount of residues which should be left for the feeding and bedding of animals (kg). Sustainable removal rate (SRR) refers to the share of residues which could be collected from the field, by considering the share of the residues which should remain in the field in order to protect the soil from wind and erosion, but also the share which is not possible to collect due to losses in the collecting process.

The amount of residues which should be left for the feeding and bedding of animals ($COMP$) is calculated according equation (5):

$$COMP = \sum N_{(a)} * SPA_{(a)} * s_{(a)} \quad (5)$$

Where $N_{(a)}$ stands for the number of animals (-) in the region, $SPA_{(a)}$ for annual requirements of straw per animal (kg/year) and $s_{(a)}$ for the share of animals to which $SPA_{(a)}$, refers, since not all farms use a straw for livestock production (%).

2.1.4 Assessment of biogas and methane potential

Biogas potential of the considered feedstocks is based on a technical (or theoretical potential, for livestock and industrial by-products) of fresh feedstocks, specific biogas yield from fresh feedstock and methane content of biogas, according to equation (6):

$$E_{bio} = P_{th/tech} * y_{bio} * s_{CH_4} \quad (6)$$

where E_{bio} stands for a methane potential of the considered feedstock (m^3), y_{bio} for a specific biogas yield of specific feedstock (m^3/t) and s_{CH_4} for a share of the methane contained in biogas (%). It is important to note here that biogas yield and methane content depend not only on feedstock type but also on pre-treatment methods and anaerobic digestion conditions.

2.1.5 Seasonal assessment

The seasonal variability of biogas potential from agricultural residues and industrial by-products is identified based on the harvest months (for agricultural residues) and production months (for industrial by-products). For residues and by-products with extended harvest or production seasons spanning several months, it's assumed that production is evenly spread across those months. The occurrence months and their corresponding potentials are specified for each type of agricultural residue and industrial by-product. This is captured as an additional attribute set, with each attribute representing a month of the year and detailing the biogas potential for that specific month. These attributes can be formulated and computed in CSV files since GIS tools support the incorporation of layers in CSV format. Alternatively, these attributes can be directly inputted and computed within GIS tools using the Field Calculator.

2.1.6 GIS mapping

The viability of using residues and by-products economically is often limited by their geographic spread and their proximity to potential biogas facilities. This is particularly the case for smaller industries producing limited quantities of by-products and residues for biogas conversion, as well as regions with low agricultural production. GIS biomass mapping is a specialized application of Geographic Information System technology aimed at evaluating and analysing biomass resources. Through GIS mapping methods, biomass resources can be precisely located, measured, and mapped over a designated geographic span. A key advantage of GIS tools is their capability to merge spatial data with non-spatial attributes.

For effective GIS mapping, the following datasets are essential:

- Production or processing months of the examined feedstocks.

- Specific monthly biogas potential. For agricultural residues, this data is determined at the regional level, whereas, for industrial by-products, it's gauged for individual industrial locations due to the accessibility of data.
- Georeferenced data on region boundaries and georeferenced land use maps (for agricultural residues) or coordinates of the farms and industries in which considered residues and by-products occur (in a case when respective industries are not pre-defined in the map).

QGIS tool [81] is used to conduct the mapping process. Data on a monthly availability of biogas potential of agricultural residues, calculated in previous steps, is joined to the georeferenced layer of regions' boundaries. In order to carry out a spatial distribution of biogas potential in each region, land cover maps are used. Those maps represent georeferenced information on different types (classes) of physical coverage of the Earth's surface, e.g. grasslands, forests, croplands, lakes, wetlands, etc. [82]. Based on two layers of georeferenced information (land cover map and biogas potential at a regional level for each month of the year), a biogas potential map is developed. In order to assess the distribution of the biogas potential, the top-down approach is applied and the following equation is used:

$$\mathrm{E_{bio,fiel,m}} = \frac{\mathrm{A_{fiel}}}{\mathrm{A_k}} * \mathrm{E_{bio,k,m}} \tag{8}$$

Where $E_{bio,fiel,m}$ stands for the energy value of the biogas potential for the specific field in the specific month *m* (MJ) A_{fiel} for the area of the specific field (m^2), A_k for the total agricultural area of the region , in which specific field is located [m^2] and $E_{bio,k,m}$ for the energy value of biogas potential of the region, for the specific month (MJ). The resulting map presents geo-location of biogas potential in each month of the year.

2.1.7 Determination of optimal biogas site location

The optimal location for a biogas plant is determined using geographic and attribute (non-spatial) data. This biogas plant can be understood as a centralised production site that produces biogas from feedstock supplied by the concerned industry, farms and agricultural sites. The goal function of this optimisation is to minimize transport distance between a biogas site and concerned feedstock providers. For this optimisation, the "Mean coordinate" spatial query, available in QGIS was used. As the input data for the optimisation, biogas potential was used as the weighted factor. In a case where biogas potential is represented in both point and polygon vector layers, it is important to align the type of layers and merge those layers into one, which can be used for optimal biogas site location determination. As residues and by-products from livestock and industrial production are being generated at the specific locations, farms and industrial sites are represented in a point vector. In this work, the potential of the agricultural residues, initially represented in the polygon vector layer was transferred to the point layer by using the “Centroids” query in QGIS. The generated points can be understood as the collection sites of agricultural residues.

2.1.8 Route assessment

The "Shortest path" query in QGIS can be used to examine routes (transport distance). This query allows the automatic assessment of the shortest (or fastest, upon user preferences) route between feedstock providers and biogas plants. The input data used for this assessment includes a network layer representing transport routes (roads) in the considered area, a layer representing feedstock providers including the information a respective biogas potential and a layer including the location of the optimal biogas site location. The transport routes (road networks) can be imported to QGIS with the "QucikOSM" plugin. Specific transportation costs can be determined with equation (9):

$$C_{trans} = \frac{d * (K_{full} + K_{empty})}{B_{biogas}} * b * T \tag{9}$$

Where C_{trans} stands for specific transport cost (EUR/GJ), d for transport distance (km), K_{full} for fuel consumption of a full truck (L/km), K_{empty} for fuel consumption of empty truck (L/km), b for fuel price (EUR/L), B_{biogas} for biogas potential of transported feedstock (GJ) and T for transport cost correction factor. In this work, the assumption that T equals 3 was used. This implies that the fuel cost accounts for a third of the overall transportation cost.

2.2 Analysis of greenhouse gas emissions, greenhouse emission savings and sustainability requirements for the production and use of biogas

The method used in this work is based on the method developed by the Joint Research Centre and implemented in Directive 2018/2001 [13]. Directive 2018/2001 includes disaggregated typical and default GHG and GHG saving values for biogas used for the production of electricity and heat for wet manure, maize whole plant (maize silage) and biowaste.

The method can be used for the determination of specific GHG emissions from different solid and gaseous pathways. In this method, the Global Warming Potential (GWP) is used as the climatic metric. Global Warming Potential is a term used to describe the relative potency, molecule for molecule, of a greenhouse gas, taking into account how long it remains active in the atmosphere [83]. As defined in the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report [84], the GWP of methane is equal to 25 and for nitrous oxides is 298, for a time period of 100 years.

Biogas feedstocks analysed are those presented in the previous subsection. This method is described into more detail in Paper 3. The general equation used to calculate greenhouse gas emissions from the production and use of biogas:

$$E = e_{ec} + e_l + e_p + e_{td} + e_u - e_{sca} - e_{ccs} - e_{ccr} \tag{10}$$

Where is:

E - total emissions from the use of the fuel (gCO_2eq/MJ);

e_{ec} - emissions from the extraction or cultivation of feedstocks (gCO_2eq/MJ);

e_l - annualised emissions from carbon stock changes caused by a land-use change (gCO_2eq/MJ);

e_p - emissions from processing (gCO_2eq/MJ),

e_{td}- emissions from transport and distribution (gCO_2eq/MJ);

e_u- emissions from the fuel in use (gCO_2eq/MJ);

e_{sca} - emission reduction from soil carbon accumulation due to improved agricultural management (gCO_2eq/MJ);

e_{ccs} - emission savings from CO_2 capture and geological storage (gCO_2eq/MJ);

e_{ccr} - emission savings from CO_2 capture and replacement (gCO_2eq/MJ).

In the subsections below, each emission factor is described in greater detail.

Emissions from the extraction or cultivation of feedstocks-e_{ec}

Emissions from the collecting, drying, and storage of feedstocks, waste, leaks, as well as the production of chemicals or goods used in extraction or culture, are all included in the definition of emissions from the extraction or cultivation of feedstocks [13]. These emissions apply when agricultural feedstocks like palm, maize, sugarcane, soybean or rapeseed are extracted or grown. The emissions from the cultivation or extraction of feedstocks are regarded as zero in situations when residues, by-products, and waste materials are utilised as feedstocks for the generation of biogas [20].

Annualised emissions from carbon stock changes due to land-use change-e_l

By averaging emissions from carbon stock changes over a 20-year period, annualised emissions from carbon stock changes caused by a change in land use are determined [13]. Any alteration in the carbon stock between the classified land categories of grassland, forestland, cropland, wetland, settlements and other lands is considered as a land-use change [85]. The calculation is based on equation (11) [13]:

$$e_l = \frac{(CS_R - CS_A)}{P * 20} * 3.664 - e_B \tag{11}$$

Where is:

CS_R-the carbon stock per unit area corresponding to the reference land use. The reference land use is the land use as of January 2008 or as of 20 years prior to the receipt of feedstock, whichever was more recent. It is quantified as a mass of carbon per hectare (gC/ha), which includes both soil and vegetation;

CS_A –the quantity of carbon stored per area corresponding to the actual land use. It is quantified as a mass of carbon per hectare (gC/ha), which includes both soil and vegetation;

P- the crop's productivity, expressed as the amount of biofuel produced per hectare per year (MJ/ha*yr);

e_B-bonus of 29 gCO_2eq/MJ, applied for the case when biomass is obtained from restored degraded land (gCO_2eq/MJ);

Emissions from processing-e_p

Emissions from processing refer to emissions that result from the actual processing itself, waste generation, product leakage, and the production of chemicals and other processing-related products. In addition, regardless of whether fossil fuel inputs are burned during processing, these emissions also include CO_2 emissions proportional to their carbon content. The benchmark for measuring the amount of emissions of greenhouse gases caused by electricity not produced on a biogas site is the average emission intensity of electricity production and distribution in a given area. Regardless of whether they are processed into intermediate products before being converted into the end product, agricultural residues and industrial by-products are regarded to have zero life-cycle greenhouse gas emissions up until the collection process [13].

The agricultural residues having a bulk density of less than 0.2 tonne/m^3 require a processing step before prior transportation: baling or additional grinding or clustering. This can be represented by a single process [86], presented in Table 1.

Table 1 Emissions from bailing/processing agricultural residues

Baling/processing				
Input	**Output**	**Unit**	**Amount**	**Source**
Agri-residue	-	MJ/MJbale	1.0	[86], [87]
Diesel	-	MJ/MJbale	0.010	[86], [87]
-	Bales	MJ	1.0	[86], [88]
-	CH4	g/MJbale	1.23E-05	[86], [88]
-	N2O	g/MJbale	3.03E-05	[86], [88]

Emissions from transport and distribution-e_{td}

GHG emissions from the transport and distribution, e_{td}, should include all transport and distribution steps in the value chain. e_{td} is calculated based on the following equation [85] :

$$e_{td} = \frac{\left(d_{loaded} * K_{loaded} + d_{empty} * K_{empty}\right) * EF_{fuel}}{E_{bio.feedstock}} \tag{12}$$

Where is:

d_{loaded} - transport and distribution distance of loaded truck (km);

K_{loaded} - fuel use of loaded truck (l/km);

d_{empty} - transport and distribution distance of empty truck (km);

K_{empty}- fuel consumption of empty truck (l/km);

EF_{fuel}- fuel's emission factor (gCO_2eq/l);

$E_{bio.feedstock}$ - biogas potential of loaded biomass feedstocks (MJ).

According to the JRC report [86], it is assumed that a 40 t truck will be used to deliver the feedstock to biogas sites (27 t payload).

Biogas potential of loaded biomass feedstocks, $E_{bio.feedstock}$, is calculated by equation (13):

$$E_{bio.feedstock} = \rho_{feedstock} * V_{truck} * y_{biogas} * s_{CH_4} * LHV_{CH_4} \tag{13}$$

Where is:

$\rho_{feedstock}$ - bulk density of feedstock (t/m^3);

V_{truck}- truck load capacity (m^3);

y_{biogas} -biogas yield from 1 tonne of fresh feedstock (m^3/t);

s_{CH_4}– methane content of biogas (%);

LHV_{CH_4}-methane lower heating value (MJ/m^3).

It is important to highlight, that for feedstocks with a bulk density greater than 0.75 t/m^3, the feedstock load is constrained by weight, while for bulk densities less than 0.75 t/m^3, it is volume constrained. The bulk density of agricultural residues can be increased by 8-12 times at different bailing/ briquetting process parameters [89].

Emissions from the fuel in use-e_u

Emissions of the fuel (biogas) in use, e_u, are considered to be zero for biofuels (biogenic CO_2 combustion emission). However, e_u factor should take into account the emissions of non-CO_2 greenhouse gases (CH_4 and N_2O) of the fuel in use [13], [90].

Emission savings from soil carbon accumulation via improved agricultural management-e_{sca}

Emission savings from soil carbon accumulation via improved agricultural management refers to the practice that results in an increase in soil carbon. Those savings can be calculated only in case of improved manure management, shifting to minimal or zero-tillage; use of compost or improved crop rotations [91]. To assess those savings, equation (11) can be used, where 20 years should be replaced by the period of time (in years) of the actual period.

Emission savings from CO_2 capture and geological storage-e_{ccs}

Emission savings from CO_2 capture and geological storage e_{ccs} include averted emissions through CO_2 capture and geological storage directly associated with the extraction, transportation, processing, and distribution of fuel [13]. They can only be considered if it can be proven that the current storage ensures that the leakage does surpass the current state of technology [85].

Emission savings from CO_2 capture and replacement-e_{ccr}

Savings on emissions from CO_2 capture and replacement (e_{ccr}), are only possible when CO_2 that comes from biomass is captured and utilised to replace CO_2 that comes from fossil fuels in the creation of goods and services for sale [5]. The savings can be included in the overall calculation, only if it can be proven that CO_2 replaces CO_2 that comes from fossil sources and is employed in the production of goods and services for commerce [85].

2.2.1 Calculation of greenhouse gas emissions from heat and electricity

In this study, the assumption used is that biogas produced from considered feedstocks will be used in cogeneration (CHP) plants, for the production of electrical and thermal energy. In order to allocate emissions to each final energy commodity, the following equations are used [13]:

$$EC_{el} = \frac{E}{\eta_{el}} \cdot \left(\frac{C_{el} \cdot \eta_{el}}{C_{el} \cdot \eta_{el} + C_h \cdot \eta_h} \right) \tag{14}$$

$$EC_h = \frac{E}{\eta_h} \cdot \left(\frac{C_h \cdot \eta_h}{C_{el} \cdot \eta_{el} + C_h \cdot \eta_h} \right) \tag{15}$$

Where is:

EC_{el} - total GHG emissions associated to electrical energy (gCO_2eq/MJ);

EC_h - total GHG emissions associated to thermal energy (gCO_2eq/MJ);

η_{el} -electrical efficiency, determined as the annual electrical energy output divided by the energy content of annual fuel input (%);

η_h - heat efficiency, determined as the annual useful thermal energy output divided by the energy content of the annual fuel input (%);

C_{el} - a fraction of exergy in the electricity (-) For the electricity, the fraction of exergy is set to 100%;

C_h - a fraction of exergy in the useful heat, calculated as Carnot efficiency (-). It is defined as:

$$C_h = \frac{T_h - T_0}{T_h} \tag{16}$$

Where is:

T_h - temperature of the useful heat at the point of delivery (K);

T_0 - environmental temperature, set at 273.15 K (K).

2.2.2 Calculation of greenhouse gas emissions savings from heat and electricity generated from biogas

The following equations define how greenhouse gas emissions from heat and electricity for respective fossil fuel comparators in order to calculate GHG savings obtained from heat $GHG\ SAVINGS,_{heat}$ and electricity $GHG\ SAVINGS,_{electricity}$ generated from biogas:

$$GHG\ SAVINGS,_{heat} = \frac{EC_{F(h)} - EC_h}{EC_{F(h)}} \tag{17}$$

$$GHG\ SAVINGS,_{,electricity} = \frac{EC_{F(el)} - EC_{el}}{EC_{F(el)}} \tag{18}$$

Where is:

$EC_{F(h)/F(el)}$ - total emissions from the fossil fuel comparator for useful thermal energy/electrical energy (gCO_{2eq}/MJ);

$EC_{h/el}$ - total emissions from the useful thermal energy/electrical energy generated from biogas (gCO_{2eq}/MJ);

The values of fossil fuel comparators equal 183 gCO_{2eq}/MJ for electrical energy and 80 gCO_{2eq}/MJ for useful thermal energy.

2.2.3 Sustainability requirements

In addition to the GHG emissions saving requirements, additional requirements must be met to ensure the sustainable utilization of biomass for biogas production. Those requirements are defined in numerous directives and legislation documents, such as in Directive 2018/2001. There, it is stated that biogas feedstocks should not be produced from raw materials obtained from land with a high biodiversity value, such as primary forests, areas for the protection of rare or endangered ecosystems or species, highly biodiverse grasslands, and wetlands, etc. and must adhere to the criteria for forestry, land use, and land-use change (LULUCF) [13].

2.3 Optimisation of biomass supply network with P-graph approach that meets greenhouse gas saving and sustainability requirements

Selection of the approach used for the optimisation of biomass supply network that meets greenhouse gas saving and sustainability requirements may depend based on the seasonality of biomass supply network and number of options in the biomass supply network. In the scope of this thesis, two approaches within these objectives have been developed.

The first approach is aimed for more complex biomass supply networks, as it is aimed to include the seasonal variation of feedstock production, as well as different types of options in the biomass supply network, which result in different GHG emission production. This approach is explained in detail in Paper 4. On the other hand, the second approach tailored for feedstocks that are available year-round and have a limited set of options influencing GHG emission. This approach is explained in detail in Paper 5. The first approach is more appropriate for the feedstocks that occur in rural areas, while the second is more appropriate for feedstocks that occur in urban areas. In both approaches, the first part of the approach is focused on the GIS mapping, while the second part is focused on P-graph based optimisation. Due to higher complexity of the first approach, it will be explained in detail in the following paragraphs, while the main difference between those two approaches will be outlined at the end of this section.

In the first approach, input data obtained with GIS tools on seasonal feedstock availability and transport distance, as well as the requirements on greenhouse gas savings and sustainability were used as the input data for the optimisation of the biomass supply network with the P-graph approach. Here, the objective is to develop a mathematical model that defines an economically optimal structure which satisfies the sustainability requirements and limits the GHG emissions of the final optimal and sub-optimal structures, while considering the seasonality of feedstock supply and biogas demand. The method is graphically represented in the flowchart presented in Figure 3.

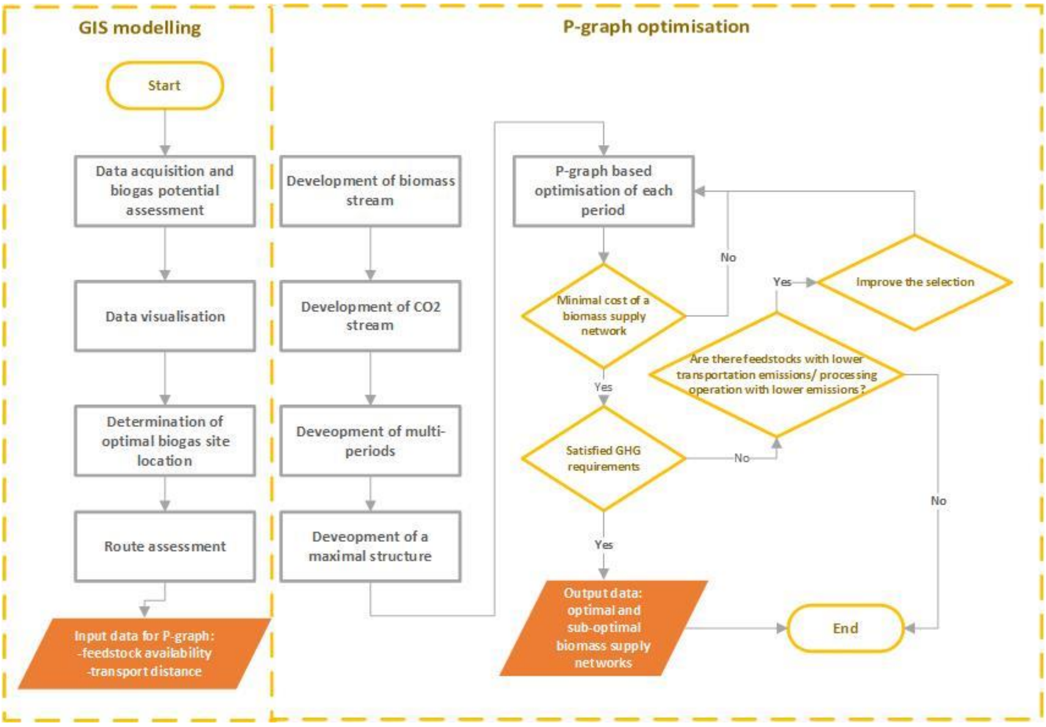


Figure 3 Flowchart representation of the method for the first approach

2.3.1 P-graph approach and P-graph based algorithms

Due to the combinatorial nature of the problem, biogas production can be accomplished by a wide range of alternative structures. The determination of the optimal network structure is most frequently referred to as process-network synthesis (PNS) flowsheet design. The P-Graph framework is a highly effective tool designed to address the challenges associated with solving Process Network Synthesis (PNS) problems [92]. P-graphs are bipartite graphs composed of material and operating unit nodes, with material flow represented by arcs. Figure 4 presents an example of a P-graph.

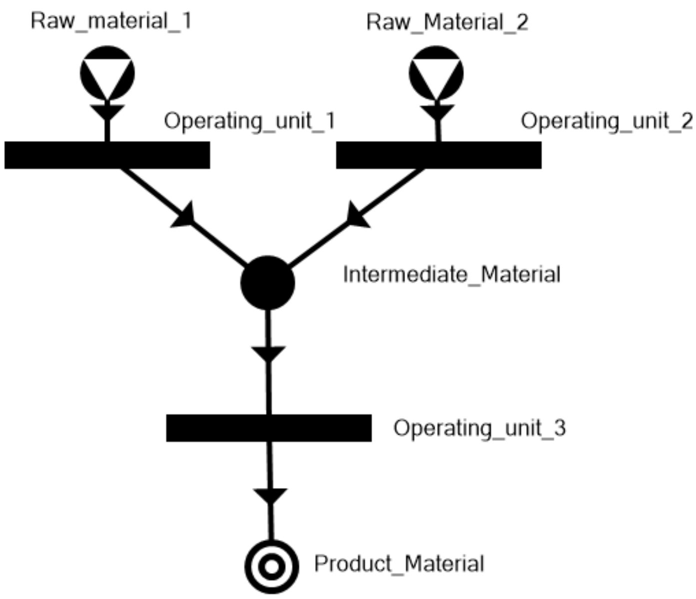


Figure 4 Example of a P-graph

In P-graphs, raw materials are used as the input for the operating units, which correspond to a unit that perform different types of operations. As can be seen from Figure 4, an output material is an intermediate material, which is used as the input for the operating unit that produces as output Product material (final product). It is worth noting that while Figure 4 portrays a fundamental P-graph instance, real-world scenarios tend to exhibit significantly greater complexity.

For solving P-graph based problems, different software has been developed, such as P-graph Studio. Within those tools, there are integrated combinatorial algorithms that enable the identification of solution, optimal and sub-optimal structures for these problems. Determination of the feasible structures is being performed in three major steps.

In the first step, the maximal structure of feasible solutions for biogas production is developed. The maximal structure comprises all the combinatorically feasible structures capable of yielding the specified products from the specified raw materials. The feasible solution structure generated by process-network synthesis must have several basic features that are taken as axioms, the introduction of which improves the efficiency of the combinatorial search during the process. In the P-graph-based methods, the algorithm MSG (Maximal Structure Generation) yields the maximal structure, i.e., the superstructure, for the Process Network Synthesis (PNS) problem. MSG Algorithm is a polynomial algorithm based on the axioms which define representations of the final product, interim products, raw materials, operating units and arcs. Those axioms are explained in detail by Friedler et al. [93]. The maximal structure will be analysed in the second step. Here, algorithm SSG (Solution Structure Generation) will be used for the generation of all the solution structures representing the combinatorically feasible flowsheets from the maximal structure. Algorithm SSG systematically and combinatorically selects a series of active sets and carries out decision mappings. Finally, ABB (Accelerated Branch and Bound) algorithm will be used to generate the n-best feasible solution structures. Algorithm ABB is a branch and bound algorithm for solving combinatorial problems. It traverses the maximal structure, keeping track of all partial solutions in corresponding tree branches and bounding until it finds a branch whose objective function is better than the current best solution.

2.3.2 Biomass supply network design

The first step in creating a P-graph for a biomass supply network is to identify the potential feedstocks that can be used in the considered area and to map out the transportation network. This data (type of feedstock, technical potential, biogas potential transport distance) were exported from the GIS tool in the previous steps.

Material nodes are representing raw materials (feedstock), interim materials and the final product (biogas). Operating unit nodes are representing biomass transport, biomass processing and anaerobic digestors. Anaerobic digesters are enclosed structures where the anaerobic breakdown of raw material (feedstock) takes place. The biomass supply network developed in this paper is presented in simplified form (for only one input raw material) in Figure 5.

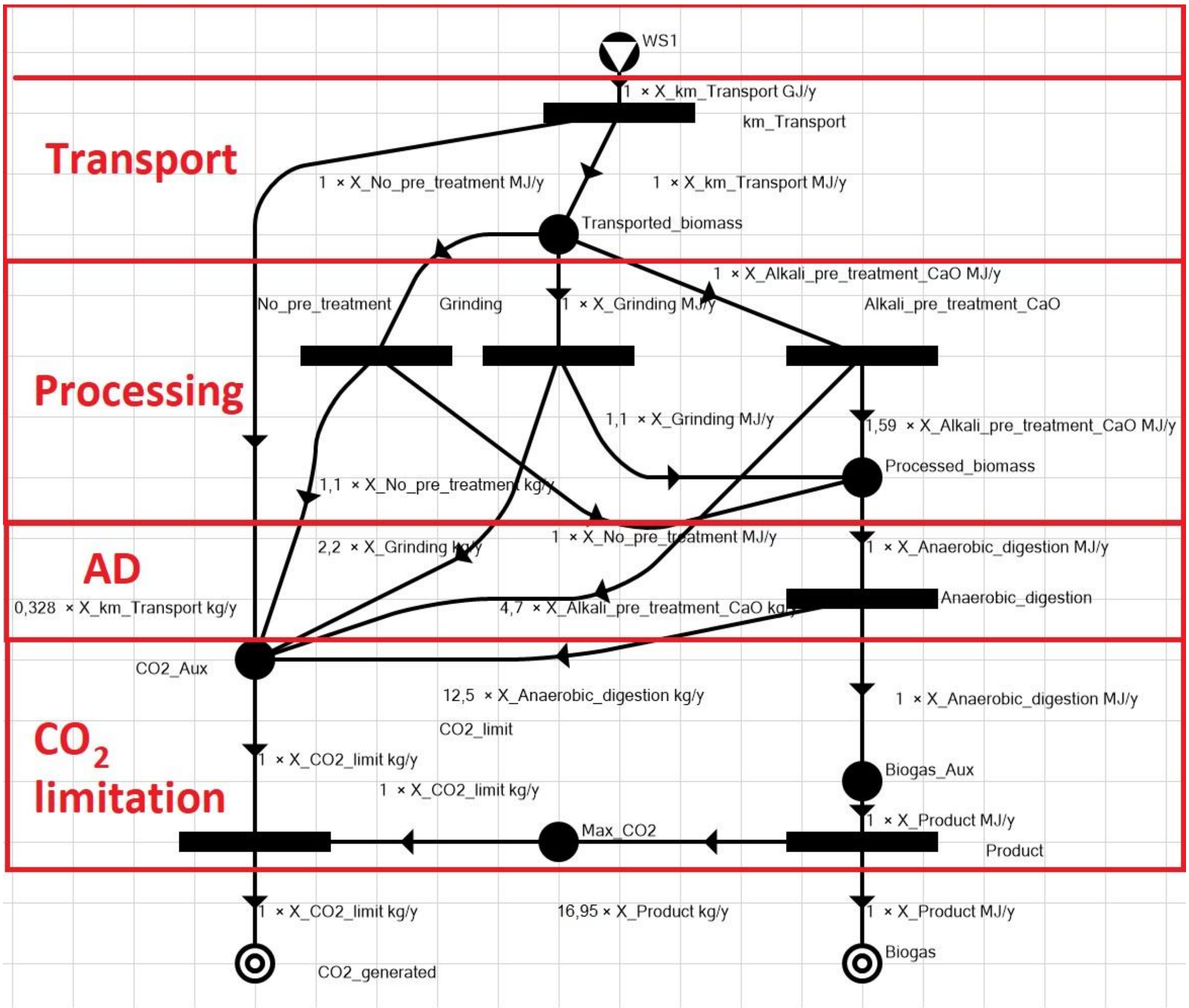
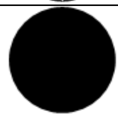


Figure 5 PNS network of the utilisation of wheat straw for biogas production

Elements included in the PNS network are presented in Table 2.

Table 2 P-graph representation of elements included in the PNS network

Element	**P-graph representation**
Raw materials: Feedstock collection sites	
Intermediate products: Transported biomass/Processed biomass Auxiliary products: Maximal allowed CO_2 (Max CO_2), Summarised CO_2 generation (CO_2_Aux), Summarised Biogas production (Biogas_Aux)	
Operating units: Transport; Pre-processing: No pre-treatment/ Grinding and bailing/ Alkali pre-treatment; Anaerobic digestion (AD) Auxiliary units: CO_2 limitation (CO_2 limit)/ Biogas limitation (Product)	
Final products: Biogas/ Generated CO_2 (CO_2 gen)	

As illustrated in Figure 5, the PNS network developed for the utilization of considered feedstocks (with wheat straw taken as an example) incorporates two primary streams: the biomass stream and the CO_2 stream. In terms of the biomass stream, it pertains to the feedstock being transported, processed, and then used in an anaerobic digester for biogas production. Conversely, the CO_2 stream addresses the emissions related to biomass transport and processing, as well as the emissions stemming from biogas utilization (here depicted as emissions linked to the anaerobic digester). In developing the CO_2 streams, special care was taken to ensure that all greenhouse gas emissions (comprising both CO_2 and non-CO_2 emissions) were accounted for and evaluated in accordance with the method laid out in Directive 2018/2001 [94]. This guarantees that the resultant structures (both optimal and suboptimal) align with the 80% GHG savings stipulated in Directive 2018/2001, which will be legally mandatory for biogas facilities commencing operations from 2026 onwards. The model employs a threshold value, specifically the calculated net emissions of 16.95 gCO2/MJ for biogas [95].

Within Figure 5, the phases of biomass and CO_2 streams are denoted by red squares. Each phase encompasses operating units, with the subsequent outputs characterized as intermediary materials, except for biogas, which stands as the final product. Two particularly noteworthy segments of the PNS network are the feedstock processing phase and the section indicating CO_2 limitations. In the context of wheat straw processing, three alternatives are showcased: nonprocessing (only encompassing field baling), grinding, and alkali pre-treatment using CaO. Each choice influences the biogas yield derived from the respective feedstock. As demonstrated by Van Fan et al. [78], grinding enhances lignocellulosic waste's biogas yield by 10%, whereas alkali pre-treatment boosts it by 59% in comparison to the no pre-treatment scenario. However, both grinding and alkali pre-treatment also increase pre-processing costs and linked GHG emissions. The arcs in the diagram highlight these variances, with alkali pre-treatment bearing the highest expenses and emissions. For cases like this, where final cost and total GHG emissions depend on numerous factors, it is very beneficial to conduct a P-graph optimisation. Lastly, it's crucial to emphasize that the biogas potential of feedstocks aligns with the standard biogas yield (in situations without pre-treatment) and not the feedstock's inherent energy value derived from its chemical composition.

Part of the PNS network representing CO_2 limitation sets the threshold for GHG generation (and resulting savings) of the production of biogas and its use. For this purpose, two auxiliary products (intermediate materials) are included in the PNS network design- CO_2_Aux and Biogas_Aux. The maximal flow of those two auxiliary products is set to zero, indicating that they are completely consumed. As can be seen from Figure 5, CO_2_Aux summarizes all of the G emissions generated by processes represented by operating units. Biogas_Aux is used for setting the threshold (maximum) on GHG emissions that the use of fuel (biogas) can generate to be in line with the GHG savings. This limit is represented in PNS Network as Max_CO_2. During the optimisation process, if CO_2_Aux emissions are higher than Max_CO_2 emissions, the P-Graph Studio makes a new iteration to find a structure whose emissions are lower than Max_CO_2.

The cost of a PNS network includes the sum of the cost of the raw materials, the cost of the transport and processing cost. As the main objective is to evaluate the economic viability of different structures, so the costs of anaerobic digestion are excluded, given they're presumed consistent across all feedstocks in question. The goal function of the optimisation is to minimise the cost of a biogas supply network (structure). The optimal solution is the one that ensures the necessary biogas output at the lowest cost while adhering to the set GHG emission generation limits. Alongside the optimal solution, the top n- best solutions will also be ranked.

2.3.3 Multi-period P-graph Optimisation

To incorporate the seasonal variation of biomass supply and biogas demand during the year, the model was extended to multi-periods. The multi-period P-graph modelling allows dividing a year into custom-selected periods of time, which can be of arbitrary length. A multi-period optimisation approach, provides more reliable data compared to a single-period model, as it takes into account fluctuations of inputs (feedstocks) supply during the year, as well as differences in output (biogas) demand throughout the year. For the considered problem, periods are selected based on the availability of considered feedstock types. Hence, months with equal feedstock supply are grouped into the same period. The multi-period extension was implemented by configuring the Multiperiodic settings of the PNS network using P-Graph Studio.

2.3.4 Utilisation of GIS tools for obtaining input data for P-graph optimisation that meets GHG saving requirements.

This subsection describes the second approach referenced earlier in this section and showcased in Paper 5. A graphical representation of this method can be seen in Figure 6.

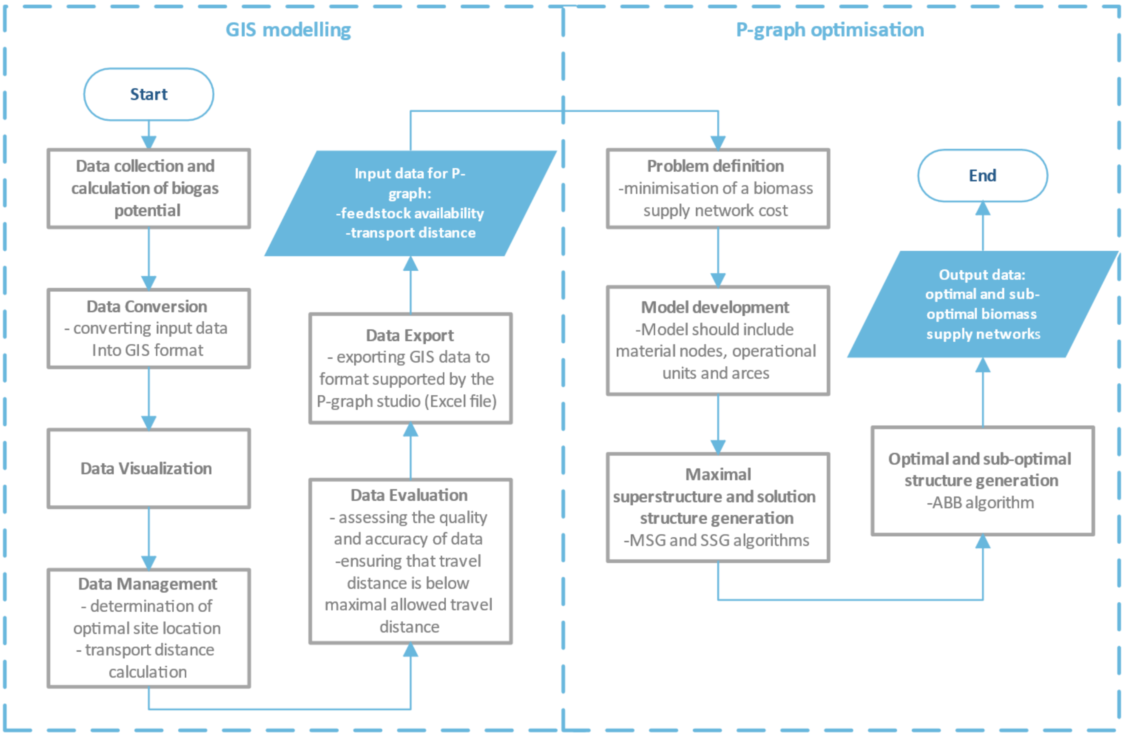


Figure 6 Flowchart representation of the method for the second approach

As can be seen from Figure 6, this approach integrates many elements and steps highlighted in the first approach. The primary distinction is that the P-graph approach formulated here focuses solely on the biomass stream, excluding the GHG emission stream. Instead of incorporating the GHG emission stream in the structure development, the step to limit maximum GHG emissions in the biomass supply network is executed during the initial GIS mapping phase, *Data evaluation* step. In the scope of this step, the evaluation of maximal transport distance is implemented, to ensure that minimum GHG emission savings will be reached in biogas production and use.

As highlighted in the *Introduction* and prior sections, from 2026 onwards, biogas plants must achieve a minimum GHG savings of 80% compared to a fossil fuel comparator. In scenarios where GHG savings solely depend on transport distance, with other categories maintaining a constant value (typically in cases with fewer options in the biomass supply chain), the minimum GHG savings will be attained if the maximum transport distance remains under the values determined by the method discussed in the previous subsection, titled *Analysis of Greenhouse Gas Emissions, Emission Savings, and Sustainability Requirements for Biogas Production and Use*. To guarantee adherence to this transport distance threshold, it's essential to assess the distance for each feedstock provider site under consideration. It's also crucial to recognize that the maximum transport distance varies among feedstock types and should be determined in accordance with the methodology outlined in Directive 2018/2001. This assessment is carried out in two stages. Initially, data regarding the maximum permissible transport distance (calculated for each feedstock category) is linked to each feedstock category. Subsequently, feedstocks are grouped and compared to the maximal distance constraint using GIS queries like "Select Features Using an Expression" and the "Field Calculator".

Data gathered using the GIS tool will subsequently be employed as input for the P-graph optimization. Hence, GIS data should be exported to a data format supported by the P-graph studio, which is Excel file format. The first step for this is exporting data from QGIS to comma-separated values (CSV) file format, which is a plain text format that stores tabular data with each row representing a feature and each column representing an attribute. This process allows users to extract and transfer attribute data from spatial layers in QGIS for further analysis or sharing with other software or users.

3 SELECTED RESULTS AND DISCUSSION

This section presents the main results obtained in the scope of this doctoral thesis. They are based on the five published papers, available in the Annex of the thesis. Three of them are published in Q1 journals, one in Q2 journal and one in Q3 journal.

3.1 GIS mapping of the spatial distribution and seasonal variation of biogas production potential from agricultural residues, livestock production and municipal biowaste

GIS mapping of the spatial distribution and seasonal variation of biogas production potential from agricultural residues, livestock production and municipal biowaste was applied in the case study for the Republic of Croatia. Prior to GIS mapping, biogas potential was assessed for each Croatian county (NUTS3 region). Data provided by Paying Agency for Agriculture, Fisheries and Rural Development [96] and the Croatia Bureau of Statistics [97] was used for calculating theoretical biomass potential in each of the regions. The input data for assessing the potential of manure was taken from the register of domestic animals [98] and the list of utilised agricultural land and numbers of poultry of private households [99]. In order to assess the spatial distribution of the biogas potential, CORINE land Cover map [100], which defines 42 different land classes was used to detect agricultural land, urban areas and dump sites.

The results shown that manure and municipal biowaste have nearly continuous production during a year. Therefore, their seasonal variation can be neglected and biogas potential from municipal biowaste and manure for one average month is presented in Figure 7.

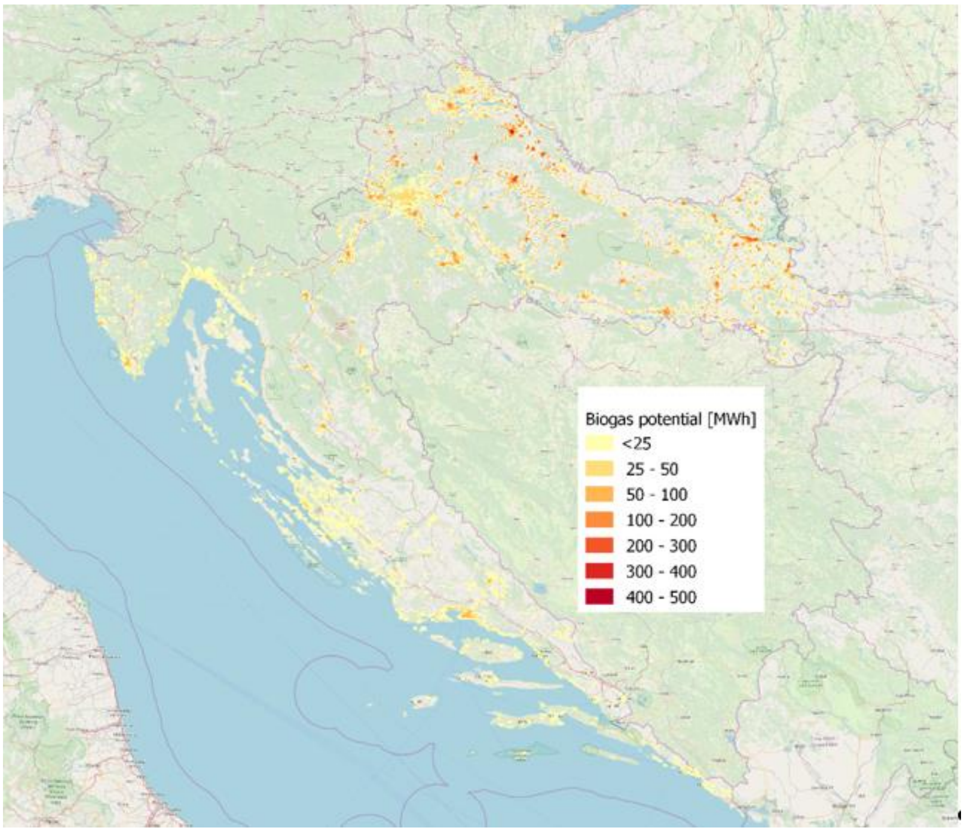


Figure 7 Biogas potential from municipal biowaste and manure for one average month (per 1 km^2)

Figure 7 clearly shows that biogas potential from biowaste and manure is mostly located in the continental part of Croatia, in rural areas. This can be confirmed by the fact that the city of Zagreb, which has by far the greatest population in Croatia, has the lowest density of biogas potential and lowest total biogas potential. On the other hand, biogas potential in Adriatic part of Croatia mostly follows the population density.

From Figure 7, it is evident that the biogas potential derived from biowaste and manure is predominantly concentrated in the continental region of Croatia, specifically in its rural zones. This observation aligns with the fact that despite Zagreb, Croatia's most populous city, it reflects the lowest biogas potential both in terms of density and overall volume. Conversely, in the Adriatic region of Croatia, the biogas potential largely mirrors the population density.

The agricultural by-products examined in this study are predominantly lignocellulosic biomass, including maize stover, wheat straw, barley straw, oat straw, triticale straw, rapeseed straw, and soya-beans straw. These specific lignocellulosic materials are available for only three months annually. Consequently, the spatial distribution of their biogas potential was assessed for each month of their production and is presented in Figure 8.

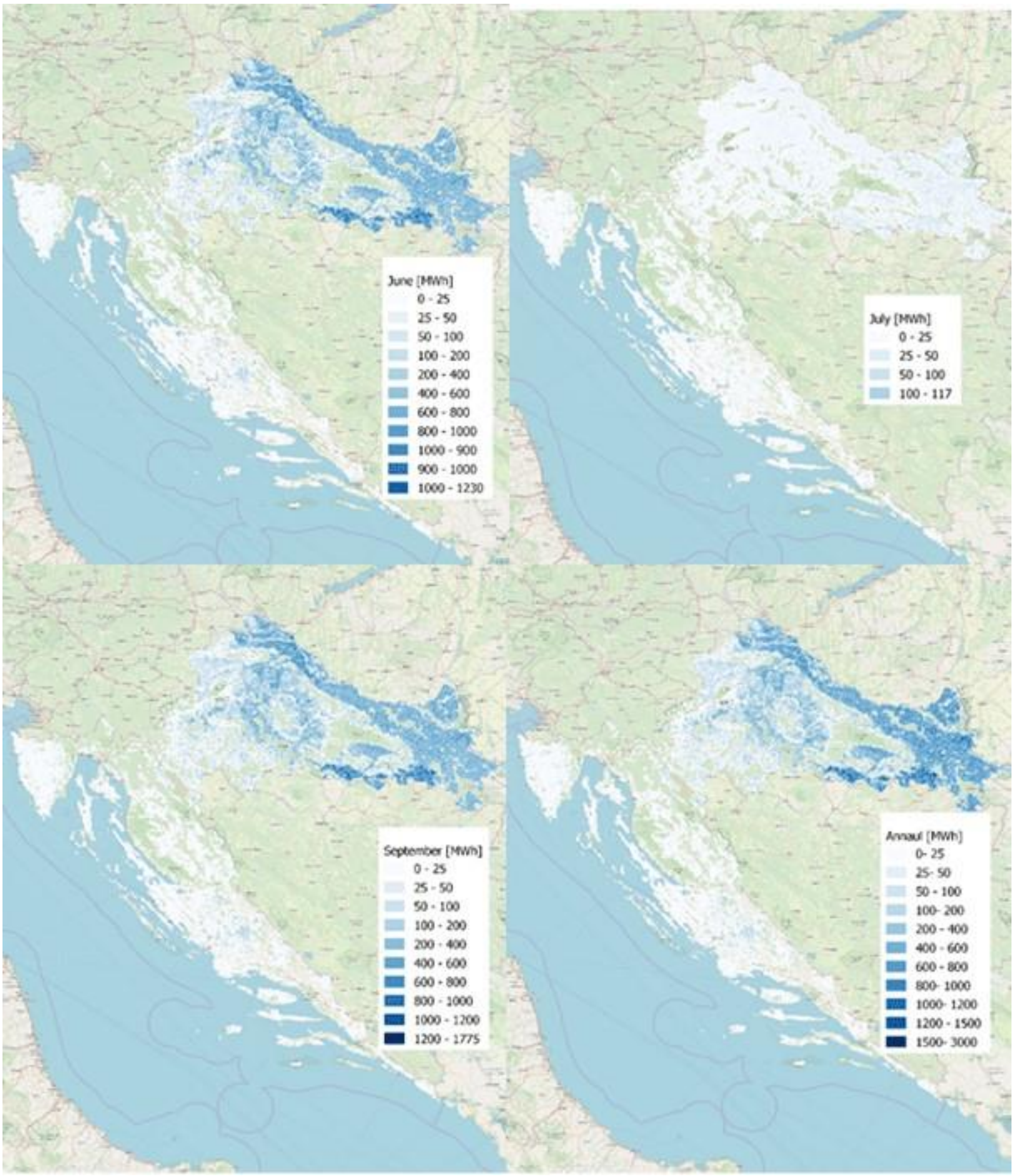


Figure 8 Annual and monthly biogas potential from lignocellulosic agricultural residues (per 1 km^2)

As can be seen from Figure 8, it is evident that Croatia possesses substantial biogas production potential from lignocellulosic agricultural by-products. Yet, comparing the potential presented in Figure 7, to that in Figure 8 it becomes apparent that utilisation of lignocellulosic agricultural residues requires extensive storage capabilities, given that these feedstocks are produced only over a three-month span each year.

To demonstrate the advantage of the proposed approach and calculate the influence of the seasonality of the residual biomass generation, it was compared with currently used approaches. Therefore, required storage capacities were assessed for two examples selected from the area presented in Figure 8, for which spatial and seasonal assessment was conducted in the previous steps. For both examples, storage facility capacity is calculated for the lignocellulosic agricultural residues which are being produced in the area of 90 km^2 (a grid encompassing 90 cells. To mitigate the risk of feedstock supply shortages, the minimum end-of-month stockpile is designated to meet the feedstock needs for at least one and a half months. Moreover, for both case studies, the year-round demand for feedstocks is presumed to be consistent. One of these case studies, situated in Varaždin county (in northern Croatia), is elaborated upon here and visualized in Figure 9.

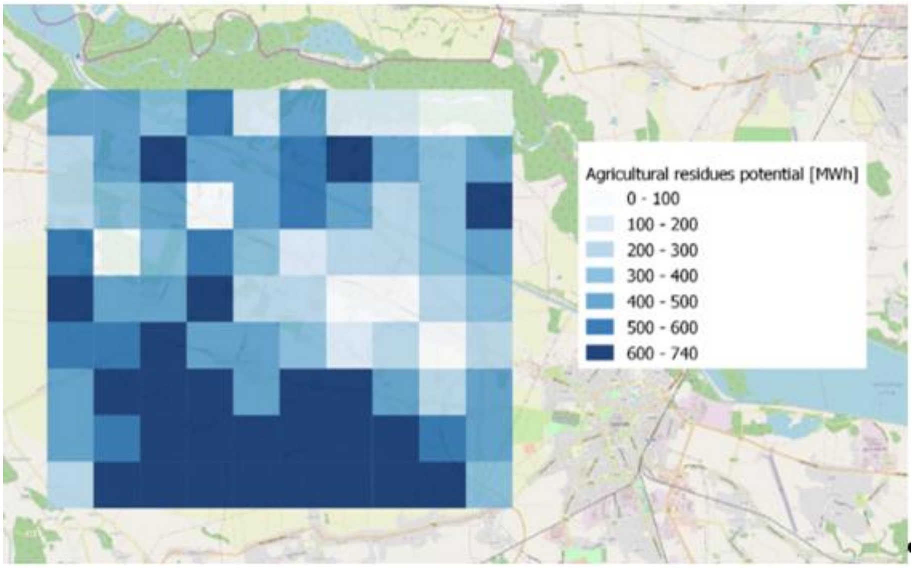


Figure 9 Annual potential of agricultural residues- Varaždin county Example 1)

In the given example, annual technical potential equals 14105 tonnes of agricultural resides, which has the biogas potential of 0.143 PJ (39952 MWh). Annual variations of biomass potential (supply) and stored quantities are presented in Table 3.

Table 3 Seasonal variation of biomass potential (supply) and stored amount

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Supply [t]	-	-	-	-	-	1,774	508	-	11,823	-	-	-
Demand [t]	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1175	1,175	1,175	1,175
Stored amount [t]	7,708	6,532	5,357	4,181	3,006	3,605	2,937	1,762	12,410	11,234	10,059	8,883

*feedstock amount stored at the end of the month

As it is shown in Table 3, the peak feedstock storage size is in September and it equals 12410 t. Thus, 12410 t can be considered as the required storage facility capacity. In other cases where biomass availability is assessed at the annual basis and there is no information on the seasonal variation, it is assumed that the value of necessary storage facility capacity is the same as the annual biomass potential. When comparing the storage facility calculated with this approach to the one related to the annual assessment, it is evident that for the given example, application of seasonal assessment results in 12% lower storage facility capacity. In other examples, the application of seasonal assessment may result in u to 40% lower storage facility capacity [101].

Similar research was performed developed by Popp et al. [102] where authors investigated the monthly availability of crop, horticultural and forestry residues, enabling seasonal assessment of biomass potential on a regional basis. Due to the wide geographic distribution of the agricultural residues and the low energy density of the considered feedstocks, information on the biomass monthly availability on a regional level is often not sufficient for the determination of the feasibility of biogas utilization.

Assessment of the storage facility capacity is a part of the optimization of biomass supply chain in some of the research works, such as research work developed by Ahlgren et al [45]. In this work, authors have determined the storage facility capacity in accordance with the price of the land unit where the storage unit is planned to be built, but with the constraint that capacities of all storage facilities should equal to the annual biomass potential in the considered regions. As it is shown from the example given above, the approach presented in this research work results with lower storage facility capacity, due to better insight into the biomass availability. This shows the importance of including integrated seasonal and spatial variation in the assessment of the potential of lignocellulosic residues available for biogas production, in order to have more accurate input data for the feasibility projects for biogas utilization.

3.2 GIS mapping of the spatial distribution and seasonal variation of biogas production potential from industrial residues and by-products

GIS mapping of the spatial distribution and seasonal variation of biogas production potential from industrial by-products and residues was performed for biogas feedstocks, which occur in sugar refineries, wineries, tomato and olive oil industry (olive oil mills). The presented method was demonstrated in the case study of Istria county and Osijek- Baranja county. Istria County is the westernmost county of the Republic of Croatia and the largest peninsula of the Adriatic. Meanwhile, Osijek-Baranja County lies in the northeastern segment of Croatia. These counties were chosen to showcase a range of industrial productions. The spatial and seasonal distribution of biogas potential in Istria county is presented in Figure 10.

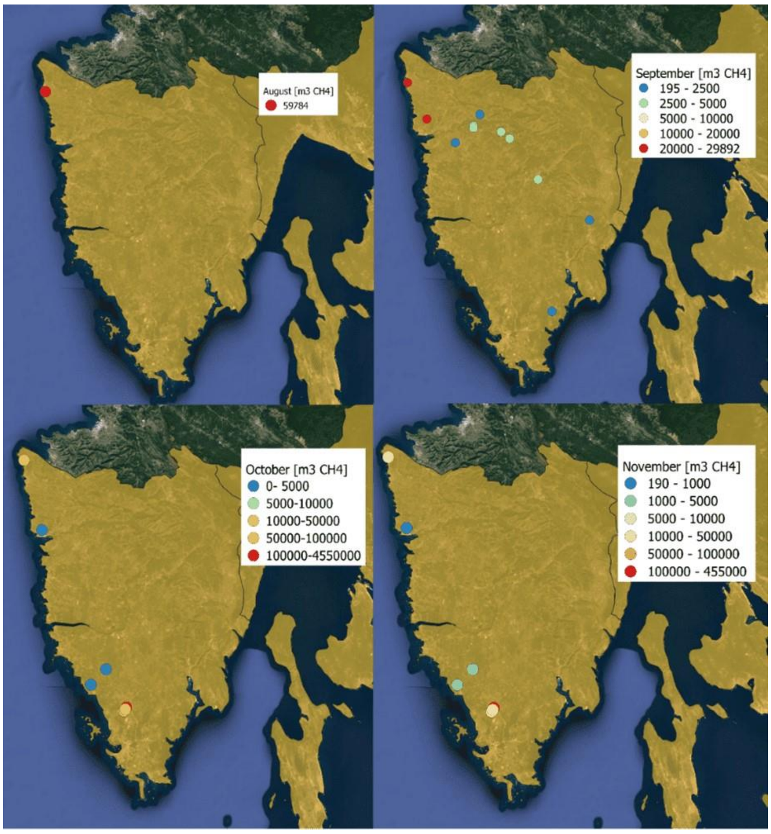


Figure 10 Biogas potential from wineries, olive mills and tomato industry in August, September, October and November

Within this county, the vegetable processing facility, along with several olive oil mills and wineries, presents significant biogas production potential. By-products from the vegetable sector (specifically from tomato processing) are generated in August and September. By-products from wineries are available in September, while residues and by-products from the olive oil industry emerge in October and November. Drawing upon the yearly biogas potential and mapping industries with the highest biogas potential, potential biogas production sites were identified. As outlined in the Methods section, the GIS tool was deployed to determine optimal locations of potential biogas sited. This is presented in Figure 11. When defining the biogas sites locations, suitable locations for biogas plant installations were those situated in the radius of 20 kilometres from the industrial site, maximising the potential for feedstock utilisation and minimizing transport distances. Figure 11 also clearly depict which industries are considered as viable to provide their residues and by-products as feedstocks for biogas production.

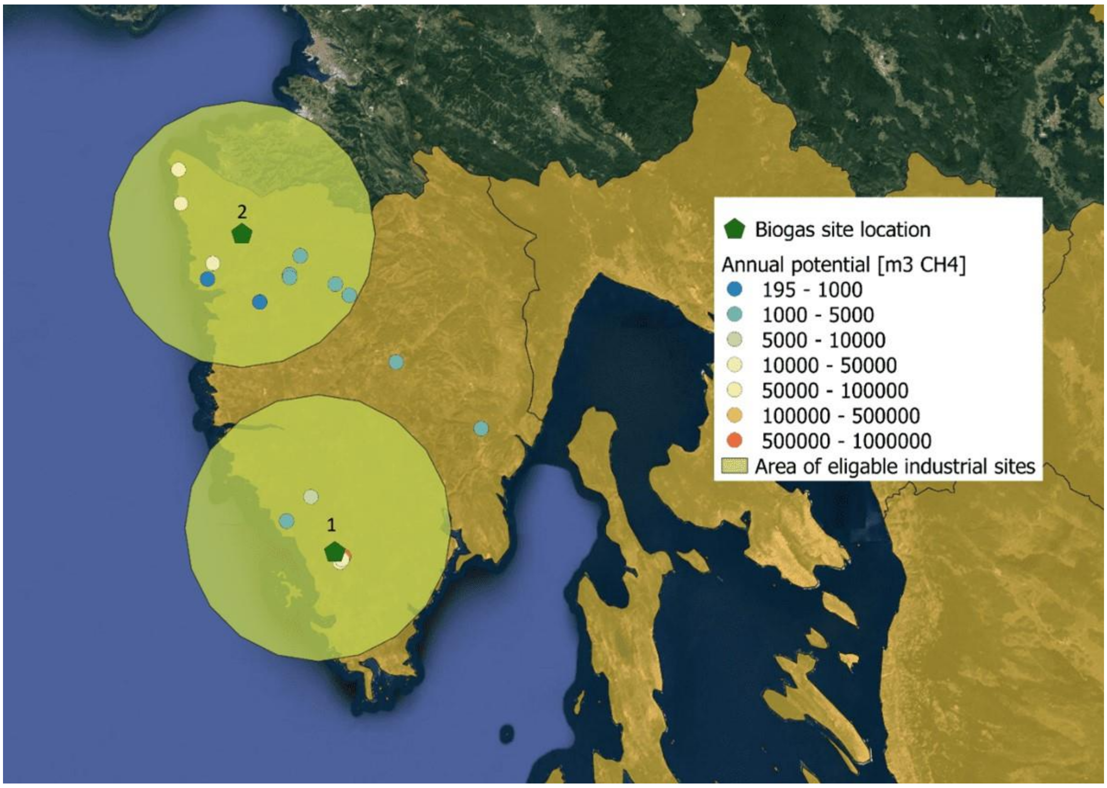


Figure 11 Annual biogas potential and optimal locations of biogas sites

The results of the spatial and seasonal biogas potential assessment from industrial residues and by-products in Osijek-Baranja county are presented in Figure 12. The considered by-products from the wine industry occur in September and from the sugar industry in September, October and November. The left part of Figure 12 presents the spatial distribution of biogas potential from by-products that occur in September (grape pressings and sugar beet pulp). Since the biogas potential from sugar beet pulp is equal in November and October, this potential is presented in one figure (right part of Figure 12).

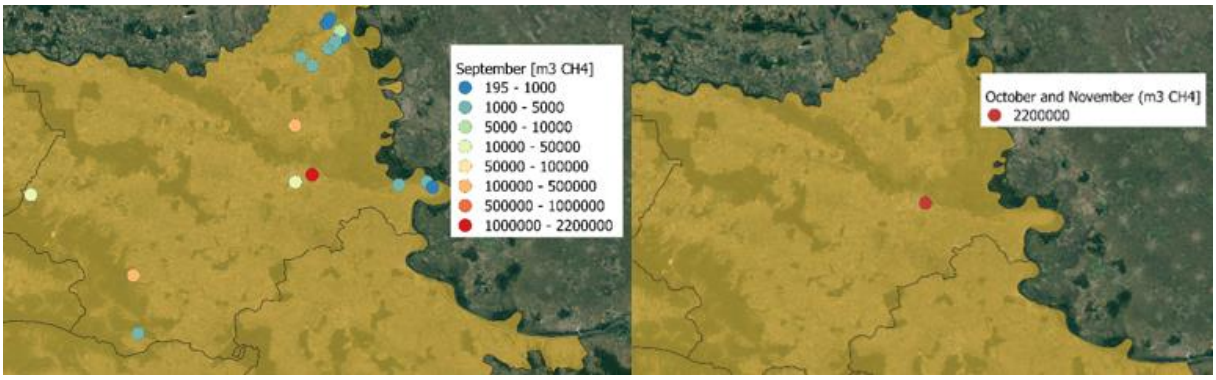


Figure 12 Biogas prom from sugar refinery and wineries in September (left) and from sugar refinery in October and November (right)

Like in the previous case, locations of optimal biogas sites were determined based on the annual biogas potential and locations of industries with the greatest biogas potential, locations of optimal biogas sites were determined. Those locations and eligible industries are presented in Figure 13.

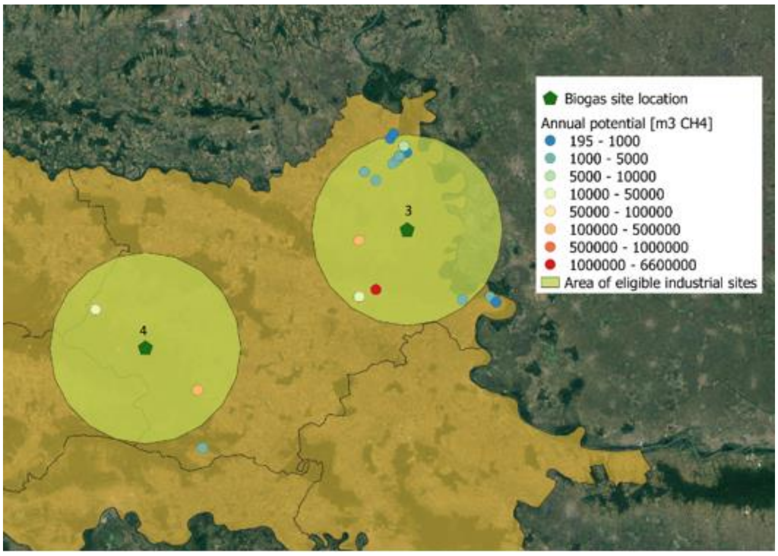


Figure 13 Annual biogas potential and potential biogas sites

As can be seen from the results conducted for the case studies, industrial residues and by-products have significant potential to be utilised for biogas production. The combined biogas potential across the four potential sites amounts to 8,119,280 m^3 CH_4. Yet, as evident from Figure 10 and Figure 12, these feedstocks are available only for a few months each year. Given this data, it prompts the question of the economic feasibility of employing these feedstocks for biogas generation, as many studies advocate. To provide a clearer understanding of how seasonality affects the economic viability of using industrial residues and by-products at biogas facilities, the annual load factors for each potential biogas cluster Figure 10 and Figure 12 was assessed.

In the first scenario, it is assumed that there is no storage capability for feedstock. Hence, the feedstock must be used for biogas production in the month of its occurrence. This means the biogas plant's potential is tied to the highest biogas potential available in a particular month. In contrast, the second scenario presumes there is sufficient feedstock storage, allowing feedstocks to be stored for a maximum of six months. This six-month timeframe is chosen based on findings indicating that extending storage might cause the feedstock to degrade [103], potentially leading to a pH shift in the anaerobic digester. These two scenarios represent two opposite ends of the spectrum: the first where feedstock storage is not an option, and the second where feedstock can be stored for the maximum possible duration before any alteration in its biological properties. For this study, these two extreme conditions will delineate the range of the annual load factor, depending on storage duration. The outcomes of these scenarios are detailed in Table 4.

Table 4 Biogas plant capacity and load factor for two scenarios (without feedstock storage and with 6-month feedstock storage)

Biogas plant (cluster)	Industry	P_{biogas} (kW)		$f_{an.load}$ (-)	
		Without storage	6-month storage	Without storage	6-month storage
1	Olive oil mills, wineries	6,658	1,665	0.16	0.66
2	Wineries, olive oil mils, vegetable industry	1,008	185	0.18	0.82
3	Wineries, sugar refinery	32,824	10,400	0.24	0.74

4	Wineries	2,633	376	0.1	0.58

As it can be seen in Table 4, load factors for case study biogas sites, for the case where there is no feedstock storage capacity, are ranging from 0.1-0.24. These values indicate that case study biogas sites will be operating at full load from 720-2,100 hours. It is worth mentioning that the biogas plants which are nowadays in operation have a high annual load factor. Stürmer et al. [104] conducted research on 291 biogas plants with different capacities and from different European countries and concluded that those biogas plants operate on an annual basis from 6,096 to 8,421 full load hours. In addition, Hublin et al. [105] calculated on a Croatian case study, that a biogas plant with an annual load factor of 0.82, that use cow manure and whey as biogas feedstock has a payback period of 9.9 years. Hence, it can be concluded that biogas plant with load factor ranging from 0.1-0.24 cannot be considered as economical feasible.

One of the possibilities to increase the number of full load hours is by integrating feedstock storage. As can be seen from Table 4, 6-month storage would in the case of pilot biogas sites lead to load factors from 0.58-0.82 (equivalent to 5,080-7,180 hours). However, it must be noted here that investment in 6-month storage leads to additional investments cost, which could strongly affect a payback period. Furthermore, storage of considered feedstock requires special attention, as improper storage practices may lead to deterioration, mould formation and pests occurrence.

Another possibility to increase load factor is to use feedstock from diverse industries and from those in which feedstock for biogas production is generated in a longer period, as it can be seen on example for biogas site 2 and 3 (Table 4).

3.3 Analysis of greenhouse gas emissions, greenhouse emission savings and sustainability requirements for the production and use of biogas

Based on the method described above, GHG emissions and savings of GHG emissions from biogas used in CHP engines after being produced from various kinds of agricultural and industrial residues and by-products, as well as municipal biowaste, are computed. Here, it is presumed that a CHP engine meets the electricity and heat needs in the biogas generation process (referred to as Case 1 in Directive 2018/2001). As described in detail in Paper 3, the emissions from the extraction or cultivation of raw material and annualised emissions from carbon stock changes, can be considered as zero, as agricultural land is not required for the production of the examined feedstocks.

The emissions from processing differs based on the processing techniques. For instance, emissions from bailing agricultural residues equal 1.1 gCO_{2eq}/MJ. For industrial by-products and municipal biowaste, values used for calculation are set in accordance with the default values for biowaste, in both open digestate and closed digestate configurations. Due to high emission contribution to emission generation due to open digestate (21.8 gCO_{2eq}/MJ), none of the considered feedstock can achieve the required GHG savings for a case of open digestate. Regarding the emissions from the fuel in use (biogas), biogenic CO_2 combustion emissions are considered to be zero for biogas. However, the fuel's typical non-CO_2 emissions are equal to 8.9 gCO_2/MJ, while default values are anticipated to be 40% higher than the typical values [13].

Emissions from transport and distribution are interesting GHG emission category, as they are a function of transport distance. Therefore, those values are presented in diagrams in Figure 14 for unbaled agricultural residues, in Figure 15 for baled agricultural residues and in Figure 16 for municipal biowaste and industrial by-products.

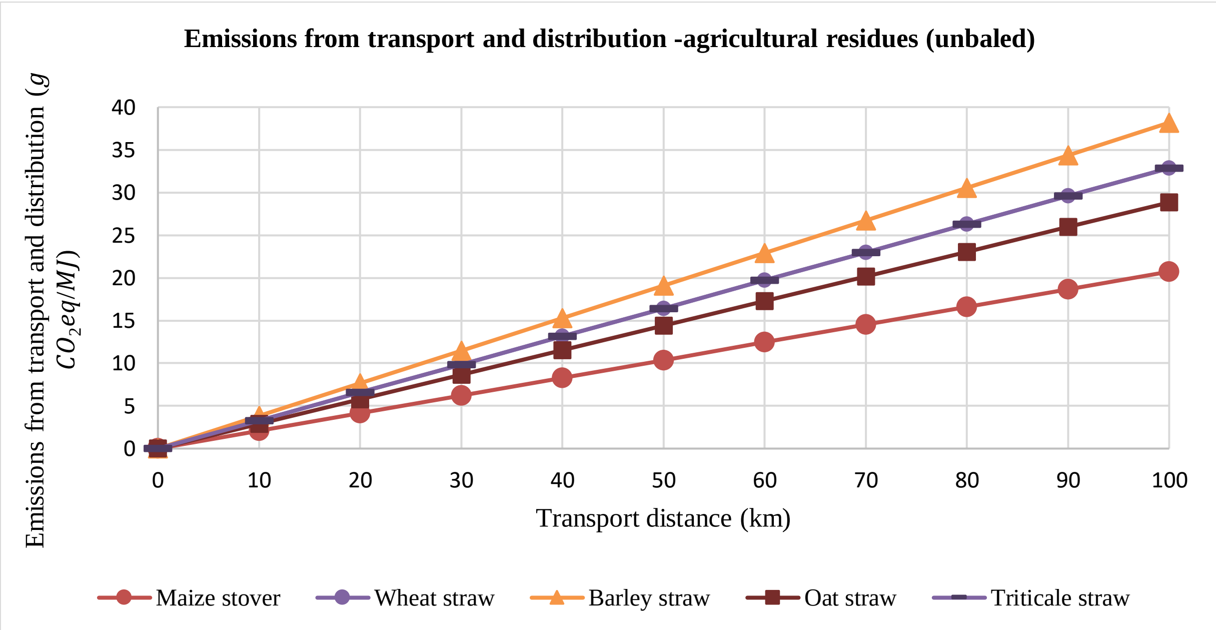


Figure 14 Emissions from transport and distribution -agricultural residues (unbaled)

Unbaled agricultural residues have a considerable increase in transport emissions per kilometre, as shown in Figure 14, due to their low bulk density. Barley straw has the highest increase in emissions per kilometre due to its lowest density. The emissions from transportation and distribution, for barley straw, outweigh the emissions from all other factors when the transport distribution is 33 km or greater. On the other hand, maize stover has the lowest rise in emissions per kilometre due to its higher density and higher biogas yield. In the case of a transport distribution of 60 km or more, emissions from transportation outweigh other sources of emissions in this situation.

The baling of agricultural residues increases the bulk density 8 times [89]. Due to the shape of a bale, it is assumed that bales could fill up to 80 % of the truck storage space. Emissions of transport and distribution for baled agricultural residues are presented in Figure 15.

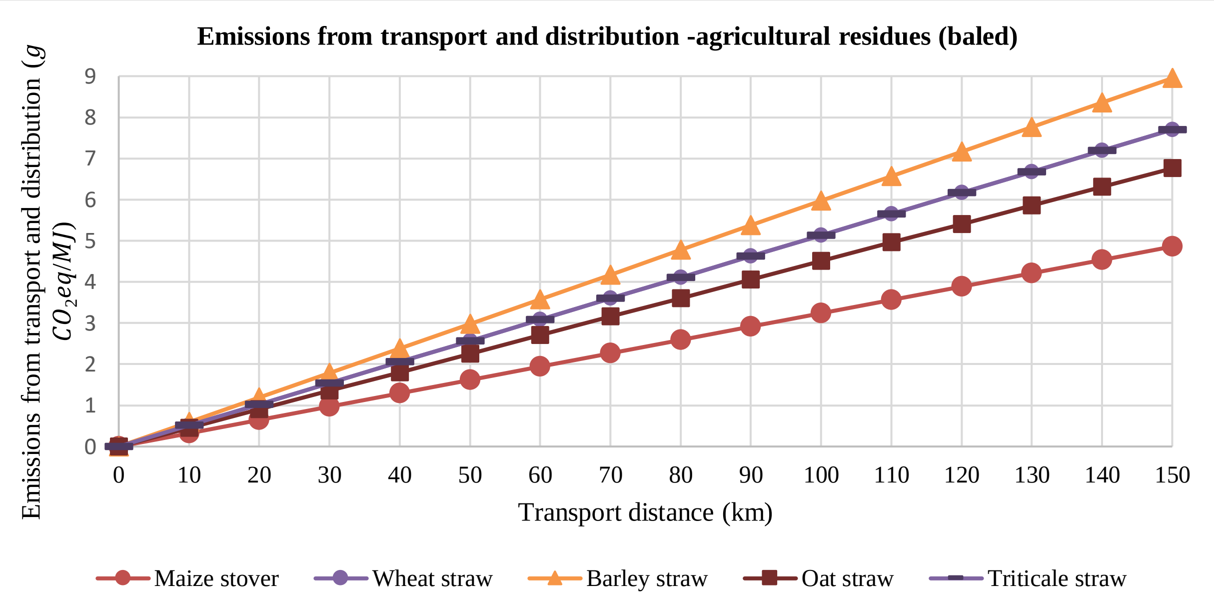


Figure 15 Emissions from transport and distribution -agricultural residues (baled)

The baling of agricultural by-products increases the bulk density and, as a result, greatly slows the increase in emissions from transit and distribution while slightly increasing the emissions of feedstock processing. Baled agricultural residues have a five-fold lower impact on distribution and transportation-related specific emissions than in the previous case.

Figure 16 shows the emissions from the distribution and transport of municipal biowaste and industrial by-products as a function of distribution and transport.

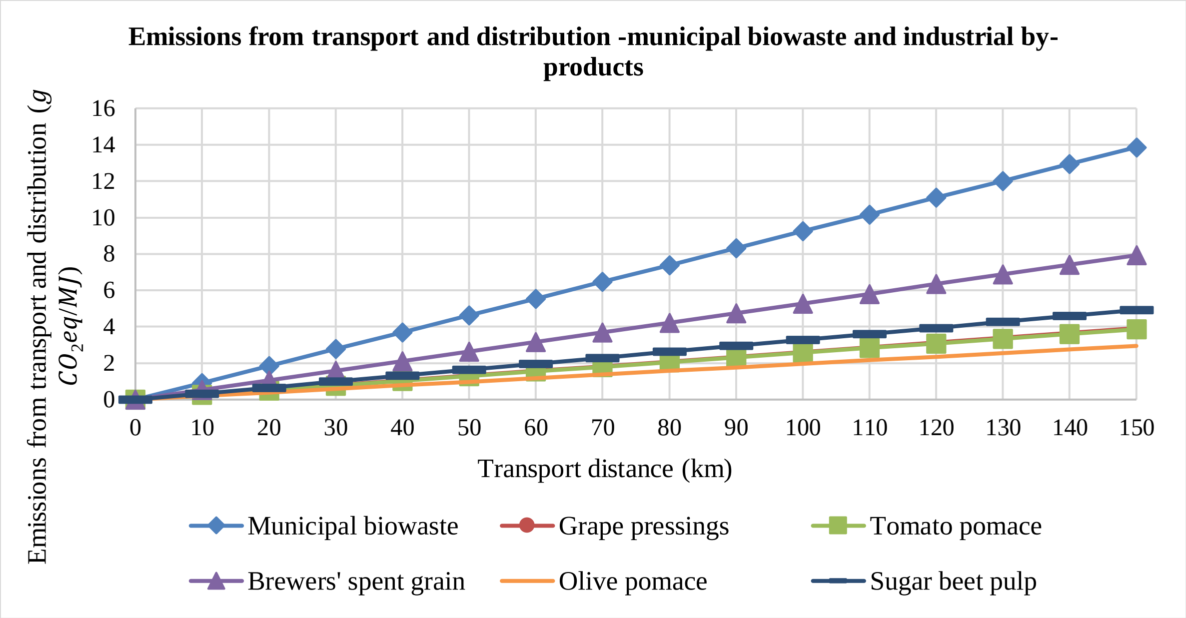


Figure 16 Emissions from transport and distribution -municipal biowaste and industrial by-products

Brewers' spent grain has the biggest rise in specific transport emissions among the analysed industrial by-products, whereas olive pomace has the lowest increase, as can be seen in Figure 16. Although, compared to the agricultural residues, those numbers are still much lower (even with the second case). This is caused by the industrial by-products' greater bulk density. Municipal biowaste, on the other hand, has a significant increase in emissions per kilometre due to lower bulk density.

Biogas plants with CHP engines must achieve both GHG savings for electricity and usable heat to comply with Directive 2018/2001. It is evident from the computed GHG reductions for heat and electricity production that the percentage of GHG savings for the generation of electricity is lower. As a result, the benchmark for calculating the maximum transport distance is 80% of GHG savings for electricity production. GHG savings of unbaled agricultural residues are presented in Figure 17.

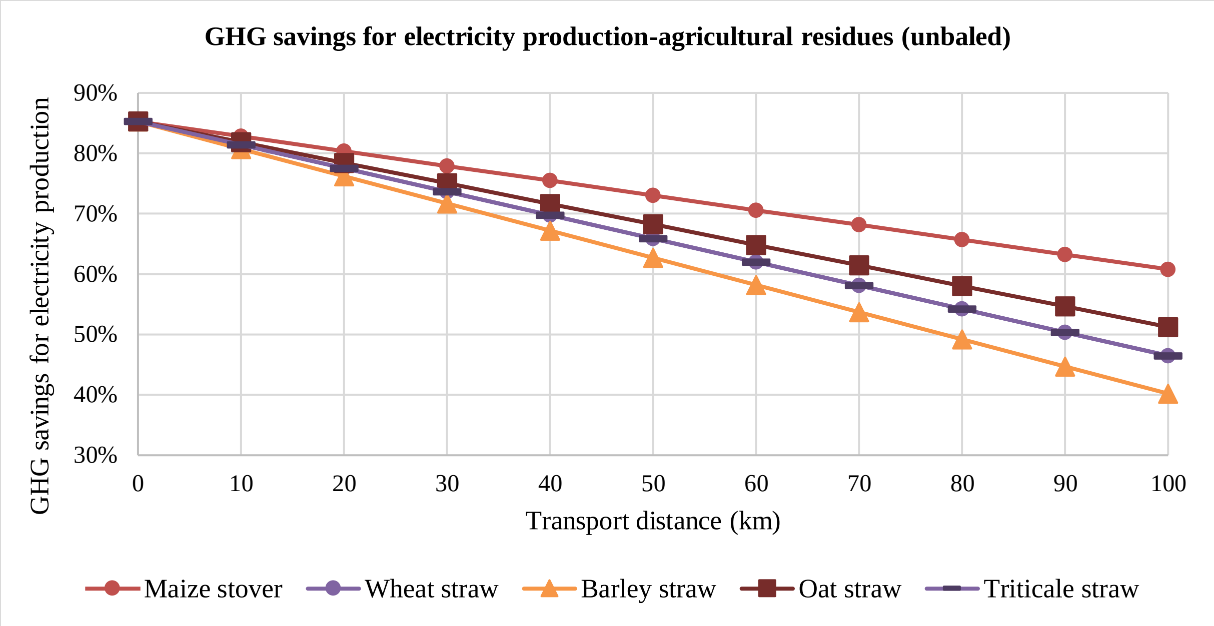


Figure 17 GHG savings for electricity production-agricultural residues (unbaled)

Based on the presented diagrams, the maximum travel distance to achieve 80% of GHG savings from the utilisation of biogas can be determined. As explained above, bailing of agricultural residues will result in five-fold lower impact, so their maximal transport distance may be five times higher. Those values are defined in Table 5.

Table 5 Maximum travel distance, for achieving 80% of GHG savings- unbaled and baled agricultural residues

	Maize stover	Wheat straw	Barley straw	Oat straw	Triticale straw
D, max-unbaled (km)	21	14	12	15	14
D, max-baled (km)	104	65	55.5	74	65

As can be seen from Figure 17 and Table 5, the greatest travel distance of unbaled agricultural residues is fairly low, ranging from 12 km for barley straw to 21 km for maize stover. Even with increased processing emissions, bailed agricultural residues have significantly higher maximal travel distance, which range from 65 km for wheat and triticale straw to 104 km for maize stover.

Figure 18 shows GHG savings of municipal biowaste and industrial residues as a function of transport distance.

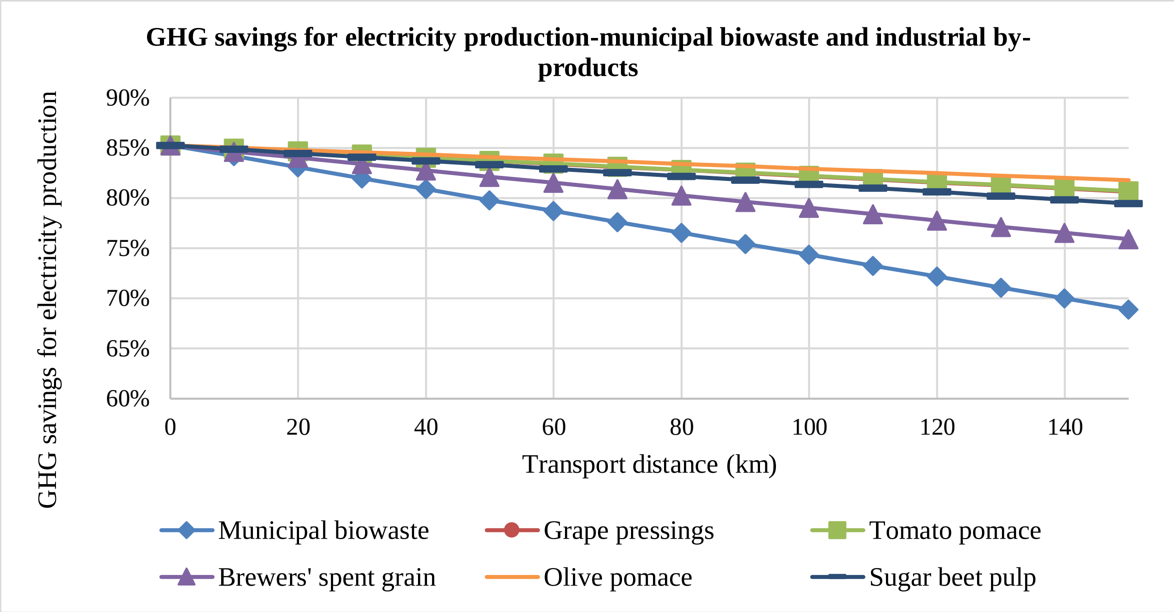


Figure 18 GHG savings for electricity production-municipal biowaste and industrial by-products

It is important to note that Figure 18 presents only the case with closed digestate, as the considered feedstocks cannot achieve 80% of GHG savings in the event of an open digestate. The maximum travel distance for municipal biowaste equals 48 km, while for the industrial by-products ranges from 84 km for brewers' spent grain to 227 km for olive pomace. Those values can be determined from the diagram presented in Figure 18 and are defined in Table 6. Compared to agricultural residues, industrial by-products can achieve the required savings with a higher transport distance, due to higher bulk density.

Table 6 Maximum travel distance, for achieving 80% of GHG savings-municipal biowaste and industrial by-products

	Municipal biowaste	**Grape pressings**	**Tomato residues**	**Brewers' spent grain**	**Olive pomace**	**Sugar beet pulp**
D, max (km)	48	170	173	84	227	136

The examined feedstocks are a sustainable option for producing biogas as they do not require agricultural land (unlike the already widely used maize silage) and hence satisfy the sustainability requirements defined in Directive 2018/2001. These requirements state that biogas feedstocks should not be produced from raw materials obtained from land with a high biodiversity value, such as primary forests, areas for the protection of rare or endangered ecosystems or species, highly biodiverse grasslands, and wetlands, etc. and must adhere to the criteria for forestry, land use, and land-use change (LULUCF).

The results show that emissions from transportation and distribution have a substantial impact on total emissions and the resulting reductions in greenhouse gas emissions for the cases and feedstock groups discussed.

The maximum transport distance between the examined feedstocks greatly varies, as shown by the results. Given that waste materials are preferred as biogas feedstock, it is clear that the scope of the analysed feedstocks must be enlarged, and the maximum distance for each of the examined feedstocks must be defined.

3.4 P-Graph model for the economical optimisation of biomass supply network that meets requirements on greenhouse gas emissions savings - a case study of rural areas

Based on the results of the GIS mapping, GHG saving and sustainability requirements, input data were obtained for the development of the P-graph mathematical model. The presented method was demonstrated in the case study of the rural area of Osijek-Baranja County. According to the method provided in the *Method* section, the seasonal and spatial variation of biogas potential from wheat straw, manure, grape pressings, and sugar beet pulp was determined for the farms, wineries, sugar factories, and wheat straw collection sites. In accordance with the optimal location, the transport distance between industry/farm/collection sites and the optimal location of the biogas site was calculated as represented in Figure 19.

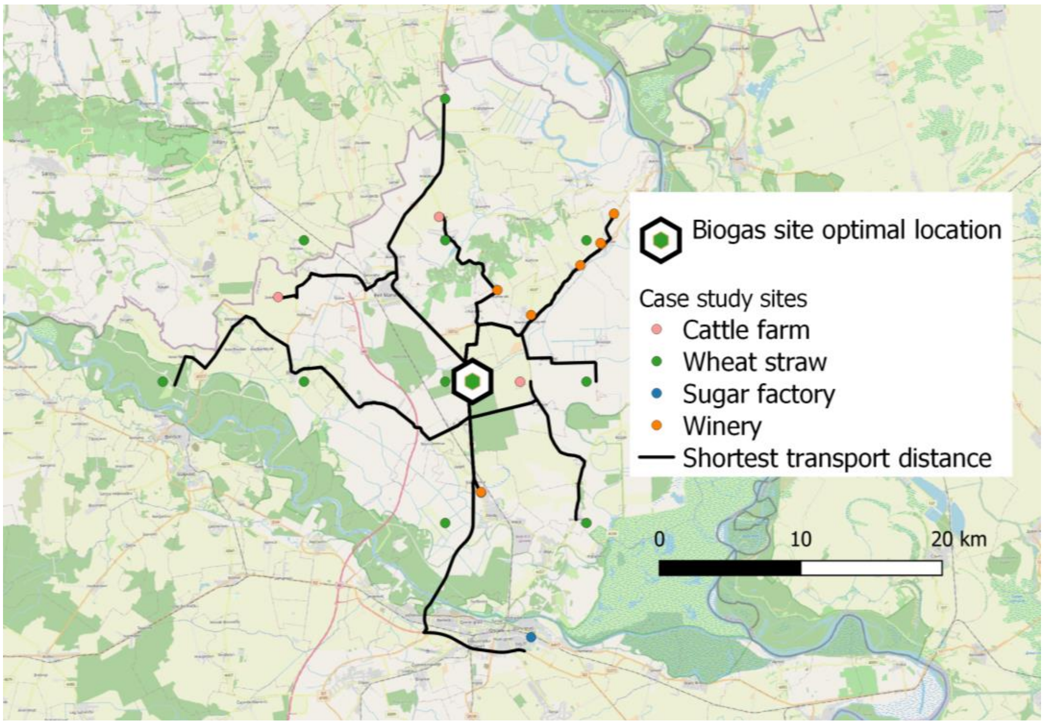


Figure 19 Transport road route and optimal biogas site location

In accordance with the resulting GIS layer (Figure 19), the P-graph representation and the maximal structure are developed. The data set obtained with QGIS includes biogas potential and transport distance for 18 feedstock-providing sites. The P-graph representation of the maximal structure of the case study is represented in Figure 20. As described in the method, the material nodes are represented by raw materials (wheat straw- WS, cattle manure-CF, wineries-W, sugar factory- SF) and the final product (biogas). Operating unit nodes are representing anaerobic digestors.

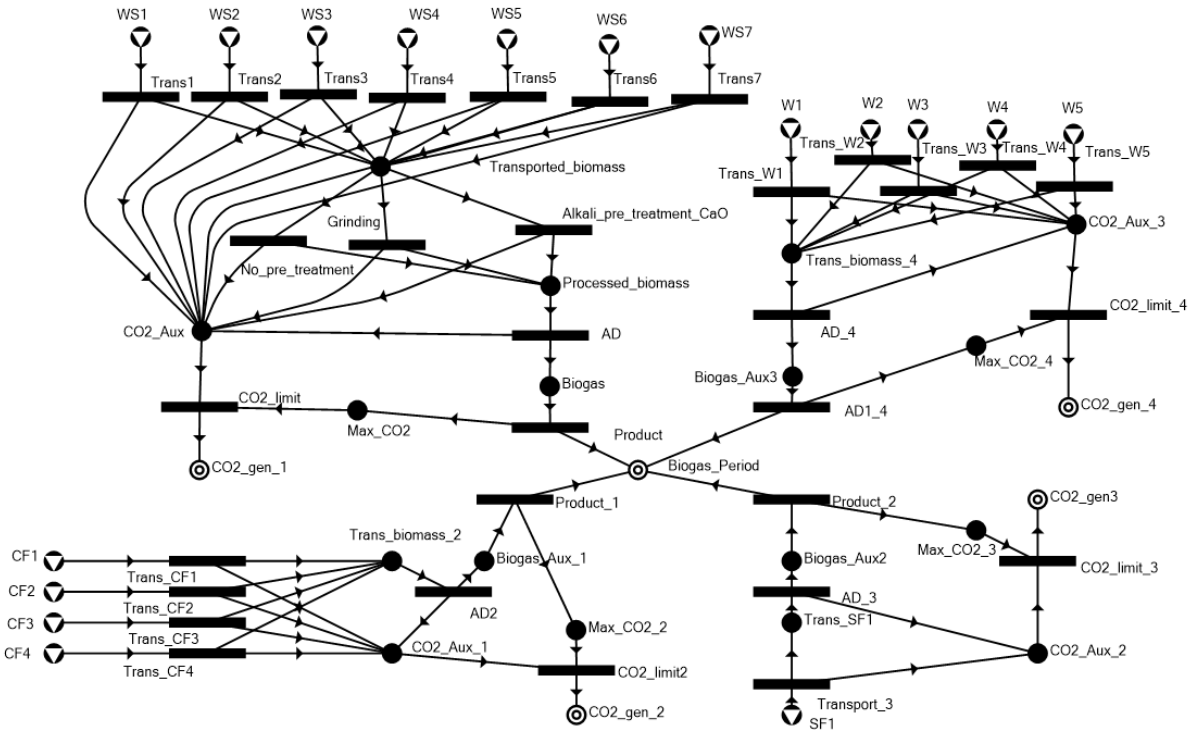


Figure 20 P-graph representations of the maximal structure of the case study

As described in the *Method* section, based on maximal structure, all feasible structures were defined. For the optimal solution, the objective was to minimise the cost of the biomass supply network and to limit the associated GHG emissions below the given threshold. This was done for two cases, both having the required biogas production of 120,000 GJ/y, but in the first case the optimisation is performed on an annual level, while in the second case, the multiperiod approach was implemented to include the seasonal variation of feedstock supply. The biogas production of 120,000 GJ corresponds to the production of anaerobic digestors which deliver biogas to CHP with 1.5 MW_{el}. The optimal structure for annual biogas production of 120,000 GJ/y is presented in Figure 21.

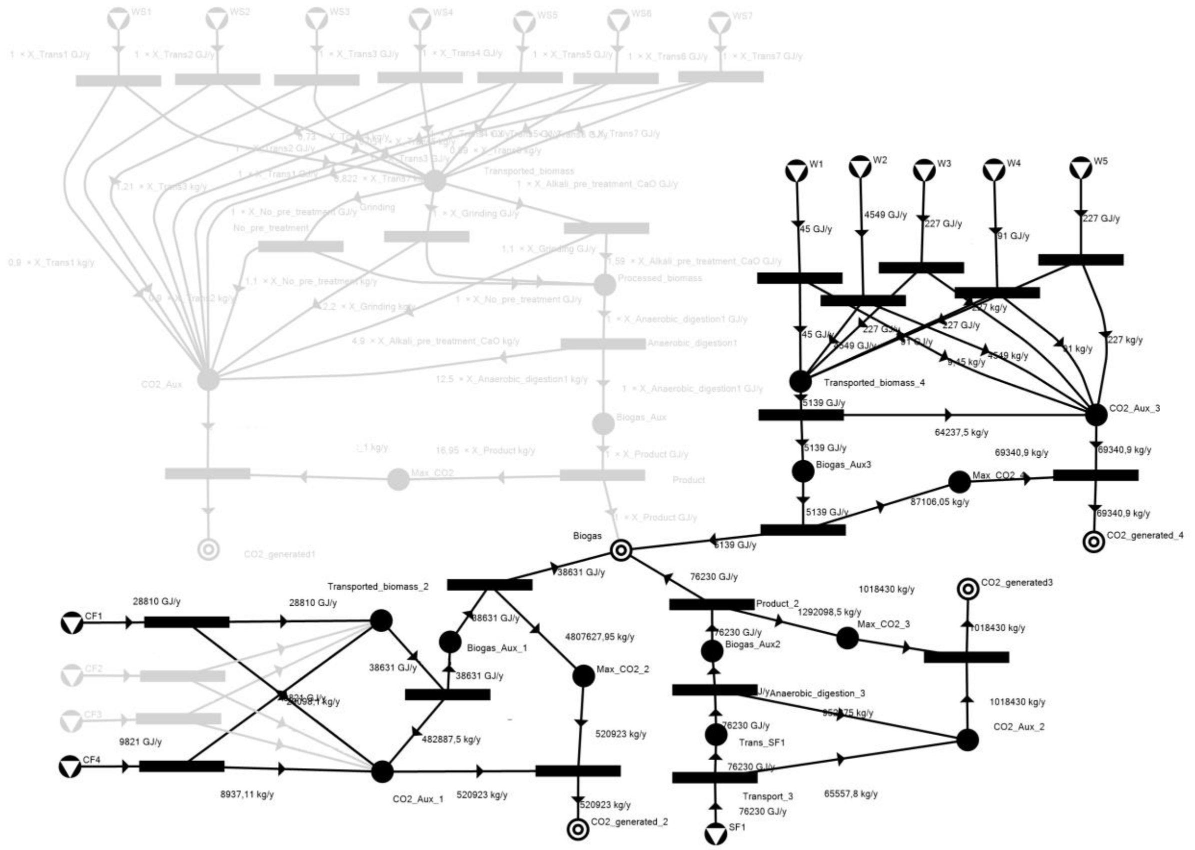


Figure 21 The optimal structure for annual biogas production of 120,000 GJ/y

The cost of the biomass supply network (including feedstock and transport costs) is 292,016 EUR. This equals 2.43 EUR/GJ. The data from the optimal structure are presented in Table 7, to improve the visibility of the numbers presented in Figure 21.

Table 7 The optimal structure for annual biogas production of 120 000 GJ/y

Abbreviation	CF1	CF4	SF1	W1	W2	W3	W4	W5
Delivered feedstock (GJ)	28,810	9,821	76,230	45	4,549	227	91	227

As can be seen from the results, the model first selects the wineries and sugar factory, after that cattle farms and finally wheat straw. It is interesting to see that the model would select sugar beet pulp prior to the manure from the further farms, as feedstock cost is lower for manure. The reason for this selection is the relatively low bulk density of the biogas potential of manure, compared to the bulk density of a biogas potential of sugar beet pulp. Hence, higher transport may surpass the difference in feedstock cost.

GHG emissions linked to each stage of the production and use of biogas are assessed. Contribution to GHG emission generation is presented in Table 8 by each feedstock group, as well as the GHG savings compared to the fossil fuel comparators (for both heat and electricity).

Table 8 GHG emission generation- case 1 (annual assessment)

Feedstock	Wheat straw	Manure	Sugar beet pulp	Grape pressings
Biogas produced from feedstock (GJ)	-	38,631	76,230	5,139
Associated GHG emissions (kg CO_{2eq})	-	520,923	1,018,430	68,769
Associated GHG emission savings (kg CO_{2eq})	-	4,143,757	-	-
Neto GHG emissions (kg CO_{2eq})	-	-3,622,834	1,018,430	68,769
Specific GHG emissions (kg CO_{2eq}/GJ)	-	-93.8	13.36	14
GHG savings compared to fossil fuel comparator for heat, Case 1, closed digestate	-	210.70%	84.25%	83.49%
GHG savings compared to fossil fuel comparator for electricity, Case 1, closed digestate	-	165.35%	90.70%	90.26%

As can be seen from the specific GHG emissions presented in Table 8, GHG emissions are below the threshold (which is set to 16.95 kg CO_2/GJ biogas), which can be considered as a confirmation that the developed model presented as feasible structures only those which fulfil GHG savings.

Integration of GHG emissions limitation, in line with Directive 2018/2001, represents an added value and a step beyond the current state of the art in P-graph optimisation. To enhance the understanding of GHG emission limitation and improve the visibility of Figure 21, part of the PNS network (for the case of optimal structure) whose function is to limit GHG emission is presented enlarged in Figure 22.

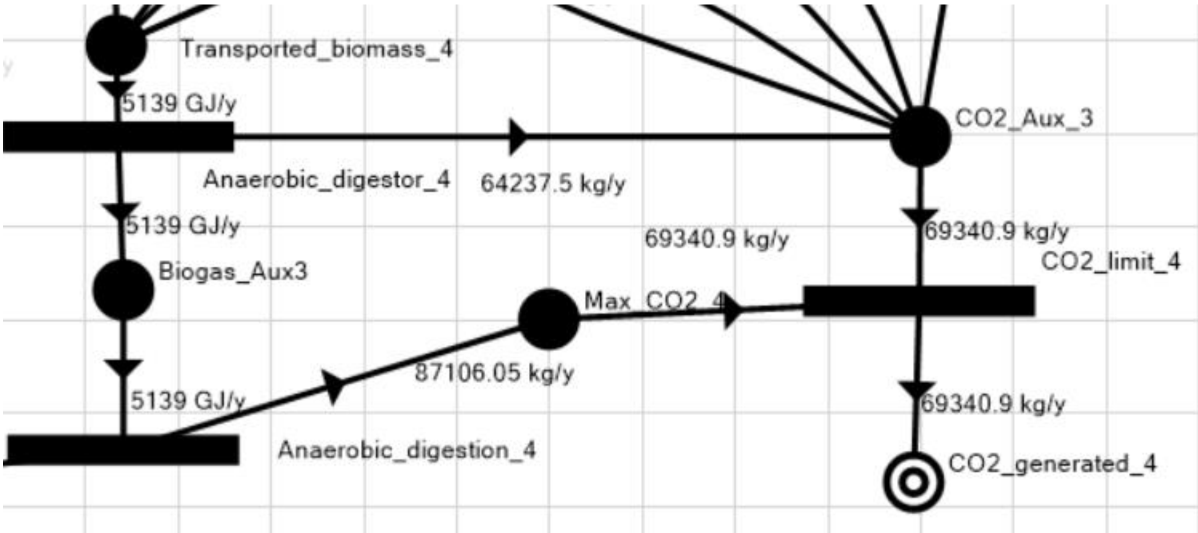


Figure 22 GHG emission limitation in PNS network (optimal structure)

For the given example, the selected feedstock group is grape pressings. CO_2_Aux is an auxiliary node represented as an interim product, whose main objective is to summarise GHG emissions that occur in biogas production and use lifecycle. This value equals 69 340.9 kg/y in Figure 22. The obtained value is then compared with the maximal allowed GHG emissions. The maximum allowed GHG emissions are calculated based on the biogas generation (GJ), obtained from the auxiliary node Biogas_Aux, which is multiplied by the specific limitation of GHG emissions per GJ of biogas. This value equals 87106.05 kg/y in Figure 22. Those two values meet at node Max_CO_2. For the case where GHG emissions that occur in a biogas lifecycle are higher than the maximum values, the model makes a new iteration and searches for a new economically optimal structure whose GHG emissions are below the given limit.

To enhance the accuracy of the results, the seasonal aspect of biomass production was integrated into the model. To integrate this, a year was divided into several periods, each representing certain months.

For each considered period, the assumption is used that biomass available for biogas production is the one generated in the specific period (months). There is a threefold reason for this. The first one is that some of the considered feedstock cannot be stored for a longer period of time, due to potential changes in feedstock conditions, which could result in the adverse performance of biogas production. The second is that seasonal feedstock storage may result in additional methane emissions generated during the storage period, which could result in exceeding the threshold of GHG emissions, due to the high global warming potential of methane. The final one is the cost of the investment and maintenance of the seasonal storage.

For the considered biogas production, in case the required biogas production exceeds the biogas potential contained in the biomass, the assumption was used that this gap will be covered with the wheat straw, due to favourable storage properties. Required biogas production, for the case of the annual production of 120, 000 GJ is presented in Table 9.

Table 9 Required biogas production in the concerned periods

Period/ month	1/ January-May	2/ June-July	3/ August	4/ September	5/ October-November	6/ December
Required biogas production (GJ)	55,848	19,029	0	11,096	22,561	11,466

As can be concluded from Table 9, annual maintenance of the biogas site is scheduled for August. The optimal structure for each period is presented in Figure 23, Figure 24, Figure 25, Figure 26 and Figure 27.

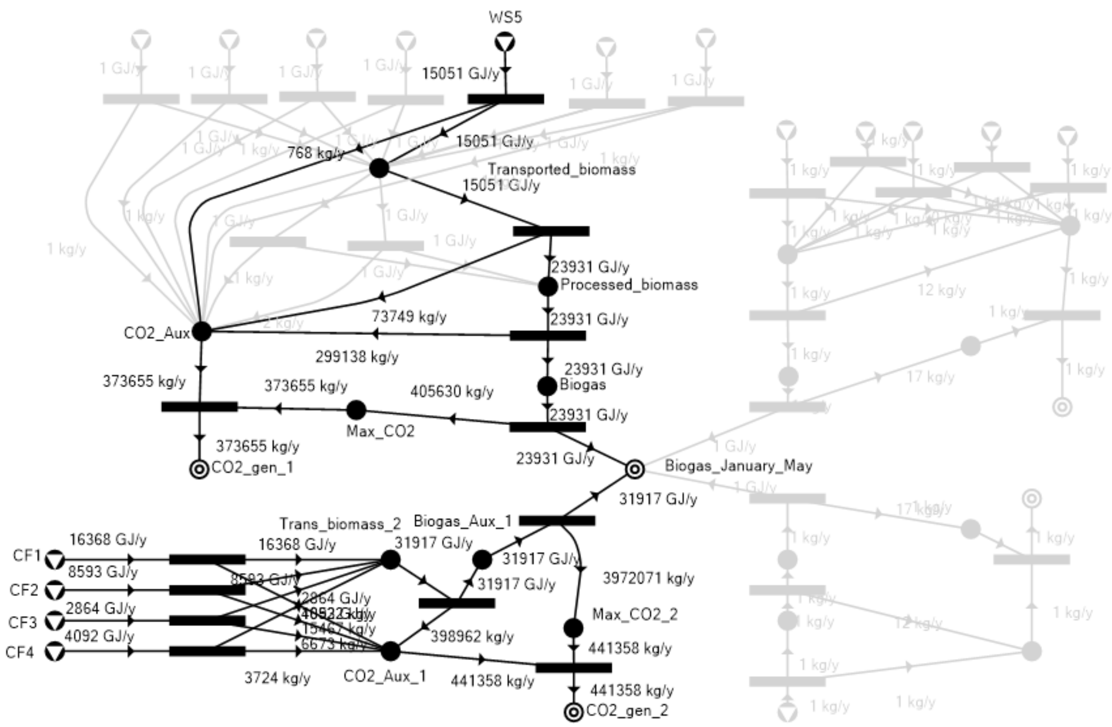


Figure 23 Optimal structure of biogas production from January until May

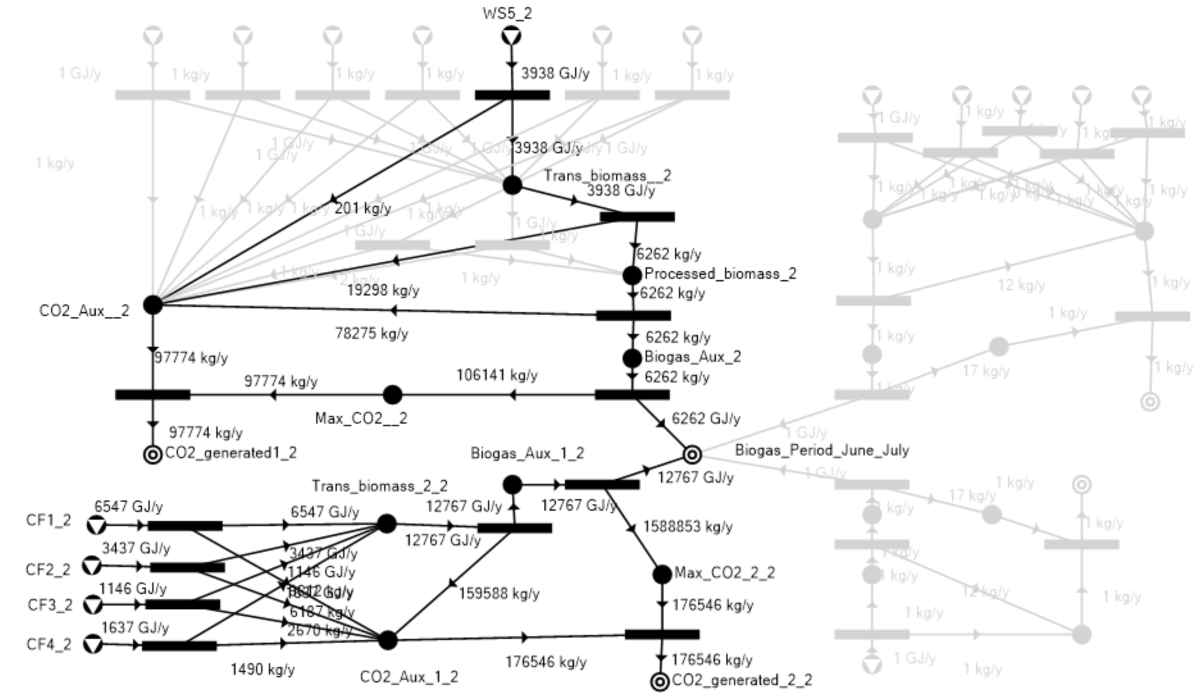


Figure 24 Optimal structure of biogas production in June and July

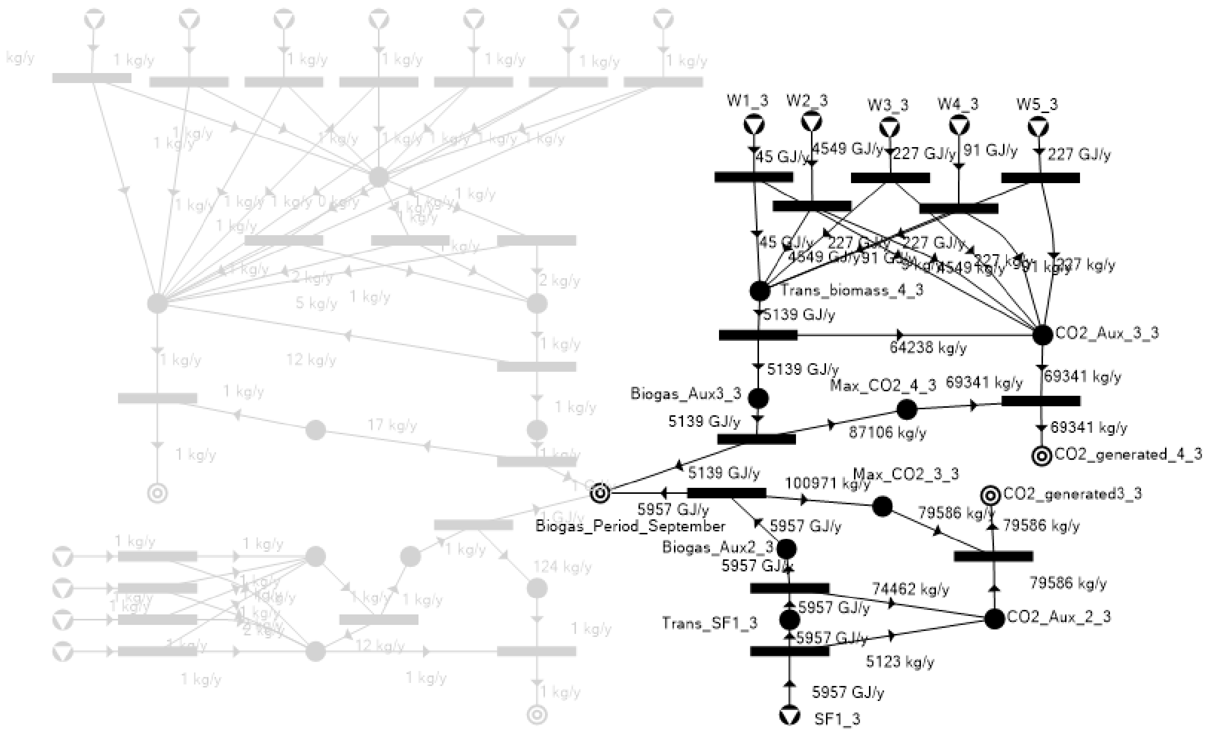


Figure 25 Optimal structure of biogas production in September

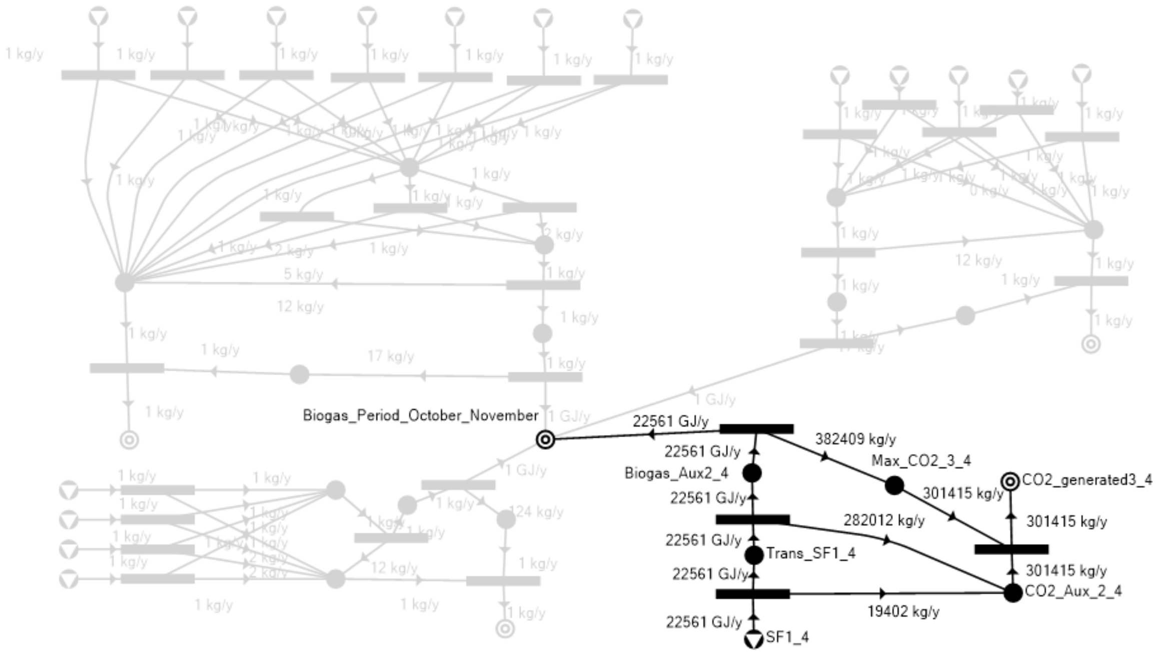


Figure 26 Optimal structure of biogas production in November and October

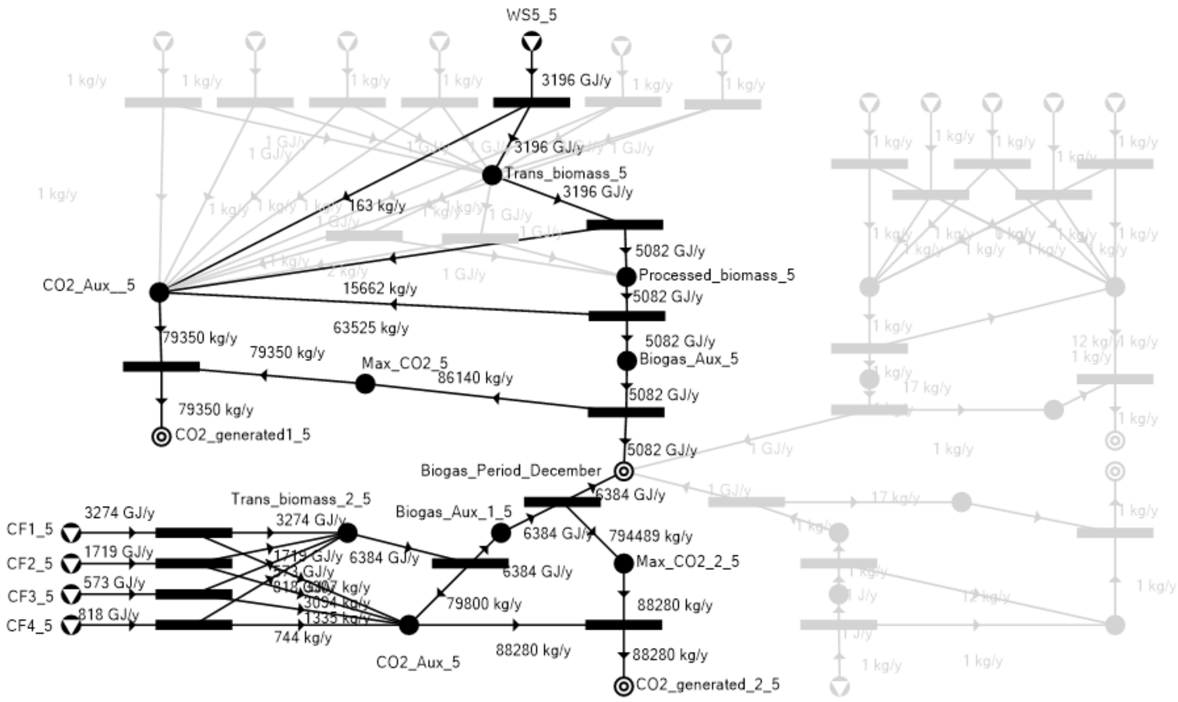


Figure 27 Optimal structure of biogas production in December

The values on delivered feedstock presented in optimal structures are listed also in tables provided in Paper 4, to improve the visibility of the numbers. The optimal structures of periods from January-May, June-July and December are very similar. Although in the second period, there is a significant biogas potential of the wheat straw sites, the model will select manure, even from further farms. The total values of delivered feedstocks significantly differ from the first case where the optimal structure was defined on an annual basis. Compared to the first case where the model did not commit the wheat straw sites, in the second case wheat straw contributes to 18.3 % of biogas production. Furthermore, in the second case, the contribution of manure is significantly (34%) higher, even from the more distant farms, which would result in higher transport costs. On the other hand, most of the potential of sugar beet pulp was untapped in the second case (63%). Those changes negatively affected the total cost of the biomass supply network, which equals 733,684 € (6.11 €/GJ biogas). The increase in biomass supply cost is, to some extent, an expected result, as during the periods in which feedstock with high energy density and low prices were not available, the model committed the sites with higher transport distances and/or sites with higher feedstock and processing costs. Furthermore, although the multi-period approach resulted in less favourable results in terms of cost, it can be stated that this approach results in more accurate results and provides insights into the sensitivity of the cost of biomass supply network for the case where economically favourable feedstocks are not available, since they are being generated in a very short period of time during the year.

As for the first case, the contribution to GHG emission generation, as well as GHG savings compared to fossil fuel comparators, is presented in Table 10 for each feedstock group.

Table 10 GHG emission generation -case 2 (multi-period assessment)

Feedstock	Wheat straw	Manure	Sugar beet pulp	Grape pressings
Biogas produced from feedstock (GJ)	35,275	51,068	28,518	5,139
Associated GHG emissions (kg CO2eq)	550,778	706,184	381,001	69,341
Associated GHG emissions savings (kg CO2eq)	-	5,477,813	-	-
Net GHG emissions (kg CO2eq)	550,778	-4,771,628	381,001	69,341

Specific GHG emissions (kg CO_{2eq}/GJ)	15.61	-93.44	13.36	13.49
GHG savings compared to fossil fuel comparator for heat, Case 1, closed digestate	89.12%	210.22%	84.25%	84.08%
GHG savings compared to fossil fuel comparator for electricity, Case 1, closed digestate	81.57%	165.06%	90.70%	90.60%

As in the first case, specific GHG emissions were below the given threshold, which can be considered as a confirmation that the model successfully limits the GHG emissions in both single-period optimisation and multi-period optimisation. It is also interesting to note that, due to the higher contribution of manure to biogas production, the net GHG emissions are significantly lower for the second case. As mentioned earlier, when defining the optimal structure, P-graph Studio defines and ranks sub-optimal structures as well. Hence, the results could be used for the development of a Pareto front that would define both the cost of the structure and the generated GHG emissions.

The developed model does not automatically prioritize the structures with the lowest GHG emissions, as it considers GHG emissions savings as constraints, not as the variable to be minimised. Although this may be considered as the limitation of the model, the minimisation of the GHG emissions was not selected as the target group of this model is the biogas industry, whose objective is commonly to fulfil the requirements given by the legislation and to minimise the cost of the biomass supply network. However, in case if biogas industry would receive some additional incentive to future reduce GHG savings, or in general decides to achieve savings higher than the given threshold, the developed model easily allows the comparison of the cost and GHG emissions of optimal and sub-optimal structures, thus enabling efficient assessment of trade-offs. Based on the obtained results, it can be assumed that for the case of the minimisation of GHG emissions, the model would prioritize manure as the feedstock (even from more allocated farms), due to the high GHG emission savings resulting from the improved manure management.

As the developed model determined economically optimal and sub-optimal structures, simultaneously limiting GHG emissions in both single-period optimisation and multi-period optimisation, it can be stated that the hypothesis of this thesis is confirmed.

3.5 GIS and P-Graph model for the economical optimisation of biomass supply network that meets requirements on greenhouse gas emissions savings - a case study of urban areas

The second approach presented in the *Method* section is tested at the case study of the city of Zagreb. The biogas potential from spent grain industrial biowaste, oil, and fat was determined for the considered supermarkets (SH), fast food chains (FF) and breweries (BR). The location of the biogas site was selected following the location of the existing composting plant and landfill. In accordance with the selected location, the transport distances between supermarkets, fast food restaurants, breweries and the biogas site were determined, as represented in Figure 28. The transport distance was calculated for each site that provides feedstock to the biogas plant. In this analysis, the assumption was made that trucks would be utilized for transporting the feedstock. The selected roads, determined as the shortest routes, are permissible for truck travel. Moreover, only feedstocks with transport distances below the maximum allowed distance (to achieve the necessary GHG savings) were considered for future evaluation.

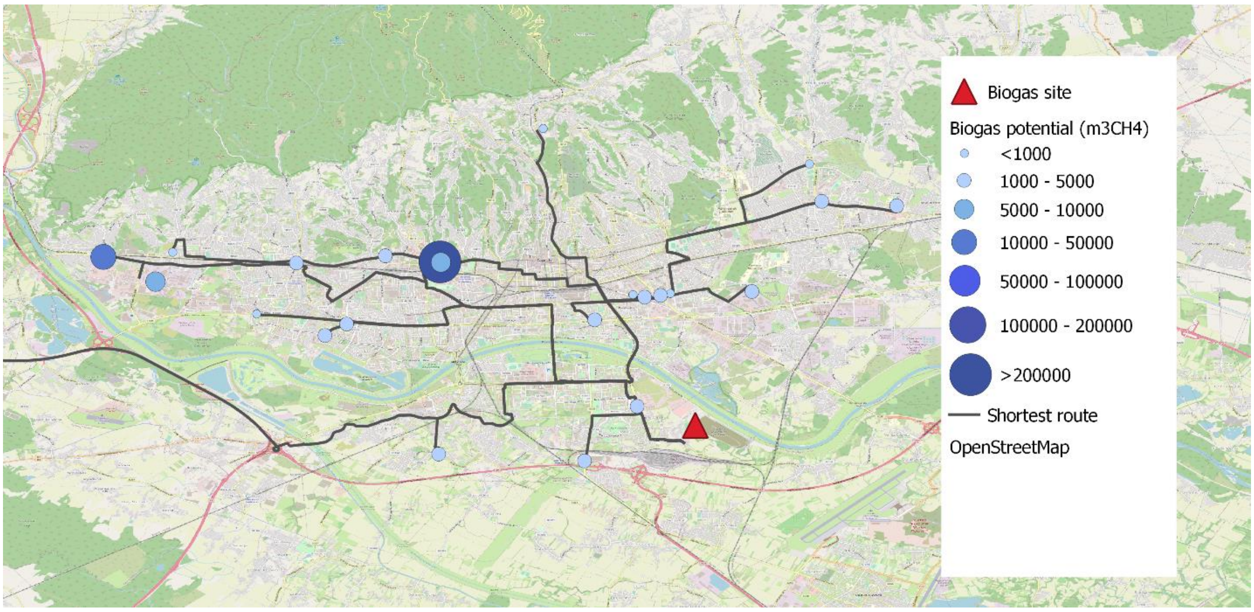


Figure 28 Transport road route and optimal biogas site location

As seen in Figure 28, biogas potential significantly varies between different feedstock-providing sites. For the considered case study, the greatest biogas potential comes from breweries. As explained in the Method section, GIS data represented in Figure 28 were converted to a format supported by P-graph Studio (Excel file) and used as the input data for P-graph-based optimisation.

Based on the maximum structure and input data obtained from the GIS tool, optimal and suboptimal structures were defined. Here, the objective function is to minimise the cost of the biomass supply network. Additionally, it is important to note that the main purpose of this structure optimization is to compare the economic viability of the utilization of different biomass supply structures. Consequently, only the costs that differ between different biomass supply structures are included in this analysis. The optimal structure for annual biogas production of 36,000 GJ/y is shown in Figure 29.

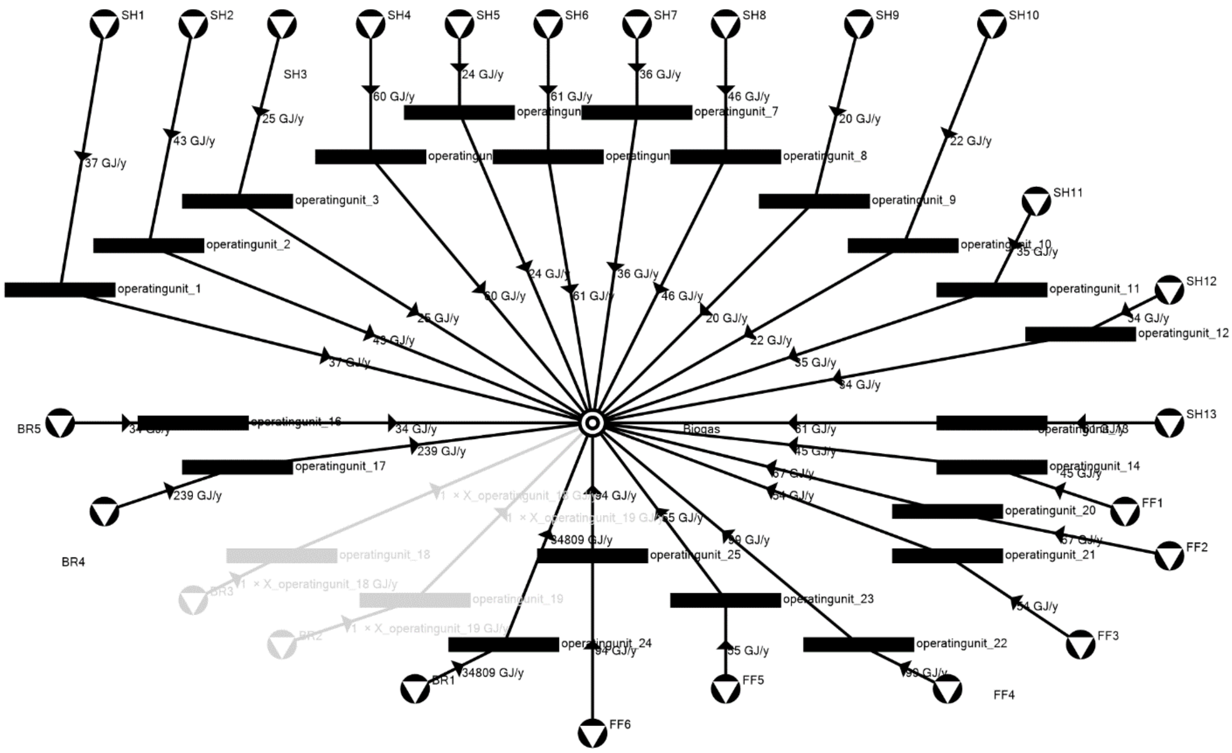


Figure 29 The optimal structure for annual biogas production of 36,000 GJ/y

The cost of the biomass supply chain (including feedstock and transport costs) is 448,080 EUR. This equals 12.44 EUR/GJ. As seen in Figure 29, feedstock sites that provide waste materials (supermarkets and fast-food restaurants) are prioritised as feedstock suppliers. However, their contribution is relatively low due to the limited potential of these sites and the biogas production is mostly based on the feedstocks that occur in breweries. Consequently, this supplier significantly influences the average price of the biomass supply chain. The increased demand for biogas would lead to a further rise in the specific cost of the biogas supply network, due to the high price of the breweries' spent grain 33 €/t [106]. Thus, it can be inferred that the economic viability of biogas production in urban areas should rely on waste materials to enhance its feasibility.

The combination of integrating GIS mapping of biomass potential and employing a P-graph framework for optimizing biomass supply networks, implemented in this work and tested in the case study proved to be effective. This approach improves the accuracy of input data and consequently results, in comparison with other studies that consider biomass potential to be generated from a single site [69] or clustered into zones [107].

Furthermore, the elimination of feedstock suppliers with high greenhouse gas (GHG) emissions from transport during the initial step of GIS mapping has demonstrated its efficiency by ensuring that the utilization of the final product, namely biogas, achieves at least the minimum GHG savings. However, this approach's applicability is limited to cases where the transport distance is the sole factor affecting GHG emissions, and typical values can be used to calculate other GHG-related factors. For more complex situations where total GHG emissions may vary based on selections made within the supply chain network, a more intricate integration of GHG emissions savings, such as the one presented in the first approach, is required.

4 CONCLUSIONS AND FUTURE WORK

The European Commission has set ambitious commitments for transitioning to a circular bioeconomy, reducing greenhouse gas emissions, and exponentially increasing biomethane production to rapidly reduce dependence on fossil fuels. Biogas production through anaerobic digestion offers a promising solution to these challenges. It recovers the energy stored in biomass and produces renewable energy in the form of biogas and biomethane. Additionally, it contributes to the recycling of nutrients and micronutrients and offers a threefold reduction in greenhouse gas emissions.

The three objectives of this thesis are: to determine the spatial distribution and to calculate the influence of the seasonality of the residual biomass generation: to define constraints for which different types of industrial by-products and agricultural residues meet sustainability and greenhouse gas emissions saving criteria; and to prove the value of the use of graph theory approach in modelling a residual biomass supply network, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria.

The hypothesis of this research is that a economically feasible residual biomass supply network for biogas production, that meets sustainability and greenhouse gas emissions saving criteria, could be determined with graph theory approach. The hypothesis of this doctoral thesis has been confirmed.

The doctoral thesis was based on five papers published journals indexed in CC database, available in the Annex of the thesis. Paper 1 and Paper 2 introduce novel GIS approaches that integrated the seasonality of the residual biomass generation in the process of GIS mapping. Paper 1 focuses on the agricultural residues and municipal biowaste as the feedstocks for the biogas production, while Paper 2 investigate industrial residues and by-products. The influence of seasonality was calculated in both cases, with different parameters. Paper 1 shows that for the lignocellulosic agricultural residues, that application of seasonal assessment approach leads to lower storage facility capacity requirements. For the considered examples, it resulted in 12%

and 40% lower storage facility capacity requirements, compared with annual assessment approach. In Paper 2, the influence of seasonality on the economic viability of industrial residues and by-products utilisation for biogas production was calculated through assessment of annual load factors for two scenarios- the scenario without feedstock storage and the scenario with 6-month case storage. For the first scenario, annual load factors for case study biogas sites are ranging from 0.1-0.24. As biogas sites are operating at multiple higher load factor, it can be concluded that seasonality highly affects the economic viability of biogas site operation. For the scenario where 6-month feedstock storage is available, the annual load factors increase to 0.58-0.82. However, it should be noted that those storages may highly affect total investment and require special attention, due to possible issues which occur in case of improper feedstock storage. As it is presented in the results, spatial and seasonal assessment of the potential of lignocellulosic agricultural residues and industrial residues and by-products available for biogas production provide more accurate input data. Therefore, it can be concluded is a strong need to include seasonal variations in the potential assessment of the considered feedstocks.

The objective of Paper 3 was to conduct an analysis of greenhouse gas emissions and specific greenhouse gas emissions savings, as well as sustainability requirements for biogas production that uses as feedstock agricultural residues (wheat straw, barley straw, oat straw, triticale straw and maize stover) municipal biowaste and industrial by-products (grape pressings, tomato residues, brewers' spent grain, olive pomace, sugar beet pulp). The method is based on the method outlined in Directive 2018/2001. This method is a GHG accounting method, which includes numerous emission factors for biogas pathways for electricity and heating production. The emissions are calculated/determined for each of the emission factors, except for the emissions from transport and distribution which are calculated and presented as a function of a transport distance. According to calculations, the maximum travel distance for considered residues was defined. The results of this paper demonstrated that transportation emissions have a significant impact on biogas production's potential to achieve the required greenhouse gas emissions savings. A substantial difference in the results further supports the need to increase the number of feedstocks for which default and typical values are available.

In the scope of Paper 4 the mathematical model for residual biomass supply network modelling from sustainability, greenhouse gas emissions savings and economic point of view was developed. This paper presents a novel multi-period P-graph-based model for optimizing biomass supply networks, which goes a step further in integrating environmental constraints in the PNS network. The model developed in this work enables the economical optimisation of a biomass supply network, while simultaneously limiting the CO_2 emissions that the biomass supply network can generate, in line with the EU Directive 2018/2001 requirements. Furthermore, through the extension of the model to multi-periods, the developed model is enabled to consider the seasonality of biomass supply during the year. The study also demonstrates the linkage between GIS mapping and route assessment with the graph theory approach for biomass supply network optimisation. Paper 5 also integrates GIS mapping and graph theory approaches. The approach developed in the scope of this paper is more appropriate for feedstocks which occur during the whole year and have a limited number of options which may contribute towards GHG production. In this paper, the GIS tool is used to evaluate the suitability of feedstocks for biogas production based on their transport distance and the maximum allowable greenhouse gas (GHG) emissions allocated for feedstock transportation, in line with the requirements outlined in Directive 2018/2001. These limitations are incorporated as part of the input data to develop the maximal structure and optimize the biomass supply network using a p-graph approach. Both Paper 4 and Paper 5 proved the value of the use of the graph theory approach in modelling residual biomass supply network, for different types of case study areas, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria.

To enhance the economic feasibility of biogas production, it is crucial to explore additional sources of waste materials and prioritize the utilization of such materials in biogas production. The future should aim to expand the range of eligible feedstocks for biogas production, thus enhancing the economic feasibility of this process. The impact of co-digestion of those feedstocks should be also examined. Furthermore, the possibilities of transforming digestate (by-product of biogas production), into a range of high-value products, such as biofertilizers and biopesticides, should be thoroughly examined.

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6 CURRICULUM VITAE

Ana Kodba (maiden Lovrak), mag. Ing. Mech. was born on May 3rd 1992 in Koprivnica, Croatia. After finishing the high school "Gimnazija Dr. Ivana Kranjčeva" in Đurđevac, she enrolled Faculty of Mechanical Engineering and Naval Architecture University of Zagreb, where she finished the undergraduate studies in mechanical engineering in 2015, and the graduate studies in 2017. During her graduate studies, she took part in the Master program in the Electric Power Industry at Universidad Pontificia Comillas, Madrid. She is alumni member of the Student association for promoting energy efficiency and consulting, in which she actively participated during her studies. In 2017 she enrolled in a PhD programme under the mentorship of assoc prof. Tomislav Pukšec. Since 2018, she has been employed at the Department of Energy and Power Engineering, as a Research and Project Assistant. Her primary field of PhD research involves economical and sustainable utilization of agricultural residues and industrial by-products for biogas production, GHG savings assessment, GIS mapping of livestock manure, agricultural residues, industrial residues and by-products, underutilised biomass feedstocks and biomass supply network optimisation. She is the first author of 5 scientific papers (indexed CC bases) and 10 conference papers presented at international scientific conferences. She participated in the preparation, implementation and coordination of projects within Horizon Europe, Horizon2020, Interreg Adrion, Erasmus and national programs (Croatian Science Foundation). She attended numerous international scientific conferences. She participates in the organisation of SDEWES conference series as a part of the local organising committee. She serves as a Reviewer for Renewable and Sustainable Energy Reviews, Journal of Sustainable Development of Energy, Water and Environment Systems, Energy and Applied Energy journal.

List of published scientific journal papers:

- **Lovrak, Ana**; Pukšec, Tomislav; Duić, Neven. A Geographical Information System (GIS) based approach for assessing the spatial distribution and seasonal variation of biogas production potential from agricultural residues and municipal biowaste // Applied Energy, 267 (2020), 115010, 12 doi:10.1016/j.apenergy.2020.115010
- **Lovrak, Ana**; Pukšec, Tomislav; Grozdek, Marino; Duić, Neven. An integrated Geographical Information System (GIS) approach for assessing seasonal variation and

spatial distribution of biogas potential from industrial residues and by-products // Energy (Oxford), 239 (2021), 122016, 12 doi:10.1016/j.energy.2021.122016

- **Kodba, Ana**; Pukšec, Tomislav; Duić, Neven. Analysis of Specific Greenhouse Gas Emissions Savings from Biogas Production Based on Agricultural Residues and Industrial By-Products // Energies, 16 (2023), 3721, 15 doi:10.3390/en16093721
- **Kodba, Ana**; Pukšec, Tomislav; Duić, Neven. P-Graph approach for the economical optimisation of biomass supply network that meets requirements on greenhouse gas emissions savings - A case study of rural areas // Journal of Cleaner Production, 416 (2023), 137937, doi: 10.1016/j.jclepro.2023.137937
- **Kodba, Ana**, Pukšec Tomislav; Duić, Neven. P- Graph approach for the optimisation of biomass supply network for biogas production in urban areas // Optimisation and Engineering, (2023), doi: 10.1007/s11081-023-09819-7

7 SUMMARY OF PAPERS

PAPER 1

A. Lovrak, T. Pukšec, and N. Duić, A Geographical Information System (GIS) based approach for assessing the spatial distribution and seasonal variation of biogas production potential from agricultural residues and municipal biowaste // Applied Energy, 267 (2020), 115010, 12 doi:10.1016/j.apenergy.2020.115010

This paper presents a Geographical Information System (GIS) based approach for the assessment of the spatial distribution of the biogas production potential from agricultural residues, by taking into consideration the seasonal variation of biomass production, in order to assess the influence of biomass seasonality. This paper contributes to the first objective of this doctoral thesis (to determine the spatial distribution and to calculate the influence of the seasonality of the residual biomass generation) and to the first scientific contribution (the novel approach for the assessment of seasonality of technical potential of agricultural residues and industrial by-products for biogas production). The presented approach was tested in a case study of Croatia and the final results are representing the seasonal and spatial distribution of biogas potential at the spatial level of 1 km x 1 km. The results show that there is a strong need to include the influence of seasonality in the assessment of biogas potential for the lignocellulosic agricultural residues. The benefits are demonstrated in two examples that resulted in 12% and 40% lower storage facility capacity by using the proposed approach, compared to the currently used approaches.

In this paper, Ana Lovrak (Kodba) developed the method, performed the computations and wrote the manuscript. Tomislav Pukšec encouraged Ana Lovrak to enhance the description of the research gap, update the literature review, validate the benefits of the proposed method and to improve clarification of the originality of the paper. All authors discussed the results and provided critical feedback and helped shape the research and manuscript.

PAPER 2

A. Lovrak, T. Pukšec, M. Grozdek and N. Duić, An integrated Geographical Information System (GIS) approach for assessing seasonal variation and spatial distribution of biogas potential from industrial residues and by-products // Energy, 239 (2021), 122016, 12 doi:10.1016/j.energy.2021.122016

This paper presents a Geographical Information System (GIS) based approach approach for the assessment of biogas potential from industrial residues and by-products, by taking into consideration spatial and seasonal variation of feedstock production. This paper contributes to the first objective of this doctoral thesis (to determine the spatial distribution and to calculate the influence of the seasonality of the residual biomass generation) and to the first scientific contribution (the novel approach for the assessment of seasonality of technical potential of agricultural residues and industrial by-products for biogas production). This approach was tested through the case study of two Croatian counties. The results are presenting the spatial distribution and seasonal variation of the biogas potential from residues and by-products of considered industries. The results proved the hypothesis that there is a strong need to include a seasonal aspect when defining the biomass potential viable for biogas production, due to the low annual load factor calculated for potential biogas sites, which range from 0.1-0.24 for the case when feedstock storage is not available.

In this paper, Ana Lovrak (Kodba) developed the method, performed the computations and wrote the manuscript. Tomislav Pukšec encouraged Ana Lovrak to enhance the description of the research gap and to improve clarification of the originality of the paper. He supervised the findings of the work. All authors discussed the results and provided critical feedback and helped shape the research and manuscript.

PAPER 3

A. Kodba, Pukšec, Tomislav; Duić, Neven Analysis of Specific Greenhouse Gas Emissions Savings from Biogas Production Based on Agricultural Residues and Industrial By-Products // Energies, 16 (2023), 3721, 15 doi:10.3390/en16093721

The aim of this study was to analyse specific greenhouse gas emissions savings for a variety of agricultural residues, industrial by-products, and municipal biowaste. This paper contributes to the second objective of this doctoral thesis (to define constraints for which different types of industrial by-products and agricultural residues meet sustainability and greenhouse gas emissions saving criteria) and the second scientific contribution of this doctoral thesis (defined constraints for which different types of industrial by-products and agricultural residues meet sustainable and greenhouse gas emissions saving criteria) This study defined sustainability criteria and delivered values for the maximum transport distance of agricultural residues and industrial by-products to achieve the greenhouse gas (GHG) emissions saving requirement (80%), compared with fossil fuel comparator, defined by Directive 2018/2001. The obtained results can be used as the constraints in the optimisation of the biomass supply chains for the feedstocks considered in this work.

In this paper, Ana Kodba made the literature review, performed the computations and wrote the manuscript. Tomislav Pukšec critically reviewed the results of the paper and supervised the findings of the work. All authors discussed the manuscript and provided critical feedback and helped shape the research and manuscript.

PAPER 4

A. Kodba, T Pukšec and N. Duić, P-Graph approach for the economical optimisation of biomass supply network that meets requirements on greenhouse gas emissions savings - A case study of rural areas // Journal of Cleaner Production, 416 (2023), 137937, doi: 10.1016/j.jclepro.2023.137937

This work presents a novel P-graph-based model for optimizing a biomass supply network. The objective of this optimization is twofold: to find the most cost-effective biomass supply network with a minimum cost, while also fulfilling the required sustainability and greenhouse gas (GHG) emissions savings defined in Directive 2018/2001 (80% savings compared to fossil fuel comparators) for the use of biogas. Additionally, seasonal variation in biomass supply was integrated into the model by using a multiperiod approach. This paper contributes to the third objective of this doctoral thesis (to prove the value of the use of graph theory approach in modelling residual biomass supply network, for different types of case study areas, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria) and to the third scientific contribution (the mathematical model for residual biomass supply network modelling from sustainability, greenhouse gas emissions savings and economic point of view). Furthermore, in the scope of this paper, a hypothesis of this doctoral thesis has been confirmed (the hypothesis of this research is that economically feasible residual biomass supply network for biogas production, that meets sustainability and greenhouse gas emissions saving criteria, could be determined with graph theory approach).
The approach was tested in a case study located in a rural area. This model can benefit a wide range of stakeholders, including biogas plant operators, policymakers, researchers, and energy regulatory authorities.

In this paper, Ana Kodba made the literature review, developed the method, performed the computations and wrote the manuscript. Tomislav Pukšec was involved in method development, critically reviewed the results of the paper and supervised the findings of the work.. All authors discussed the manuscript and provided critical feedback and helped shape the research and manuscript.

PAPER 5

A Kodba, T. Pukšec and N. Duić-P- Graph approach for the optimisation of biomass supply network for biogas production in urban areas // Optimisation and Engineering, (2023), doi: 10.1007/s11081-023-09819-7

This paper introduces a model that focuses on the economic optimization of a biomass supply network for biogas production in urban areas. The selected feedstocks considered in the model are biowaste and residues sourced from restaurants, shops, and the food and beverage industry. This paper contributes to the third objective of this doctoral thesis (to prove the value of the use of graph theory approach in modelling residual biomass supply network, for different types of case study areas, which is economically feasible, but also meet sustainability and greenhouse gas emissions saving criteria) and to the third scientific contribution (the mathematical model for residual biomass supply network modelling from sustainability, greenhouse gas emissions savings and economic point of view). This study employs an enhanced GIS-based approach that integrates greenhouse gas (GHG) requirements by incorporating a maximal allowed transport distance. These GHG-based requirements align with the specifications outlined in Directive 2018/2001.

In this paper, Ana Kodba made the literature review, developed the method, performed the computations and wrote the manuscript. Tomislav Pukšec was involved in method development, critically reviewed the results of the paper and supervised the findings of the work. All authors discussed the manuscript and provided critical feedback and helped shape the research and manuscript.

PAPER 1

A Geographical Information System (GIS) based approach for assessing the spatial distribution and seasonal variation of biogas production potential from agricultural residues and municipal biowaste

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ABSTRACT

Bioenergy can be produced from a wide range of feedstocks and can be utilised for production of renewable electricity, thermal energy, chemicals or transportation fuels. Anaerobic digestion technology (AD) for biogas production has an important role in achieving circular economy goals, as it may not only recover the energy contained in the biomass but also contribute to nutrient recovery and reduction of greenhouse gas emissions. The expansion of biogas production promotes the need for assessment of the technical potential of biomass, which is available for biogas production and is not in the competition with other purposes. This research work presents a Geographical Information System (GIS) based approach for the assessment of the spatial distribution of the biogas production potential by taking into consideration seasonal variation of biomass production, in order to assess the influence of biomass seasonality. The method developed in this research work is based on a combination of statistical and spatial explicit methods. The presented approach was tested in a case study of Croatia and the final results are representing the seasonal and spatial distribution of biogas potential at the spatial level of 1 km x 1 km. The results show that there is a strong need to include the influence of seasonality in assessment of biogas potential for lignocellulosic agricultural residues. The benefits are demonstrated in two examples that resulted in 12% and 40% lower storage facility capacity by using the proposed approach, compared to currently used approaches.

KEYWORDS

GIS, biogas, agricultural residues, seasonal variation

INTRODUCTION

Energy produced from biomass can be in form of bioliquids, biogas or solid biomass and may represent one of major options for substituting fossil fuels in the energy mix [1]. AD technology has a high potential for significant reduction of waste through the generation of high value products [2]- biogas, which can be used for electrical and thermal energy production, transport

or as the substitute for natural gas (if upgraded to biomethane) and digestate, which is suitable as a fertiliser for agricultural production, due to high ammonium-N/total N ratio [3].
Those advantages have been recognised by the European Commission, which has regarded in EU waste legislation [4] AD as a recycling operation in the waste hierarchy.
The number of European biogas plants has increased steadily over the past decade. By the end of 2017, there were 17783 biogas plants and 540 biomethane plants in operation in Europe [5]. This has resulted with a significant increase in food and feed crops (mostly maize silage) utilization for biogas production, due to high biogas yields and favourable support. However, utilization of feedstocks grown on agricultural land indicates that bioenergy production may be in competition with alternative demands for food and material [6] and leads to negative environmental impacts due to direct and indirect land use change.
In December 2018 the revised Renewable Energy Directive entered into force, which set up the targets and constraints for future biogas utilization in transport, as well in electricity, heating and cooling production. The new directive defines numerous sustainability and GHG emission criteria that biogas used in transport, electricity, heating and cooling production must fulfil. Furthermore, it sets a target that the contribution of advanced biofuels and biogas produced from the feedstock such as algae, straw, animal manure, husks, industrial waste etc., should be at least 3.5% in 2030.
New directives and concerns about the sustainability of the biogas production have resulted in increased interest in underestimated feedstocks for biogas production, such as lignocellulosic agricultural residues. Their utilisation does not bring ethical conflicts [7] and can lead to a significant improvement in the environmental sustainability of energy production [8]. In order to define the perspective of shifting to renewable energy systems, first step is to estimate the potential of domestic renewable sources [9].
There have been numerous studies on the assessment of the biomass technical potential. Two main groups of commonly used methods for potential-focused approaches are statistical analysis (non-spatial specific), which relies on statistical data to assess the potential of biomass for energy utilization and other uses, and spatially explicit analysis, which combines spatially explicit data and land use [10].
In the past years, application of GIS tools has been recognised as very useful for biomass potential mapping, as it gives valuable insights into the spatial distribution of the biomass potential and enables optimisation of bioenergy production plants. In the work [11], a GIS tool was used for assessing the spatial distribution of agroforestry residues annual potential and in the work [12] it was used for the assessment of the spatial distribution of annual sustainable crop residue potentials. Spatial distribution of annual biogas potential from non-woody biomass of conservation areas and roadsides for biogas was assessed with a GIS tool in the work [13]. Authors of the work [14] used a GIS tool to assess the annual theoretical and technical potential of chicken manure from various rearing systems in Polish provinces. In the work [15], author presented the method for assessing the annual economic potential of biomass supply from crop residues, in which he used a GIS based approach to identify the areas in China that are likely to produce crop residue. In the work [16], authors have presented a GIS-based combined approach for the determination of the most cost-effective investments in biomass sector. The proposed approach included GIS mapping of annual biomass potential and defining both storage and plant locations. Similarly, in the work [17], authors used a GIS tool to assess annual potential of corn stover, switchgrass and miscanthus in order to assess biofuel production potential and suitable biorefinery locations in the USA, while in the work [18], authors assessed annual potential of food waste, cattle slurry and wheat straw to locate bio-energy facilities. Annual potential of agricultural waste, co-products and by-products was assessed in the work [19], for the 28 member countries of the European Union. Works such as [20] have already shown that the application of GIS tools enables assessment of biomass transportation cost. In the work [21], authors used a GIS tool to assess the annual biogas potential of citrus pulp, olive pomace, whey,

poultry litter, cattle manure and corn silage. In accordance with the results, the authors conducted an economic assessment which allowed them to determine the size and location of four biogas plants in Sicily. The same feedstocks were considered in the work [22], in which authors developed a GIS-based spatial index of feedstock-mixture availability for anaerobic co-digestion. The developed spatial index describes the availability of the specific feedstock in each municipality, in accordance to annual production of respective feedstock and enables identification of municipalities which are most suitable for biogas production. Authors of the work [23] used a GIS tool to identify financially viable locations for biomethane injection to a natural gas network, in accordance with the spatial distribution of the annual potential from grass silage and cattle slurry. In the work [24], a GIS tool was used for annual biomass potential assessment in India, for which authors developed land use maps for the selected pilot regions. Authors of the work [25] developed a regional GIS based method to analyse suitable locations and capacities of biogas plants, based on theoretical annual potential of various biomass resources, as well as transportation distances. Similar to this, authors of the work [26] developed a model to solve the multi-criteria decision problem of identifying the most suitable location for biogas plant, taking into consideration annual potential of slurry, population density, distance to heat plants and transportation-optimal sites.

In general, the efficiency of the waste-to-energy technologies is strongly affected by the distance of the biomass supply and the rate available during the year [25]. Seasonal availability of the cereal and horticultural crops, as well as residual forest biomass on the administrative region level, for the Party of General Pueyrredón (Argentina), was investigated in the work [27].

Seasonality of biomass production affects requested storage facility capacity and consequently, the cost of the logistic supply chain. Authors of the work [28] developed a model to maximize the profit of the biomass supply chain. In this research, one of the variables included in the optimization was a unit land cost, which was used to determine capacity and locations (counties) in which a storage facility would be most economically feasible to install. Total storage capacity for all considered regions was determined in accordance with the annual biomass availability (corn silage, layer hen manure, broiler hen manure and cattle manure).

As can be seen from the literature review, considerable amount of research has been conducted on the development of GIS based approaches for assessment of the biogas potential available on an annual basis. However, generation of agricultural residues is not continuous during the year and since those feedstocks have a low energy density, there is a need for significant storage capacities in case of large time gap between supply and demand. Considering this and the fact that a GIS approach that integrates the seasonal and spatial distribution of biogas potential from agricultural residues and municipal biowaste has not been presented in the previous research, this research work aims to address this research gap. It can be assumed that the integrated assessment of the spatial and seasonal variation could give better insight into the biogas potential and feasibility of its utilization.

METHOD

This work focuses on the assessment of the biogas potential from municipal biowaste and agricultural residues, derived from plants (maize stover, wheat straw, barley straw, oat straw, triticale straw, rapeseed straw, soya-beans straw, sugar beet tops, damaged vegetables) and livestock (manure). As the technologies used to produce biogas are strongly influenced by the structure of the feedstock, the considered feedstocks are divided in two groups: lignocellulosic and non-lignocellulosic biomass. Figure 1 illustrates the biomass classification used in this research work.

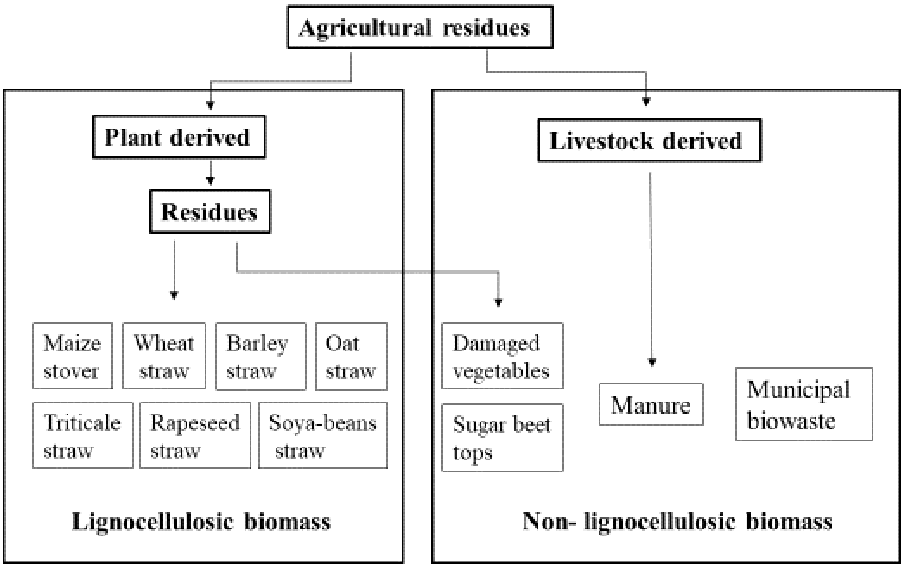

Figure 1 The classification of biomass used in this work

The developed method is based on the combination of statistical and spatial explicit methods. The developed method is divided into the following main steps:

- Biomass technical potential assessment at regional level;
- Energy valorisation of the technical potential;
- Seasonal assessment;
- GIS mapping.

In the next sub-sections, more detailed elaboration of the mentioned steps will be provided.

Biomass technical potential assessment at regional level

In order to assess the technical potential of biomass available for biomass production, this research work aims to investigate the part of the theoretical potential (total production of residues) which is available due to competition with other uses (food, feed, land protection etc.) The technical potential assessment is conducted at regional level, by using the bottom-up approach. The process itself is handled in two steps:

1. theoretical biomass potential assessment at regional level

Theoretical potential of residues from plant production is defined as the annual production of residues generated during agricultural production. As it is shown in equation (1), it is a function of agricultural production and residue to product ratio:

$$P_{th,pl(i,k)} = M_{(i,k)} * RPR_{(i)} \tag{1}$$

where $P_{th.pl(i,k)}$ stands for the theoretical potential of residues from the agricultural category i in the region k [kg], $M_{(i,k)}$ for the production of the agricultural category i in the region k [kg] and $RPR_{(i)}$ for the residue-to-product ratio for the agricultural category i [kg/kg]. RPR factors are obtained from the literature and their values for the considered agricultural categories are presented in Table 1.

Table 1 Residue-to-product ratio for different types of agricultural residues

Biomass type	Residue i	$RPR_{(i)}$	Source
Lignocellulosic biomass	Maize stover	196%	[29]
	Wheat straw	128%	[29]
	Barley straw	135%	[29]
	Oat straw	128%	[29]
	Triticale straw	128%	[29]
	Rapeseed stalk	186%	[30]
	Soya-beans straw	153%	[29]
Non-lignocellulosic biomass	Damaged vegetables	153%	[29]
	Sugar beet tops	20%	[29]

Total production $M_{(i,k)}$ of the specific agricultural category is estimated according to equation (2):

$$M_{(i,k)} = y_{(i,k)} * A_{(i,k)} \tag{2}$$

where $y_{(i,k)}$ stands for the average biomass yield of the agricultural category i in the region k (kg/m^2) and $A_{(i,k)}$ for utilised agricultural land for production of category i in the respective region k (m^2).

Biomass yield $y_{(i,k)}$ represents the amount of biomass produced per unit area (1 m^2). Some of the agricultural cultures have a significant variation of the yield, mostly due to weather and climate conditions, as well as soil properties. For this reason, this approach takes into consideration minimum and maximum yield in the last five years, or more precisely, the average value of those two extremes for each considered region (equation 3).

$$y_{(i,k)} = \frac{y_{MIN(i,k)} + y_{MAX\,(i,k)}}{2} \tag{3}$$

where $y_{(i,k)}$, $y_{MIN\,(i.k)}$ and $y_{MAX\,(i,k)}$ stand respectively for average, minimum and maximum biomass yield of the agricultural category i in the last five years, for the region k (kg/m^2).

In the case of livestock derived residues, the theoretical potential of manure is estimated according to equation (4):

$$P_{th,liv(k,l)} = N_{(k,l)} * MPH_{(k,l)} \tag{4}$$

where $P_{th,liv(k,l)}$ stands for the theoretical potential of manure generated in the region k, for the livestock l [kg], $N_{(k,l)}$ for the number of heads of livestock l in the region k [head] and $MPH_{(k,l)}$ for manure per head ratio (Table 2); annual manure production per livestock type l [kg/head].

Table 2 Manure per head ratio for different livestock

L	Cattle	Dairy cow	Pig	Sheep	Poultry
$MPH_{(l)}$ [kg/head]	12,300	18,830	1,200	400	95
Source	[31]	[32]	[31]	[33]	[31]

2. technical biomass potential assessment at regional level

Technical potential is defined as the part of the theoretical potential which is available due to competition with other uses (food, feed, land protection etc.). The assessment of this potential is based on the previously calculated theoretical potential, sustainable removal rates and competitive uses (for livestock production), according to equation (5);

$$P_{tech\,(i,k)} = P_{th,pl(i,k)} * SRR_{(i)} - COMP_{(k)} \tag{5}$$

where $P_{tech\,(i,k)}$ stands for the technical potential of residues of the agricultural category i in the region k [kg], $P_{th,pl(i,k)}$ for the theoretical potential of residues from the agricultural category i in the region k [kg], $SRR_{(i)}$ for a sustainable removal rate for the agricultural category i [%] and $COMP_{(k)}$ for the amount of residues which should be left for the feeding and bedding of animals in the region k [kg].
Sustainable removal rate ($SRR_{(i)}$) refers to the share of residues which could be collected from the field, by considering the share of the residues which should remain in the field in order to protect the soil from wind and erosion, but also the share which is not possible to collect due to losses in the collecting process. Sustainable removal rates ($SRR_{(i)}$) for considered agricultural categories are listed in Table 3.

Table 3 Sustainable removal rates $SRR_{(i)}$ for considered agricultural categories

i	Wheat, barley, rye, oats etc.	Maize	Sunflower	Rapeseed	Sugar beet	Vegetable
$SRR_{(i)}$	40%	50%	50%	50%	50%	90%
Source	[34]	[34]	[34]	[34]	[30]	[30]

In addition to the residues, which should be left in the field, a part of residues (straw) should be used for competitive purposes, mostly livestock production. The amount of residues required for competitive purposes is calculated according to equation (6):

$$COMP_{(k)} = \sum N_{(a,k)} * SPA_{(a)} * s_{(a)} \tag{6}$$

Where $COMP_{(k)}$ stands for the amount of residues which should be left for the feeding and bedding of animals in the region k [kg], $N_{(a,k)}$ for the number of animals a (-) in the region k, $SPA_{(a)}$ for annual requirements of straw per animal a (kg/year) and $s_{(a)}$ for the share of animals to which $SPA_{(a)}$, refers, since not all farms use a straw for livestock production (%). The values of these parameters are listed in Table 4.

Table 4 The values of the parameters for calculating the competitive use of straw for cattle, pig and sheep [35]

a	Cattle	Pig	Sheep
$SPA_{(a)}$[kg/year]	548	183	37
$s_{(a)}$ [%]	25	12.5	

Energy valorisation of the technical potential

Once the technical biomass potential is assessed, energy potential can be estimated from specific methane yield of fresh feedstocks and lower heating value of methane, according to equation (7):

$$E_{bio\,(i,k)} = P_{tech\,(i,k)} * y_{CH_4,i} * LHV_{CH_4} \tag{7}$$

where $E_{bio\,(i,k)}$ stands for the energy value of biogas potential from residues of the agricultural category i (or municipal biowaste) in the region k [MJ], $y_{CH_4,i}$ for the methane yield from 1 kilogram of fresh feedstock [m^3/kg] and LHV_{CH_4} for methane lower heating value [MJ/m^3]. Since the methane yield of lignocellulosic agricultural residues highly depends on the used pre-treatment method, Table 5 lists the methane yield for the respective pre-treatment method.

Table 5 Specific methane yield from lignocellulosic agricultural residues

Residue i	Pre-treatment method	Methane yield [m3/kg]	Source
Maize stover	Pre-treated with 6% NaOH	0.315	[36]
Wheat straw	Pre-treated with 10% NaOH 100 C	0.305	[37]
Barley straw	Extrusion	0.305	[38], [39]
Oat straw	Steam fermention	0.195	[40]
Triticale straw	Pre-treated with with N-methylmorpholine-N-oxide	0.203	[41], [42]

Rapeseed stalk	wet oxidation pretreatment	0.28	[43]
Soya-beans straw	Trichoderma reesei RUT C30	0.08	[44], [45]

Seasonal assessment

Seasonality of feedstocks' availability is assessed according to the months of harvesting/ occurring of the considered feedstocks. Seasonal assessment of plant derived agricultural residues is calculated form the agricultural crops' harvest calendar. For the municipal biowaste, statistical data on monthly production is used.

Both for the lignocellulosic and non-lignocellulosic biomass the potential for the biogas production is assessed for each month of the year, according to equation (8):

$$E_{bio\ (k,m)} = \sum_i E_{bio\ (i,k,m)} \tag{8}$$

where $E_{bio\ (k,m)}$ stands for the energy value of biogas potential in the region k in the month m [MJ] and $E_{bio\ (i,k,m)}$ for the energy value of biogas potential in the region k, in the month m, for the specific commodity (residue or biowaste) i [MJ].

GIS mapping

In order to perform GIS mapping, the following set of data is necessary: monthly availability of the biogas potential of lignocellulosic and non-lignocellulosic biomass at regional level, georeferenced data on region boundaries and georeferenced land use maps. QGIS tool is used to conduct the mapping process.

Data on monthly availability of biogas potential of lignocellulosic and non-lignocellulosic biomass, calculated in previous steps, is joined to the georeferenced layer of regions' boundaries. In order to carry out a spatial distribution of biogas potential in each region, land cover maps are used. Those maps represent georeferenced information on different types (classes) of physical coverage of the Earth's surface, e.g. grasslands, forests, croplands, lakes, wetlands [46]. Based on two layers of georeferenced information (land cover map and biogas potential at regional level for each month of the year), a biogas potential map is developed. In order to assess the distribution of the biogas potential, the top-down approach is applied and the following equation is used:

$$\mathrm{E_{bio,fiel,m}} = \frac{\mathrm{A_{fiel}}}{\mathrm{A_k}} * \mathrm{E_{bio,k,m}} \tag{9}$$

Where $E_{bio,fiel,m}$ stands for the energy value of the biogas potential for the specific field in the specific month m [MJ], A_{fiel} for the area of the specific field [m^2], A_k for the total agricultural (or urban) area of the region k, in which specific field is located [m^2] and $E_{bio,k,m}$ for the energy value of biogas potential of the region k, for the specific month m [MJ].

CASE STUDY

The presented method was applied in the case study for the Republic of Croatia, for evaluating the spatial distribution and seasonal variation of biogas production potential from agricultural residues, livestock production and municipal biowaste.

According to Eurostat, in 2016 Croatia had 134 460 agricultural holdings (or farms), working 15 460 km^2 of utilised agricultural area, what is around one quarter (27.7%) of the total land area of Croatia [47]. Croatia's territory is classified in 21 administrative regions (20 counties and the city of Zagreb), which are grouped in 2 statistic regions (Continental and Adriatic Croatia) [48].

Prior to GIS mapping, biogas potential was assessed for each Croatian county (NUTS3 region). Data provided by Paying Agency for Agriculture, Fisheries and Rural Development [49] and the Croatia Bureau of Statistic [50] was used for calculating the theoretical biomass potential in each of the regions. The input data for assessing the potential of manure was taken from the register of domestic animals [51] and the list of utilised agricultural land and number of cottages and poultry of private households [52]. When assessing the seasonality of residue generation, data from Table 6 is used.

Table 6 Residue generation month

Biomass type	Residue	Residue generation month	Source
Lignocellulosic biomass	Maize stover	September	[53]
	Wheat straw	June	[54]
	Barley straw	June, July	[55]
	Oat straw	June	[54]
	Triticale straw	July	[55]
	Rapeseed stalk	June	[54]
	Soya-beans straw	September	[53]
Non-lignocellulosic biomass	Damaged vegetables	August, September	[53]
	Livestock manure	Whole year	[54]
	Municipal biodegradable waste	Whole year	[54]
	Sugar beet tops	September	[54]

In order to assess the spatial distribution of the biogas potential, CORINE land Cover map [56], which defines 42 different land classes was used to detect agricultural land, urban areas and dump sites. Figure 2 shows the land cover of Croatia.

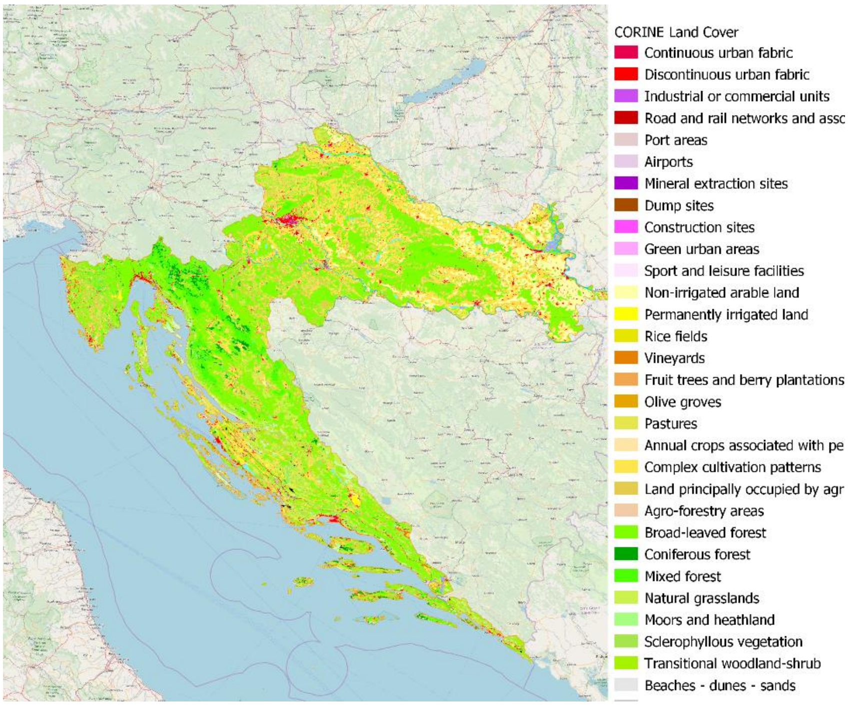

Figure 2 CORINE land Cover -Croatia

RESULTS

The biogas potential from the considered lignocellulosic and non-lignocellulosic feedstocks was calculated at regional level and the seasonal and spatial distribution assessment of the biogas potential at the spatial level of 1 km x 1 km was conducted, as described in the previous sections.

Biogas technical potential assessment at regional level

Non-lignocellulosic biomass

On the national level, the technical potential of non-lignocellulosic biomass available for biogas production is assessed as 3321 GWh (11.96 PJ). Figure 3 presents the energy value of technical potential of non-lignocellulosic biomass available for biogas production for each considered region (Croatian county). It also clearly shows that the highest contribution comes from cattle and dairy cow manure. Osijek- Baranja, Koprivnica-Križevci and Bjelovar-Bilogora counties have the highest potential from all Croatian counties.

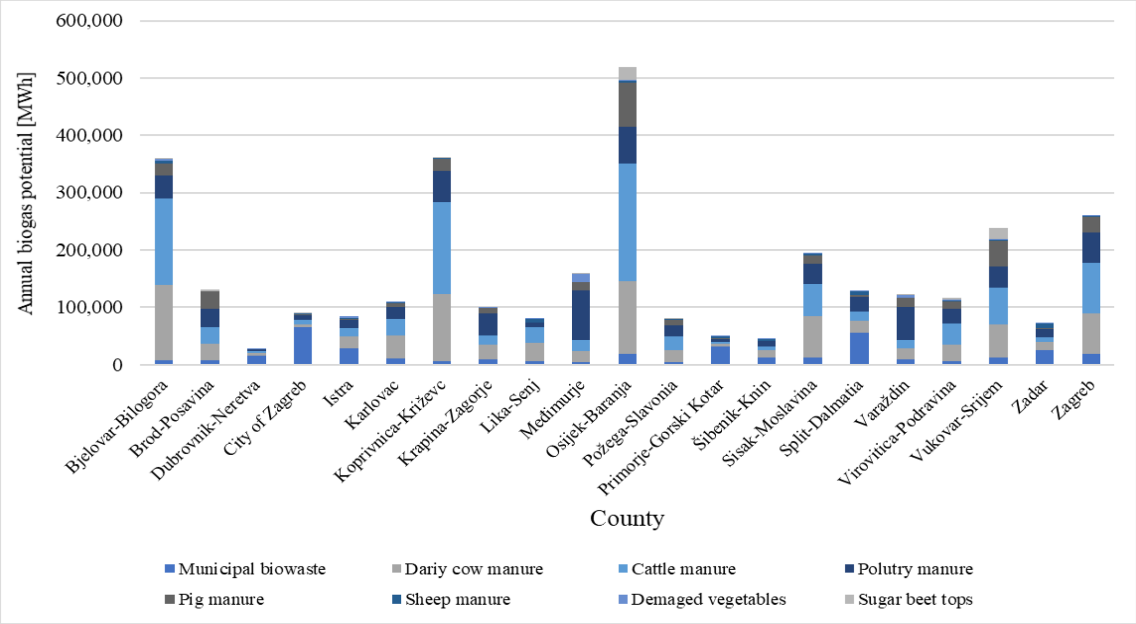

Figure 3 Results of technical potential assessment of non-lignocellulosic biomass available for biogas production

<u>Lignocellulosic biomass</u>

The technical potential of lignocellulosic biomass available for biogas production, which occurs during agricultural production is shown in Figure 4. On the national level, the total technical potential available for biogas production equals 6679 GWh (24 PJ). In all regions, maize stover contributes with the highest share, followed by wheat straw. As it was the case with the non-lignocellulosic biomass, Osijek-Baranja County again has the highest annual technical potential from agricultural production. This correlation can be explained by the fact that in Croatia around 80 % of farms have livestock [57].

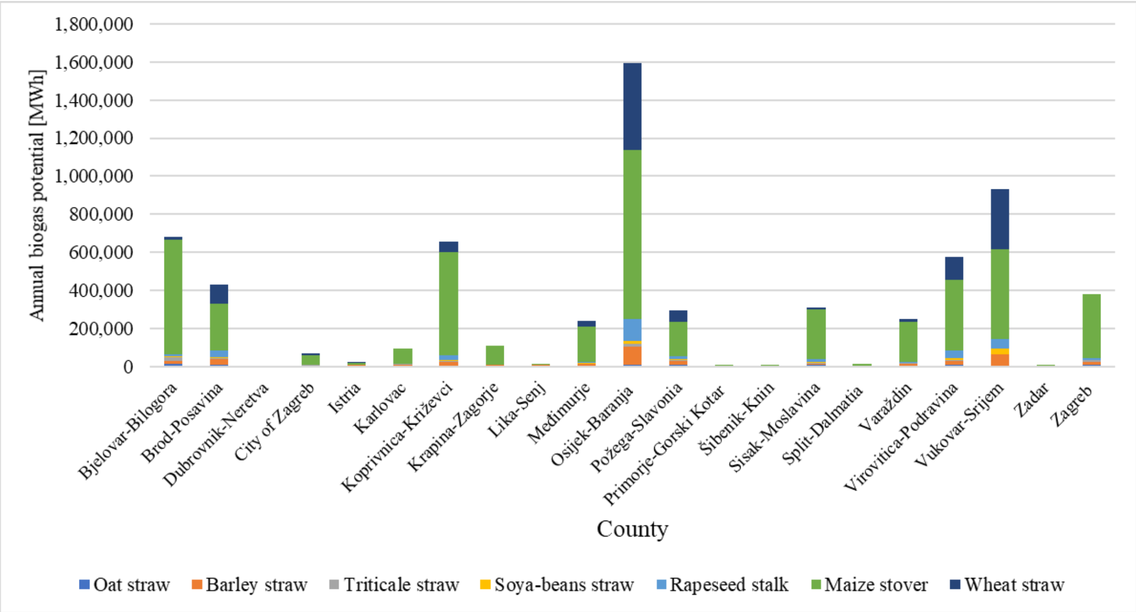

Figure 4 Results of technical potential assessment of lignocellulosic biomass available for biogas production

Seasonal assessment

The monthly availability of biomass potential was determined in accordance with the technical potential assessed at regional level and data on harvesting periods.

Non-lignocellulosic biomass

Table 7 presents the aggregated biogas production potential from non-lignocellulosic biomass for each month of the year for each of the Croatian counties. As it is shown in Table 7, the generation of non-lignocellulosic biomass does not have significant variation during the year. This is due to the nearly continuous production of manure and municipal biowaste, which has a significant share in non-lignocellulosic biomass potential. Therefore, seasonal variations of the considered non-lignocellulosic biomass can be neglected.

Lignocellulosic biomass

Table 8 presents biogas production potential from non-lignocellulosic biomass for each month of the year for each of the Croatian counties. As is it shown in Table 8, considered lignocellulosic feedstocks occur only during three months of the year. Thus, the spatial distribution of the biogas potential was evaluated for each month of its generation.

Table 7 Biogas production potential from non-lignocellulosic biomass

County	January [MWh]	February [MWh]	March [MWh]	April [MWh]	May [MWh]	June [MWh]	July [MWh]	August [MWh]	September [MWh]	October [MWh]	November [MWh]	December [MWh]
Bjelovar-Bilogora	22,355	22,355	22,355	22,355	22,355	22,355	22,355	22,784	25,032	22,355	22,355	22,355
Brod-Posavina	8,159	8,159	8,159	8,159	8,159	8,159	8,159	8,363	10,229	8,159	8,159	8,159
Dubrovnik-Neretva	2,038	2,038	2,038	2,038	2,038	2,038	2,038	2,239	2,201	2,038	2,038	2,038
City of Zagreb	6,883	6,883	6,883	6,883	6,883	6,883	6,883	7,076	7,169	6,883	6,883	6,883
Istria	5,661	5,661	5,661	5,661	5,661	5,661	5,661	7,161	7,119	5,661	5,661	5,661
Karlovac	6,957	6,957	6,957	6,957	6,957	6,957	6,957	7,111	7,989	6,957	6,957	6,957
Koprivnica-Križevci	22,623	22,623	22,623	22,623	22,623	22,623	22,623	22,867	23,308	22,623	22,623	22,623
Krapina-Zagorje	6,372	6,372	6,372	6,372	6,372	6,372	6,372	6,386	6,596	6,372	6,372	6,372
Lika-Senj	5,130	5,130	5,130	5,130	5,130	5,130	5,130	5,162	6,762	5,130	5,130	5,130
Međimurje	9,100	9,100	9,100	9,100	9,100	9,100	9,100	9,531	22,276	9,100	9,100	9,100
Osijek-Baranja	31,318	31,318	31,318	31,318	31,318	31,318	31,318	32062	54,159	31,318	31,318	31,318
Požega-Slavonia	5,011	5,011	5,011	5,011	5,011	5,011	5,011	5363	7,147	5,011	5,011	5,011
Primorje-Gorski Kotar	3,692	3,692	3,692	3,692	3,692	3,692	3,692	3701	3,796	3,692	3,692	3,692
Šibenik-Knin	3,024	3,024	3,024	3,024	3,024	3,024	3,024	3046	3,076	3,024	3,024	3,024
Sisak-Moslavina	12,384	12,384	12,384	12,384	12,384	12,384	12,384	12535	12,715	12,384	12,384	12,384
Split-Dalmatia	9,098	9,098	9,098	9,098	9,098	9,098	9,098	9273	9,465	9,098	9,098	9,098
Varaždin	7,422	7,422	7,422	7,422	7,422	7,422	7,422	8148	11,881	7,422	7,422	7,422
Virovitica-Podravina	7,113	7,113	7,113	7,113	7,113	7,113	7,113	8652	9,783	7,113	7,113	7,113
Vukovar-Srijem	13,849	13,849	13,849	13,849	13,849	13,849	13,849	14911	32,811	13,849	13,849	13,849
Zadar	5,018	5,018	5,018	5,018	5,018	5,018	5,018	5332	5,635	5,018	5,018	5,018
Zagreb County	16,543	16,543	16,543	16,543	16,543	16,543	16,543	16732	17,170	16,543	16,543	16,543

Table 8 Biogas production potential from lignocellulosic biomass

County	January [MWh]	February [MWh]	March [MWh]	April [MWh]	May [MWh]	June [MWh]	July [MWh]	August [MWh]	September [MWh]	October [MWh]	November [MWh]	December [MWh]
Bjelovar-Bilogora	-	-	-	-	-	51,596	23,818	-	60,7543	-	-	-
Brod-Posavina	-	-	-	-	-	156,991	21,205	-	252,630	-	-	-
Dubrovnik-Neretva	-	-	-	-	-	7	2	-	342	-	0	0
City of Zagreb	-	-	-	-	-	12,481	2,841	-	50,480	-	-	-
Istria	-	-	-	-	-	5,255	4,653	-	9,348	-	-	-
Karlovac	-	-	-	-	-	5,709	6,331	-	82,851	-	-	-
Koprivnica-Križevci	-	-	-	-	-	91,916	15,758	-	546,327	-	-	-
Krapina-Zagorje	-	-	-	-	-	4,055	3,770	-	101,689	-	-	-
Lika-Senj	-	-	-	-	-	4,810	5,180	-	5,005	-	-	-
Međimurje	-	-	-	-	-	47,273	7,725	-	185,605	-	-	-
Osijek-Baranja	-	-	-	-	-	627,659	59,533	-	906,401	-	-	-
Požega-Slavonia	-	-	-	-	-	98,965	13,508	-	182,916	-	-	-
Primorje-Gorski Kotar	-	-	-	-	-	477	450	-	6,801	-	-	-
Šibenik-Knin	-	-	-	-	-	776	689	-	1,338	-	-	-
Sisak-Moslavina	-	-	-	-	-	29,491	8,616	-	268,990	-	-	-
Split-Dalmatia	-	-	-	-	-	1,239	1,186	-	8,350	-	-	-
Varaždin	-	-	-	-	-	28,962	8,510	-	211,276	-	-	-
Virovitica-Podravina	-	-	-	-	-	178,600	15,579	-	383,519	-	-	-
Vukovar-Srijem	-	-	-	-	-	396,688	32,350	-	501,805	-	-	-
Zadar	-	-	-	-	-	2,125	2,055	-	5,695	-	-	-
Zagreb	-	-	-	-	-	22,684	14,738	-	343,882	-	-	-

GIS mapping

The final results present georeferenced maps of the seasonal and spatial distribution of the biogas potential at the spatial level of 1 km x 1 km. These maps were developed using open-source QGIS software.

Non-lignocellulosic biomass

As mentioned above, the seasonal variation of the considered non-lignocellulosic biomass can be neglected. Thus, the spatial distribution of non-lignocellulosic biomass was evaluated for one average month and is presented in Figure 5.

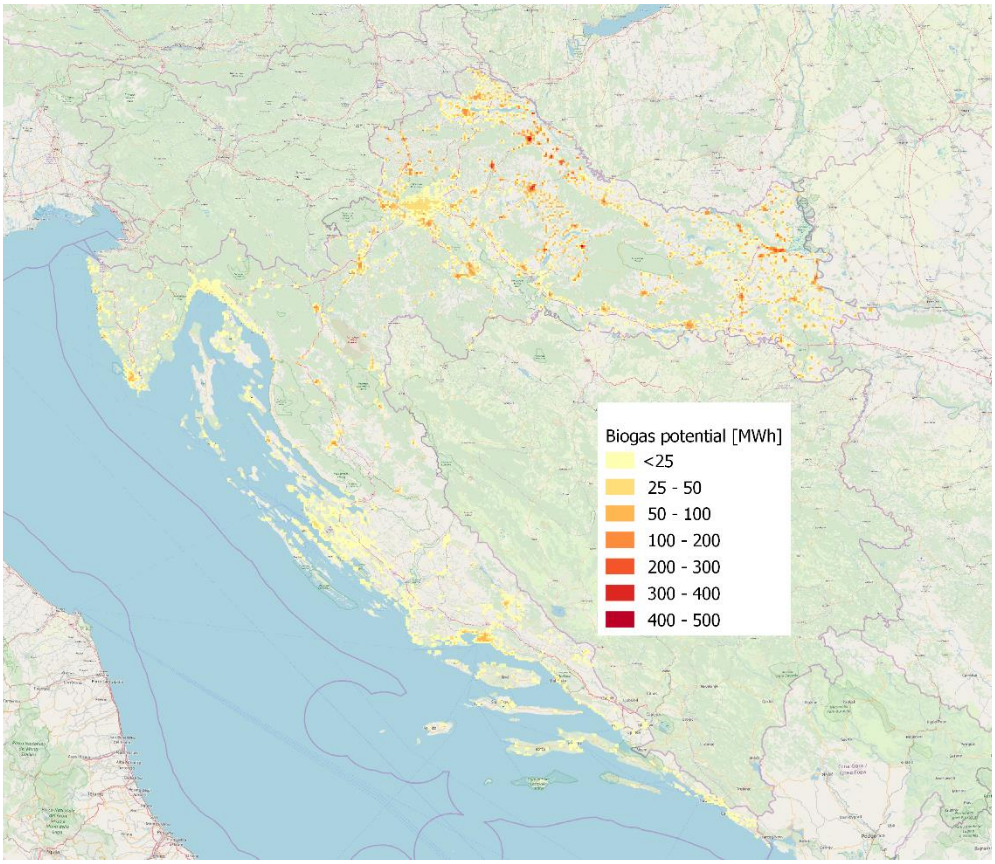

Figure 5 Biogas potential from municipal biowaste and non-lignocellulosic agricultural residues for one average month

Figure 5 clearly shows that biogas potential from biowaste and manure is mostly located in the continental part of Croatia, in rural areas. This can be confirmed by the fact that the city of Zagreb, which has by far the greatest population in Croatia, has the lowest density of biogas potential and lowest total biogas potential. On the other hand, biogas potential in Adriatic part of Croatia mostly follows the population density.

Lignocellulosic biomass

The spatial distribution of the biogas potential was evaluated for each month of its generation. As it is shown in Figure 6, the seasonal variation of the biogas potential significantly differs between the counties. Furthermore, Figure 6 clearly shows that the peak potential is in September. The results obtained for lignocellulosic biomass implies that utilization of lignocellulosic biomass for biogas production requires significant storage capacities.

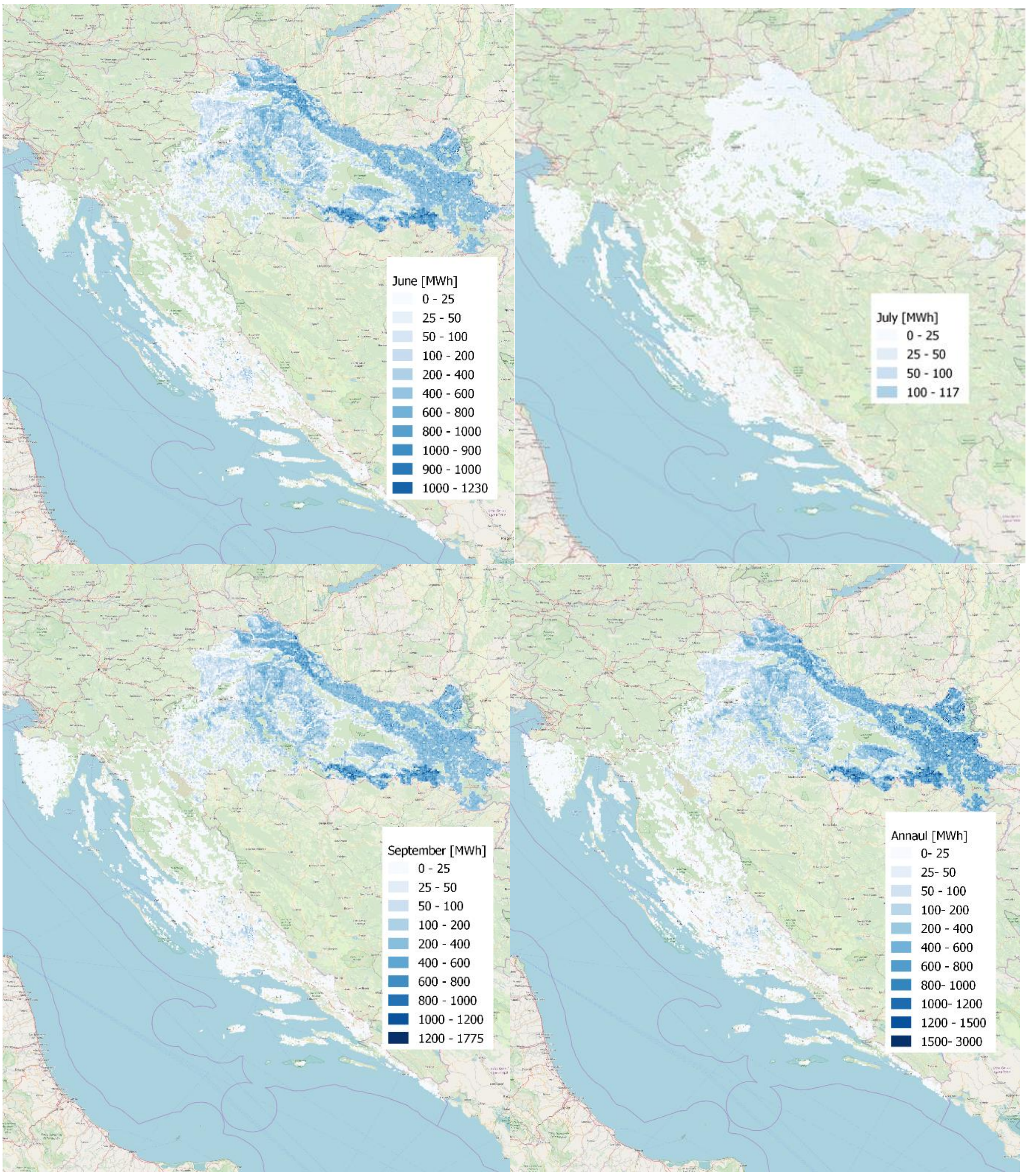

Figure 6 Annual and monthly biogas potential from lignocellulosic agricultural residues

The advantage of integrated seasonal and spatial mapping is the possibility of storage facility capacity assessment. In order to prove the benefits of the proposed approach, it was compared with currently used approaches. Therefore, required storage capacities were assessed for two examples selected from the area presented in Figure 6, for which spatial and seasonal assessment was conducted in the previous steps. For both examples, storage facility capacity is calculated for the lignocellulosic agricultural residues which are being produced in the area of

90 km^2 (grid with 90 cells). In order to handle supply risk of feedstock, the minimum stored amount at the end of one month is set to be sufficient to cover the feedstock demand for at least one and a half month. Furthermore, the demand for the feedstocks is expected to be continuous during the year, for both examples.

The first example is located in Varaždin county (northern Croatia) and presented in Figure 7.

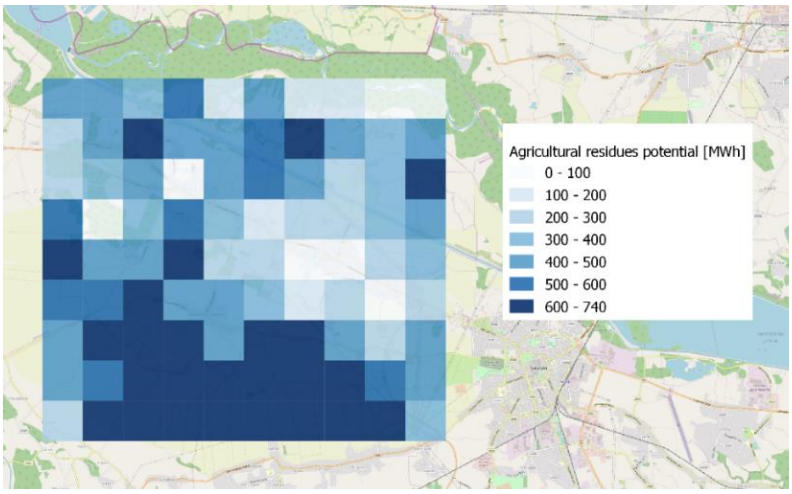

Figure 7 Annual potential of agricultural residues- Varaždin county (Example 1)

In the first example, annual technical potential equals 14105 tonnes of agricultural residues, which has the biogas potential of 0.143 PJ (39952 MWh). Annual variations of biomass potential (supply) and stored quantities are presented in Table 9.

Table 9 Seasonal variation of biomass potential (supply) and stored amount (Example 1)

	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Supply [t]	-	-	-	-	-	1,774	508	-	11,823	-	-	-
Demand [t]	11,75	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1175	1,175	1,175	1,175
Stored amount* [t]	77,08	6,532	5,357	4,181	3,006	3,605	2,937	1,762	12,410	1,1234	10,059	8,883

*feedstock amount stored at the end of the month

As it is shown in Table 9, the maximum stored feedstock amount is in September and it equals 12410 t. Thus, 12410 t can be considered as the necessary storage facility capacity. In other cases where biomass availability is assessed at the annual basis and there is no information on the seasonal variation, it is assumed that the value of necessary storage facility capacity is the same as the annual biomass potential. By comparing the storage facility calculated with this approach to the one related to the annual assessment, it is shown that the application of seasonal assessment results in 12% lower storage facility capacity. The second example is located in Brod-Posavina county (eastern Croatia) and is shown in Figure 8.

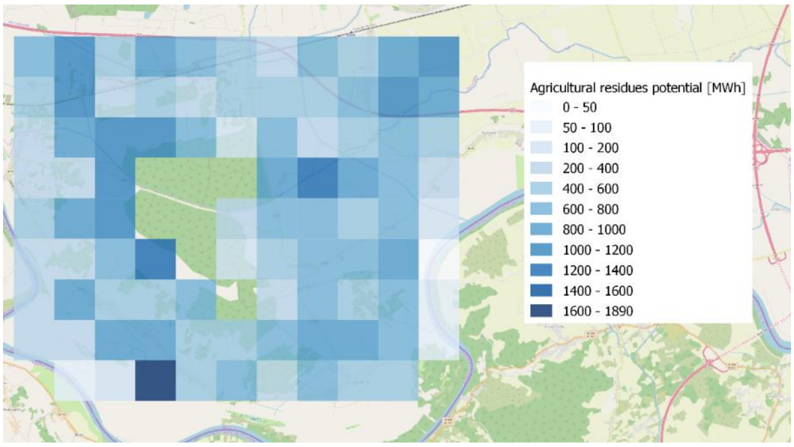

Figure 8 Annual potential of agricultural residues- Brod-Posavina county (Example 2)

In the second example, annual technical potential equals 20579 tonnes of agricultural residues, which has the biogas potential of 0.198 PJ (55017 MWh). Annual variations of the biomass potential (supply) and stored amounts are presented in Table 10.

Table 10 Seasonal variation of biomass potential (supply) and stored amount for example 2

	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Supply [t]	-	-	-	-	-	4,105	670	-	1,5803	-	-	-
Demand [t]	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175	1,175
Stored amount* [t]	7,708	6,532	5,357	4,181	3,006	3,605	2,937	1,762	12,410	11,234	10,059	8883

*feedstock amount stored at the end of the month

As in the previous example, the maximum stored feedstock amount is in September. However, in this example, the difference between biomass potential in September and in other summer months is not so significant. When comparing the storage facility capacity determined with this approach, and the one related to the annual assessment, it is shown that the application of seasonal assessment results in 40% lower storage facility capacity for this specific example.

DISCUSSION

The developed method can be used for local, regional and national planning of biogas production projects, supply chain risk management and storage facility capacity assessment. The implementation of those projects have a potential to increase local renewable production, but also provide biological stabilization of manure, agricultural residues and municipal biowaste by AD and therefore decrease related GHG emissions that would otherwise occur without AD [58]. Furthermore, one of the positive externalities beyond renewable energy production and GHG reduction is increased soil organic matter, due to continuous return of digestate. This results in increased food and feed production, compared to the case prior to bioenergy production [59].

In the previous research works, the authors used GIS tools for the assessment of the spatial distribution of the annual biomass technical assessment. As it is shown by the results obtained in this research work, this gives a sufficient insight for the feedstocks with near continuous monthly production, such as manure and municipal biowaste, what is not the case with lignocellulosic biomass.

The method developed in the work [27] which investigates the monthly availability of crop, horticultural and forestry residues, enables seasonal assessment of biomass potential on a regional basis. Due to wide geographic distribution of the agricultural residues and low energy density of the considered feedstocks, information on the biomass monthly availability on regional level is often not sufficient for the determination of the feasibility of biogas utilization. Therefore, the added value of the integrated approach presented in this research work is that it provides better insight into the biogas potential availability and required storage facility capacity. Assessment of the storage facility capacity is a part of the optimization of biomass supply chain in some of the research works, such as the work [28]. In this work, authors have determined the storage facility capacity in accordance with the price of the land unit where the storage unit is planned to be built, but with the constraint that capacities of all storage facilities should equal to the annual biomass potential in the considered regions. As it is shown from the two examples given in the section above, the approach presented in this research work results with lower storage facility capacity, due to better insight into the biomass availability. This shows the importance of including integrated seasonal and spatial variation in the assessment of the potential of lignocellulosic residues available for biogas production, in order to have more accurate input data for the feasibility projects for biogas utilization.

CONCLUSION

This paper presents a Geographical Information System (GIS) based approach for evaluating the spatial distribution and seasonal variation of biogas production potential. In detail, the biogas production potential was assessed in accordance with the technical potential which was calculated at regional level and takes into consideration the sustainable removal rate of biomass, as well as competitive purposes. This approach is used for the case study of Croatia. Furthermore, the potential of agricultural residues and organic municipal biowaste was assessed in order to define the potential for biogas production.

The results at national level show that the annual potential for biogas production from manure, damaged vegetable and municipal biowaste equals 11.96 PJ, while the potential of lignocellulosic agricultural residues is 24 PJ. The use of the GIS tool proved to be beneficial for the seasonal assessment as it enabled fast and accurate seasonal assessment. The results proved that seasonal variations of the potential of non-lignocellulosic agricultural residues and municipal biowaste can be neglected since the generated feedstocks which make the most significant share of considered feedstocks (manure and biowaste) have near-continuous generation during the whole year. It is not the case with the generation of lignocellulosic agricultural residues, which have a significant variation during the year. In this research work, it is shown in two examples that application of seasonal assessment approach leads to lower storage facility capacity requirements. For the considered examples, it resulted in 12% and 40% lower storage facility capacity requirements, compared with annual assessment approach. It also proved that seasonal variation of biogas potential from non-lignocellulosic biomass does not follow the same trend between the counties. As it is presented in the results, spatial and seasonal assessment of the potential of lignocellulosic agricultural residues available for biogas production provide more accurate input data. Therefore, there is a strong need to include seasonal variations in potential assessment of the considered feedstocks. The developed method can be used for development of a GIS based decision support system, that could be used for the national, local and regional development of biogas production. The integrated spatial and seasonal assessment gives planners and investors a detailed and clear view on the distribution of the biogas potential at high spatial level and monthly availability. As further research, this method can be extended to include more feedstocks feasible for biogas production but barely valorised, such as industrial residues and by-products.

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PAPER 2

An integrated Geographical Information System (GIS) approach for assessing seasonal variation and spatial distribution of biogas potential from industrial residues and by-products

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ABSTRACT

Biogas production through anaerobic digestion technology offers numerous benefits as it may not only recover a part of the energy contained in the biomass but also contributes to circular economy targets. Concerns about biogas production from feed and food crops raise the need for the assessment of biogas potential produced out of biomass, which is not in competition with the other purposes, such as potential of industrial residuals and by-products. This research presents the approach for the assessment of biogas potential from industrial residues and by-products, by taking into consideration spatial and seasonal variation of feedstock production.

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In this work, considered feedstocks are those which occur in sugar, wine, vegetable and olive oil industries. This approach was tested through the case study of two Croatian counties. The results are presenting the spatial distribution and seasonal variation of the biogas potential from residues and by-products of considered industries. The results proved the hypothesis that there is a strong need to include a seasonal aspect when defining the biomass potential viable for biogas production, due to the low annual load factor calculated for potential biogas sites, which range from 0.1-0.24 for the case when feedstock storage is not available.

Keywords: biogas, GIS, seasonality, industrial by-products and residues

HIGHLIGHTS

- A novel approach for integrated assessment of seasonal and spatial distribution of biogas potential
- The influence of seasonality on the cost-effectiveness of biogas production is assessed
- The results proved a strong need to include seasonal aspect when defining biogas potential

INTRODUCTION

Degradation of organic materials under anaerobic conditions by microorganisms results in the production of biogas, renewable fuel used for production of electricity, heat or biomethane-biogas cleaned of impurities, which can be used as a natural gas substitution.

The EU policies on renewable energy production introduced various support schemes that encouraged the increase of biogas production [1]. By the end of 2018, there were 18,802 biogas plants and 610 biomethane plants in operation in Europe [2]. Those plants use, to the greatest extent, maize silage as a substrate for biogas production [3], due to high biomass and biogas yields, as well as feedstock storability [4]. However, utilization of feedstocks that have been grown on agricultural land has caused concerns over the negative environmental impact due to direct and indirect land-use change. Direct land-use change is defined as the land-use change that occurs when biogas feedstock cultivation displaces a prior crop that was cultivated on that land, for other use (i.e. food or feed production). Thus, a direct connection can be made between biogas production and land-use change [5]. On the other hand, indirect land-use change occurs when the cultivation of crops for biogas (or bioliquids, biomass) production displaces the traditional production of crops for food and feed purposes, which result in additional demand on land. This increasing pressure on land can lead to the extension of agricultural land into areas with high carbon stock such as wetlands, peat land and forests, thus causing additional greenhouse gas emissions [6].

In order to diminish this negative environmental impact, residual resources are expected to have increased utilization due to lower environmental impact [7]. In 2018, the revised Renewable Energy Directive [8] has set minimum GHG savings, which biogas used for electricity, heat and cooling production has to compile, and limits the use of maize silage. It has been proven by researchers that alternative substrates like industrial by-products yield better prospects and lower production costs [9]. In addition, Korberg et al. [10] concluded that free feedstock for biogas generation brings significant energy system cost reductions.

Besides the contribution of biogas to renewable energy generation, biogas generation from wastes must be viewed from the standpoint of bio circular economy and sustainable development. While generating renewable energy and minimising environmental impacts of various types of waste materials, biogas generation through anaerobic digestion technology (AD) meets requirements related to waste treatment. In addition, digestate obtained through AD technology is suitable as agricultural fertiliser, due to the high ammonium-N/total N ratio [11]. Residues and by-products which occur in some industries for food and beverage production, such as industries for sugar, wine, olive oil and vegetable production, are noted as feasible feedstock for biogas generation. This has been proven in various experimental studies, such as the study [12] where Al Afif et al. concluded that the quality of biogas produced from olive mill solid waste was sufficient for all experiments. Furthermore, Duarte et al [13] concluded in their experimental research that industrial residues such as residues from vegetable and fruit industry are promising co-digestion substrates due to the positive synergetic effect demonstrated in increased biogas yield.

In this context, the assessment of biogas potential of residues and by-products captivates the attention of many researchers. Moreda et al. [14] calculated the yearly methane potential of numerous agricultural residues and by-products from agro-industrial production in Uruguay. In this review work, Moreda et al detected residues and by-products from a brewery, dairy, fish, malting, poultry, rice, sausage, slaughterhouse, tannery, wine and wool scouring industry as viable for biogas production and assessed respective annual potential on the national level. Similar to this, Kythreotou et al. [15] assessed the annual biogas potential of several potential sources for biogas production, such as biodegradable fractions of municipal solid waste, residues from food and beverage industry and sewage sludge. Assessment of the biogas potential from manure and slaughterhouse by-products was conducted in the work [16]. In this work, Mahmoud Ali et al. used the GIS tool to present the distribution of annual biogas potential from the above-mentioned feedstocks, between Mauritania's provinces. In another work [17], Pereira et al. calculated the economic potential for electricity generation from vinasse in accordance with the annual potential of biogas from vinasse, obtained from sugarcane processing. The economic potential was calculated with a GIS tool, for each municipality of the state of São Paulo. Höhn et al. [18] developed GIS-based methods for the analysis of suitable biogas plant location considering the spatial variation of annual biogas potential from numerous agricultural residues, industrial by-products, municipal biowaste, wastewater sludge and energy crops. In the work [19], the annual potential of biogas and second-generation biomethane was calculated for the territory of Sicily, for numerous feedstocks: pomace, olive residue, slaughter, waste, pulp, cattle slurry, pig slurry, straw from cereal crops and many other agricultural residues and industrial by-products, within the territory of Sicily. In accordance with the annual potential of residues from palm oil industry, Loong Lam et al. [20] developed an environmental strategy for a sustainable supply chain.

As can be seen from the literature review, application of the GIS tool has been recognised as very beneficial for biomass potential mapping, as it can give valuable insights into the spatial distribution of the biomass potential and provide input data for identification of the optimal location for new biogas plant sites, sustainability assessment, techno-economic studies of biomass supply chains and supply risks management. Furthermore, interest in the utilization of novel feedstock for biogas production, such as industrial by-products for biogas production is increasing, due to several environmental and economic benefits.

Up until now, biogas potential from industrial residues and by-products was assessed on an annual basis. However, industrial production of some commodities, such as wine, sugar, mashed tomatoes and olive oil is not continuous during the year. This represents an additional

challenge in the utilization of those feedstocks, as it brings several constraints in energy planning. The contribution of this work is to develop a method that would enable the integration of seasonal and spatial assessment, developed to be used with a GIS tool.
The hypothesis of this research is that the assessment of the spatial and seasonal variation of biogas potential from industrial by-products could give better insight into the economic viability and feasibility of its utilization. This approach will be presented and validated in the following sections.

METHOD

The approach presented in this work exploits the spatial distribution of biogas potential from industrial residues and by-products and integrates seasonal (monthly) variation of potential generation. A part of this method is based on previously published work [21] by authors, in which spatial and seasonal assessment was conducted for agricultural residues.

This work aims to present an integrated approach for assessment of the spatial and seasonal variation of the potential of the industrial residues and by-products, but also prove its value through the calculation of seasonality and its influence on the economic viability of biogas plant operation. Here, it is important to mention that the scope of this research is set from feedstock determination/production to the assessment of biogas production potential. However, in real-life applications, the final disposal of residues and by-products does not end with the potential production of biogas, as this problem is much more complex.

The method used in this work contains several steps, which are presented in Figure 1. Each step is described in more detail in the sections below.

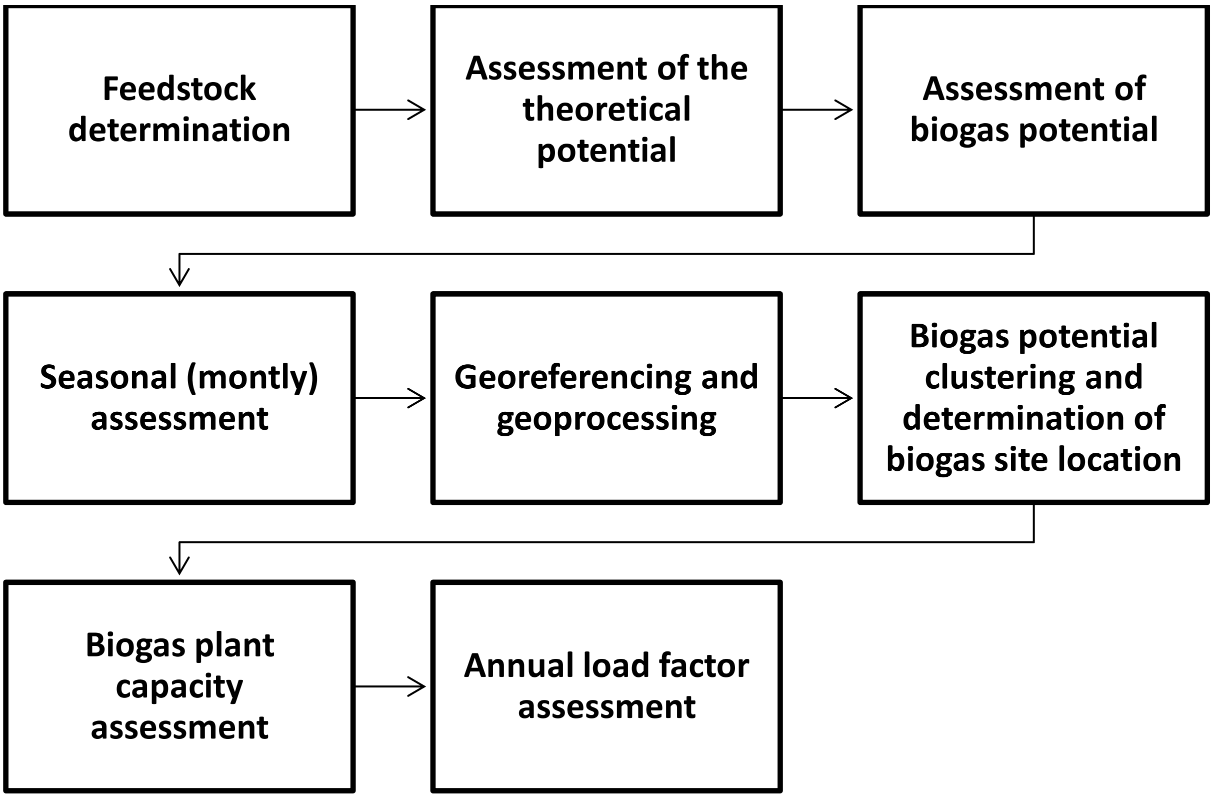

Figure 1 Steps of the method

Feedstock determination

The feedstocks considered in this work are industrial residues and by-products, which occur in sugar refineries, wineries, tomato and olive oil industry (olive oil mills).

Grape pressings occur during the grape crushing and pressing. This step is done after the collection of grapes in vineyards, in order to separate a liquid from grape marc. Grape pressings include the skins and pulp, seeds and stems.

Sugar beet pulp is the fibrous by-product obtained after water extraction of sugar contained in the root of the sugar beet [22]. Sugar beet is a vegetable cultivated for the extraction of crystallized sugar.

Oliva pomace and olive mill wastewater (OMW) are residue and by-product which occur as a result of olive milling for olive oil production. Olive pomace is the solid residue obtained during pressing or centrifugation [23].

Residues and by-products which occur during olive oil production are suitable for biogas production due to relative high biogas yield. Those residues are high-strength organic effluents whose disposal can degrade soil and water quality [24]. Thus, biogas production via AD technology is beneficial from the point of waste management, in addition to energy recovery. However, there are certain negative features for anaerobic digestion, as OMW are deficient in nitrogen, and inhibitory effects due to low pH could create problems in anaerobic digestion [25]. Therefore, there is a need for pre-treatments and the share of OMW should be low in co-digestion with other feedstocks for biogas production.

Tomato waste is a residue that occurs during the industrial processing of fresh tomatoes into mashed tomatoes, juice, sauces, food additives, etc.

Assessment of the theoretical potential of considered feedstocks

The theoretical potential of by-products is defined as the annual production of industrial by-products and residues. As can be seen from equation (1), it is a function of the amount of processed commodities and residue to the processed commodity ratio:

$$P_{ind} = M_{p.com} * RPPC \tag{1}$$

where P_{ind} stands for the theoretical potential of residues and by-products from industrial production (t), $M_{p.com}$ for the amount of processed commodities (t) and $RPPC$ for the residue to processed commodity ratio for a specific commodity (t/t). RPPC factors for the considered commodities are presented in Table 1.

Table 1 RPPC factors for the considered commodities

Industry	**Processed commodity**	**By-product**	**RPPC** (t/t)	**Reference**
Sugar	Sugar beet	Sugar beet pulp	0.25	[26]
Wine	Grape	Grape pressings	0.22	[27]

Olive oil production	Olive	Olive mill wastewater	1.25	[28]
		Olive pomace	0.55	[29]
Tomato	Tomato	Tomato residues	0.15	[29]

Assessment of biogas and methane potential

Biogas potential of the considered feedstocks is based on theoretical potential of fresh feedstocks, specific biogas yield from fresh feedstock and methane content of biogas, according to equation (2):

$$E_{ind} = P_{ind} * y_{com} * s_{CH_4} \tag{2}$$

where E_{ind} stands for a methane potential of residues and by-products from industrial production (m^3), y_{com} for a specific biogas yield of specific industrial by-product (m^3/t) and s_{CH_4} for a share of the methane contained in biogas (%). Specific biogas yield and share of the methane contained in the biogas obtained from the specific industrial by-products are given in Table 2. It is important to note here that those parameters are not obtained from the same basis of anaerobic digestion conditions, as can be seen in Table 2.

Table 2 Specific values for the calculation of biogas and methane potential from industrial by-products

By-product	y_{com} (m^3/t*)	s_{CH_4} (%)	**Anaerobic digestion conditions**			**Reference**
			pH (-)	**Temperature** (°C)	**Retention time** (days)	
Sugar beet pulp	96	50	5.18 ± 0.07	ND	ND	[30]
Grape pressings	160	80	ND	ND	21	[31]
Olive pomace	121	71	ND	ND	27	[32]
Olive mil wastewater	57.1**	**	ND	35	ND	[25]
Tomato residues	94	53	ND	ND	32	[33]

ND- not defined in the referenced literature
*values of specific biogas yield, y_{com}, are given per 1 tone of fresh matter.
** In literature, the value of biogas yield is given in specific methane yield (m^3 CH_4/t)

Seasonal assessment

Seasonality of the biogas potential of grape pressings, sugar beet pulp, olive pomace, OMW and tomato residues is determined in accordance with the month(s) of processing. In the case of tomato, olive and sugar industry, in which the production season lasts several months, the assumption is used that production is equally distributed during those months. Months of occurring and respective potential is determined for each industry and written as an additional set of attributes, where each attribute presents one month in a year and includes information of biogas potential in a specific month. Those attributes can be written and calculated in csv documents since GIS tools enable adding layers written in csv format. Furthermore, those attributes can be added and calculated directly in GIS tools by using the Field calculator.

Geocoding and geoprocessing

The economic feasibility of residues and by-products utilization is often constrained by the geographical distribution of the potential and the distance to potential biogas plants. This especially relates to smaller industries, with a small amount of by-products and residues available for biogas production. The prior step to geoprocessing is geocoding, which is the process of converting addresses into geographic coordinates. Geoprocessing enables visualisation of sources of the biogas potential, distance determination between several points, density analysis etc. GIS tools are used for geospatial information processing (geoprocessing) and can be applied to a wide range of various problems. One of the main advantages provided by GIS tools is the possibility to link non-spatial attributes with spatial information. When using a GIS tool for seasonal and spatial assessment of the biogas potential, the following information is required:

- Coordinates of the industries in which considered residues and by-products occur (in a case when respective industries are not pre-defined in the map);
- Biogas potential in a specific month;
- Months of processing commodities.

Biogas potential clustering and determination of biogas site location

GIS tools enable assessment of the areas with high concertation of biogas potential. The prior step (Geocoding and geoprocessing) will result in a point vector layer, where each point represents one industry site and includes attributes listed in the subsection Geocoding and geoprocessing. This spatial and non-spatial information are used in defining the biogas plant location. Those biogas plant locations can be understood as the centralised processing sites, which use residues and by-products from nearby industries to produce biogas. When defining the biogas plant location, the objective is to maximise the biogas potential which can be utilised and to minimise the transportation distance. For the purpose of this research, we used the assumption that the maximum air distance from the industrial site to the biogas plant is 20 kilometres. Therefore, the first step was to define the area within a radius of 20 kilometres from the industrial site. This was done in the GIS tool, with the Vector Spatial Analysis tool, by performing a "Buffer" spatial query, which resulted in buffered polygons, with a radius of 20 kilometres. The next step is to define biogas plant location, which can utilise maximal potential but is also close enough to each industrial site. Suitable areas for locating biogas sites are defined as intersections of buffered polygons, by performing the "Intersection" spatial query. As those intersections are representing area (and not a point) suitable for locating biogas sites, the final step was to define optimal locations, which are close to the industries with the greatest biogas potential, in order to minimise transportation cost and related greenhouse gas emissions. This was performed with the "Mean coordinate" spatial query. Once the potential site locations are determined, industrial sites within a radius of 20 kilometres from potential biogas site are considered viable to provide their by-products and residues for biogas production. As described in the "Geocoding and geoprocessing" sub-section, for each industrial site is defined the biogas potential and months of occurrence. Therefore, this approach integrates spatial and seasonal distribution, as the final map presents geo-location of biogas potential in each month of the year.

Biogas plant capacity assessment

In this work, the capacity of biogas plants is defined in accordance with feedstock supply. More precise, biogas potential is determined as it is a function of biogas potential of the specific cluster and time duration in which specific feedstock should be utilised, as described in equation (3):

$$P_{biogas} = \frac{E_{ind} * H_{d,CH4}}{N_{hours}} \quad (3)$$

Where P_{biogas} stands for a capacity of biogas plant (kW), E_{ind} for methane potential of residues and by-products from industrial production (m^3), $H_{d,CH4}$ for the specific lower heating value of methane (kWh/m^3) and N_{hours} for a number of hours (time duration) in which specific feedstock should be utilised (h).

Time duration in which specific feedstock should be utilised, N_{hours}, is limited by two constraints:

- Available storage capacity;
- The time period in which feedstock can be stored (storability of feedstock).

It is important to mention that industrial residues and by-products are more challenging to store, in comparison with some other conventional feedstocks for biogas production (such as maize silage). Improper storage of grape pressings, sugar beet pulp, tomato and olive mill residues may lead to degradation of feedstock: deterioration, mould formation and pests occurrence. Furthermore, it has been noted, that storage of olive pomace for 7 months causes an increase in triterpenic acids and other bioactive compounds [34]. Since biogas production is very sensitive to pH change, the assumption was used that considered residues and by-products can be stored for up to six months.

In this work, capacity was assessed for two scenarios, which present two extremes. In the first scenario, the assumption is used that there is no feedstock storage capacity and therefore feedstock for biogas production has to be utilised in the month of its occurrence. Thus, the potential of the biogas plant is determined by the maximal biogas potential in a specific month. In the second scenario, the assumption is used that feedstock for biogas production can be stored for up to six months. Those two scenarios are selected to have two extreme cases-the first in which there is no possibility to store feedstock and the second in which feedstock is stored for as long as possible, prior to a change of the bioactive compounds. The latter scenario will give a maximum annual load factor, which can be obtained by the utilisation of the considered feedstocks. For the purpose of this research, those two extremes will present the annual load factor ranges, as a function of the storage time. However, in a real-life application, the selected storage time may be in between, as the investment cost of the 6-month storage can offset the financial benefits of the produced biogas.

Annual load factor assessment

Annual load factor is a measure of the utilisation rate. In this work, annual load factor is used as a measure of utilisation of biogas plants which use industrial residues and by-products as feedstock for biogas production. This factor determines to a great extent the payback period of the specific plant. Thus, it is used in this work for the calculation of the influence of the seasonality of the industrial residues and by-products on the economic viability of biogas plant operation.

Annual load factor is a ratio of average load factor and peak load. In the case of a biogas plant, it can be considered as the ratio of biogas produced in one year and maximal amount of biogas that could be produced in one year, if a biogas plant operated at full capacity all 8,760 hours of the year, as described in equation (4):

$$f_{an.load} = \frac{E_{ind} * H_{d,CH4})}{P_{biogas} * 8760\ h} \tag{4}$$

Where $f_{an.load}$ stands for annual load factor (-), E_{ind} for methane potential of residues and by-products from industrial production (m^3), $H_{d,CH4}$ for the specific lower heating value of methane (kWh/m^3), for a number of hours (time duration) in which specific feedstock should be utilised (h) and P_{biogas} stands for a capacity of biogas plant (kW).

Annual load factor can be also represented by annual full load hours.

CASE STUDY

The presented method was demonstrated in the case study of Istria county and Osijek- Baranja county, which are presented in Figure 2. Istria County is the westernmost county of the Republic of Croatia and the largest peninsula of the Adriatic. Osijek-Baranja county is a county situated in the north-eastern part of the country. Those counties are selected to give diversity in industrial production. Both counties have intensive use of land for agricultural production.

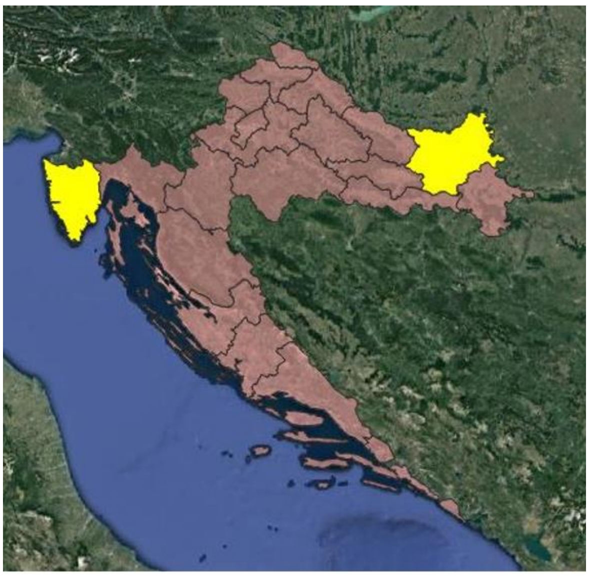

Figure 2 Case study area- Istria county (left) and Osijek- Baranja county (right)

Istria County

Istria County has a Mediterranean climate, suitable for olive oil and wine production. For the case study, nine wineries, six olive oil mills and one vegetable factory, which process tomatoes, were selected. Those industries are listed in Table 3, with respective annual processing amounts and respective commodity production.

Table 3 Annual grape processing in selected wineries, olive oil mills and vegetable industry[35], [36]

Industry	Processed commodity	Annual processed commodity (t)	Final product	Annual final product production (l)
Oil mill 1	Olive	7,140	Olive oil	1,000,000
Oil mill 2		43		6,000
Oil mill 3		464		65,000
Oil mill 4		21		3,000
Oil mill 5		200		-*
Oil mill 7		200		28,000
Vegetable factory 1	Tomato	12,000	Mashed tomato, juice, sauces	-*
Winery 1	Grape	108	Wine	70,000
Winery 2		92		60,000
Winery 3		138		90,000
Winery 4		54		35,000
Winery 5		7		4,500
Winery 6		154		100,000
Winery 7		770		500,000
Winery 8		58		37,500
Winery 9		154		100,000

*no data available

Since in the publicly open reports there are no data on the annual grape processing in each of the selected wineries, the assumption was used that one litre of wine requires 1.54 kilograms of grapes. Similar to this for olive oil production, the assumption was used that one litre of olive oil requires 7.14 kilograms of olives [37].

Osijek-Baranja County

Due to favourable climate and soil conditions, wine production is among the most represented economic activities in Osijek-Baranja county. For the case study, nineteen wineries have been selected. Those wineries are listed in Table 4, with respective annual wine production and annual grape processing amount. Grape harvesting and processing are done in September.

Table 4 Annual grape processing in selected wineries [36]

Industry	Annual wine Production (l)	Annual grape processing (kg)
Winery 10	30,000	46,150
Winery 11	300,0000	4,615,400
Winery 12	150,000	230,770
Winery 13	6,000	9,230
Winery 14	133,000	204,600
Winery 15	60,000	92,300
Winery 16	14,000	21,550
Winery 17	15,000	23,075
Winery 18	10,000	15,383
Winery 19	3,500,000	5,384,167
Winery 20	30,000	46,150
Winery 21	37,500	57,688
Winery 22	20,000	30,767
Winery 23	900,000	1,384,500
Winery 24	35,000	53,842
Winery 25	150,000	230,750
Winery 26	60,000	92,300
Winery 27	50,000	76,917
Winery 28	350,000	538,417

One of two Croatian sugar refineries is situated in the capital city of Osijek- Baranja county. This sugar refinery produces 70,000 tons of sugar annually by processing 550,000 tons of sugar beet [38]. The average sugar production campaign length in Croatia is three months [39].

RESULTS

The biogas potential from grape pressings, sugar beet pulp, olive pomace, OMW and tomato residues was calculated for the considered wineries, sugar refinery, olive oil mills and vegetable industry as described in the Method section.

Istria county

The results of the calculations of biogas potential from industrial by-products and residues in Istria County are presented in Table 5.

Table 5 Biogas potential from industrial residues and by-products in Istria County

Industry	Industrial by-product	Biogas potential (m^3 CH_4)			
		August	September	October	November
Oil mill 1	Olive pomace	-	-	168,684	168,684
	OMW	-	-	286,180	286,180
Oil mill 2	Olive pomace	-	-	786	786
	OMW	-	-	1,717	1,717
Oil mill 3	Olive pomace	-	-	8,518	8,518
	OMW	-	-	18,602	18,602
Oil mill 4	Olive pomace	-	-	393	393
	OMW	-	-	859	859
Oil mill 5	Olive pomace	-	-	3,671	3,671
	OMW	-	-	8,016	8,016
Oil mill 6	Olive pomace	-	-	3,669	3,669
	OMW	-	-	8,013	8,013
Vegetable factory 1	Tomato residues	59,784	29,892	-	-
Winery 1	Grape pressings	-	3,032		-
Winery 2	Grape pressings	-	2,599	-	-
Winery 3	Grape pressings	-	3,899	-	-
Winery 4	Grape pressings	-	1,516	-	-
Winery 5	Grape pressings	-	195	-	-
Winery 6	Grape pressings	-	4,332	-	-
Winery 7	Grape pressings	-	21,660	-	-
Winery 8	Grape pressings	-	1,624	-	-
Winery 9	Grape pressings	-	4,332	-	-

As can be seen from Table 5, the vegetable factory, as well as several olive oil mills and wineries have a significant potential for biogas production. The considered by-products from the vegetable industry (tomato processing) occur in August and September, from wineries occur in September, while from the olive oil industry residues and by-products occur in October and November. The spatial and seasonal distribution of biogas potential is presented in Figure 3.

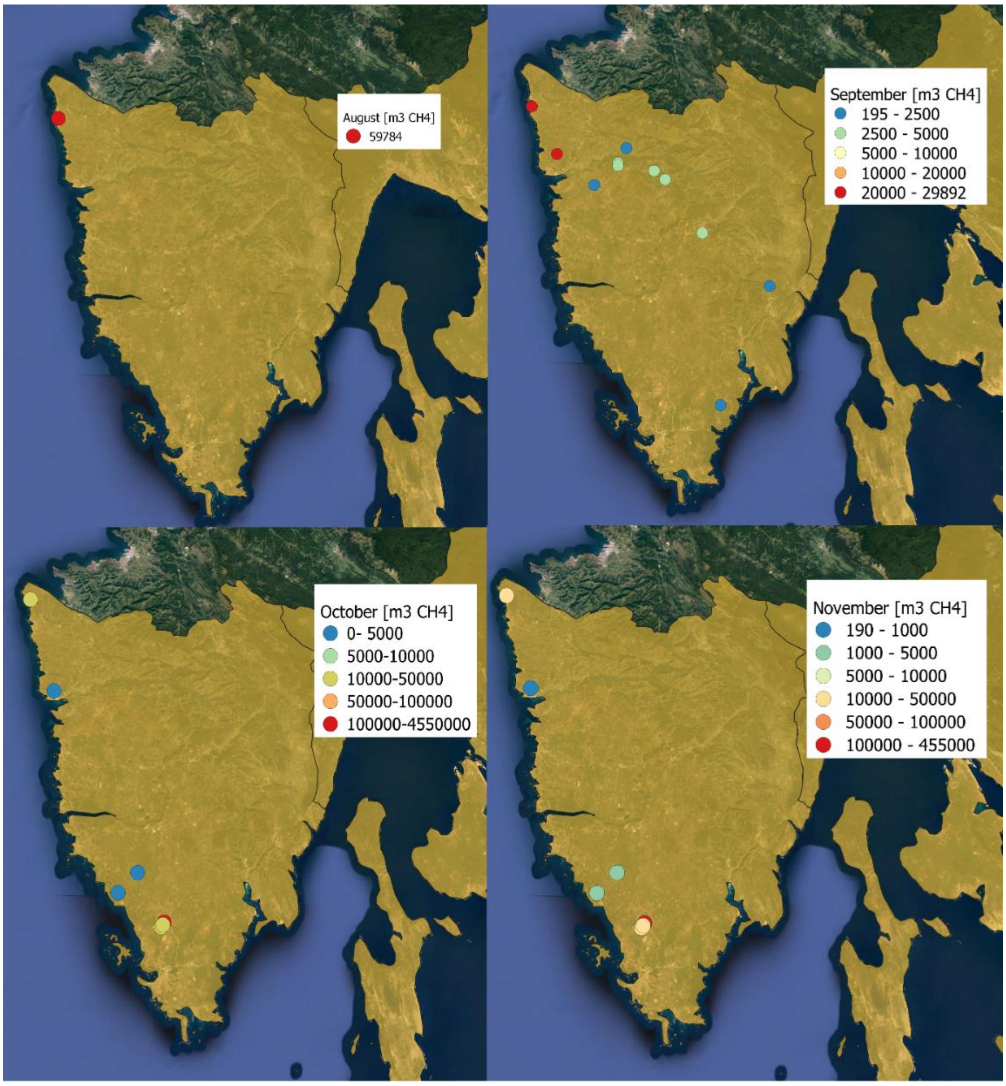

Figure 3 Biogas potential from wineries, olive mills and tomato industry in August, September, October and November

Based on the annual biogas potential and locations of industries with the greatest biogas potential, locations of potential biogas sites were determined. The determined biogas site locations for Istria County are represented in Figure 4, together with the annual biogas potential of considered industries. As described in the Method section, the GIS tool was used to define optimal biogas sites. When defining the biogas sites, suitable locations for biogas plant installations were those which are in the radius of 20 kilometres from the industrial site, which can utilise the maximum potential and where the transport distance was minimised. Figure 4 also clearly depict which industries are considered as viable to provide their residues and by-products as feedstocks for biogas production.

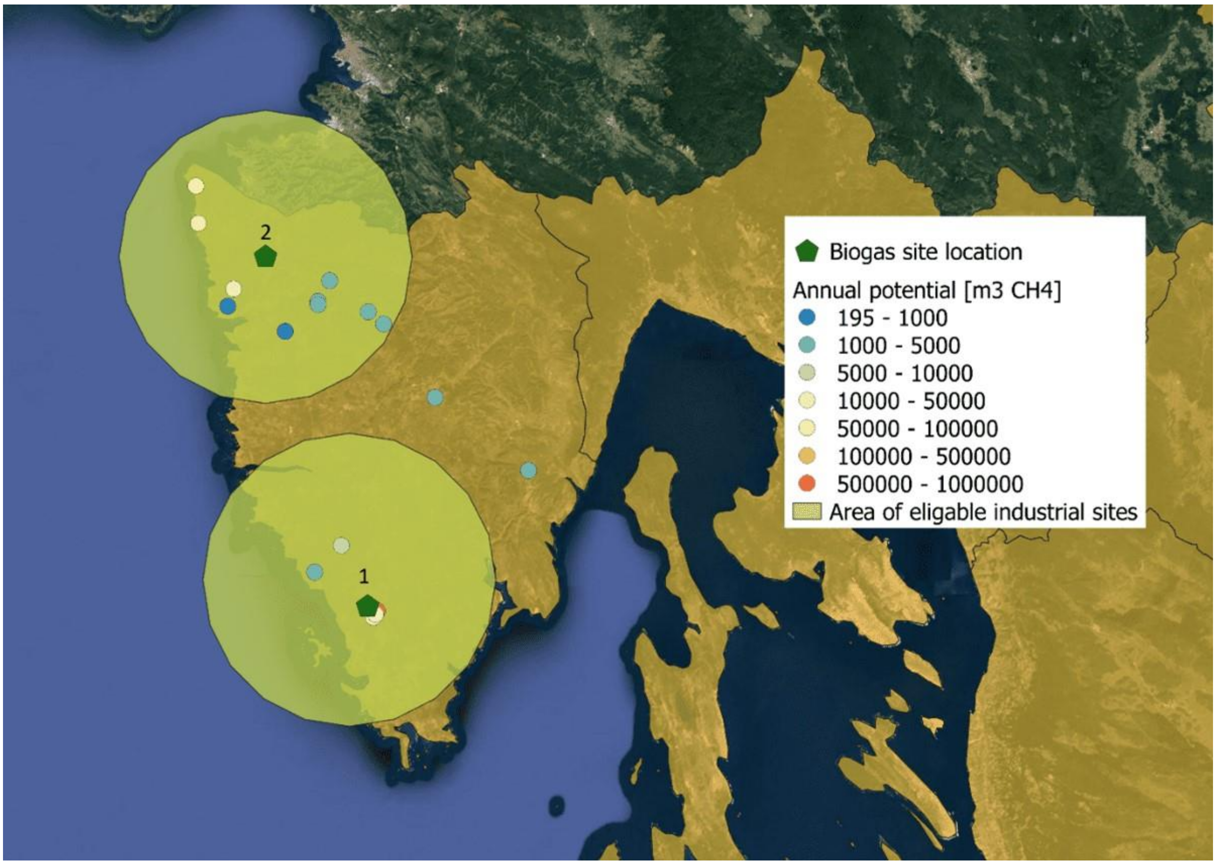

Figure 4 Annual biogas potential and potential biogas sites

Osijek Baranja County

The results of the biogas potential assessment from industrial residues and by-products in Osijek-Baranja county are presented in Table 6.

Table 6 Biogas potential from industrial residues and by-products in Osijek-Baranja county

Industry	Industrial by-product	Biogas potential (m^3 CH_4)		
		September	October	November
Winery 10	Grape pressings	1,300	-	-
Winery 11	Grape pressings	129,969	-	-
Winery 12	Grape pressings	6,498	-	-
Winery 13	Grape pressings	260	-	-
Winery 14	Grape pressings	5,762	-	-
Winery 15	Grape pressings	2,599	-	-
Winery 16	Grape pressings	607	-	-
Winery 17	Grape pressings	650		
Winery 18	Grape pressings	433		
Winery 19	Grape pressings	151,618		
Winery 20	Grape pressings	1,300		
Winery 21	Grape pressings	1,624		
Winery 22	Grape pressings	866		

Winery 23	Grape pressings	38,988		
Winery 24	Grape pressings	1,516		
Winery 25	Grape pressings	6,498		
Winery 26	Grape pressings	2,599		
Winery 27	Grape pressings	2,166		
Winery 28	Grape pressings	15,162		
Sugar refinery	Sugar beet pulp	2,200,000	2,200,000	2,200,000

As can be seen from Table 6, the sugar refinery and some of the wineries have a significant potential for biogas production. As expected, winery 11 has by far the highest potential for biogas production from grape pressings. The considered by-products from the wine industry occur in September and from the sugar industry in September, October and November.

The spatial and seasonal distribution of biogas potential is presented in Figure 5. The left part of Figure 5 presents the spatial distribution of biogas potential from by-products that occur in September (grape pressings and sugar beet pulp). Since the biogas potential from sugar beet pulp is equal in November and October, this potential is presented in one figure (right part of Figure 5).

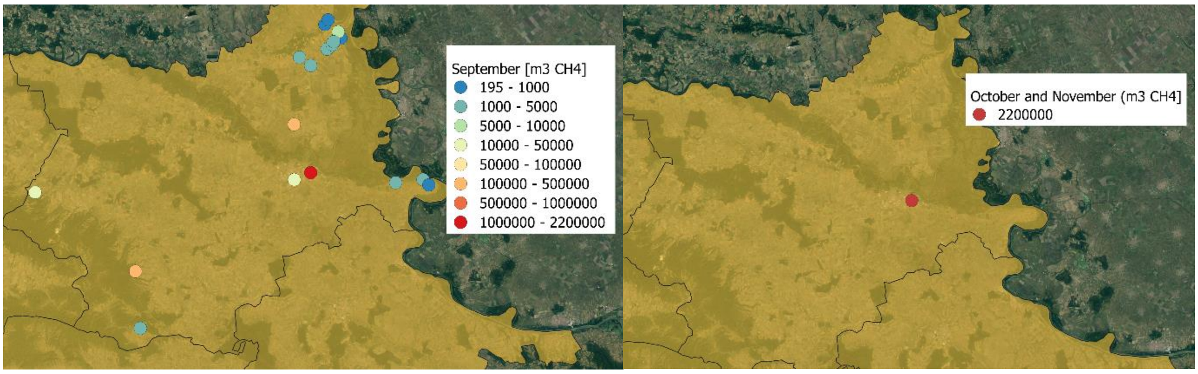

Figure 5 Biogas from sugar refinery and wineries in September (left) and from sugar refinery in October and November (right)

Based on the annual biogas potential and locations of industries with the greatest biogas potential, locations of potential biogas sites were determined. The determined biogas site locations for Osijek-Baranja county are represented in Figure 6, together with the annual biogas potential of considered industries. Figure 6 also clearly depict which industries are considered as viable to provide their residues and by-products as feedstocks for biogas production.

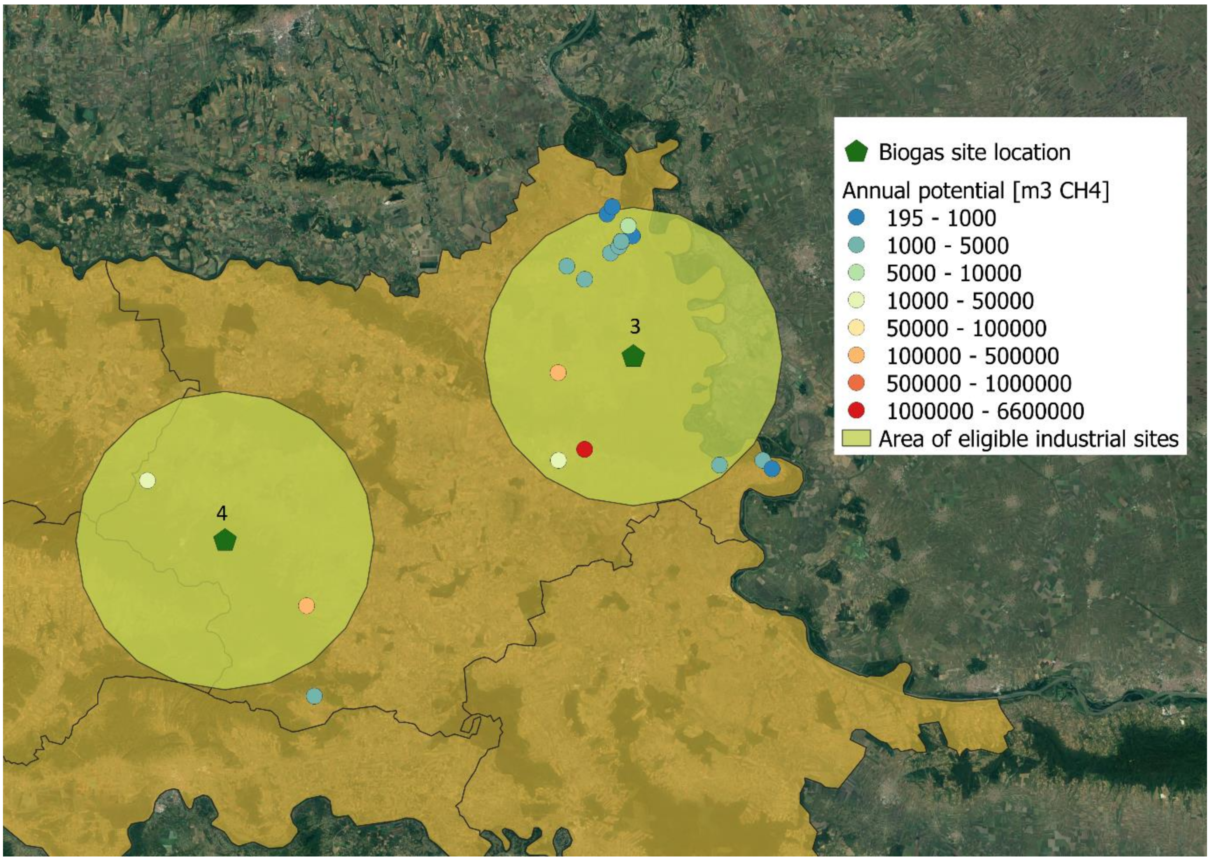

Figure 6 Annual biogas potential and potential biogas sites

Buffered layers presented in Figure 4 and Figure 6 are used to assess the seasonal variation of each biogas site. The final results are aggregated and presented in Table 7. Here, seasonal variation is represented for biogas sites which are presented in Figure 4 and Figure 6.

Table 7 Seasonal variation of biogas potential in four potential biogas sites

Biogas site		**Industry**	**Annual biogas potential** (m^3 CH_4)	**August** (m^3 CH_4)	**September** (m^3 CH_4)	**October** (m^3 CH_4)	**November** (m^3 CH_4)
1		Olive oil mills	964,172	-	-	482,060	482,060
2		Wineries, olive oil mils, vegetable industry	187,946	59,784	72,988	27,587	27,587
3		Wineries, sugar refinery	6,776,560	-	2,376,560	2,200,000	2,200,000
4		Wineries	190,605	-	190,605	-	-

As can be seen in Table 7, the seasonal variation of biogas potential differs for each considered biogas site.

Biogas plant capacity and annual load factor

As described in the Method section, biogas plant capacity and annual load factors were calculated for two scenarios. For the first scenario assumption was used that there is no feedstock storage capacity and therefore feedstock for biogas production has to be utilised in the month of its occurrence. For the second scenario, the assumption was used that there is feedstock storage that enables storage of feedstocks for up to six months. The results are given in Table 8.

Table 8 Biogas plant capacity and load factor for two scenarios (without feedstock storage and with 6-month feedstock storage)

Biogas plant (cluster)	Industry	P_{biogas} (kW)		$f_{an.load}$ (-)	
		Without storage	6-month storage	Without storage	6-month storage
1	Olive oil mills, wineries	6,658	1,665	0.16	0.66
2	Wineries, olive oil mils, vegetable industry	1,008	185	0.18	0.82
3	Wineries, sugar refinery	32,824	10,400	0.24	0.74
4	Wineries	2,633	376	0.1	0.58

DISCUSSION

This research presented a method based on an integrated GIS approach for the assessment of the seasonal and spatial distribution of biogas potential from industrial residues and by-products. As can be seen from the results conducted for the case studies, industrial residues and by-products have significant potential to be utilised for biogas production. Total biogas potential, which could be used in 4 potential biogas sites, equals 8,119,280 m^3 CH_4. The potential biogas sites are determined in accordance with the methodology elaborated in the section above. It is important to note that localisation of biogas plant is very complex, not only due to technical reasons, which are addressed in this research (maximisation of the potential, minimisation of the transport distance), but also due to social reasons, such as "not in my back yard" (NIMBY) phenomenon. This phenomenon refers to individuals who recognize the greater benefits of a facility but show a protectionist attitude when the object is proposed in their “neighbourhood”.

Potential of industrial residues and by-products was studied in numerous papers, such as the work [40], in which Francesca et al. calculated the annual potential of olive pomace available for biogas production on a municipal level. Based on annual feedstock potential, some of the authors calculated electrical and thermal energy which could be produced from biogas generated from industrial residues and by-products. Ulusoy et al. [33] calculated that on an annual basis, 14,175,000 kWh of electrical energy and 150,660,000 kWh of thermal energy can be produced from biogas generated from 90,000 tonnes of tomato waste. Similar to this, Ulosoy et al. [24] calculated that on an annual basis, 74,959,780 kWh of electrical energy and 71,961,390 kWh of thermal energy could be produced from biogas generated from 1,260,000 tons of OMW. Results of this research prove that feedstocks which are nowadays regarded as waste can significantly contribute to an increase of renewable energy production.

However, as can be seen in Figure 3 and Figure 5 those feedstocks occur only in a few months of the year. Based on the results presented in Figure 3 and Figure 5, the question which arises is whether it would be economically viable to use this feedstock for biogas production, as recommended in many studies.

To give better insight into the influence of seasonality on the economic viability of industrial residues and by-products utilisation in biogas sites, annual load factors are assessed for each potential biogas cluster. As it can be seen in Table 8, load factors for case study biogas sites, for the case where there is no feedstock storage capacity, are ranging from 0.1-0.24. These values indicate that case study biogas sites will be operating at full load from 720-2,100 hours. It is worth mentioning that the biogas plants which are nowadays in operation have a high annual load factor. In the work [41], Stürmer et al. conducted research on 291 biogas plants with different capacities and from different European countries and concluded that those biogas plants operate on an annual basis from 6,096 to 8,421 full load hours. In addition, Hublin et al. [42] calculated on a Croatian case study, that a biogas plant with an annual load factor of 0.82, that use cow manure and whey as biogas feedstock has a payback period of 9.9 years.

One of the possibilities to increase the number of full load hours is to include feedstock storage. As can be seen from Table 8, 6-month storage would in the case of pilot biogas sites lead to load factors from 0.58-0.82 (5,080-7,180 hours). However, it must be noted here that investment in 6-month storage leads to additional investments cost, which could strongly affect a payback period. Furthermore, storage of considered feedstock requires special attention, as improper storage may lead to deterioration, mould formation and pests occurrence.

Another possibility to increase load factor is to use feedstock from diverse industries and from those in which feedstock for biogas production is generated in a longer period, as it can be seen on example for biogas site 2 and 3 (Table 8).

To investigate the influence of the seasonality on the payback period of biogas plants that use industrial by-products and residues for biogas production, the payback period is calculated for five different load factors:0.1, 0.24, 0,58, 0.82 and 0.9. For this purpose, we selected as a referent a biogas plant with 1 MW_{el}, which sells net electricity to the electric grid and net thermal energy to the district heating grid.

For payback period calculation, which refers to a number of years it takes to recover the cost of an investment, we used the following equation (5) [42] :

$$I = \sum_{t=1}^{PB} C_t \tag{5}$$

Where I is a capital investment (€), C_t is a net annual cash flow in year t (€) and PB is a payback period of the investment.

For the calculation of the initial investment, we used the following assumptions:

- Biogas CHP engine cost- 1,000,000 €;
- Feedstock preparation equipment costs-600,000 €;
- Civil works-500,000 €;
- Plant’s regulation system cost-300,000 €;

- Cost of connecting the plant to the local district heating network-750,000 €;

Net annual cash flow, C_t is the difference between annual income and annual expense. When calculating the annual income, we assumed that the income is generated by selling electricity to an electric grid and by selling heat to a district heating grid. The annual expenses consist of operation and maintenance costs, as well as corporate tax.

More precisely, the following assumptions were used:

- Electrical efficiency, η_{el} is 40 % and heat efficiency, η_{el} is 43%;
- The electrical energy is sold to the grid at the referent price of 140 €/MWh$_{el}$ [43] and the thermal energy is sold to the district heating system at the price of 20 €/MW$_{h}$;
- The cost of feedstock is neglected, as those feedstocks are nowadays regarded as waste;
- Digestate will not be sold and the cost of thermal treatments, required before digestate utilisation in agriculture is not included;
- Operations and maintenance (O&M) cost 6% of the total investment. In the case of load factor <0.5, O&M cost is 4% of the total investment;
- 10% of produced electrical energy and 15% of produced thermal energy is used for biogas plant operation;
- Depreciation of the equipment, connection to the district heating plant and civil work is 15 years;
- Corporate tax is 20%.

The payback periods, for each considered load factor, are presented in Table 9.

Table 9 Payback period dependence on the load factor

Load factor, $f_{an.load}$ (-)	0.9	0.82	0.58	0.24	0.1
Payback period (year)	3.9	4.4	6.5	15	44.1

The results presented above confirm that seasonality of biogas potential from industrial by-products and residues has a significant influence on the economic viability of biogas utilization and therefore it is beneficial to include this aspect in the potential assessment.

CONCLUSION

The approach presented in this work exploits the spatial distribution of biogas potential from industrial residues and by-products and integrates seasonal (monthly) variation of potential generation. The developed method demonstrates how seasonal assessment can be integrated into GIS assessment of biogas potential from industrial residues and by-products.

The presented approach was applied to the wine, sugar, vegetable and olive oil industry and tested at two case study area-Istria and Osijek-Baranja County. The presented method contains eight steps and was used for defining biogas potential available to be utilised in potential biogas sites, whose locations are set in areas with a higher concentration of biogas potential.

The results show that considered industries generate a substantial amount of residues and by-products (grape pressings, sugar beet pulp, tomato waste, olive pomace and OMW) suitable for biogas production. For the industries located nearby potential biogas sites (in a radius of 20 km), total biogas potential equals 8,119,280 m^3CH_4. Feedstock considered in potential assessment occurs from one (grape pressings) to three months (sugar beet pulp).

The influence of seasonality on the economic viability of industrial residues and by-products utilisation for biogas production was assessed in the case biogas sites, through assessment of annual load factors for two scenarios- the scenario without feedstock storage and the scenario with 6-month case storage. For the first scenario, annual load factors for case study biogas sites are ranging from 0.1-0.24. As biogas sites are operating at multiple higher load factor, it can be concluded that seasonality highly affects the economic viability of biogas site operation. For the scenario where 6-month feedstock storage is available, the annual load factors increase to 0.58-0.82. However, it should be noted that those storages may highly affect total investment and require special attention, due to possible issues which occur in case of improper feedstock storage.

The results presented in this work confirm the hypothesis that the integrated assessment of the spatial and seasonal variation of biogas potential from industrial by-products and residues give better insight into the economic viability and feasibility of its utilization.

ACKNOWLEDGEMENT

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PAPER 3

energies

Article

Analysis of Specific Greenhouse Gas Emissions Savings from Biogas Production Based on Agricultural Residues and Industrial By-Products

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Abstract: The aim of this study was to analyse specific greenhouse gas emissions savings for a variety of agricultural residues, industrial by-products, and municipal biowaste. One of the most viable alternatives to fossil fuels is bioenergy, particularly biogas produced by the anaerobic digestion of renewable feedstocks. The revised Renewable Energy Directive (D 2018/2001) recognizes that biogas production from agricultural residues, livestock production, and industrial by-products is an acknowledged greenhouse gas mitigation technology in cases where their use results in a certain level of specific greenhouse gas savings. This study delivered values for the maximum transport distance of agricultural residues and industrial by-products to achieve the greenhouse gas (GHG) emissions-saving requirement defined by Directive 2018/2001. It analysed the greenhouse gas emissions reduction for numerous feedstocks for which Directive 2018/2001 has not defined the default and typical values but which could be used as sustainable substitutes for currently dominantly used maize silage in biogas production. The results obtained in this work define the maximum transport and distribution distance for which biogas produced from considered feedstocks achieved required specific greenhouse gas emissions savings (80%), compared with fossil fuel comparator. The obtained results can be used as the constraints in the optimisation of the biomass supply chains for the feedstocks considered in this work.

Keywords: greenhouse gas emission saving; biogas; agricultural residues; industrial by-products

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1. Introduction

Biogas production has been on the rise in Europe in the last two decades as countries seek to reduce their dependence on fossil fuels and meet climate change targets. Biogas plants can range greatly in terms of installed capacity, from micro plants in developing countries, small-scale plants used on farms, and large-scale plants used in centralised systems in cities [1]. Although mostly used to generate electricity and heat, biogas can also be utilised to create biomethane, a fuel that can replace natural gas in transportation and industrial applications. Biofuels are the only commercially viable alternative already utilized for transport and industrial needs [2].

Up to 72% of the feedstocks used for biogas production come from the agricultural sector [3], primarily maize silage. The competitive use of biogas feedstocks with food and feed production has raised not only environmental but also socioeconomic concerns, reflected in new sustainability requirements defined by European Union (EU) legislation [4]. The revised Renewable Energy Directive, which came into effect in December 2018, has established sustainability and the greenhouse gas (GHG) emission-savings criteria with which biogas used in transport, electricity, heating, and cooling production, must comply. The new Directive enhances the sustainability requirements for biogas feedstocks and adds new requirements for specific greenhouse gas emission savings from biogas production, which biogas facilities must adhere to in order to contribute to renewable energy goals and qualify for government funding. As one of these criteria, the Directive states that the GHG savings from the use of biomass for heating, cooling, and electricity production must be at least 70% for plants that started to work in 2021 and 80% for plants starting operation in 2026 [5]. The Directive also defines typical and default values for GHG savings for the three mostly used biogas feedstocks: manure, biowaste, and maize whole plant (maize silage).

A significant amount of sustainable feedstocks and a thorough understanding of the sustainable potential of biomass supply are necessary to achieve GHG savings [6]. The utilisation of materials previously regarded as waste, such as agricultural and industrial residues and by-products, is receiving increased attention, as it not only improves the sustainability of biogas production but also improves waste management and resource efficiency.

Research Problem and Literature Review

The environmental sustainability of biomass utilisation for energy purposes has raised significant concerns. It has been reported that biomass utilisation may result in unsolved challenges and trade-offs concerning the accounting of GHG and non-GHG emissions [7]. The environmental sustainability of biomass utilisation is a complex problem which depends on various factors such as the feedstock type, feedstock preprocessing and processing technology, transportation and distribution distance, emissions from the fuel in use, etc. Because of its high complexity, the importance of this problem has increased in the last decades. Hence, a significant number of research papers have investigated various types of environmental sustainability performance of biomass utilisation for energy production. Some of them are presented in the following paragraphs.

Hamelin et al. [8] performed the life cycle assessment of biogas production based on manure and the following co-substrates: straw, garden waste, food waste, energy crops, and animal urine and faeces. The results, given in $kgCO_2eq$ per functional unit, prioritised source-segregated solid manure as co-substrates, followed by straw and biowastes, while energy crops were identified as co-substrates whose utilisation would result in adverse environmental impacts. In their recent work, Meng et al. [9] examined the viability of total or partial replacement of peat by maize straw biogas residues and manure biogas residues. The results showed that a biogas plant that produced 10,000 m^3 biogas daily could achieve savings of 439.4 tonnes/year of CO_2 through the proposed replacement. Den Boer et al. [10] calculated that using kitchen waste for biogas production could lead to 680,000 tCO_2eq savings per year.

The transport distance of the biomass supply and biomass availability throughout the year have a significant impact on the energy conversion efficiency and GHG reductions in anaerobic digestion (AD) technology [11]. Anaerobic digestion (AD) is a collection of processes by which microorganisms break down biodegradable material in the absence of oxygen. The results of AD are biogas and digestate. Berglund et al. [12] performed the energy life cycle analysis of eight feedstocks for biogas production. The results showed that the difference between energy output and input was positive in the cases of transport distances less than 700 km for slaughterhouse waste, 580 km for municipal organic waste, 240 km for straw, 220 for pig manure, and 200 km for cow manure. In their study, Uusitalo et al. [13] concluded that using biogas to produce heat and electricity leads to greater GHG reductions than composting feedstock, yet not as high as in the case of its utilisation for transport. In the work of Balcioglu et al. [14], the authors calculated that if 60% of cattle manure and all available chicken manure in Turkey were co-digested with other waste feedstock, this could lead to annual GHG emissions reduction of up to 2.5%. Wąs et al. [15] assessed the GHG mitigation potential of biogas production that uses agricultural waste and manure as biogas feedstock in Ukraine. Results indicated that the theoretical potential of GHG savings ranged between 5% to 6.14%, while technical potential varied between 2.3% to 2.8% of total GHG emissions. Tamburni et al. [16] calculated that biogas production from agricultural waste could result in GHG emission savings of up to 3,000,000 $MgCO_2eq$ in the Emilia Romagna region.

As can be seen from the literature review, environmental sustainability and GHG savings have been studied extensively, obtaining results in different forms using a variety of methods. However, there are still numerous feedstocks recognised as novel feedstocks for biogas production (mostly so-called waste materials), but the constraints to achieving required GHG savings still need to be defined, as there are limited data on GHG emissions from the biogas production chain that can serve as typical and defaults limits. To address this gap, the research object of this study was to define the maximal transport distance of various novel biomass feedstock that complies with the GHG savings of 80%, compared with fossil fuels, as required in Directive 2018/2001. This was calculated for agricultural residues, municipal biowaste, and industrial by-products. This work hypothesised that all considered feedstocks would achieve the requested GHG savings (80%) for transport and distribution distances up to 50 km. The method used for the calculation is presented in the section below.

2. Method

The method used in this work is based on the method developed by the Joint Research Centre and implemented in Directive 2018/2001 [5]. Directive 2018/2001 includes disaggregated typical and default GHG and GHG saving values for biogas used to produce electricity and heat for wet manure, maize whole plant (maize silage), and biowaste.

The method can be used for the determination of specific GHG emissions from different solid and gaseous pathways. In this method, the Global Warming Potential (GWP) is used as the climatic metric. Global Warming Potential is a term used to describe the relative potency, molecule for molecule, of a greenhouse gas, taking into account how long it remains active in the atmosphere [17]. As defined in the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report [18], the GWP of methane is equal to 25, and for nitrous oxides is 298 for a period of 100 years.

Biogas feedstocks analysed in this work are:

- **Agricultural residues:** straws (wheat straw, barley straw, oat straw, and triticale straw), and maize stover;
- **Industrial residues:** grape pressings, tomato pomace, brewers' spent grain, olive pomace, and sugar beet pulp;
- **Municipal biowaste:** The method is described in detail in the following subsections.

2.1. Calculation of Greenhouse Gas Emissions from the Production and Use of Biogas

The general equation used to calculate greenhouse gas emissions from the production and use of biogas is:

$$E = e_{ec} + e_l + e_p + e_{td} + e_u - e_{sca} - e_{ccs} - e_{ccr} \quad (1)$$

where

E is the total emissions from the use of the fuel (gCO_2eq/MJ);

e_{ec} is emissions from the extraction or cultivation of feedstocks (gCO_2eq/MJ);

e_l is annualised emissions from carbon stock changes caused by a land-use change (gCO_2eq/MJ);

e_p is emissions from processing (gCO_2eq/MJ);

e_{td} is emissions from transport and distribution (gCO_2eq/MJ);

e_u is emissions from the fuel in use (gCO_2eq/MJ);

e_{sca} is emission reduction from soil carbon accumulation due to improved agricultural management (gCO_2eq/MJ);

e_{ccs} is emission savings from CO_2 capture and geological storage (gCO_2eq/MJ);

e_{ccr} is emission savings from CO_2 capture and replacement (gCO_2eq/MJ).

In the subsections below, each emission factor is described in greater detail.

2.1.1. Emissions from the Extraction or Cultivation of Feedstocks (e_{ec})

Emissions from collecting, drying, and storing feedstocks, waste, leaks, and the production of chemicals or goods used in extraction or culture were all included in the definition of emissions from the extraction or cultivation of feedstocks [5]. These emissions apply when agricultural feedstocks like palm, maize, sugarcane, soybean, or rapeseed are extracted or grown. The emissions from the cultivation or extraction of feedstocks were regarded as zero when residues, by-products, and waste materials were utilised as feedstocks for biogas generation [19].

2.1.2. Annualised Emissions from Carbon Stock Changes Due to Land-Use Change (e_l)

By averaging emissions from carbon stock changes over a 20-year period, we determined the annualised emissions from carbon stock changes caused by a change in land use [5]. Any alteration in the carbon stock between the classified land categories of grassland, forestland, cropland, wetland, settlements, and other lands was considered a land-use change [20]. The calculation was based on Equation (2) [5]:

$$e_l = \frac{(CS_R - CS_A)}{P * 20} * 3.664 - e_B \tag{2}$$

where

CS_R is the carbon stock per unit area corresponding to the reference land use. The reference land use is the land use as of January 2008 or 20 years prior to receiving feedstock, whichever was more recent. It is quantified as a mass of carbon per hectare (gC/ha), which includes both soil and vegetation;

CS_A is the quantity of carbon stored per area corresponding to the actual land use. It is quantified as a mass of carbon per hectare (gC/ha), which includes both soil and vegetation;

P is the crop's productivity, expressed as the amount of biofuel produced per hectare per year (MJ/ha·year);

e_B is a bonus of 29 gCO_2eq/MJ, applied when biomass is obtained from restored degraded land (gCO_2eq/MJ).

2.1.3. Emissions from Processing (e_p)

Emissions from processing refer to emissions that result from the actual processing itself—waste generation, product leakage, and the production of chemicals and other processing-related products. In addition, regardless of whether fossil fuel inputs were burned during processing, these emissions also included CO_2 emissions proportional to their carbon content. The benchmark for measuring the emissions of greenhouse gases caused by electricity not produced on a biogas site was the average emission intensity of electricity production and distribution in a given area. Regardless of whether or not they were processed into intermediate products before being converted into the end product, agricultural residues and industrial by-products were regarded as having zero life cycle greenhouse gas emissions until the collection process [5].

The agricultural residues having a bulk density of less than 0.2 tonne/m^3 required a processing step before prior transportation: baling, additional grinding, or clustering. This is represented by a single process [19] in Table 1.

Table 1. Emissions from bailing/processing agricultural residues.

Baling/Processing				
Input	**Output**	**Unit**	**Amount**	**Source**
Agri-residue	-	MJ/MJ_{bale}	1.0	[19,21]
Diesel	-	MJ/MJ_{bale}	0.010	[19,21]
-	Bales	MJ	1.0	[19,22]

Table 1. *Cont.*

Baling/Processing				
Input	**Output**	**Unit**	**Amount**	**Source**
-	CH_4	g/MJ_{bale}	1.23×10^{-5}	[19,22]
-	N_2O	g/MJ_{bale}	3.03×10^{-5}	[19,22]

2.1.4. Emissions from Transport and Distribution (e_{td})

GHG emissions from the transport and distribution (e_{td}) should include all transport and distribution steps in the value chain. e_{td} was calculated from the following equation [20]:

$$e_{td} = \frac{\left(d_{loaded} * K_{loaded} + d_{empty} * K_{empty}\right) * EF_{fuel}}{E_{bio.feedstock}} \tag{3}$$

where

d_{loaded} is the transport and distribution distance of a loaded truck (km);
K_{loaded} is fuel use of loaded truck (L/km);
d_{empty} is the transport and distribution distance of an empty truck (km);
K_{empty} is the fuel consumption of an empty truck (L/km);
EF_{fuel} is fuel's emission factor (gCO_2eq/l);
$E_{bio.feedstock}$ is the biogas potential of loaded biomass feedstocks (MJ).

Typical values for transport and distribution calculation are given in Table 2. According to the JRC report [19], it was assumed that a 40 t truck would be used to deliver the feedstock to biogas sites (27 t payload).

Table 2. Typical e_{td} values calculation [19].

	K_{empty} [L/km]	K_{loaded} [L/km]	EF_{fuel} [gCO_2eq/l]
Value	0.3	0.35	3157

The Biogas potential of loaded biomass feedstocks is calculated by Equation (4):

$$E_{bio.residues} = \rho_{residues} * V_{truck} * y_{biogas} * s_{CH_4} * LHV_{CH_4} \tag{4}$$

where

$\rho_{residues}$ is bulk density of residues (t/m^3);
V_{truck} is truck load capacity (m^3);
y_{biogas} is biogas yield from 1 tonne of fresh feedstock (m^3/t);
s_{CH_4} is methane content of biogas (%);
LHV_{CH_4} is methane lower heating value (MJ/m^3).

Typical values for calculating the biogas potential of loaded biogas feedstocks for the considered feedstocks are given in Table 3.

Table 3. Typical values for the calculation of biogas potential.

Biogas Feedstock	$\rho_{residues}$ (t/m^3)	y_{biogas} (m^3/t)	s_{CH_4} (%)
Maize stover	0.060 [23]	276 [24]	55 [24]
Wheat straw	0.043 [25]	125 [24]	52.5 [24]
Barley straw	0.037 [25]	125 [24]	52.5 [24]
Oat straw	0.049 [25]	125 [24]	52.5 [24]
Triticale straw	0.043 [25]	125 [24]	52.5 [24]

Table 3. *Cont.*

Biogas Feedstock	$\rho_{residues}$ (t/m^3)	y_{biogas} (m^3/t)	s_{CH_4} (%)
Grape pressings	0.525 [26]	150 [24]	52.5 [24]
Tomato pomace	0.73 [27]	94 [28]	53 [29]
Brewers' spent grain	0.45 [30]	89 [31]	62 [31]
Olive pomace	0.9 [32]	121 [33]	71 [33]
Sugar beet pulp	0.561	96 [34]	50 [34]

It is important to highlight that for feedstocks with a bulk density greater than 0.75 t/m^3, the feedstock load was constrained by weight, while for bulk densities less than 0.75 t/m^3, it was volume constrained. The bulk density of agricultural residues could be increased 8–12 times at different bailing/briquetting process parameters [25].

2.1.5. Emissions from the Fuel in Use (e_u)

Emissions of the fuel (biogas) in use (e_u) were considered to be zero for biofuels (biogenic CO_2 combustion emission). However, e_u factor should take into account the emissions of non-CO_2 greenhouse gases (CH_4 and N_2O) of the fuel in use [5,35].

2.1.6. Emission Savings from Soil Carbon Accumulation via Improved Agricultural Management (e_{sca})

Emission savings from soil carbon accumulation via improved agricultural management refers to the practice that results in an increase in soil carbon. Those savings could be calculated only in cases of improved manure management, shifting to minimal or zero tillage, use of compost, or improved crop rotations [36]. To assess those savings, Equation (2) can be used, replacing the 20 years with the appropriate number of years for a given period.

2.1.7. Emission Savings from CO_2 Capture and Geological Storage (e_{ccs})

Emission savings from CO_2 capture and geological storage (e_{ccs}) include averted emissions through CO_2 capture and geological storage directly associated with fuel extraction, transportation, processing, and distribution [5]. They could only be considered if proven that the current storage ensures that the leakage does surpass the current state of technology [19].

2.1.8. Emission Savings from CO_2 Capture and Replacement (e_{ccr})

Savings on emissions from CO_2 capture and replacement (e_{ccr}) were only possible when CO_2 that comes from biomass was captured and utilised to replace CO_2 that comes from fossil fuels in the creation of goods and services for sale [5]. The savings could be included in the overall calculation only if proven that CO_2 replaces CO_2 that comes from fossil sources and is employed in the production of goods and services for commerce [19].

2.2. Calculation of Greenhouse Gas Emissions from Heat and Electricity

In this study, the assumption was that biogas produced from considered feedstocks would be used in cogeneration (CHP) plants to produce electrical and thermal energy. In order to allocate emissions to each final energy commodity, the following equations were used [5]:

$$EC_{el} = \frac{E}{\eta_{el}} \cdot \left(\frac{C_{el} \cdot \eta_{el}}{C_{el} \cdot \eta_{el} + C_h \cdot \eta_h} \right) \tag{5}$$

$$EC_h = \frac{E}{\eta_h} \cdot \left(\frac{C_h \cdot \eta_h}{C_{el} \cdot \eta_{el} + C_h \cdot \eta_h} \right) \tag{6}$$

where:

EC_{el} is total GHG emissions associated with electrical energy (gCO_2eq/MJ);

EC_h is total GHG emissions associated with thermal energy (gCO_2eq/MJ);

η_{el} is electrical efficiency, determined as the annual electrical energy output divided by the energy content of annual fuel input (%);

η_h is heat efficiency, determined as the annual useful thermal energy output divided by the energy content of the annual fuel input (%);

C_{el} is a fraction of exergy in the electricity (-). For electricity, the fraction of exergy is set to 100%;

C_h is a fraction of exergy in the useful heat, calculated as Carnot efficiency (-). It is defined as:

$$C_h = \frac{T_h - T_0}{T_h} \tag{7}$$

where:

T_h is the temperature of the useful heat at the point of delivery (K);

T_0 is environmental temperature, set at 273.15 K (K).

2.3. Calculation of Greenhouse Gas Emissions Savings from Heat and Electricity Generated from Biogas

The following equations define greenhouse gas emissions from heat and electricity for respective fossil fuel comparators in order to calculate GHG savings obtained from heat $GHG\ SAVINGS_{,heat}$ and electricity $GHG\ SAVINGS_{,electricity}$ generated from biogas:

$$GHG\ SAVINGS_{,heat} = \frac{EC_{F(h)} - EC_h}{EC_{F(h)}} \tag{8}$$

$$GHG\ SAVINGS_{,electricity} = \frac{EC_{F(el)} - EC_{el}}{EC_{F(el)}} \tag{9}$$

where

$EC_{F(h)/F(el)}$ is total emissions from the fossil fuel comparator for useful thermal energy/electrical energy (gCO_2eq/MJ);

$EC_{h/el}$ is total emissions from the useful thermal energy/electrical energy generated from biogas (gCO_2eq/MJ).

The values of fossil fuel comparators were equal to 183 gCO_2eq/MJ for electrical energy and 80 gCO_2eq/MJ for useful thermal energy.

3. Results and Discussion

As previously noted, the savings of GHG emissions from biogas used in CHP engines after being produced from various kinds of agricultural and industrial residues and by-products, as well as municipal biowaste, are computed. Here, it was presumed that a CHP engine meets the electricity and heat needs in the biogas generation process (referred to as Case 1 in Directive 2018/2001). The temperature at the delivery site was considered to be 80 °C (353.15 K), and the heat efficiency and power efficiency were presumed to be 40% and 36%, respectively. These figures were chosen in accordance with the operating values of the biogas plants in operation.

3.1. Emissions from the Extraction or Cultivation of Raw Material (e_{ec})

Since the direct use of agricultural land is the creation of an agricultural product that is not used for the generation of biogas, the utilisation of considered feedstocks did not generate emissions from extraction or cultivation.

3.2. Annualised Emissions from Carbon Stock Changes (e_l)

Because agricultural land is not required to produce the examined feedstocks, their utilisation did not result in land-use change. Hence, e_l could be neglected (taken to be zero), as those emissions were allocated to the cultivated agricultural goods.

3.3. Emissions from Processing (e_p)

Two cases of agricultural residues were considered. In the first case, the residues were baled to improve their bulk density; this was not performed in the second case. The values used for the calculation were obtained from Table 1 and from default values of processing emissions for agricultural residues with density > 0.2 t. For industrial by-products and municipal biowaste, values used for calculation were set in accordance with the default values for biowaste in both open digestate and closed digestate configurations. The values for both groups of feedstocks are presented in Tables 4 and 5.

Table 4. Emissions from processing and agricultural residues.

e_p (gCO_2eq/MJ)	Maize Stover	Wheat Straw	Barley Straw	Oat Straw	Triticale Straw
Unbaled	0	0	0	0	0
Baled	1.1	1.1	1.1	1.1	1.1

Table 5. Emissions from processing, municipal biowaste industrial by-products.

e_p (gCO_2eq/MJ)	Municipal Biowaste	Grape Pressings	Tomato Pomace	Olive Pomace	Brewers' Spent Grain	Sugar Beet Pulp
Open digestate	0	0	0	0	0	0
Close digestate	21.8	21.8	21.8	21.8	21.8	21.8

3.4. Emissions from the Fuel in Use (e_u)

As previously stated, biogenic CO_2 combustion emissions were considered to be zero for biogas. Regarding biogas, the fuel's typical non-CO_2 emissions were equal to 8.9 gCO_2/MJ, while default values were anticipated to be 40% higher than the typical values [21].

3.5. Emission Savings from Soil Carbon Accumulation via Improved Agricultural Management (e_{sca})

For the considered feedstocks, it was presumed that they do not fulfil the requirements needed to include this factor in the GHG savings, defined in the Method section.

3.6. Emission Savings from CO_2 Capture and Geological Storage (e_{ccs}) and Replacement (e_{ccr})

As most biogas plants operating nowadays do not have a CO_2 capture storage and replacement system, it was assumed that no GHG savings come from CO_2 capture storage and replacement.

3.7. Emissions from Transport and Distribution (e_{td})

As can be seen from Equation (3), GHG emissions from transport and distribution were a function of transport distance. Those values are presented in diagrams in Figure 1 for unbaled agricultural residues, Figure 2 for baled agricultural residues, and Figure 3 for municipal biowaste and industrial by-products.

Unbaled agricultural residues had a considerable increase in transport emissions per kilometre, as shown in Figure 1, because of their low bulk density. Barley straw had the highest increase in emissions per kilometre because of its lowest density. The emissions from transportation and distribution for barley straw outweighed the emissions from all other factors when the transport distribution was 33 km or greater. On the other hand, maize stover had the lowest rise in emissions per kilometre because of its higher density and higher biogas yield. In the case of distributions of 60 km or more, transportation emissions outweighed other sources of emissions in this situation.

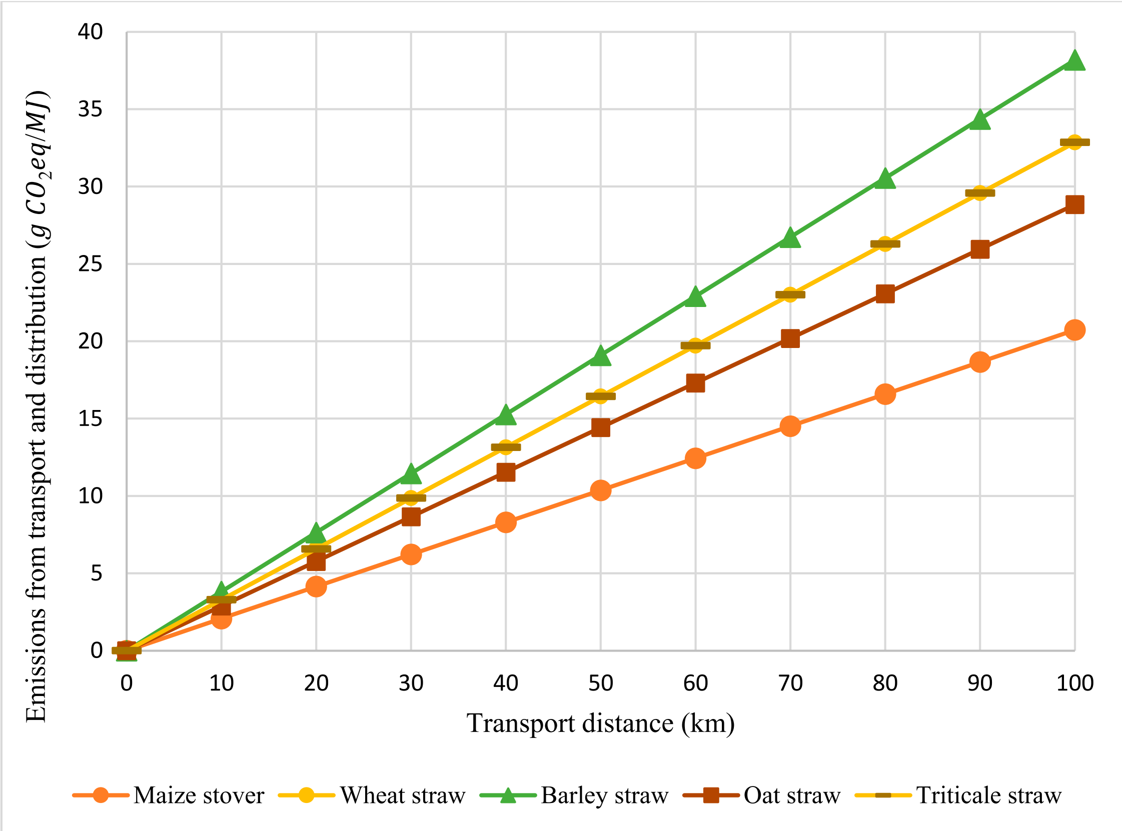

Figure 1. Emissions from transport and distribution of agricultural residues (unbaled).

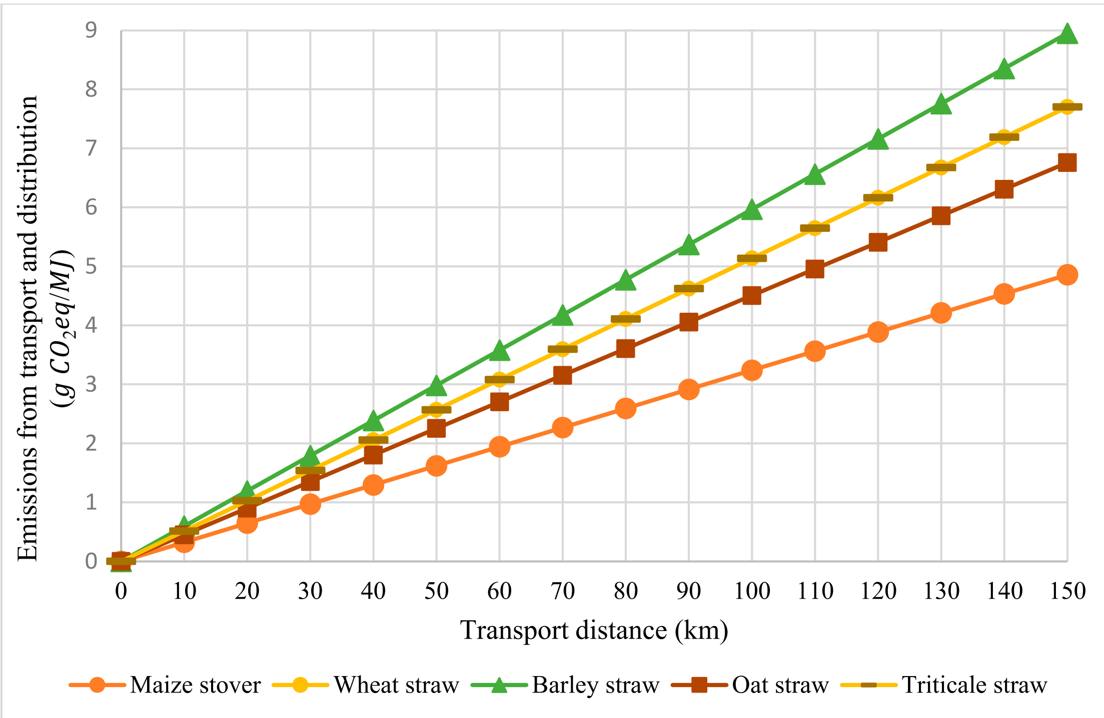

Figure 2. Emissions from transport and distribution of agricultural residues (baled).

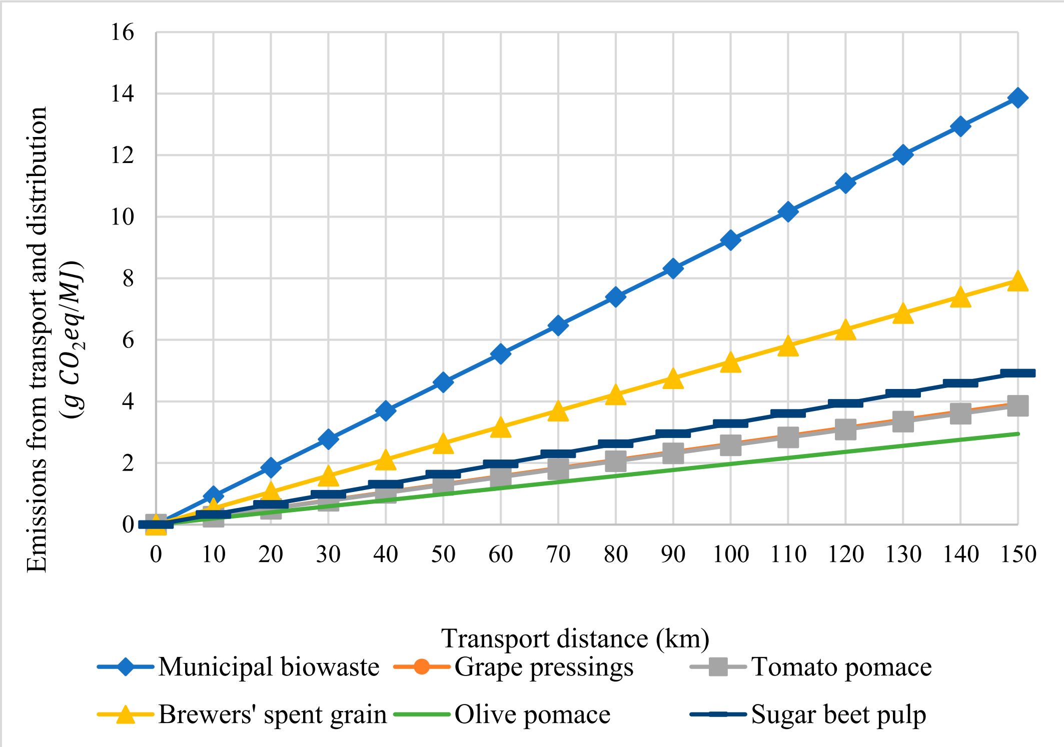

Figure 3. Emissions from transport and distribution -municipal biowaste and industrial by-products.

The baling of agricultural residues increases the bulk density eight times [25]. Because of the shape of a bale, it was assumed that bales could fill up to 80% of the truck storage space. Emissions of transport and distribution for baled agricultural residues are presented in Figure 2.

The baling of agricultural by-products increased the bulk density and, as a result, greatly slowed the increase in emissions from transit and distribution while slightly increasing the emissions of feedstock processing. Baled agricultural residues had a five-fold lower impact on distribution and transportation-related specific emissions than in the previous case.

Figure 3 shows the emissions from the distribution and transport of municipal biowaste and industrial by-products as a function of distribution and transport.

Brewers' spent grain had the most significant rise in specific transport emissions among the analysed industrial by-products, whereas olive pomace had the lowest increase, as can be seen in Figure 3. Nevertheless, compared with the agricultural residues, those numbers are still much lower (even with the second case). This is caused by the industrial by-products' greater bulk density. Municipal biowaste, on the other hand, had a significant increase in emissions per kilometre due to lower bulk density.

3.8. Greenhouse Gas Emissions Savings from Heat and Electricity Generated from Biogas as a Function of Transport Distance

Biogas plants with CHP engines must achieve both GHG savings for electricity and usable heat to comply with Directive 2018/2001. It is evident from the computed GHG reductions for heat and electricity production that the percentage of GHG savings for the generation of electricity was lower. As a result, the benchmark for calculating the maximum transport distance was 80% of GHG savings for electricity production.

GHG savings of unbaled agricultural residues are presented in Figure 4 as a function of transport distance.

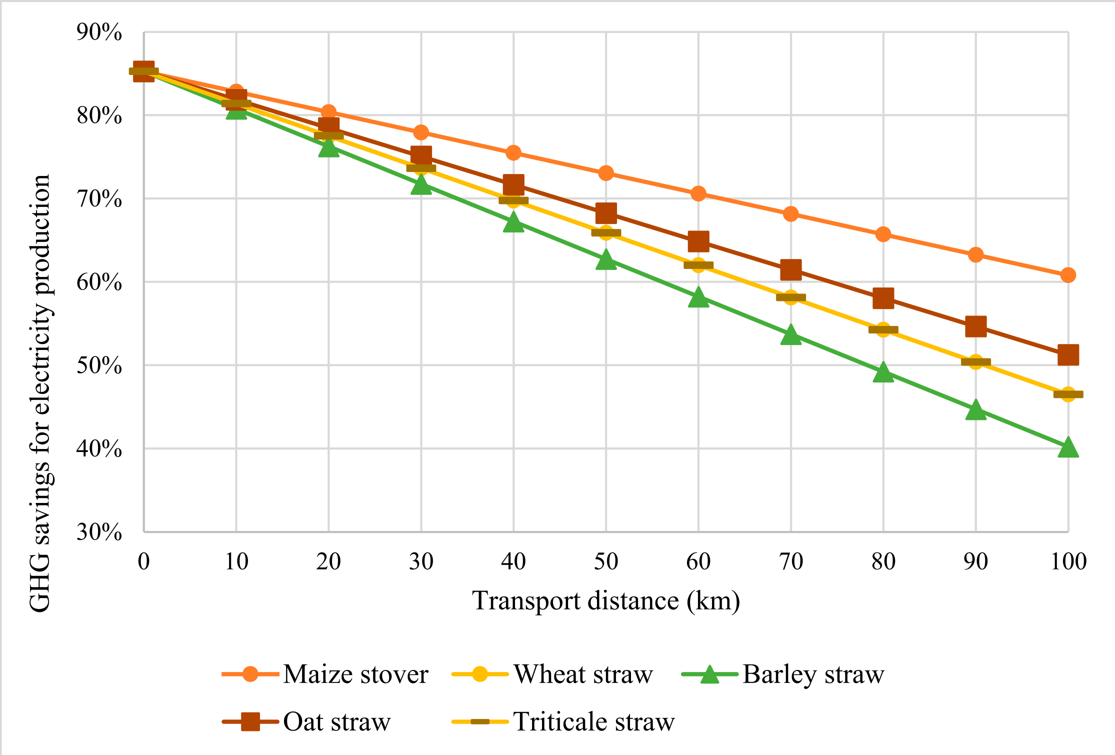

Figure 4. GHG savings for electricity production-agricultural residues (unbaled).

From the diagram presented in Figure 4, the maximum travel distance to achieve 80% of GHG savings from the utilisation of biogas, which uses unbaled agricultural residues as feedstock, can be determined. Those values are defined in Table 6. The greatest travel distance of unbaled agricultural residues was fairly low, ranging from 12 km for barley straw to 21 km for maize stover, as shown in Figure 4 and Table 6.

Table 6. Maximum travel distance for achieving 80% of GHG savings using unbaled agricultural residues.

	Maize Stover	Wheat Straw	Barley Straw	Oat Straw	Triticale Straw
$D,\ max$ (km)	21	14	12	15	14

Figure 5 shows GHG savings of baled agricultural residues as a function of transport distance.

The baling of agricultural residues greatly enhanced specific GHG savings as a function of transport distance, as predicted from the data shown in Figure 2, although it increased processing emissions. Hence, for baled agricultural leftovers, the maximum travel distance ranged from 65 km for wheat and triticale straw to 104 km for maize stover. Those values can be determined from the diagram presented in Figure 5 and are defined in Table 7.

Table 7. Maximum travel distance for achieving 80% of GHG savings using baled agricultural residues.

	Maize Stover	Wheat Straw	Barley Straw	Oat Straw	Triticale Straw
$D,\ max$ (km)	104	65	55.5	74	65

Figure 6 shows GHG savings of municipal biowaste and industrial residues as a function of transport distance.

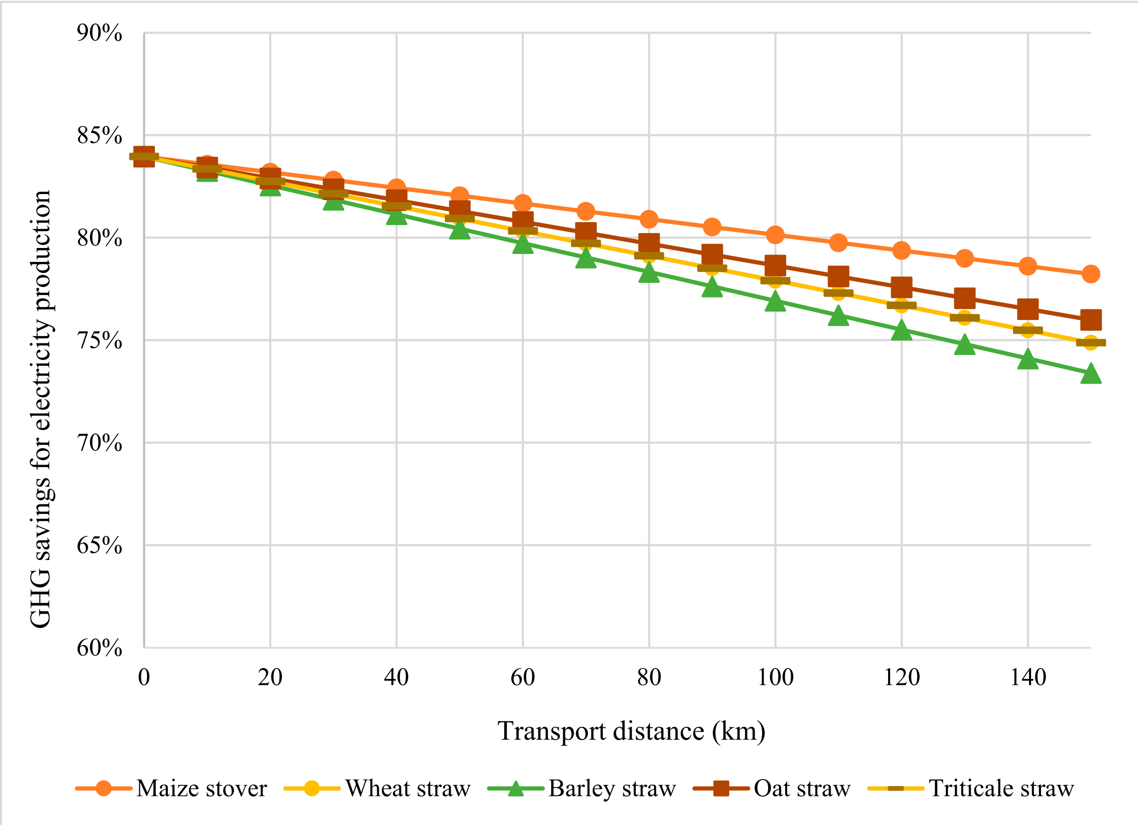

Figure 5. GHG savings for electricity production from agricultural residues (baled).

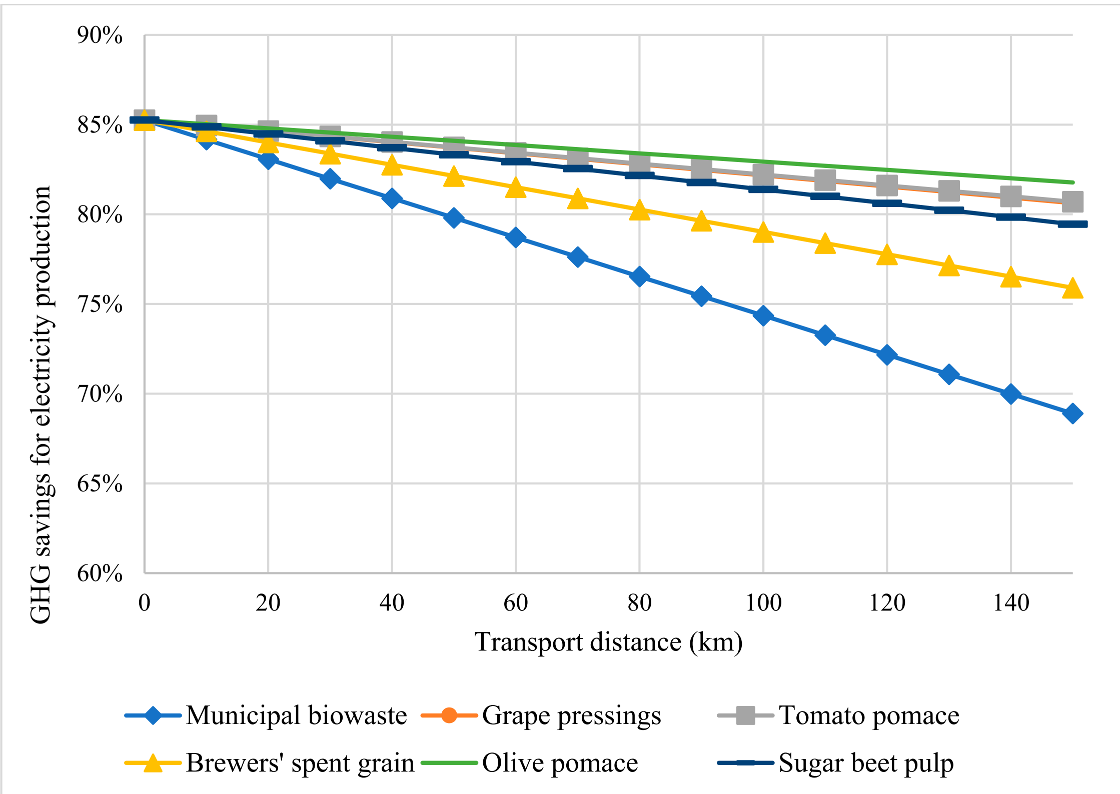

Figure 6. GHG savings for electricity production-municipal biowaste and industrial by-products.

It is important to note that Figure 6 presents only the case with closed digestate, as the considered feedstocks could not achieve 80% of GHG savings in the event of an open digestate. The maximum travel distance for municipal biowaste was 48 km, while for the industrial by-products, it ranged from 84 km for brewers' spent grain to 227 km for olive pomace. Those values can be determined from the diagram presented in Figure 6 and are defined in Table 8. Compared with agricultural residues, industrial by-products could achieve the required savings with a higher transport distance because of higher bulk density.

Table 8. Maximum travel distance for achieving 80% of GHG savings-municipal biowaste and industrial by-products.

	Municipal Biowaste	Grape Pressings	Tomato Residues	Brewers' Spent Grain	Olive Pomace	Sugar Beet Pulp
D, max (km)	48	170	173	84	227	136

The examined feedstocks were a sustainable option for producing biogas as they did not require agricultural land (unlike the already widely used maize silage) and satisfied the sustainability requirements defined in Directive 2018/2001. These requirements state that biogas feedstocks should not be produced from raw materials obtained from land with a high biodiversity value, such as primary forests, areas for the protection of rare or endangered ecosystems or species, highly biodiverse grasslands, wetlands, etc., and must adhere to the criteria for forestry, land use, and land-use change (LULUCF).

The results showed that emissions from transportation and distribution had a substantial impact on total emissions and the resulting reductions in greenhouse gas emissions for the cases and feedstock groups discussed in this study.

The maximum transport distance between the examined feedstocks greatly varied, as shown by the results. Given that waste materials were preferred as biogas feedstock, it is clear that the scope of the analysed feedstocks must be broadened, and the maximum distance for each of the examined feedstocks must be defined. The hypothesis of this work, stating that considered feedstocks can reach 80% GHG savings in the case of a travel distance of up to 50 km, was incorrect for municipal biowaste and unbaled agricultural residues.

4. Conclusions

This paper conducted an analysis based on the method outlined in Directive 2018/2001 of greenhouse gas emissions and specific greenhouse gas emissions savings from biogas production using agricultural residues (wheat straw, barley straw, oat straw, triticale straw, and maize stover), municipal biowaste, and industrial by-products (grape pressings, tomato residues, brewers' spent grain, olive pomace, sugar beet pulp) as feedstock. This method is a GHG accounting method, which includes numerous emission factors for biogas electricity and heating production. The emissions were calculated/determined for each emission factor, except for the emissions from transport and distribution, which were calculated and presented as a function of a transport distance. According to calculations, the maximum travel distance for unbaled agricultural residues to achieve 80% GHG savings when compared with fossil fuel comparators ranged from 12 km for barley straw to 21 km for maize stover. The low bulk density of agricultural residues was the primary cause of the short transport distance. In the case of baled agricultural residues, the maximum travel distance ranged from 65 km for wheat and triticale straw to 104 km for maize stover. The maximum travel distance for municipal biowaste was 48 km, while for industrial by-products, it was significantly higher: 170 km for grape pressings, 173 km for tomato residues, 84 km for brewers' spent grain, 227 km for olive pomace, and 136 km for sugar beet pulp. These results can be attributed to the higher bulk density of industrial residues.

Research findings demonstrate that transportation emissions have a significant impact on biogas production's potential to achieve the required greenhouse gas emissions savings.

A substantial difference in the results further supports the need to increase the number of feedstocks for which default and typical values are available. Finally, it can be stated that for municipal biowaste, unbaled wheat and barley straw, the hypothesis that examined feedstocks can reach 80% GHG savings in the case of a travel distance up to 50 km is incorrect. Researchers, policymakers, and operators of biogas facilities are anticipated to benefit from this research and use the results in future planning.

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Abbreviations

AD	Anaerobic digestion
CHP	Combined heat and power
EU	European Union
GHG	Greenhouse gas
GWP	Global Warming Potential
IPCC	Intergovernmental Panel on Climate Change
LULUCF	Land Use, Land-Use Change, and Forestry

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PAPER 4

P-Graph approach for the economical optimisation of biomass supply network that meets requirements on greenhouse gas emissions savings - a case study of rural areas

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This work presents a novel P-graph-based model for optimizing a biomass supply network. The objective of this optimization is twofold: to find the most cost-effective biomass supply network with a minimum cost, while also fulfilling the required greenhouse gas (GHG) emissions savings defined in Directive 2018/2001 (80% savings compared to fossil fuel comparators) for the use of biogas.
To achieve this goal, an extension to a P-graph-based biomass supply network was developed, which allows the optimization of the network while limiting GHG emissions associated with the use of biogas. The model includes a summary of GHG emissions for each stage of biogas production and consumption, compared to threshold values. Additionally, seasonal variation in biomass supply was integrated into the model by using a multiperiod approach.
The model was developed and solved in P-Graph Studio, with input data defined using the Geographic Information System (GIS) tool, including feedstock availability, an optimal location for a biogas site, and transportation distance. The approach was tested in a case study located in a rural area. This model can benefit a wide range of stakeholders, including biogas plant operators, policymakers, researchers, and energy regulatory authorities.

1. Introduction

A drastic acceleration of the energy transition and an increase in natural gas independence is required in light of the changing geopolitical and energy market realities. Anaerobic digestion (AD) of by-products, residues and waste materials has not only been recognised as technology for the generation of sustainable alternative fuel but is also an environmentally friendly waste treatment method [1]. Biogas production in the European Union (EU) has steadily increased during the last decade, going from 6,227 biogas plants in 2009 [2] to 20,000 in 2021 [3]. Up to 72 % of the feedstock used for biogas production comes from the agricultural sector [4], mostly from maize silage. The competitive use of biogas feedstocks with food and feed production raised not only environmental but also socio-economic concerns, reflected in new sustainability requirements, defined by EU legislation. The revised Renewable Energy Directive (D2018/2001), which came into force in December 2018, established sustainability and the greenhouse gas (GHG) emission-reduction standards that biogas used in transportation, electricity, heating, and cooling must meet. Concerning GHG savings, the Directive defines that the GHG savings from the use of biomass for electricity, heating and cooling production should be at least 70% for installations starting operation from the beginning of the year 2021 until the end of 2025 and 80 % for installations starting operation from 2026 [5]. Furthermore, the European Commission (EC) set a cap on food and feed crops toward the EU renewable objective, starting at 7% in 2021 and gradually decreasing to 0% in 2030, in order to reduce the impact of Indirect Land-Use Change (ILUC). With the given new requirements, the utilization of materials previously regarded as waste is receiving increased attention, as it not only improves the sustainability of biogas production but also improves waste management and resource efficiency. Hence, biogas production can serve as a treatment plant that converts waste resources into high-value products, consequently contributing towards the objectives of the European Circular Economy Action Plan [6]. In their paper, the European Biogas Association highlighted the untapped opportunities for GHG savings through the utilization of industrial waste, loaded with organic matter, for biogas production [7], as waste management measures have a significant effect on climate change mitigation [8]. Furthermore, lignocellulose biomass is now recognised as a significant untapped source of renewable energy, that may substantially contribute towards fulfilling the global demand for renewable energy [9].

A transition toward more sustainable biogas and biofuel production seeks more research on the more sustainable alternatives (such as residues and by-products) which should replace the currently dominant maize silage in biogas production by fulfilling the sustainability and greenhouse emission criteria [10]. The potential of biomass residues that is accessible for the production of biogas is limited by factors like their low energy density, scattered production and competitiveness with other uses [11]. Assessment of this potential is the necessary first step and it should include spatial dimension. The application of GIS technologies has been recognised as being particularly beneficial for mapping biomass potential during the past ten years since it can deliver insightful information about the spatial distribution of the biomass potential and input data for biomass potential analysis.

One of the main barriers to enhanced biomass utilisation in energy supply is the economic viability of a biomass supply network. Different methods for optimising the biomass supply network have been presented in the literature to overcome this obstacle. It has been recognised that graph theory methods are being employed more frequently to solve biomass supply network modelling issues. Graph theory is the study of graphs, which are mathematical constructions used to represent pairwise relationships between objects. Implementation of graph theory methods offers several advantages for supply network modelling. Some of the advantages of graph theory methods are a representation of decision structures (solutions), the algorithmic generation of a mathematical model and the derivation of multiple alternative solutions [12]. In comparison to the other methods, such as Mixed-Integer Linear Programming (MILP), there is a reduced complexity of the solution procedure.

1.1 Literature review

It is becoming evident that energy systems modelling is progressively embracing different types of integrative approaches [13]. Murele et al. [14] investigated the influence of the integration of biomass into coal-based energy supply networks. Results of the optimisation aimed to minimise the cost of the energy supply network, obtained through the General Algebraic Modelling System software (GAMS), indicate that a biomass fraction of 7.9% in the mixed solid fuel will provide an optimal solution, as it would result in a balanced cost decrease of the emission cost and increase of the supply network. Simon et al. [15] developed a model that simulates the supply curve of wood biomass from the sustainable management of natural forests. The findings indicate that the maximum admissible distance to the nearest transportation route and the associated transportation expenses are the two factors that exert the greatest impact on both the supply and cost of wood biomass. Rentizelas et al. [16] applied the Data Envelopment Analysis method for assessing the cost, energy and GHG emission efficiency of international biomass supply network pathways. The selection of the most efficient pathway depends on the total cost, energy consumption and emissions, as well as priorities of the decision maker. Shen et al. [17] developed a novel mathematical optimisation approach that allows the reduction of redundancy of data series to solve the multi-echelon biomass supply problem. This multi-echelon biomass supply problem includes economic, environmental and social indicators, optimised by maximising economic viability and social benefit while minimising environmental emission through a weighted-sum approach and max-min aggregation approach.

The use of P-graphs in energy system modelling has intensified during the past two decades. In their recent paper, Xu et al. [18] implemented the P-graph approach to define optimal energy export strategies of islands, whose objective is to minimise construction, operating and environmental cost (related to greenhouse gas footprint). Results showed that the best operational path and the best economical cost are in the case of export by electricity. Similar to this, the paper published in April 2023, written by Ji et al. [19] presents the implementation of the P-graph approach for the optimisation of multi-period renewable energy systems with hydrogen and battery energy storage. For the developed biomass energy supply scenario, the results show that the renewable energy systems with hydrogen storage and battery storage are, respectively, 21.5 % and 5.3 % cheaper than those without energy storage. The developed model investigates CO_2 generation and includes it in the optimisation through the cost of CO_2 emissions. Aviso et al. [20] implemented a P-graph approach to the development of optimal and sub-optimal biochar-based carbon networks. Here, the objective was to optimise the network in terms of overall carbon sequestered annually, without exceeding constraints on soil contamination. Lam et al. [21] have proposed a model to integrate palm biomass and waste motor oil into the waste-to-energy model. The method to solve the combinatorial of the biomass supply chain in Federal Land Development Authority Jengka was presented by Varbanov et al. [22]. Here, the authors have proposed possible locations for building a new biomass processing facility in the considered region, which should be used for the utilization of waste from oil palm biomass processing. Malladi et al. [23] have created a decision support tool to optimise the short-term logistics of forest-based biomass through the minimisation of the biomass logistic cost. The method to solve the combinatorial of the biomass supply chain in Federal Land Development Authority Jengka was presented by Varbanov et al. [22]. Here, the authors have proposed possible locations for building a new biomass processing facility in the considered region, which should be used for the utilization of waste from oil palm biomass processing. Malladi et al. [23] have created a decision support tool to optimise the short-term logistics of forest-based biomass through the minimisation of the biomass logistic cost. Van Fan et al. [24] applied the P-graph approach to detect cost-optimal and suboptimal pre-and post-treatment pathways for the anaerobic digestion of lignocellulosic waste. The result of the optimisation for the lignocellulosic waste showed that alkali CaO pre-treatment proved to be the cost-optimal pre-treatment option of the lignocellulosic waste, while H2S + membrane separation proved to be the cost-optimal post-treatment (biomethane upgrading) option. Benjamin [25]. developed a P-graph approach to perform a critical analysis of an integrated network of biomass processing industries under scenarios that involve both supply and demand side disturbances. This methodology enables the reduction of the net product stream output that results from the occurrence of climate change-induced events (supply-side disruptions) and seasonal fluctuations in demand, to be assessed. Vance et al. [26] implemented the P-graph method for the development of economically optimal and suboptimal structures of biomass network that includes corn silage, grass silage, corn straw and wood as feedstock material for combined heat and power (CHP) units. For the obtained results (ranked structures) ecological footprint was assessed, indicating the amount of land required to support and assimilate a given human population's consumption and wastes. The structures whose ecological footprint was lower than the given threshold were considered sustainable.

The objective of this work is to develop a novel P-graph-based method for economical optimisation of the biomass supply network, that meets requirements on greenhouse gas emissions savings and considers the seasonality of biomass availability, a P-graph based model was developed. The threshold of this requirement is defined in Directive 2018/2001 and it equals 80% savings compared to fossil fuel comparator. As represented in the literature review, studies in this field determine the economically optimal and sub-optimal biomass networks and thereafter compare the ecological footprint/environment constraints upon the optimisation process. Furthermore, the seasonality of biomass supply and biogas demand is mostly neglected, although it may have a significant impact on the viability of utilisation of feedstocks with high seasonal fluctuations, such as industrial by-products and agricultural residues. To address this research gap, the contributions which this study delivers, in comparison to earlier research are the following:

- a P-graph-based model which enables the optimisation of a biomass supply network that simultaneously limits the GHG emissions that a biomass network can generate and defines the optimal and sub-optimal economical structures, which are in line with the requirements of the GHG emission savings;
- the developed model integrates the seasonality of biomass supply, through the implementation of the multi-period approach. Hence, the limitations on greenhouse gas emissions are automated and fulfilled for each period.

2. Problem statement

To investigate the possibilities of the P-graph approach to perform an economical optimisation of the biomass supply chain, that meets requirements on greenhouse gas emissions savings and considers the seasonality of biomass availability, the P-graph-based model was developed. The main assumptions of the problem can be defined as follows.

- GHG emission-saving requirements
  i. The assumption is used that biogas produced in anaerobic digestion is future utilised in a CHP engine, which supplies the electric and heat demand of the process. This is a so-called case 1 in D2018/2001. This assumption was used for the calculation of the maximal allowed total GHG emissions.
- To ensure the compliance of resulting structures (optimal and suboptimal) with GHG saving requirements defined in Directive 2018/2001 80% GHG emission savings compared to the fossil fuel comparator), authors calculated the maximal allowed GHG emissions in their previous work [27]. For a typical case, where electrical and heat efficiency is 36% and 43% respectively, the maximal total GHG emissions equals 16.95 gCO_2/MJ biogas. In the developed model, this value is set as a threshold. Seasonality of feedstock availability
  i. Feedstock availability through a year is represented through multi-period representation. Here, the assumption is used that biomass available for biogas production is the one generated in the specific period (month/s). There is a threefold reason for this. The first one is that some of the considered feedstock can not be stored for a longer period of time, due to potential changes in feedstock conditions, which could result in the adverse performance of biogas production. The second is

that seasonal feedstock storage may result in additional methane emissions generated during the storage period, which could result in exceeding the threshold of GHG emissions, due to the high global warming potential of methane. The final one is the cost of the investment and maintenance of the seasonal storage.

ii. In case the required biogas production exceeds the biogas potential contained in the biomass, the assumption was used that this gap will be covered with the wheat straw, due to favourable storage properties.

- Cost of biomass supply network

i. The cost of anaerobic digestion is considered to be the same for each biomass supply network. Therefore, this cost is not included in the cost of the biomass supply network, as it does not differ for different structures of biomass supply networks.

ii. The cost of a feedstock, transport and processing is considered and calculated as a specific cost (cost per unit of mass or energy).

The objective function of the optimisation is to minimise the cost of the biomass supply network while fulfilling the given constraints regarding GHG savings and maximising the utilisation of seasonally available biomass. The hypothesis of this work is that an economically optimal residual biomass supply network for biogas production, that meets sustainability and greenhouse gas emissions saving criteria, could be determined with the P-graph approach.

3. Method

The method used for this work can be divided into two major sections. The first part of the research is conducted with the GIS tool and the second part of the research is conducted with the P -graph tool. The flowchart presented in Figure 1 represents the steps of the method, which are explained in detail below.

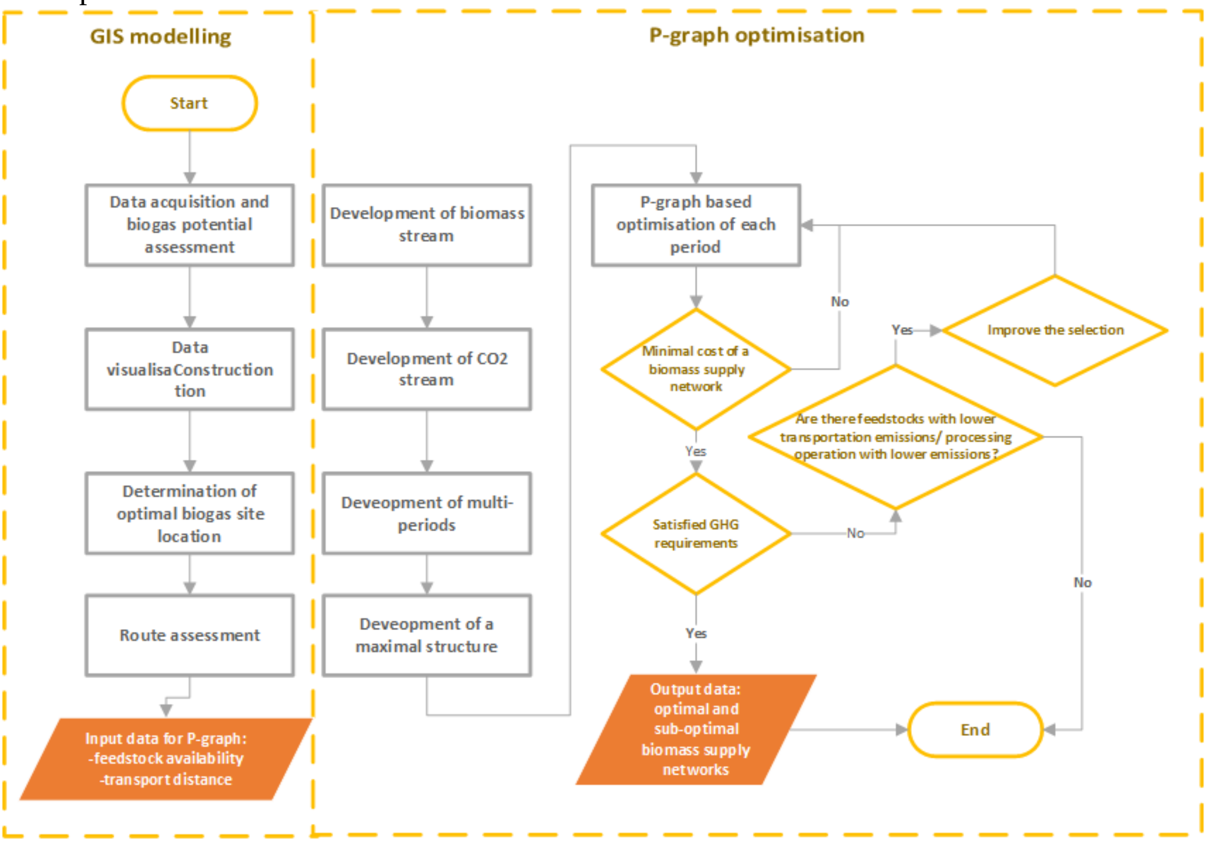


Figure 1 Flowchart representation of the method

3.1 GIS assessment of biomass and biogas potential

GIS assessment of biomass potential is the process of using a GIS tool to create maps that present the distribution and density of biomass potential in a particular area. For this purpose, the QGIS tool [28] was used. QGIS is a free and open geographic information system tool that allows users to develop, analyse, edit and print geospatial data. The conducted assessment was implemented to obtain input data for P-Graph Studio (P-graph software). It includes several steps, which are described in more detail in the subsections below.

<u>Data acquisition and biogas potential assessment</u>

Residues and by-products that are located in rural regions are the raw materials (feedstocks) taken into consideration in this work. More precisely, the following categories can be created from the examined feedstocks for this work:

- Agricultural residues (maize stover, wheat straw);
- Manure (Cattle, pig, chicken manure);
- By-products from the food industry (Grape pressings, sugar beet pulp).

To assess the biogas potential, the theoretical potential of the selected feedstock must first be assessed. This theoretical potential is based on the ratio of residue to processed commodities and the amount of processed commodities. Only a portion of the theoretical potential, also called technical potential can be utilised for the production of bioenergy due to competition with other purposes (feed, land protection, etc). Hence, a sustainable removal rate was applied for wheat straw. This factor equals 40% for wheat straw [29]. For livestock, the theoretical potential is a function of the number of livestock (head) and the amount of manure produced annually per head.

Based on the theoretical potential of fresh feedstocks, a specific biogas yield from fresh feedstock, and the methane content of biogas, the biogas potential of the evaluated feedstocks is determined. Input data for the calculation of the biogas potential from wheat straw and cow manure can be obtained from public reports and agricultural geoportals provided by a national Ministry of Agriculture and the related Agencies. To assess the biogas potential from grape pressings and sugar beet pulp, input data can be obtained through publicly available annual reports. Table 1 lists the biogas yield and biogas methane content for the considered feedstocks.

Table 1 Biogas yield and methane content of considered residues and by-products

Residues/ by-products	$\boldsymbol{y}$ **(m^3/t_{FM})**	$\boldsymbol{s_{CH_4}}$ **(%)**	**Reference**
Wheat straw	125	52.5	[30]
Grape pressings	160	80	[31]
Sugar beet pulp	96	50	[32]
Cow manure	22.1*		[33]

* In literature, the value is given for a methane yield (m^3 CH_4/t)

Previous studies [34], [35]. of the authors go into a greater level of detail on this step.

<u>Data visualisation</u>

The geographic distribution of biomass potential and the distance to a possible biogas site limit the viability of using residues and by-products from an economic and GHG savings perspective.

Data visualisation highly depends on the type of represented feedstocks. Feedstocks that are being generated at farms (manure) and in the food industry can be represented by a point vector layer since the production of feedstocks occurs at a specific location. To link the attribute (non-spatial) information on the biogas potential to spatial information, geocoding can be applied, which represents converting addresses into geographic coordinates and thus enable utilisation of the data in the GIS tool.

On the other hand, agricultural residues occur in the wider area. A top-down approach was applied to visualise and evaluate the distribution of this potential, and the potential of wheat straw was dispersed in the areas that fall under distinct land cover classifications. Data included in the GIS tool can be afterwards used for map development. GIS biomass maps can visualise information on biomass and biogas potential and be used for distance determination and optimal biogas site location determination.

Determination of optimal biogas site location

The optimal location for a biogas plant is determined using geographic and attribute (non-spatial) data. This biogas plant can be understood as a centralised production site that produces biogas from feedstock supplied by the concerned industry, farms and agricultural sites. The goal function of this optimisation is to minimize transport distance between a biogas site and concerned feedstock providers. For this optimisation, the "Mean coordinate" spatial query, available in QGIS was used. As the input data for the optimisation, biogas potential was used as the weighted factor. In a case where biogas potential is represented in both point and polygon vector layers, it is important to align the type of layers and merge those layers into one, which can be used for optimal biogas site location determination. In this work, the potential of the agricultural residues, initially represented in the polygon vector layer was transferred to the point layer by using the "Centroids" query in QGIS. The generated points can be understood as the collection sites of agricultural residues.

Route assessment

The "Shortest path" query in QGIS can be used to examine routes (transport distance). This query allows the automatic assessment of the shortest (or fastest, upon user preferences) route between feedstock providers and biogas plants. The input data used for this assessment includes a network layer representing transport routes (roads) in the considered, a layer representing feedstock providers including the information a respective biogas potential and a layer including the location of the optimal biogas site location. The transport routes (road networks) can be imported to QGIS with the "QucikOSM" plugin. Specific transportation costs can be determined with equation (1):

$$C_{trans} = \frac{d * (K_{full} + K_{empty})}{B_{biogas}} * b * T \quad (1)$$

Where C_{trans} stands for specific transport cost (EUR/GJ), d for transport distance (km), K_{full} for fuel consumption of a full truck (L/km), K_{empty} for fuel consumption of empty truck (L/km),

b for fuel price (EUR/L), B_{biogas} for biogas potential of transported feedstock (GJ) and T for transport cost correction factor. In this work, we used the assumption that T equals 3, which means that the cost of fuel is one-third of the total transport cost.

3.2 Optimisation of biomass supply network by P-graph

Due to the combinatorial nature of the problem, biogas production can be accomplished by a wide range of alternative structures. The determination of the optimal network structure is most frequently referred to as process-network synthesis (PNS) flowsheet design. The P-graph method is a graph-theoretical approach used for solving PNS problems [36]. Hence, for this step, the P-graph-based algorithms and the concomitant software (P-Graph Studio) will be used.

3.2.1 P-graph studio and P-graph based algorithms

P-graphs are bipartite graphs, each comprising material nodes (M) and operating unit nodes (O) and arcs between them. Determination of the feasible structures will be performed in three major steps.

In the first step, the maximal structure of feasible solutions for biogas production is developed. The maximal structure comprises all the combinatorically feasible structures capable of yielding the specified products from the specified raw materials. The feasible solution structure generated by process-network synthesis must have several basic features that are taken as axioms, the introduction of which improves the efficiency of the combinatorial search during the process. In the P-graph-based methods, the algorithm MSG (Maximal Structure Generation) yields the maximal structure, i.e., the superstructure, for the Process Network Synthesis (PNS) problem. MSG Algorithm is a polynomial algorithm based on the axioms which define representations of the final product, interim products, raw materials, operating units and arcs. Those axioms are explained in detail by Friedler et al. [37]. The maximal structure will be analysed in the second step. Here, algorithm SSG (Solution Structure Generation) will be used for the generation of all the solution structures representing the combinatorically feasible flowsheets from the maximal structure. Algorithm SSG systematically and combinatorically selects a series of active sets and carries out decision mappings. Finally, ABB (Accelerated Branch and Bound) algorithm will be used to generate the n-best feasible solution structures. Algorithm ABB is a branch and bound algorithm for solving combinatorial problems. It traverses the maximal structure, keeping track of all partial solutions in corresponding tree branches and bounding until it finds a branch whose objective function is better than the current best solution.

3.2.2. Biomass supply network design

The first step in creating a P-graph for a biomass supply network is to identify the potential feedstocks that can be used in the considered area and to map out the transportation network. Those data (type of feedstock, technical potential, biogas potential transport distance) were exported from the GIS tool in the previous steps.

Material nodes are representing raw materials (wheat straw, grape pressings, manure and sugar beet pulp), interim materials and the final product (biogas). Operating unit nodes are representing biomass transport, biomass processing and anaerobic digestors. Anaerobic digesters are enclosed structures where the anaerobic breakdown of raw material (feedstock) takes place. The biomass supply network developed in this paper is presented in simplified form (for only one input raw material) in Figure 2.

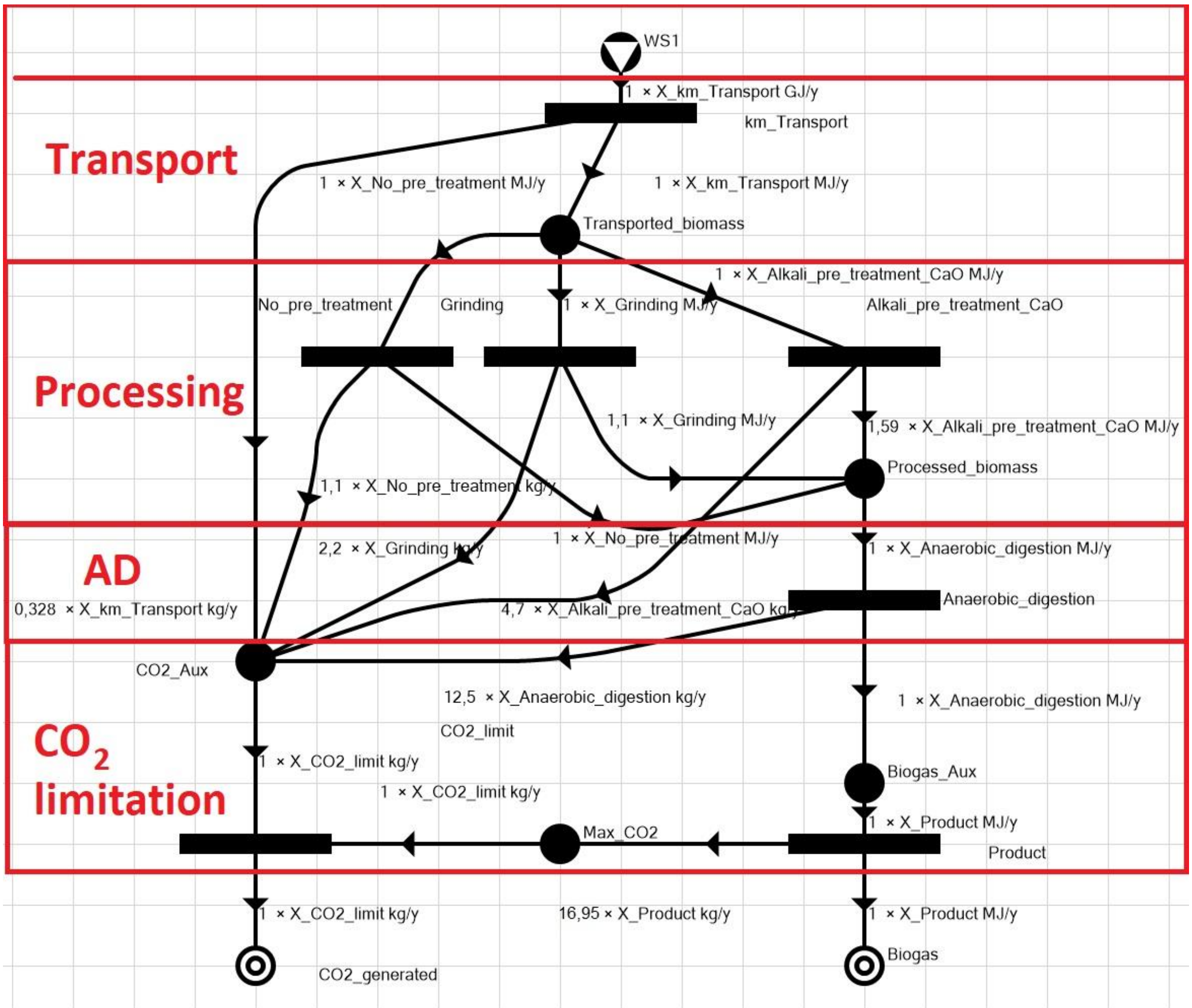
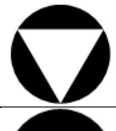
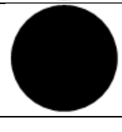


Figure 2 PNS network of the utilisation of wheat straw for biogas production

Elements included in the PNS network are presented in Table 2.

Table 2 P-graph representation of elements included in the PNS network

Element	P-graph representation
Feedstock sites: Cattle farm (CF)/ Wineries (W)/ Sugar factory (SF)/ Wheat straw collection site (WS)	
Intermediate products: Transported biomass/Processed biomass Auxiliary products: Maximal allowed CO_2 (Max CO_2), Summarised CO_2 generation (CO_2_Aux), Summarised Biogas production (Biogas_Aux)	
Operating units: Transport; Pre-processing: No pre-treatment/ Grinding and bailing/ Alkali pre-treatment; Anaerobic digestion (AD) Auxiliary units: CO_2 limitation (CO_2 limit)/ Biogas limitation (Product)	
Final products: Biogas/ Generated CO_2 (CO_2 gen)	

As can be seen from Figure 2 PNS network developed for utilisation of considered feedstocks (here wheat straw was taken as an example) includes two main streams- stream of biomass and stream of CO_2. In the context of biomass stream, it refers to the feedstock that is being transported, processed and utilised in an anaerobic digestor for biogas production. In the context of CO_2 streams, it refers to the emissions associated with the transport and processing of biomass, as well as the emissions associated with the biogas in use (here represented as emissions associated with anaerobic digestor). When developing CO_2 streams special attention was taken to ensure that all GHG emissions (including CO_2 and non-CO_2 emissions) are covered and assessed in line with the method defined in Directive 2018/2001 [38]. This ensures the compliance of resulting structures (optimal and suboptimal) with 80% GHG savings defined in Directive 2018/2001 to be legally binding for biogas sites starting operation from 2026. As a threshold, the value of 16.95 gCO_2/MJ_{biogas} net emissions is used in the model.

The stages in biomass and CO_2 streams are represented in red squares in Figure 2. All of those stages include operating units, while the resulting outputs are represented as interim materials except in the case of biogas, which is represented as the final product. Here, it is interesting to highlight part of the PNS network that represents the processing stage and part of the PNS network representing CO_2 limitation. In the processing stage for wheat straw three options are represented- no processing (only bailing at the field is included here), grinding and alkali pre-treatment (CaO). Implementation of one of those three options leads to different biogas yields obtained from the concerned feedstocks. As presented by Van Fan et al. [24], grinding will result in a 10% enhanced biogas yield of lignocellulosic waste, while alkali pre-treatment would lead to 59% higher biogas yield compared to the option without pre-treatment. On the other hand, grinding and alkali pre-treatment increases the cost of the pre-processing and associated GHG emissions. Those ratios are presented in arcs for the representing processing options of biomass stream. As expected, the cost and associated GHG emissions are the highest for alkali pre-treatment. For cases like this, where final cost and total GHG emissions depend on numerous factors, it is very beneficial to conduct a P-graph optimisation. Finally, it is important to note that the biogas potential of feedstocks refers to the reference biogas yield (in a case where there is no pre-treatment) and not the energy value of the feedstock composed in the chemical composition of the feedstock.

Part of the PNS network representing CO_2 limitation sets the threshold for GHG generation (and resulting savings) of the production of biogas and its use. For this purpose, two auxiliary products (intermediate materials) are included in the PNS network design- CO_2_Aux and Biogas_Aux. The maximal flow of those two auxiliary products is set to zero, indicating that they are completely consumed. As can be seen from Figure 2, CO_2_Aux summarizes all of the G emissions generated by processes represented by operating units. Biogas_Aux is used for setting the threshold (maximum) on GHG emissions that the use of fuel (biogas) can generate to be in line with the GHG savings. This limit is represented in PNS Network as Max_CO_2. In case if during the optimisation process, CO_2_Aux emissions are higher than Max_CO_2 emissions, the P-Graph Studio makes a new iteration to find a structure whose emissions are lower than Max_CO_2.

The cost of a PNS network includes the sum of the cost of the raw materials and the cost of the transport and processing cost. As the main objective of this work is to compare the economic feasibility of the structures, the cost of the anaerobic digestion was not considered here, as it is considered to be equal for all of the considered feedstocks. The goal function of the optimisation is to minimise the cost of a biogas supply network (structure). The optimal solution is the one which can deliver the required biogas production, for a minimal cost and by staying below the permissible limit on GHG production. In addition to an optimal solution the n- best solutions will be ranked.

3.2.3 Multi-period P-graph Optimisation

To incorporate the seasonal variation of biomass supply and biogas demand during the year, the model was extended to multi-periods. The multi-period P-graph modelling allows dividing a year into custom-selected periods of time, which can be of arbitrary length. A multi-period optimisation approach, provides more reliable data compared to a single-period model, as it takes into account fluctuations of inputs (feedstocks) supply during the year, as well as differences in output (biogas) demand throughout the year. For the considered problem, periods are selected based on the availability of considered feedstock types. Hence, months with equal feedstock supply are grouped into the same period. The multi-period extension was implemented by configuring the Multiperiodic settings of the PNS network using P-Graph Studio.

4. Case study

The presented method was demonstrated in the case study of the rural area of Osijek-Baranja County. The county is situated in the northeastern part of the country and has intensive livestock production and use of land for agricultural production. Figure 3 represents the sites and agricultural land considered in this case study.

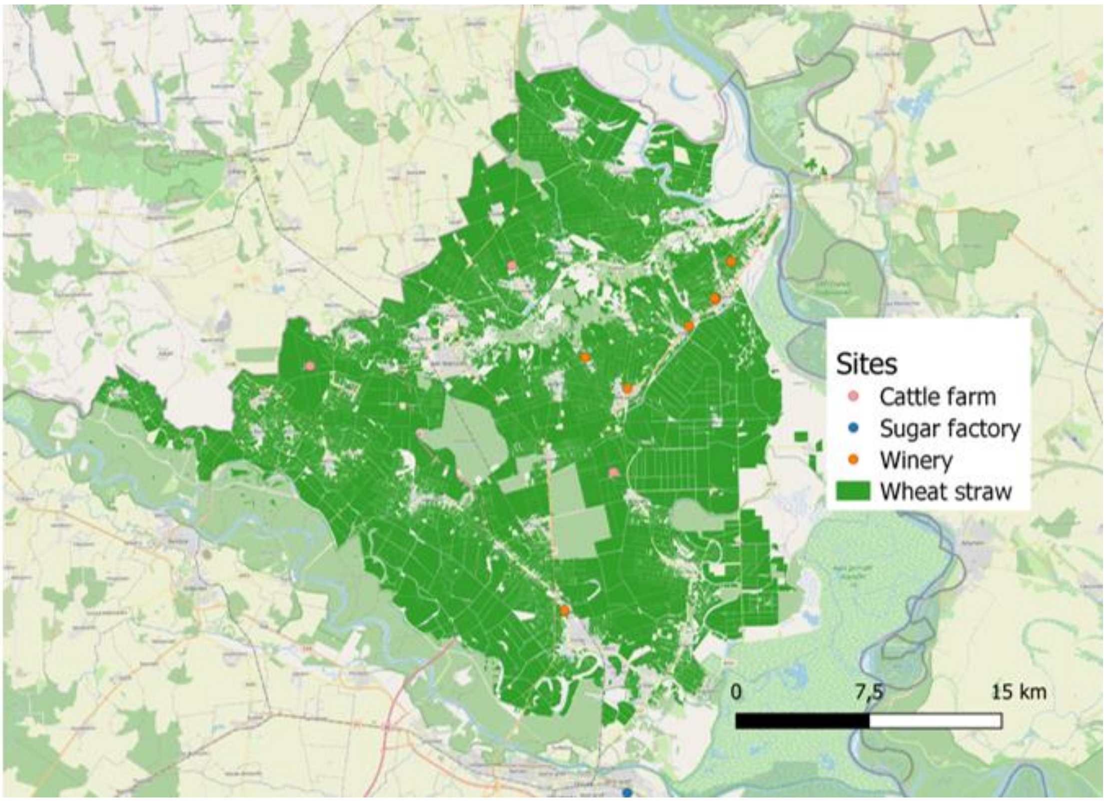


Figure 3: Case study sites and agricultural land

For the feedstock cost, the input data from Table 3 were used.

Table 3 Specific feedstock cost

Feedstock	Cattle manure	Grape pressings	Sugar beet pulp	Wheat straw
Cost (€/t)	5	5	25 [39]	25 [40]

5. Results

According to the method provided in the section above, the biogas potential from wheat straw, manure, grape pressings, and sugar beet pulp was determined for the farms, wineries, sugar factories, and wheat straw collection sites. The spatial distribution of biogas potential is presented in Figure 4.

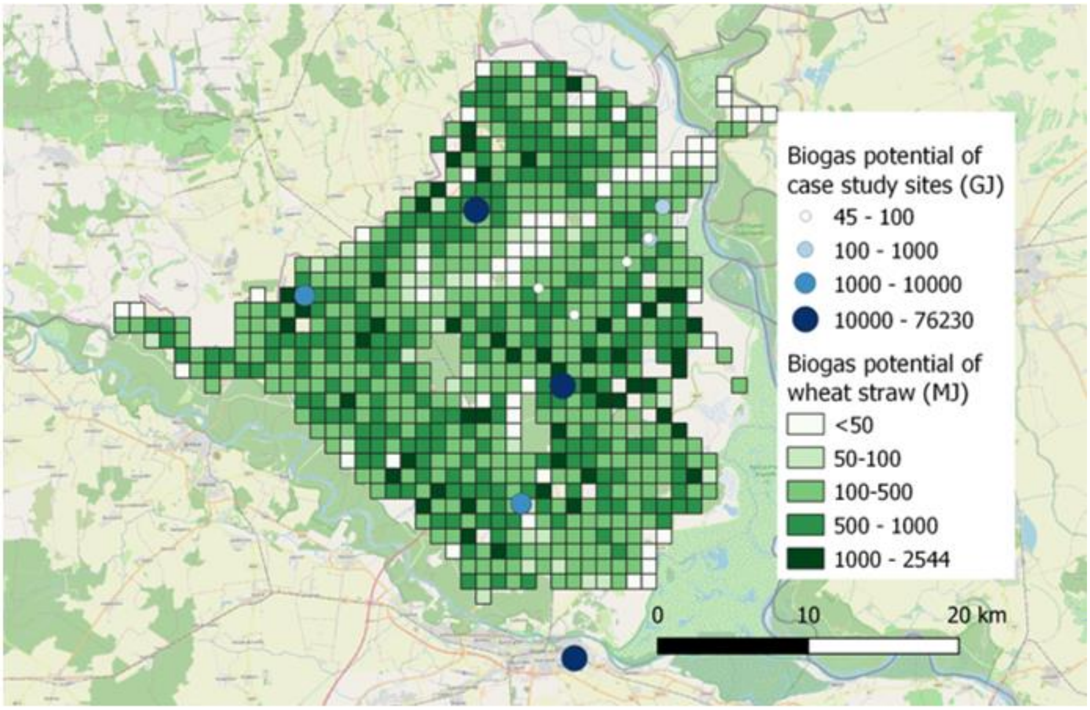


Figure 4 Biogas potential

The optimal location for the biogas site was defined based on the locations of feedstock suppliers with the highest biogas potential. In accordance with the optimal location, the transport distance between industry/farm/collection sites and the optimal location of the biogas site was calculated as represented in Figure 5.

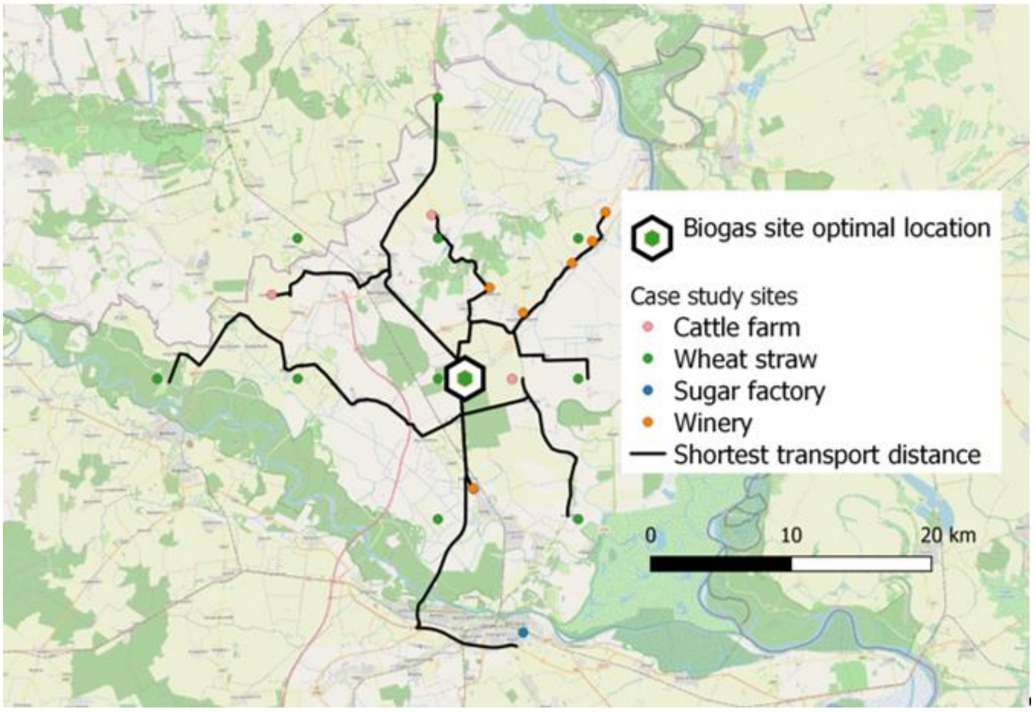


Figure 5 Transport road route and optimal biogas site location

In accordance with the resulting GIS layer (Figure 5), the P-graph representation and the maximal structure are developed. The data set obtained with QGIS includes biogas potential and transport distance for 18 feedstock-providing sites. Table 4 lists sites included in the P-graph representation and the transport distance between each feedstock-providing site and biogas site.

Table 4: P-graph legend and transport distance

Site	Cattle farm				Winery				
Abbreviation	CF1	CF2	CF3	CF4	W1	W2	W3	W4	W5
Distance (km)	9	16	21	8	8	8	17	15	19
Site	Sugar factory	Wheat straw collection site							
Abbreviation	SF	WS1	WS2	WS3	WS4	WS5	WS6	WS7	WS8
Distance (km)	26	32	17	18	24	14	1	12	16

The P-graph representation of the maximal structure of the case study is represented in. As described in the method, the material nodes are represented by raw materials (manure, industrial by-products and agricultural residues) and the final product (biogas). Operating unit nodes are representing anaerobic digestors.

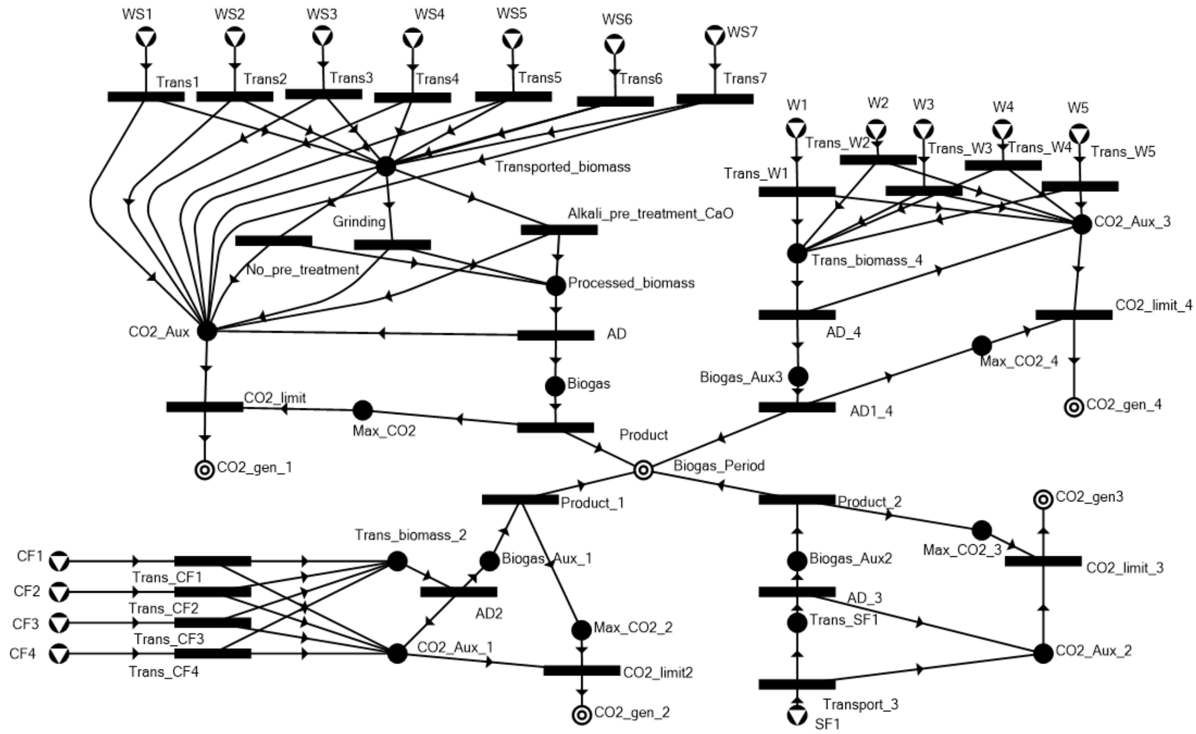
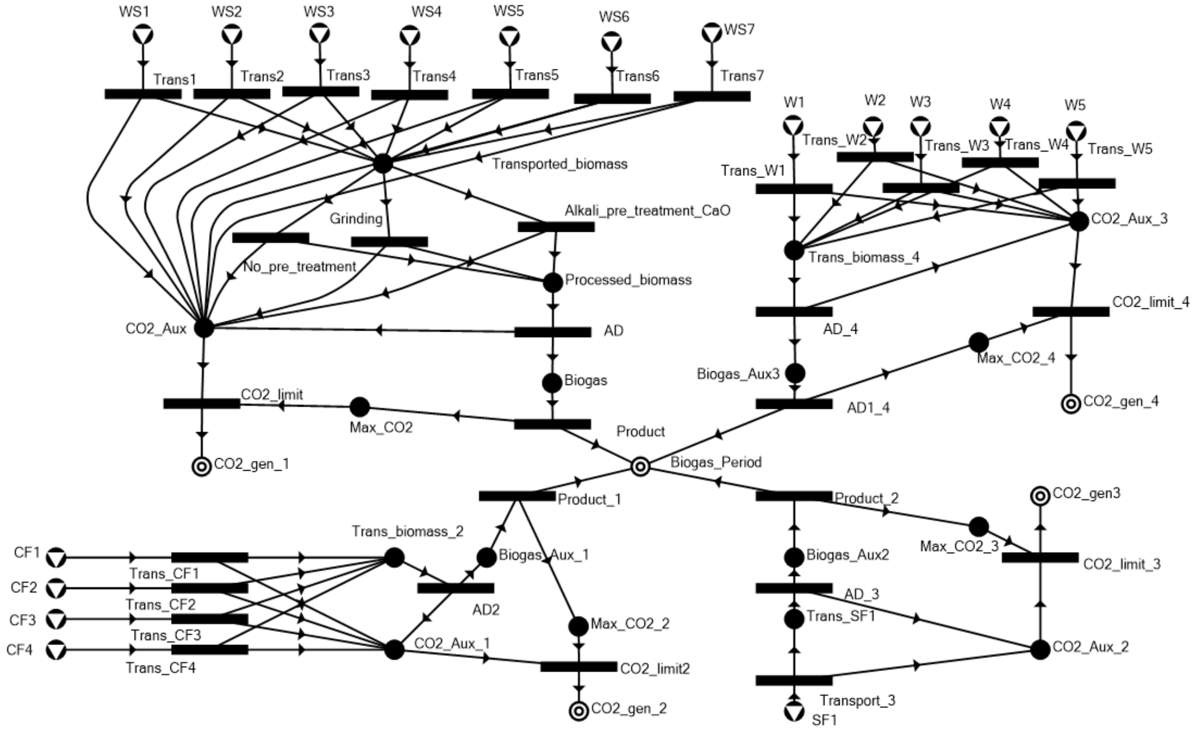


Figure 6 P-graph representations of the maximal structure of the case study

Four groups of feedstock providers can be recognised in the maximal structure. Those are wheat straw collection sites (upper left corner), cattle farms (bottom left corner), sugar factory (bottom right corner) and wineries (upper right corner). As can be seen in Figure 6



, all biomass streams lead to one final product (biogas), as this paper considers one site as a biogas production site, while CO_2 streams are set to define CO_2 generated by each feedstock group.

As described in the Method section, based on maximal structure, all feasible structures were defined. For the optimal solution, the objective was to minimise the cost of the biomass supply network and to limit the associated GHG emissions below the given threshold. This was done for two cases, both having the required biogas production of 120,000 GJ/y, but in the first case the optimisation is performed on the annual level, while in the second case, the multiperiod approach was implemented to include the seasonal variation of feedstock supply. The biogas production of 120,000 GJ corresponds to the production of anaerobic digestors which deliver biogas to CHP with 1.5 MW_{el}.

The optimal structure for annual biogas production of 120,000 GJ/y is presented in Figure 7.

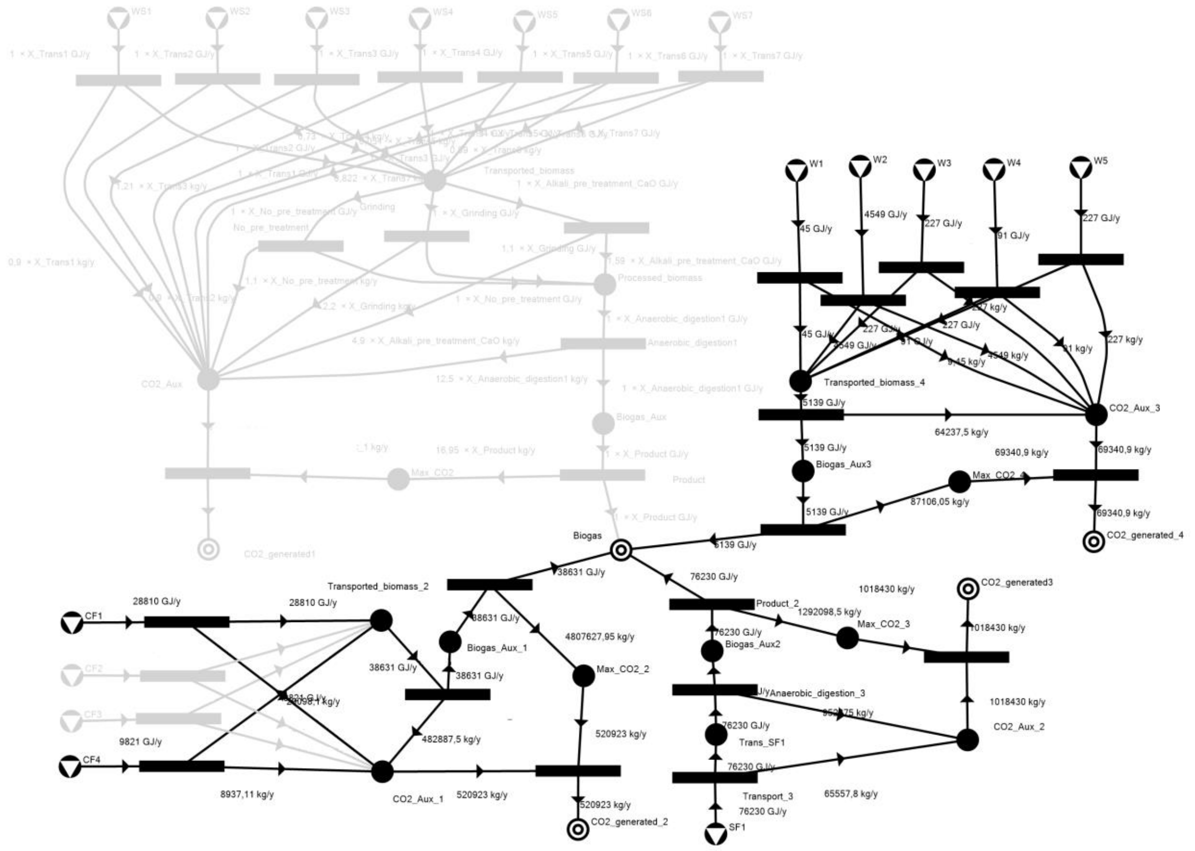


Figure 7 The optimal structure for annual biogas production of 120,000 GJ/y

The cost of the biomass supply network (including feedstock and transport costs) is 292,016 EUR. This equals 2.43 EUR/GJ. The data from the optimal structure are presented in Table 5, to improve the visibility of the numbers presented in Figure 6.

Table 5 The optimal structure for annual biogas production of 120 000 GJ/y

Abbreviation	CF1	CF4	SF1	W1	W2	W3	W4	W5
Delivered feedstock (GJ)	28,810	9,821	76,230	45	4,549	227	91	227

As can be seen from the results, the model first selects the wineries and sugar factory, after that cattle farms and finally wheat straw. It is interesting to see that the model would select sugar beet pulp prior to the manure from the further farms, as feedstock cost is lower for manure. The reason for this selection is the relatively low bulk density of the biogas potential of manure, compared to the bulk density of a biogas potential of sugar beet pulp. Hence, higher transport may surpass the difference in feedstock cost.

GHG emissions linked to each stage of the production and use of biogas are assessed. Contribution to GHG emission generation is presented in Table 6 by each feedstock group, as well as the GHG savings compared to the fossil fuel comparators (for both heat and electricity).

Table 6 GHG emission generation- case 1 (biogas production 120,000 GJ)

Feedstock	Wheat straw	Manure	Sugar beet pulp	Grape pressings
Biogas produced from feedstock (GJ)	-	38,631	76,230	5,139
Associated GHG emissions (kg CO_2eq)	-	520,923	1,018,430	68,769
Associated GHG emission savings (kg CO_2e)	-	4,143,757	-	-
Neto GHG emissions	-	-3,622,834	1,018,430	68,769
Specific GHG emissions (kg CO_2eq/GJ)	-	-93.8	13.36	14
GHG savings compared to fossil fuel comparator for heat, Case 1, closed digestate	-	210.70%	84.25%	83.49%
GHG savings compared to fossil fuel comparator for electricity, Case 1, closed digestate	-	165.35%	90.70%	90.26%

As can be seen from the specific GHG emissions presented in Table 6, GHG emissions are below the threshold (which is set to 16.95 kg CO_2/GJ biogas), which can be considered as a confirmation that the developed model presented as feasible structures only those which fulfil GHG savings.

Integration of GHG emissions limitation, in line with Directive 2018/2001, represents an added value and a step beyond the current state of the art in P-graph optimisation. To enhance the understanding of GHG emission limitation and improve the visibility of Figure 6, part of the PNS network (for the case of optimal structure) whose function is to limit GHG emission is presented enlarged in Figure 8.

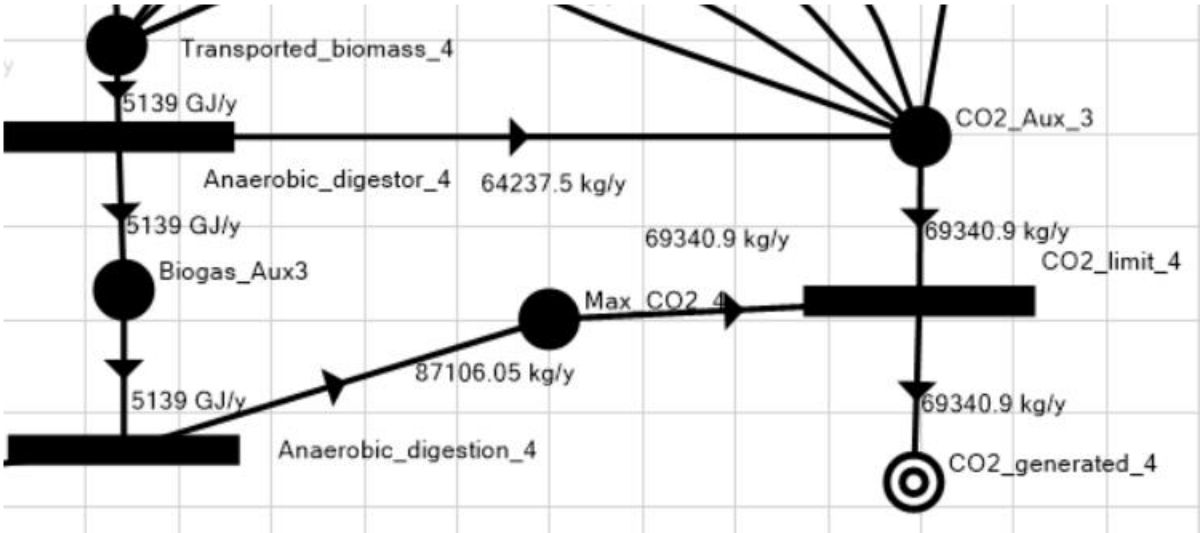


Figure 8 GHG emission limitation in PNS network (optimal structure)

For the given example, the selected feedstock group is grape pressings. CO_2_Aux is an auxiliary node represented as an interim product, whose main objective is to summarise GHG emissions that occur in biogas production and use lifecycle. This value equals 69 340.9 kg/y in Figure 8. The obtained value is then compared with the maximal allowed GHG emissions. The maximum allowed GHG emissions are calculated based on the biogas generation (GJ), obtained from the auxiliary node Biogas_Aux, which is multiplied by the specific limitation of GHG emissions per GJ of biogas. This value equals 87106.05 kg/y in Figure 8. Those two values meet at note Max_CO_2. For the case where GHG emissions that occur in a biogas lifecycle are higher than the maximum values, the model makes a new iteration and searches for a new economically optimal structure whose GHG emissions are below the given limit.

To enhance the accuracy of the results, the seasonal aspect of biomass production was integrated into the model. To integrate this, a year was divided into several periods, each representing certain months. The list of periods, corresponding months and generated types of biomass in a specific period is listed in Table 7.

Table 7 The list of periods with corresponding biomass generation

Period/ month	**1/ January-May**	**2/ June-July**	**3/ August**	**4/ September**	**5/ October-November**	**6/ December**
Wheat straw	NO	YES	NO	NO	NO	NO
Manure	YES	YES	YES	YES	YES	YES
Grape pressings	NO	NO	NO	YES	NO	NO
Sugar beet pulp	NO	NO	NO	YES	YES	NO

For each considered period, the assumption is used that biomass available for biogas production is the one generated in the specific period (months). There is a threefold reason for this. The first one is that some of the considered feedstock cannot be stored for a longer period of time, due to potential changes in feedstock conditions, which could result in the adverse performance of biogas production. The second is that seasonal feedstock storage may result in additional methane emissions generated during the storage period, which could result in exceeding the threshold of GHG emissions, due to the high global warming potential of methane. The final one is the cost of the investment and maintenance of the seasonal storage.

For the considered biogas production, in case the required biogas production exceeds the biogas potential contained in the biomass, the assumption was used that this gap will be covered with the wheat straw, due to favourable storage properties. Required biogas production, for the case of the annual production of 120, 000 GJ is presented in Table 8.

Table 8 Required biogas production in the concerned periods

Period/ month	**1/ January-May**	**2/ June-July**	**3/ August**	**4/ September**	**5/ October-November**	**6/ December**
Required biogas production (GJ)	55,848	19,029	0	11,096	22,561	11,466

As can be concluded from Table 8, annual maintenance of the biogas site is scheduled for August. The optimal structure for each period is presented in Figure 9, Figure 10, Figure 11, Figure 12 and Figure 13.

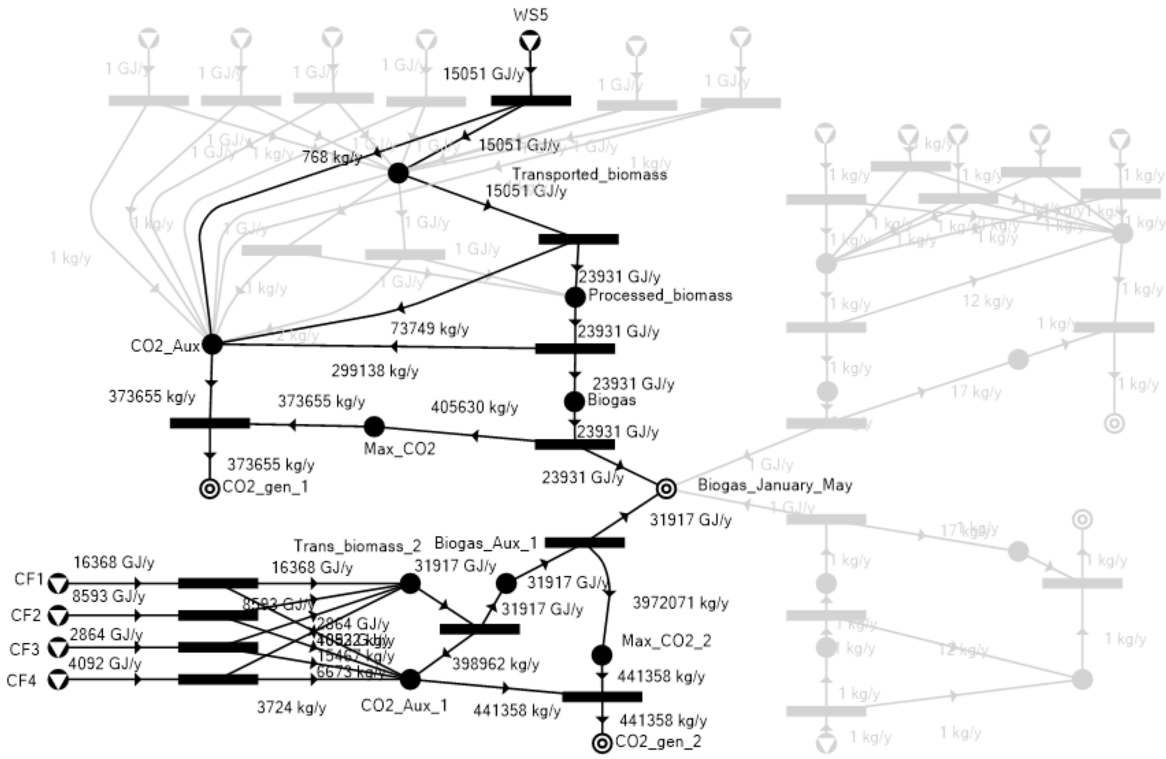


Figure 9 Optimal structure of biogas production from January until May

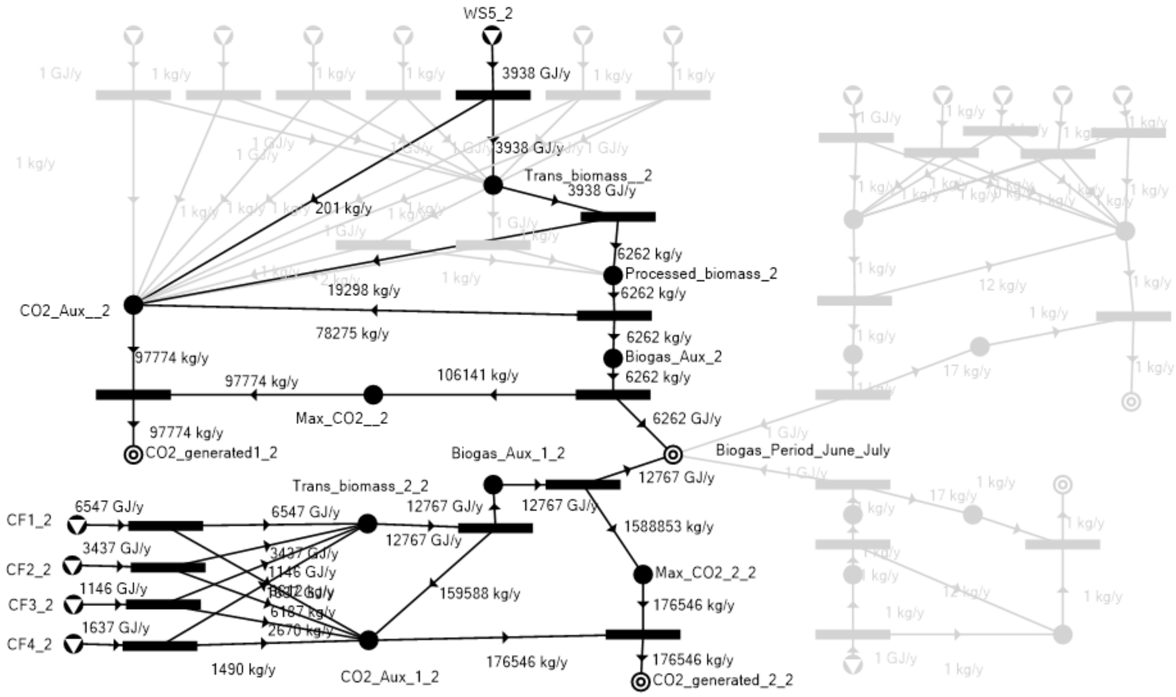


Figure 10 Optimal structure of biogas production in June and July

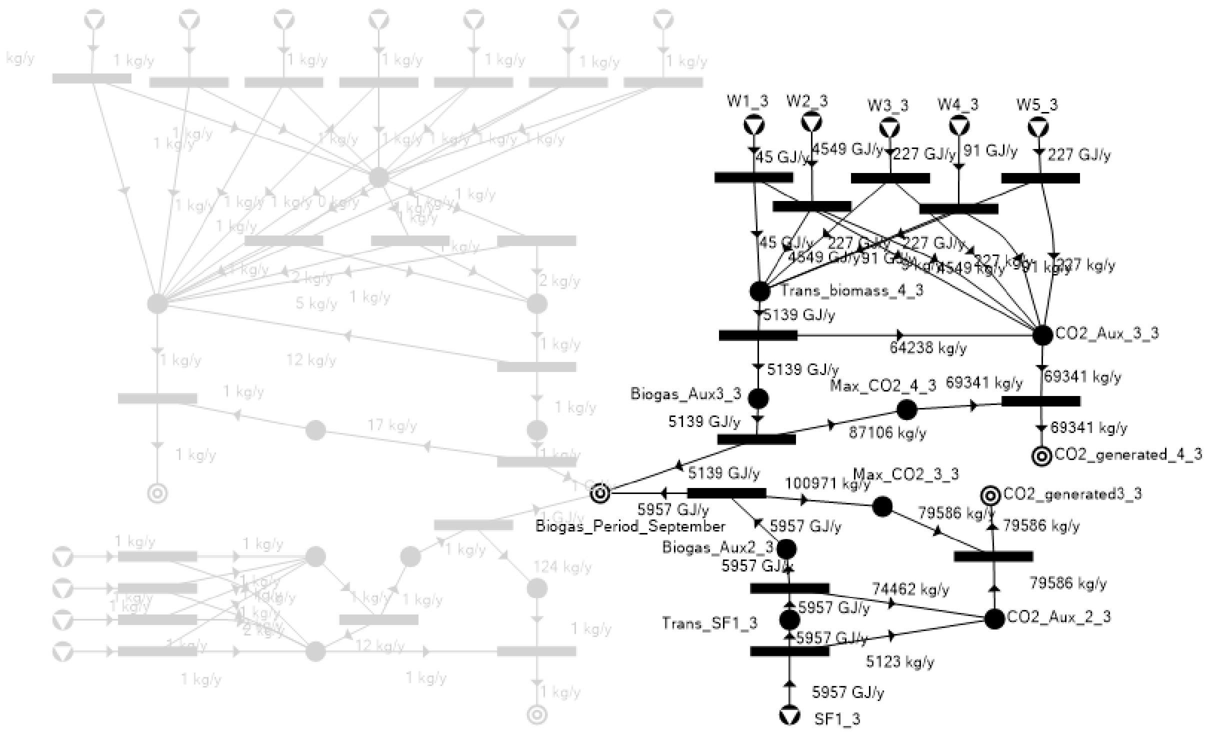


Figure 11 Optimal structure of biogas production in September

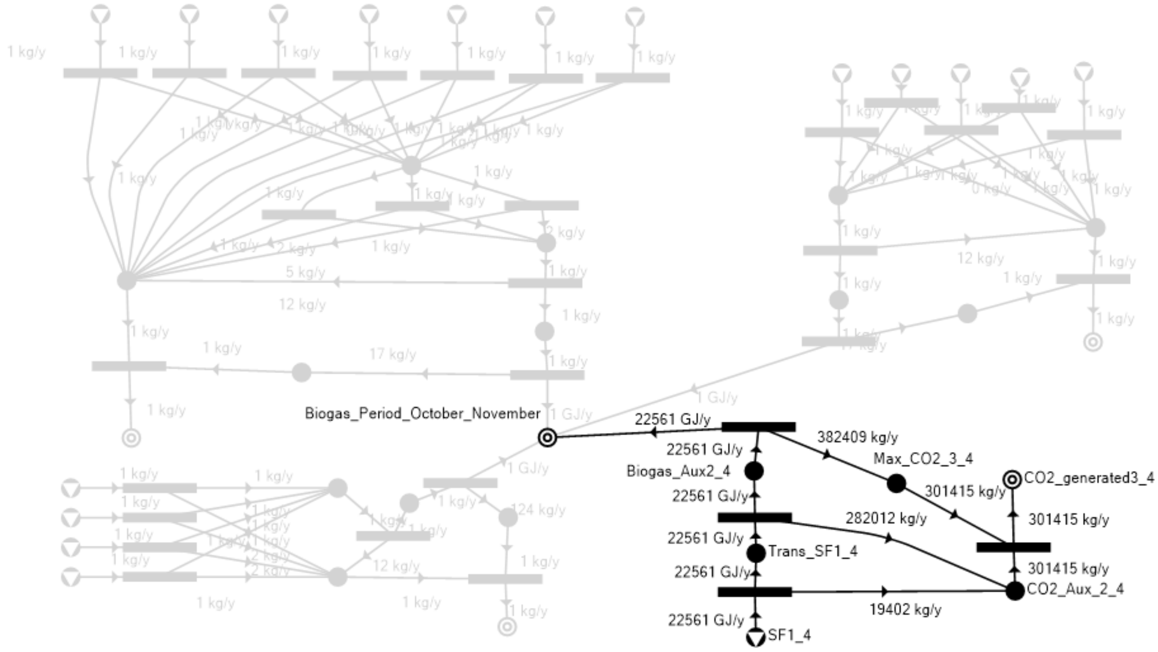


Figure 12 Optimal structure of biogas production in November and October

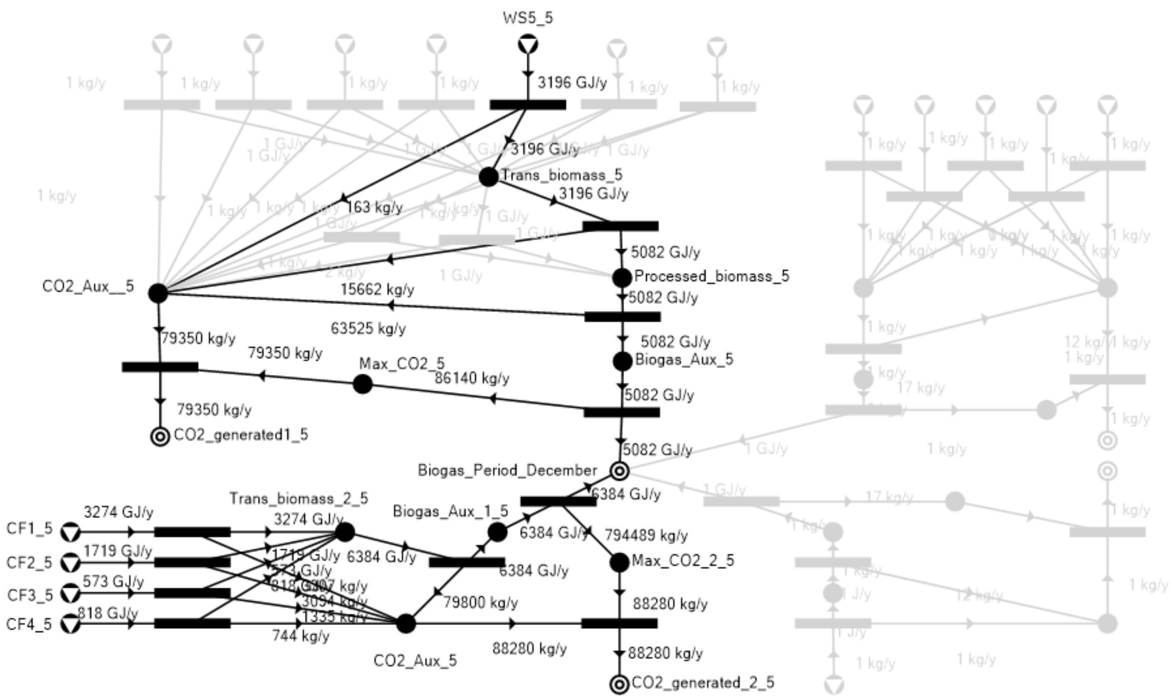


Figure 13 Optimal structure of biogas production in December

The values on delivered feedstock presented in optimal structures (Figure 9-Figure 13) provided for each period are shown in Table 9, to improve the visibility of the numbers.

Table 9 The summary of optimal structures determined for each considered period

	Delivered feedstock (GJ)						
	January-May	**June-July**	**August**	**September**	**October-November**	**December**	**Total**
WS5	15,051	3,938	-	-	-	3,196	22,185
CF1	16,368	6,547	-	-	-	3,274	26,189
CF2	8,593	3,437	-	-	-	1,719	13,749
CF3	2,864	1,146	-	-	-	573	4,583
CF4	4,092	1,637				818	6,547
SF1	-	-	-	5,957	22,561	-	28,518
W1	-	-	-	45	-	-	45
W2	-	-	-	4,549	-	-	4,549
W3	-	-	-	227	-	-	227
W4	-	-	-	91	-	-	91
W5	-	-	-	227	-	-	227

The optimal structures of periods from January-May, June-July and December are very similar. Although in the second period, there is a significant biogas potential of the wheat straw sites, the model will select manure, even from further farms. The total values of delivered feedstocks significantly differ from the first case where the optimal structure was defined on an annual basis. Compared to the first case where the model did not commit the wheat straw sites, in the second case wheat straw contributes to 18.3 % of biogas production. Furthermore, in the second case, the contribution of manure is significantly (34%) higher, even from the more distant farms, which would result in higher transport costs. On the other hand, most of the potential of sugar beet pulp was untapped in the second case (63%). Those chances negatively affected the total cost of the biomass supply network, which equals 733,684 € (6.11 €/GJ biogas). The increase in biomass supply cost is, to some extent, an expected result, as during the periods in which feedstock with high energy density and low prices were not available, the model committed the sites with higher transport distances and/or sites with higher feedstock and processing costs. Furthermore, although the multi-period approach resulted in less favourable results in terms of cost, it can be stated that this approach results in more accurate results and provides insights into the sensitivity of the cost of biomass supply network for the case where economically favourable feedstocks are not available, since they are being generated in a very short period of time during the year.

As for the first case, the contribution to GHG emission generation, as well as GHG savings compared to fossil fuel comparators, is presented in Table 10 by each feedstock group.

Table 10 GHG emission generation

Feedstock	**Wheat straw**	**Manure**	**Sugar beet pulp**	**Grape pressings**
Biogas produced from feedstock (GJ)	35,275	51,068	28,518	5,139
Associated GHG emissions (kg CO_2eq)	550,778	706,184	381,001	69,341

Associated GHG emissions savings (kg CO_2eq)	-	5,477,813	-	-
Net GHG emissions (kg CO_2eq)	550,778	-4,771,628	381,001	69,341
Specific GHG emissions (kg CO_2eq/GJ)	15.61	-93.44	13.36	13.49
GHG savings compared to fossil fuel comparator for heat, Case 1, closed digestate	89.12%	210.22%	84.25%	84.08%
GHG savings compared to fossil fuel comparator for electricity, Case 1, closed digestate	81.57%	165.06%	90.70%	90.60%

As in the first case, specific GHG emissions were below the given threshold, which can be considered as a confirmation that the model successfully limits the GHG emissions in both single-period optimisation and multi-period optimisation. It is also interesting to note that, due to the higher contribution of manure to biogas production, the net GHG emissions are significantly lower for the second case. As mentioned earlier, when defining the optimal structure, P-graph Studio defines and ranks sub-optimal structures as well. Hence, the results could be used for the development of a Pareto front that would define both the cost of the structure and the generated GHG emissions.

The developed model does not automatically prioritize the structures with the lowest GHG emissions, as it considers GHG emissions savings as constraints, not as the variable to be minimised. Although this may be considered as the limitation of the model, the minimisation of the GHG emissions was not selected as the target group of this model is the biogas industry, whose objective is commonly to fulfil the requirements given by the legislation and to minimise the cost of the biomass supply network. However, in case if biogas industry would receive some additional incentive to future reduce GHG savings, or in general decides to achieve savings higher than the given threshold, the developed model easily allows the comparison of the cost and GHG emissions of optimal and sub-optimal structures, thus enabling efficient assessment of trade-offs. Based on the obtained results, it can be considered that for the case of the minimisation of GHG emissions, the model would prioritize manure as the feedstock (even from more allocated farms), due to the high GHG emission savings resulting from the improved manure management.

As the developed model determined economically optimal and sub-optimal structures, simultaneously limiting GHG emissions in both single-period optimisation and multi-period optimisation, it can be stated that the hypothesis of this work is confirmed.

6. Conclusions

This paper presents a novel multi-period P-graph-based model for optimizing biomass supply networks, which goes a step further in integrating environmental constraints in the PNS network. The model developed in this work enables the economical optimisation of a biomass supply network, while simultaneously limiting the CO_2 emissions that the biomass supply network can generate, in line with the EU Directive 2018/2001 requirements. Furthermore, through the extension of the model to multi-periods, the developed model considers the seasonality of biomass supply during the year. The study also demonstrates the linkage between GIS mapping and route assessment with the graph theory approach for biomass supply chain

optimization. The presented model was applied to the biogas production from agricultural residues, livestock and industrial by-products, including wheat straw, sugar beet pulp, grape pressings and manure.
The model was tested in a case study of a rural area in Osijek-Baranja county, which resulted in the determination of optimal and sub-optimal economical structures that fulfil GHG savings requirements. The results indicate that the model prioritizes feedstock from wineries and sugar factories, followed by cattle farms and wheat straw. Moreover, the specific cost of the biomass supply network (feedstock, processing and transport) was calculated to be 2.62 EUR/GJ in the case of optimisation on an annual level and 5.1 EUR/GJ in the case of multi-period optimisation, due to higher demand feedstocks with higher transportation, processing and/or feedstock cost. Overall, the paper confirms the hypothesis that an economically optimal residual biomass supply network for biogas production that meets sustainability and greenhouse gas emissions saving criteria can be determined with the P-graph approach. Future research could consider expanding the scope of the analysis by incorporating a wider range of feedstock varieties.

Acknowledgements

This work has been financially supported by the Croatian Science Foundation. The authors would like to thank to Jean Pimentel from the Budapest University of Technology and Economics for his support in P-graph model development.

List of Abbreviations

ABB- Accelerated Branch and Bound
AD-Anaerobic digestion
CHP- Combined heat and power
EC-European Commission
EU- European Union
GIS -Geographic Information System
GHG-Greenhouse Gas
ILUC -Indirect Land-Use Change
MILP-Mixed-Integer Linear Programming
MSG- Maximal Structure Generation
PNS -Process-Network Synthesis
SSG-Solution structure generation

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PAPER 5

P-Graph approach for the optimisation of biomass supply network for biogas production in urban areas

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The utilization of anaerobic systems for biogas production integrates various aspects such as renewable energy generation, waste management, waste treatment, and biofertilizer production. This study introduces a model that focuses on the economic optimization of a biomass supply network for biogas production in urban areas. The selected feedstocks considered in the model are biowaste and residues sourced from restaurants, shops, and the food and beverage industry.

This study introduces two significant advancements. Firstly, it employs an enhanced GIS-based approach that integrates greenhouse gas (GHG) requirements by incorporating a maximal allowed transport distance. This integration aims to achieve minimal GHG savings from biogas usage. These GHG-based requirements align with the specifications outlined in Directive 2018/2001, which promotes the use of renewable energy sources and stipulates a minimum 80% reduction in greenhouse gas emissions from biogas plants operating from 2026, in addition to meeting environmental sustainability criteria. Secondly, the study introduces a novel approach that combines GIS mapping of biomass potential with a P-graph framework for optimizing the biomass supply network. This integration facilitates comprehensive and efficient optimization of the network for biogas production.

The model is developed and solved using P-Graph Studio, while feedstock availability and transportation distances are determined using the QGIS tool. The approach is tested under two scenarios: one with an annual production of 36,000 GJ and another with an annual production of 72,000 GJ. The p-graph approach enables the identification of the optimal economic solution for both scenarios. As the most of the biogas potential is concentrated in a single brewery, the specific cost of the biomass supply network, including feedstock and transport, remains comparable for both scenarios, with values of 12.44 EUR/GJ and 12.61 EUR/GJ for the second case.

1. Introduction

Renewable energy sources are recognised as crucial for the transition towards climate neutrality and the replacement of fossil fuels. Biogas is used as a source of heat, electricity, and transportation fuel and is becoming increasingly popular due to its ability to reduce greenhouse

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gas emissions, conserve resources, and provide a sustainable energy source. Biogas and biomethane production is steadily increasing over the last two decades [1]. Due to high yield and ease of cultivation and storage, maize silage is commonly used as a feedstock for biogas production. However, socio-economic concerns are raised due competition with land use, competition with food and feed production, as well as in the increase of the price of maize silage. Those concerns are reflected in Directive 2018/2001 which outlines several constraints for biogas production to ensure its sustainability and not having impacts on the environment and society. Among other limitations, the directive specifies that the GHG savings from the use of biomass for electricity, heating and cooling production should be at least 70% for installations starting operation from 2021 until the end of 2025 and 80 % for installations starting operation from 2026 [2]. Furthermore, the European Commission introduced a cap on food and feed crops toward the EU renewable objective, starting at 7% in 2021 and rapidly decreasing to 0% in 2030, to reduce the implications of Indirect Land-Use Change (ILUC). The given requirements, but also the increase in the price of the maize silage foster the need to shift towards sustainable alternatives of biogas feedstocks. Some examples of sustainable alternatives for maize silage are by-products from industry, supermarkets, fast food restaurants, agricultural residues and organic fractions of municipal biowaste. Those feedstocks are characterized by low energy density and in some cases, scattered feedstock generation. Hence, biomass potential assessment is an important step in the biomass-based analysis. The use of a Geographic Information System (GIS) for biomass potential mapping is recognised as beneficial as it offers several benefits, including improved decision-making due to enhanced analysis and visualization of data related to biomass potential, available infrastructure, biomass potential density analysis, identification of most promising locations for biomass processing (biogas site) and many others. In the literature, there are numerous papers that prove the benefits of GIS utilization for biogas production mapping and the untapped potential of alternative feedstocks for biogas production. Some of the most recent advancements are presented in the next paragraph.

In their recent paper, Romero et al. [3] integrated fuzzy logic with GIS to define suitable locations for a potential biorefinery implementation. Ukova [4] et al. used the GIS approach for assessing the biomass energy potential and identification of appropriate biomass conversion technologies in Nigeria. In their work, Rhofita et al. [5] performed a GIS mapping analysis of the biomass potential of agricultural and forest residues in Indonesia. Similarly to this, Chakraborty et al. [6] developed a GIS map of crop residue potential for energy utilization in biomass/biofuel power plants.

The aforementioned studies have shown that biomass may provide a significant contribution to the transition towards renewable energy solutions. However, the cost of biomass supply networks and technologies to convert biomass into useful forms of energy is often a barrier to increased utilization of biomass for biogas production. To address this barrier, significant research efforts are being made. It has been noted that graph theory methods are increasingly used in supply network optimisation problems. In mathematics and computer science, graph theory is the study of graphs, mathematical structures used to model pairwise relations between objects from a certain collection. Process-graph (or P-graph) is a unique bipartite graph representing the structure of a process system [7]. P-graph optimization can be applied to a wide range of domains and problem types. The utilization of the P-graph approach brings forth several advantages, including the unambiguous representation of decision alternatives, the generation of a mathematical model through algorithms, a decrease in solution procedure complexity, and the ability to derive multiple alternative solutions. It has been successfully used in various areas, including scheduling and resource allocation problems, task and data parallelism, parallel algorithm design, and optimization of parallel computing systems. Its versatility makes it a valuable tool for addressing different optimization challenges across different disciplines. Adonyi et al. [8] applied a p-graph framework for the optimisation of the maintenance schedule for public transportation buses. Similar to this, Bartos et al. [9] implemented a p-graph approach for the optimisation of a production line in the assembly industry. Tan et al. [10] developed a P-graph model for the synthesis of hydrogen networks. The developed model included direct reuse/ recycle and regeneration schemes. Ji et al. [11] developed a P-graph model for the optimization of hydrogen and battery energy storage. The model he developed used a multi-period modelling approach to reduce and compare costs of those two storage systems.

P-graph application is especially interesting for biomass supply network optimization problems. How et al. [12] developed a decomposition approach for a p-graph application of synthesis of multiple biomass corridors. Stile et al [13] have expanded the use of P-graph-based algorithms to assess the reliability of raw material availability. Malladi et al. [14] have developed a p-graph-based decision support tool for optimizing the short-term logistic of forest-based biomass, by minimizing the biomass logistics cost. Egieya et al [15] used a P-graph framework to optimise the integrated biopower supply network, by maximizing the economic performance. Lo et al. [16] proposed a P-graph based method that considered the incorporation of biomass supply chain uncertainties. Results have shown that a reduction in net present value (NPV) ranges from 1.39% to 12.21% when the biomass shortage scenario was included. Ondruška et al. [17] extended the application of the P-graph approach to perform resource optimization in an aquaponics facility.

Interest in biomass supply network optimization is evidently increasing. However, to exploit the potential of the waste materials for biogas production, it is crucial to link the availability of biomass and its geographical distribution, with the optimization of the economical performances of a biomass supply network. Furthermore, a limitation requested by the EU legislation [2] should not be neglected in this process and mathematical models developed for potential assessment and biomass network optimization should integrate limitations regarding the minimum GHG savings, compared to fossil fuel comparator. To address this research gap and integrate these crucial factors for the successful real-life application of waste materials for biogas production, this paper provides the following novelty:

- enhanced GIS-based approach that integrates GHG requirements in terms of maximal allowed transport distance (to achieve minimal GHG savings from the use of biogas);
- a novel approach that combines GIS mapping of biomass potential and a P-graph framework for biomass supply network optimization.

2. Method

The first part of this method is focused on conducting a GIS mapping of biomass potential. It is an important part as it enhances understanding of the availability of feedstocks for biogas production, its geographical dispersion and the transport distance between feedstock providers and biogas sites. Within the GIS mapping, an evaluation of feedstock which fulfills GHG savings of 80% will be conducted. The results of this part of the research will be used for p-graph-based optimisation of a biomass supply network, which represents the second part of this research. The method is presented in the flowchart in Figure 1 and described in the subsection below.

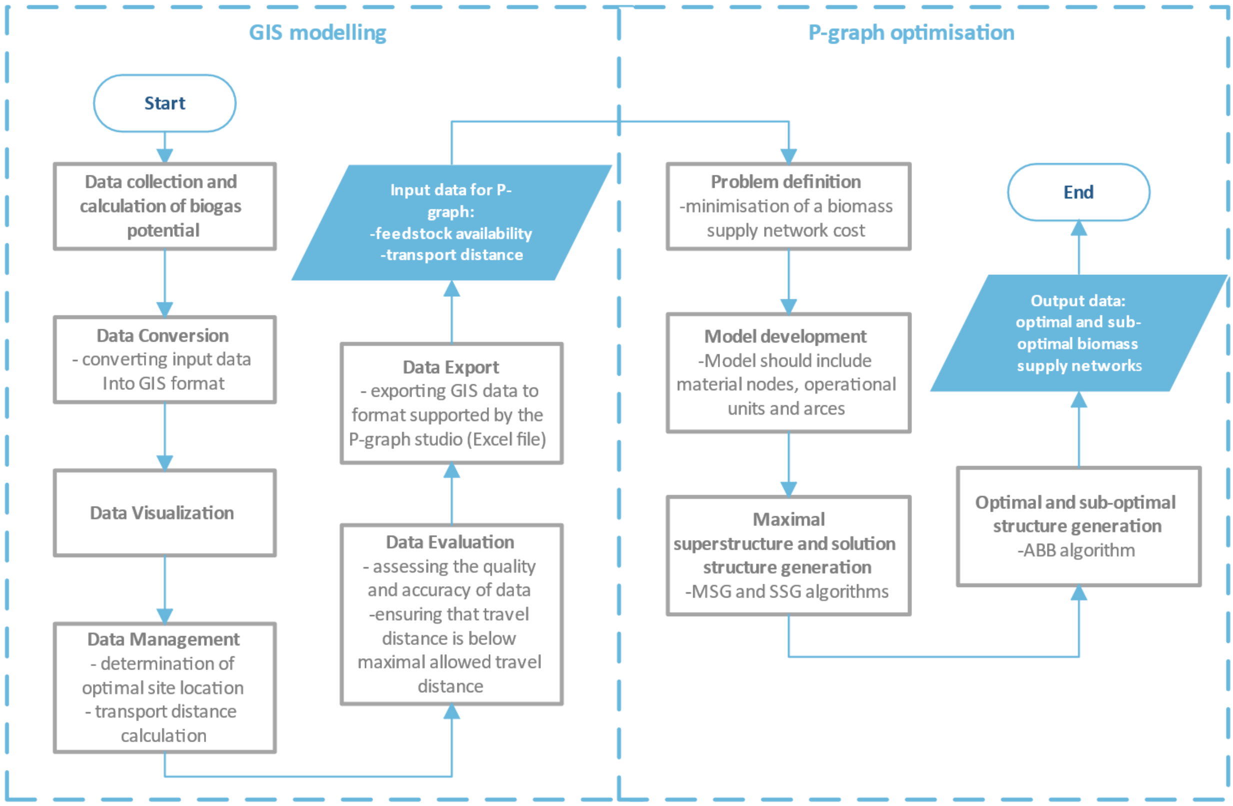

Figure 1 Flowchart representation of the method

As described in the Introduction, the method novelties of this paper include integration of GHG requirements in terms of maximal allowed GHG emissions from transport (to achieve minimal GHG savings from the use of biogas) in the GIS modelling step, as well as linkage of GIS mapping of biomass potential and a P-graph framework for biomass supply network optimization.

2.1 GIS potential assessment

The process of GIS potential assessment involves data collection, data conversion, data analysis, data visualization, and data management. The steps of this process can be tailored to meet the specific needs and requirements of different types of requirements.

2.1.1. Collection of data on biogas potential

The first step in using GIS for biomass mapping is to gather data on the potential biomass resources in the area of interest. For the urban areas, the analysed feedstock can include biodegradable waste from supermarkets, fast food restaurants and organic fraction of waste from industry, among others. This step includes identifying and quantifying the types, amounts and sources of feedstock eligible for biogas production generated in a specific area. In this step, feedstocks eligible for biogas production are determined and their respective biogas potential is assessed. In the scope of this work, the following waste materials and by-products are considered:

- Industry biowaste
- Oil and fat
- Spent grain.

The required data could be collected from waste registers, statistic registers, environmental impact assessment reports, etc. Automatised collection of information on locations of supermarkets, fast food restaurants and industries is possible via Quick OSM Plugin, which is available in the QGIS tool [18]. This Plugin enables the export of data from the Overpass server [19] that integrates data from the geographic database Open Street Maps [20].

2.1.2. Data Conversion

Once the data has been collected, it must be converted into a format that can be used in a GIS. This typically involves converting the data into a digital format, such as a shapefile or a raster image, that can be imported into the GIS software. As the considered feedstocks occur at specific locations (in industry, fast food restaurants, etc), data conversion refers to the process of taking a physical address, such as a street address and postal code and converting it into a set of geographic coordinates, such as latitude and longitude. The resulting geographic coordinates are used to locate and map addresses on a digital map.

2.1.3. Data Visualization

The results of the data conversion can be visualized using a GIS. This includes creating maps that show the distribution of biomass resources, where different colours and different sizes indicate the potential of different feedstock providers. A more detailed description of GIS mapping is provided in the authors' earlier papers [21], [22].

2.1.4. Data Management

Data collected, converted to GIS format and visualized in previous steps can be future managed to obtain the required input data for P-graph optimisation. For the cases of biogas utilization, optimal biogas site location is insightful information. Optimal biogas site location selection in GIS involves evaluating proximity to potential biogas sources. However, in urban areas, the location of a biogas plant is often constrained by the General urban plan. Therefore, for urban locations, it is advisable to set a location next to an existing landfill or composting plant. Based on the location of biogas providers and the location of the biogas site, transport distance can be assessed. This can be done via the "Shortest path" query in QGIS. The shortest path in QGIS is a route-finding analysis that determines the quickest or shortest path from a starting point (feedstock providers) to an endpoint (biogas site) through a network of interconnected lines, representing transport routes. Transport distance is an important element to consider for feedstock utilization, as it influences both associated GHG emissions and transport costs In the scope of this work, the transport distance is based on the shortest path. The transport distance is used to assess the transport cost, as presented in Equation (1):

$$C_{trans} = \frac{d\ (K_{full} + K_{empty})}{B_{biogas}} \times b \times\ T \tag{1}$$

Where C_{trans} represents specific transport cost (EUR/GJ), d transport distance (km), K_{full} fuel consumption of loaded truck (l/km), K_{empty} fuel consumption of an empty truck (l/km), b fuel price (EUR/l), B_{biogas} for biogas potential of transported feedstock (GJ) and T for transport cost correction factor. In this work, it was assumed that T equals 3, meaning that the cost of fuel is one-third of the total transport cost. This assumption is based on the calculation of the Joint Research Centre presented in their report "Estimating road transport costs between EU regions" [23]. When calculating the fuel cost, the average cost of diesel for the last two years was taken as the reference.

2.1.5. Data evaluation

GIS data evaluation is a critical step in the process of using geographic information system (GIS) technology. The evaluation process involves assessing the quality and accuracy of the data that is being used to create maps, perform analysis, or make decisions. This evaluation helps to ensure that the results of GIS analysis are accurate and reliable, and that the data being used is suitable for the intended purpose. It may also involve checking the data against other sources to validate its accuracy and identifying and correcting any errors or inconsistencies in the data.

As explained in the Introduction, GHG savings for biogas plants starting from 2026 must be at least 80% compared to fossil fuel comparator. The minimum GHG savings can be used for determining the maximal transport distance of feedstock for biogas transportation. To ensure that the transport distance is below the given limit, the transport distance should be evaluated for each considered site that provides feedstock. Additionally, is important to note that maximum transport distance differs for the different feedstock groups and should be calculated based on the Method defined in Directive 2018/2001. The evaluation of maximum transport distance is implemented in two steps. In the first step, the information on the maximum distance is associated with each considered feedstock group. The grouping of the feedstocks and comparison with the maximum travel distance can be implemented with "Select features by using an expression" and "Field calculator". The implementation of this step before the P-graph optimization eliminates the future need to include GHG limitation in p-graph optimization.

2.1.6. Data export

Data obtained via QGIS will be further as the input data for the P-graph optimization. Hence, GIS data should be exported to a data format supported by the P-graph studio, which is Excel file format. The first step for this is exporting data from QGIS to comma-separated values (CSV) file format, which is a plain text format that stores tabular data with each row representing a feature and each column representing an attribute. This process allows users to extract and transfer attribute data from spatial layers in QGIS for further analysis or sharing with other software or users. To export CSV to Excel, the obtained file can be opened directly in Excel and saved as an Excel file. Excel will provide step-by-step instructions during the import process, enabling to specify the delimiters, column formats, and other settings necessary for accurately interpreting and displaying the data from the CSV file.

2.2 P-graph

The P-Graph optimization method is a mathematical optimization technique that can be used to optimize the biomass supply network. The following is a method for using P-Graph optimization to optimize the biomass supply network:

2.2.1. Problem definition

The first step in using P-Graph optimization is to define the problem that needs to be solved. This involves identifying the objectives of the optimization, such as minimizing costs, maximizing profit, maximizing efficiency, and defining the constraints, such as available biomass resources, transportation capacity, and demand for biomass. In the scope of this work, the objective function is to minimize the cost of a biomass supply network.

2.2.2 Model development

The next step is to develop a mathematical model of the biomass supply network. This model should include all relevant variables, such as biomass production, transportation, and utilization, as well as the constraints and objectives. The P-Graph optimization method uses a graph-based representation of the network to model the relationships between the variables and the constraints. In this representation, material nodes are used to represent raw, interim and final materials, while operating unit nodes are representing transportation, processing, and anaerobic digestors. Anaerobic digesters are enclosed structures where the anaerobic break down of raw material (feedstock) takes place. Arcs in P-graphs are directed edges that represent relationships between nodes in a network graph. In a P-graph, arcs represent a one-way flow of resources, or dependencies between nodes and they can have different lengths, capacities, or costs associated with them. In this work, the model is being developed in the P-graph studio.

2.2.3 Maximal superstructure and solution structure generation

Once the model has been developed, it can be solved using P-graph-based algorithms. Algorithm Maximal structure generation (MSG) yields the maximal structure, i.e., the superstructure, for the Process Network Synthesis (PNS) problem. The generated superstructure incorporates each combinatorically feasible process structure. MSG algorithm is followed by the solution structures generator, known as algorithm SSG. The SSG exhaustively identifies all combinatorically feasible solution structures that satisfy five axioms. A detailed description of those axioms is described in the literature by professors F. Friedler and L.T. Fan [24], founders of the P-graph framework. Those feasible structures are used for future evaluation and optimization.

2.2.4 Optimal and sub-optimal structure generation

Finally, algorithm ABB (an accelerated branch and bound algorithm) will be used to generate the optimal structure together with a ranked list of suboptimal structures. The objective function here is to minimize the cost of a biomass supply network. Hence, the structure with the lowest cost to the biomass supply network is the optimal one.

3. Case study

The Croatian capital city of Zagreb served as the case study for the presented method. The locations considered for this case study are shown in Figure 2.

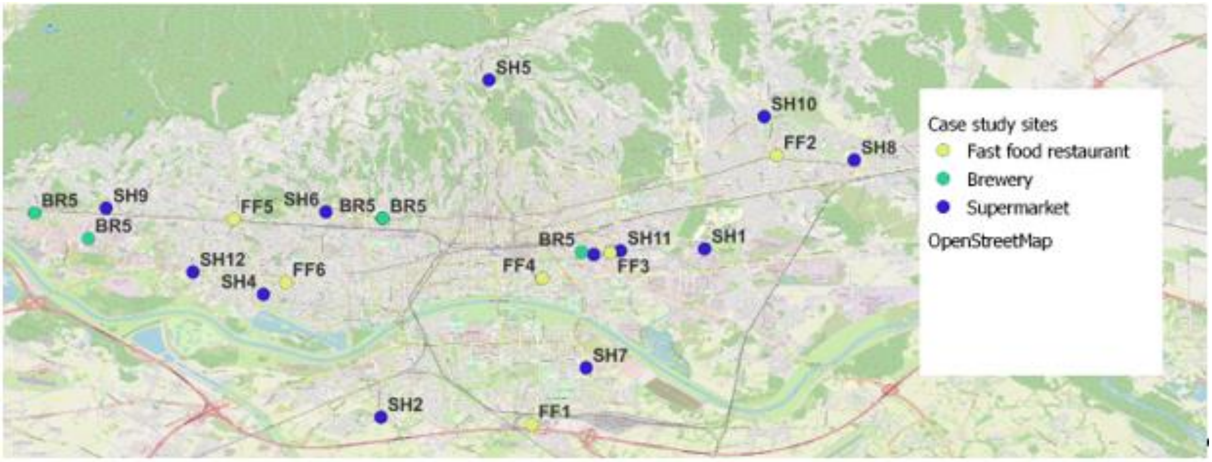

Figure 2 Case study Sites

The input data from Table 1 were applied to compute the cost of the feedstock.

Table 1 Specific feedstock cost

Feedstock	Cost (EUR/t)
Spent grain	33 [25]
Industry biowaste	0
Oil and fat	0

4. Results

According to the method stated in the Method section, the biogas potential from spent grain, industrial biowaste, oil, and fat was determined for the considered supermarkets, fast food chains, and breweries. The location of the biogas site was selected following the location of the existing composting plant and landfill. In accordance with the selected location, the transport distances between supermarkets, fast food restaurants, breweries and the biogas site were determined, as represented in Figure 3. The transport distance was calculated for each site that provides feedstock to the biogas plant. In this analysis, the assumption was made that trucks would be utilized for transporting the feedstock. The selected roads, determined as the shortest routes, are permissible for truck travel. Moreover, only feedstocks with transport distances below the maximum allowed distance (to achieve the necessary GHG savings) were considered for future evaluation.

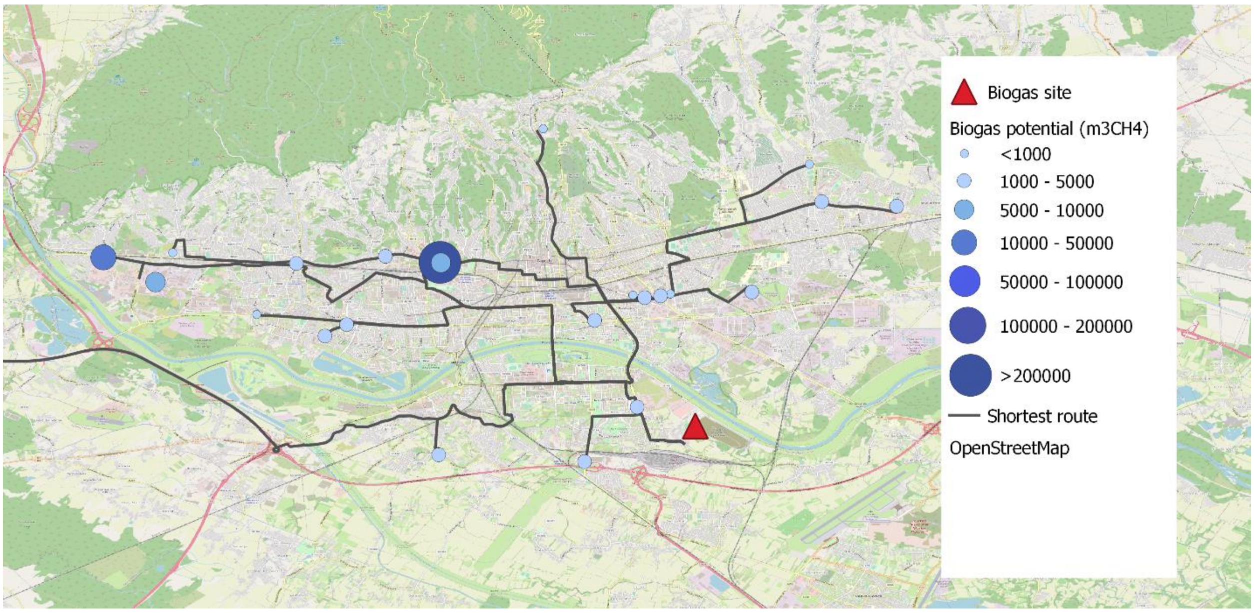

Figure 3 Transport road route and optimal biogas site location

As seen in Figure 3, biogas potential significantly varies between different feedstock-providing sites. For the considered case study, the greatest biogas potential comes from breweries. As explained in the Method section, GIS data represented in Figure 3 were converted to a format supported by P-graph Studio (Excel file) and used as the input data for P-graph-based optimisation The list of these sites, along with the respected abbreviation used for the P-graph representation and transport distance is presented in Table 2.

Table 2 P-graph, legend and transport distance

Site	Abbreviation	Distance (km)
Supermarket	SH1	9.4
Supermarket	SH2	10.5
Supermarket	SH3	33.4
Supermarket	SH4	12.9
Supermarket	SH5	12.2
Supermarket	SH6	11.8
Supermarket	SH7	1.9
Supermarket	SH8	15.2
Supermarket	SH9	17.6
Supermarket	SH10	14.4
Supermarket	SH11	7.3
Supermarket	SH12	14.3
Supermarket	SH13	6.7
Fast food restaurant	FF1	4.3
Fast food restaurant	FF2	13.5
Fast food restaurant	FF3	7.1
Fast food restaurant	FF4	7.5

Fast food restaurant	FF5	14.5
Fast food restaurant	FF6	12.4
Brewery	BR1	10.8
Brewery	BR2	18.5
Brewery	BR3	18.9
Brewery	BR4	10.8
Brewery	BR5	6.8

Figure 4 displays a P-graph representation of the case study's maximal structure. As described in the method, the material nodes are represented raw materials (feedstock) and the final product (biogas). Operational units are representing feedstock transportation. As this analysis assumes that the specific cost of anaerobic digestion will not differ between the considered feedstock materials, the cost of the anaerobic digestion was not a variable (and operating unit) included in the determination of the minimal biomass supply network cost.

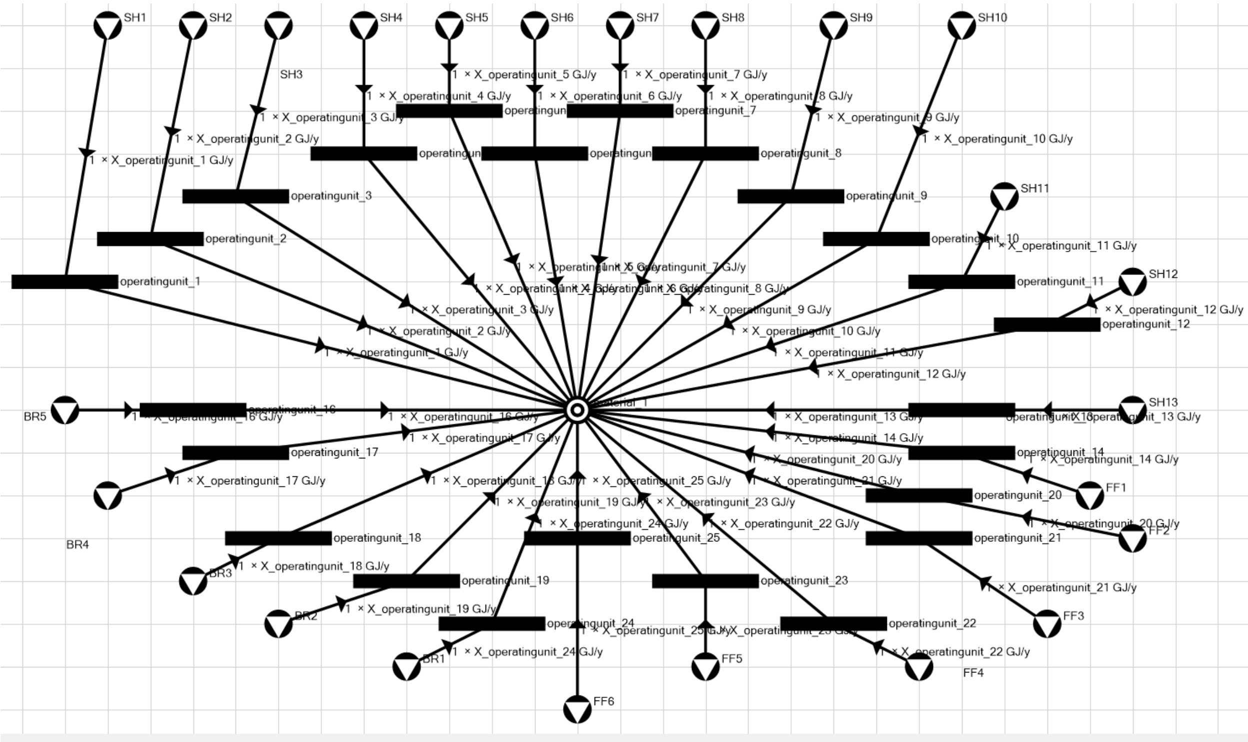

Figure 4 P-graph representations of the maximal structure of the case study

Based on the maximum structure and input data obtained from the GIS tool, optimal and suboptimal structures were defined. Here, the objective function is to minimise the cost of the biomass supply network. Additionally, it is important to note that the main purpose of this structure optimization is to compare the economic viability of the utilization of different biomass supply structures. Consequently, only the costs that differ between different biomass supply structures are included in this analysis.

The optimal and suboptimal structures were defined for two cases which correspond to the biogas production required to power a Combined Heat and Power (CHP) engine with electric power of 0.5 MW_{el} and 1 MW_{el}. For the first case, this corresponds to annual biogas demand of 36,000 GJ/y, and for the second to annual biogas demand of 72,000 GJ/y. The base for two demand-side scenarios is to quantify the influence on the price for the cases with different demands. The optimal structure for annual biogas production of 36,000 GJ/y is shown in Figure 5.

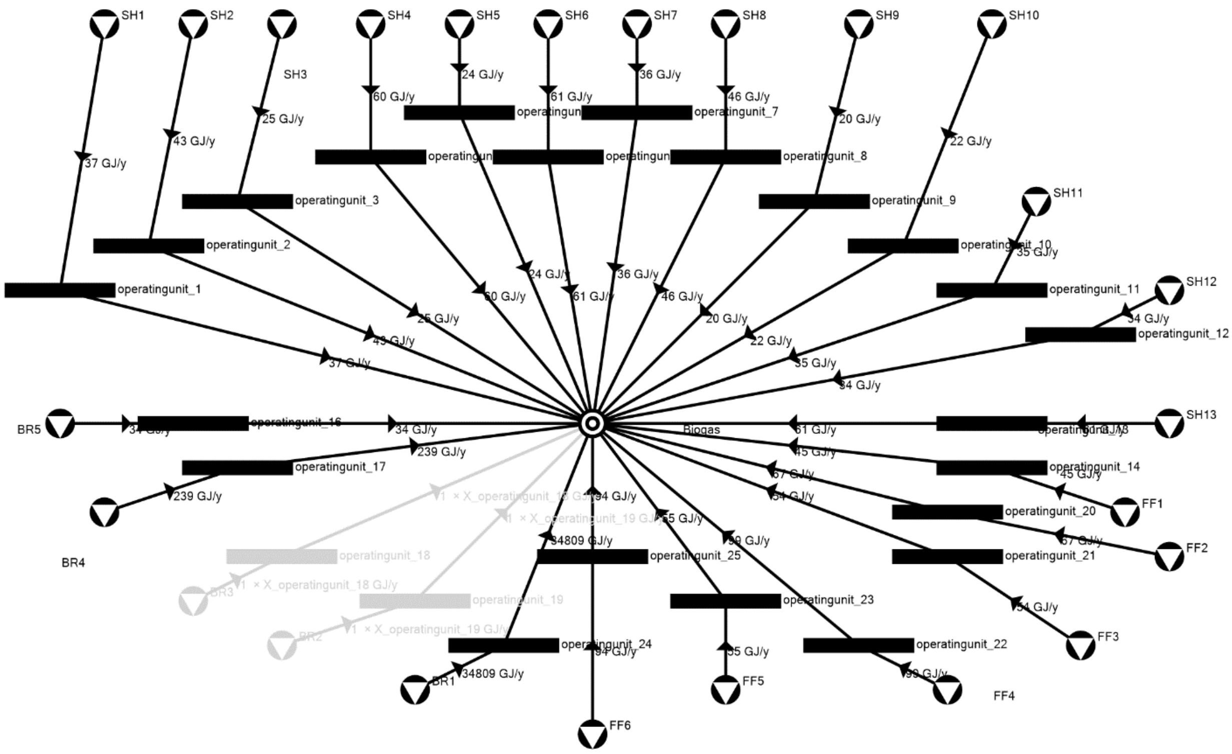

Figure 5 The optimal structure for annual biogas production of 36,000 GJ/y

The cost of the biomass supply chain (including feedstock and transport costs) is 448,080 EUR. This equals 12.44 EUR/GJ. The data from the optimal structure (Figure 5) are presented in Table 3, to improve the visibility of the numbers. As seen in Figure 5, feedstock sites that provide waste materials (supermarkets and fast-food restaurants) are prioritised as feedstock suppliers. The optimal structure for annual biogas production of 72,000 GJ/y is presented in Figure 6.

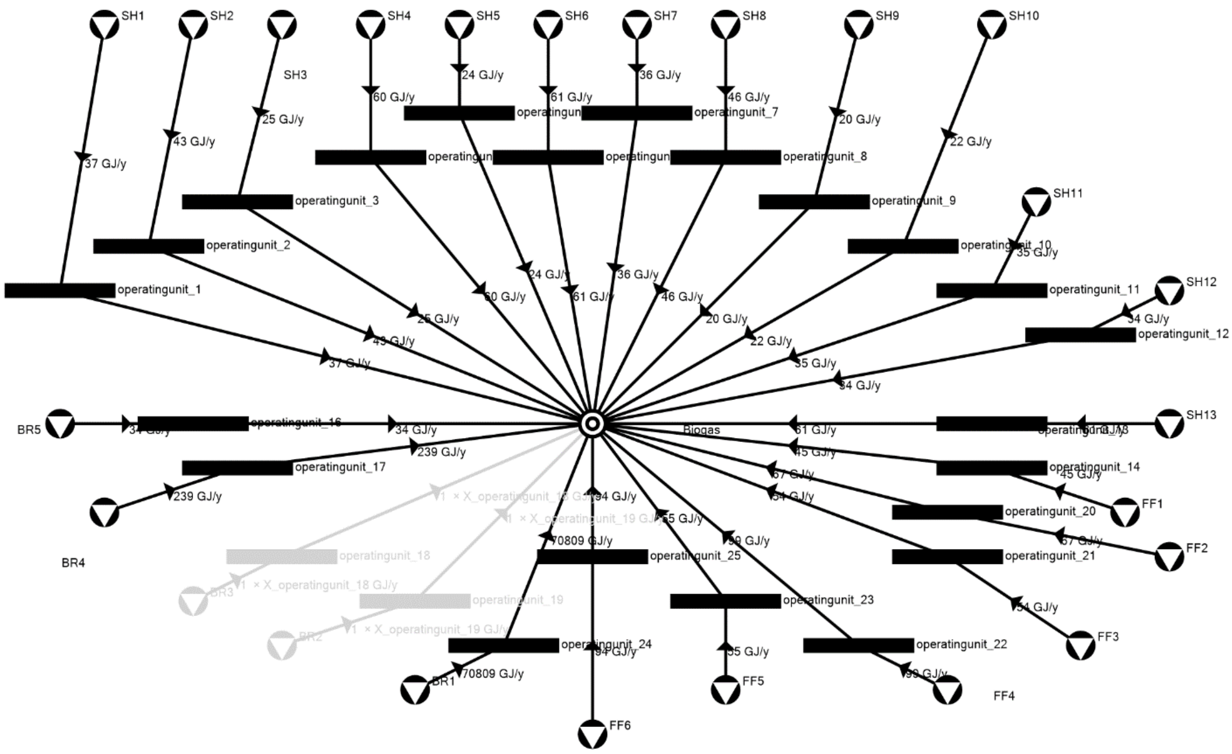

Figure 6 The optimal structure for annual biogas production of 72,000 GJ/y

The cost of the biomass supply chain (including feedstock and transport costs) is 907,593 EUR. This equals 12.61 EUR/GJ. The data from the optimal structure (Figure 6) are presented in Table 3 to improve the visibility of the numbers.

Table 3 The optimal structure for annual biogas production of 36,000 GJ/y (S1) and 72,000 GJ/y (S2)

Site		SH1	SH2	SH3	SH4	SH5	SH6	SH7	SH8	SH9	SH10	SH11
S1	**Delivered feedstock (GJ/y)**	37	43	25	60	24	61	36	46	20	22	35
S2		37	43	25	60	24	61	36	46	20	22	35
Site		SH12	SH13	FF1	FF2	FF3	FF4	FF5	FF6	BR1	BR4	BR5
S1	**Delivered feedstock (GJ/y)**	34	62	45	67	54	99	55	94	34809	239	34
S2		34	62	45	67	54	99	55	94	70809	239	34

The utilization of waste materials from supermarkets and fast-food establishments is evident in both scenarios, as indicated in Table 3. However, their contribution is relatively low due to the limited potential of these sites. It is noteworthy that the specific cost of biomass supply is slightly higher in the second case, primarily because of a dominant supplier (brewery) with the highest biogas potential. Consequently, this supplier significantly influences the average price of the biomass supply chain. The increased demand for biogas leads to a further rise in the specific cost of the biogas supply network. Thus, it can be inferred that the economic viability of biogas production in urban areas should rely on waste materials to enhance its feasibility.

The combination of integrating GIS mapping of biomass potential and employing a P-graph framework for optimizing biomass supply networks, implemented in this work and tested in the case study proved to be effective. This approach improves the accuracy of input data and consequently results, in comparison with other studies that consider biomass potential to be generated from a single site [11] or clustered into zones [26].
Furthermore, the elimination of feedstock suppliers with high greenhouse gas (GHG) emissions from transport during the initial step of GIS mapping has demonstrated its efficiency by ensuring that the utilization of the final product, namely biogas, achieves at least the minimum GHG savings. However, this approach's applicability is limited to cases where the transport distance is the sole factor affecting GHG emissions, and typical values can be used to calculate other GHG-related factors. For more complex situations where total GHG emissions may vary based on selections made within the supply chain network, a more intricate integration of GHG emissions savings limitations is required.

5. Conclusions

The method employed in this study integrates GIS mapping and graph theory approaches. GIS mapping is utilized to assess the availability and geographical distribution of feedstocks for biogas production, as well as to determine the transport distance between feedstock providers and the biogas facility. Additionally, the GIS tool is used to evaluate the suitability of feedstocks for biogas production based on their transport distance and the maximum allowable greenhouse gas (GHG) emissions allocated for feedstock transportation, in line with the requirements outlined in Directive 2018/2001. These limitations are incorporated as part of the input data to develop the maximal structure and optimize the biomass supply network using a p-graph approach. The objective of this optimization is to minimize the overall cost of the biomass supply network.
The proposed approach is implemented and tested in a case study conducted in an urban area in Zagreb, focusing on biowaste, residues, and by-products from supermarkets, fast food restaurants, and breweries. Two scenarios are considered, one with an annual production of 36,000 GJ and the other with an annual production of 72,000 GJ. The P-graph approach enabled the identification of the optimal economic solution for both cases. Since the majority of the biogas potential is concentrated in a single brewery, the specific cost of the biomass supply network (including feedstock and transport) remains similar for both scenarios, at 12.44 EUR/GJ and 12.61 EUR/GJ for the second case. The results reveal that the specific cost of the biomass supply network is relatively high, primarily due to the significant contribution of spent grain in biogas production. This research is expected to be beneficial to decision-makers and biogas plant operators in the development of the biogas industry. To enhance the economic feasibility of biogas production, it is crucial to explore additional sources of waste materials and prioritize the utilization of such materials in biogas production. Future studies should aim to expand the range of eligible feedstocks for biogas production, thus enhancing the economic feasibility of this process.

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