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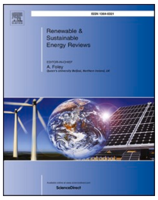

The importance of high crop residue demand on biogas plant site selection, scaling and feedstock allocation – A regional scale concept in a Hungarian study area

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ABSTRACT

In regions characterised by intensive agriculture, livestock manure is a commonly used feedstock for biogas production. Due to its expensive transportation, manure sources are often the sole criteria during biogas plant site selection, regarding feedstock supply. Encouraging biogas plant operators to use larger amounts of crop residues in the feedstock is favourable from an energy management viewpoint, but its spatial projection on resource logistics and its significance on biogas plant selection is less investigated. In this study, scenarios were created with different feedstock compositions considering constant manure and varying crop residue ratios. Based on their potential biogas yields and the location of livestock farms, a manure source-oriented site selection and facility scaling was made in a Hungarian study area. The applied GIS-based feedstock allocation and logistic analysis defined the crop acquisition possibilities and optimal transportation routes, assuming multiple resource-competitive biogas plants. The results indicate that feedstock composition can indirectly impact the site selection procedure and supply security if high crop residue demand is considered. Resource acquisition possibilities and economic feasibility are significantly affected by the location and density of the proposed biogas plants and their relative position to the crop supply areas. Due to the geographical heterogeneity of the supply side and the demand points, the transportation costs of crop residues and the digestate exceed those of the manure in all scenarios, which draws attention to the importance of spatial availability of crop residues during biogas plant site selection and scaling.

1. Introduction

1.1. Biogas production and its role in sustainable energy systems

In 2019, the European Commission presented the European Green Deal, which urges comprehensive structural changes, especially in the energy sector [1]. Among the efforts, continuous promotion of renewable energy sources is required, which is expected to result in a further expansion of sustainable energy generation technologies [2,3]. Accordingly, the ongoing capacity growth of intermittent renewable energy sources are generating an increased demand towards balancing capacities in order to maintain the stability of the power system [4].

Flexible power generation technologies like biogas plants can partly perform this task, while their purpose is not limited to energy production only. Besides electricity and heat production, a biogas plant has an important role in agriculture and waste management by treating hazardous organic substances and generating valuable fertilisers for farming [5]. The biogassing process is a good example of sector coupling by its contribution to the establishment of a circular economy.

In line with the EU-level ambitions, national energy and climate plans of the EU member states should also increase the interest of investors in biogas energy projects [6,7]. However, if several new actors emerge, complex environmental management and energy planning must be carried out first on different geographical scales, taking into account both regional and local conditions [8], especially with regard to the procurement of raw materials as feedstock for biogas production.

Site selection and plant scaling is initially driven by the spatial distribution and characteristics of different energy sources. Biomass, among other renewable energy sources, has the unique feature to be spatially allocated via transportation; therefore, site selection and sizing of biogas plants are slightly flexible. Since transportation of organic

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Abbreviations

AD plant	Anaerobic Digestion plant
CapEx	Capital Expenditure
CHP	Combined Heat and Power
CM	Cow Manure
CR	Crop Residue
CRM_{VS}	Crop Residue Multiplier
CRS	Crop Residue Share
CS	Corn Stalk
CSTR	Continuously Stirred Tank Reactor
d	distance
Dig	Digestate
DM	Dry Matter
E_e	Electricity Production
GIS	Geographical Information System
LM	Livestock Manure
LU	Livestock Unit
MC	Methane Content
OpEx	Operational Expenditure
PS	Pig Slurry
$Revenue_e$	Electricity Revenue
T	Transportation
PMY	Potential Methane Yield
UMY	Ultimate Methane Yield
VS	Volatile Solid
WS	Wheat and other cereal (triticale, oat, barley and rye) straw

materials to the anaerobic digestion (AD) plants has high cost and environmental impacts, consideration of supply-side installation factors in site selection can make the biogas technology more sustainable [9–11].

AD plants have certain flexibility to utilise different digestible materials which can be provided by point sources with high spatial energy density, e.g. the organic fraction of municipal solid waste on landfills [12] and sludge from wastewater treatment plants [13]. Globally, most of the biogas is produced from agricultural wastes, like livestock manure (LM) from intensive livestock farming sites, which are also point sources [14,15]. In regions affected by intensive arable farming, LM is usually treated together with crop residues (CR) or energy crops during co-digestion. Nevertheless, the cultivation of dedicated energy crops, such as silage maize, may lead to unfavourable agricultural practices and negative environmental effects, i.e. monocultural crop production and excessive use of pesticides [16,17]. Energy crop cultivation also induces tensions between food industry and the energy sector [18]. Hence, organic residues, like food waste and environmentally less harmful crop alternatives should be preferred as feedstocks [19–22]. In contrast to LM, CRs from arable lands are spatially extended, covering larger areas, which entails lower spatial energy density and the necessity of its transportation after harvest and collection. If several AD plants that use CR are in operation in a particular region, the supply areas of the facilities can overlap, generating supply chain difficulties and distribution conflicts.

1.2. Biogas potential and resource management with GIS support

Geographical Information System (GIS) is a tool often used by energy planners to examine spatial aspects of supply, distribution and demand sides of energy management. It is beneficial to apply it especially for renewable energy potential calculations and site selections for such complex spatial problems as biomass utilisation. Its application has a long tradition in the field of energy planning, on different geographical scales [23–25].

Studies on national or even continental scales help strategic decision making and energy roadmapping, although their resolution and application possibilities on smaller scales are limited [26–30]. The methods at a national level usually apply aggregated statistical data and use centroids to define supply areas of bioenergy facilities, resulting in coverage and feedstock distribution uncertainties at the regional level [31,32].

Studies on regional and sub-regional levels are able to provide more detailed spatial modelling, including precise bioenergy facility site selections by using high quality data and sophisticated GIS analysis. Some studies use Euclidean distances for substrate logistics modelling without taking the existing road network into account, which may lead to less accurate results. Regional models are usually supported by multi-criteria decision analysis, applying several environmental, social and techno-economic factors [33–36].

Through an LM transportation optimisation model, Thompson et al. [37] showed how spatial patterns of AD plants from a given area change by their growing number. Site selection was driven by land suitability restrictions, but in terms of the optimal distribution of one type of raw material only. Sultana and Kumar [38] examined the surroundings of possible biomass processing sites to define their individual supply areas. The allocation analysis of biomass to pellet plants showed that transporting straw to the closest facility is the most cost-effective solution, resulting in supply areas without overlaps. Similar outcomes were published by Höhn et al. [39] for biogas facilities. Bojesen et al. [40] draws attention to the importance of resource shortages, which may occur if AD plant service areas overlap and underlines the necessity of central planning and coordination in order to avoid supply conflicts.

A regional biomass allocation analysis was carried out by Panichelli and Gnansounou [41], where woody biomass allocation was made after examination of spatial distribution and transportation possibilities. The authors highlight allocation difficulties since the facility supply areas in the study region overlap significantly. Possible feedstock acquisition difficulties also were reported by Sliz-Szkliniarz and Vogt [42] in a theoretical regional biogas concept focusing on a province in Poland. Designated AD plant locations of livestock farms with given amounts of LM provide the basis of the concept. Proposing co-digestion with energy crops, supply shortages could occur in some cases due to limited crop production or demand competition between biogas facilities close to each other.

Several research articles deal with spatial biomass distribution, however, allocation and supply area overlapping problems are not considered with enough attention. Thus, possible conflicts between biomass processing plants remain hidden. Also, transportation and disposal of the digestate are generally neglected in the literature. However, feedstock acquisition and digestate management affects the nexus between the AD plant and its environment from a resource logistic and spatial energy planning viewpoint.

1.3. The effect of co-digestion on CH_4 yields

Several quantitative and qualitative factors need to be taken into account during the planning process of AD plants. On the one hand, plant capacity is determined by the amount of feedstock that can be delivered to the biogas facility for energy recovery. On the other hand, besides the conditions during the biogas process, co-digesting different types of organic materials has a significant influence on the expected biogas yield and its methane content [43].

Regarding agricultural waste, several articles focus on special feedstock compositions that can be applied in certain geographical areas only [44–46]. LM from intensive animal husbandry is a commonly utilised feedstock. Its co-digestion with CR is beneficial for biogas production since manure is characterised by a wide range of nutrients, while plants could improve the C/N ratio [47].

In addition to dry matter (DM) and volatile solid (VS) content measurements of specific organic materials, several research articles put emphasis on the altering effect of co-digestion by applying different mixing ratios of the feedstock. Measurement results of biogas production performance are reported for cow manure (CM) mixed with cereal straws (WS) [48–51], and with corn stalks (CS) [52–56]. Several studies deal with pig slurry (PS) co-digestion with WS [57–60], and with CS [61–63]. Most of these articles conclude that higher crop share in the feedstock results in increased biogas yield and methane content; however, negative effects may also occur in some cases [64]. Biogas yield values range widely in the aforementioned studies, mainly due to advancing biogas technology, different pre-treatment methods and digesting conditions. By applying feedstock pre-treatment, e.g. mechanical or chemical methods, biogas production can be enhanced by increasing the specific surface of biodegradable compounds [65–67].

According to Ref. [68], applying biogas performance values from laboratory batch tests on full-scale anaerobic digesters should be interpreted with caution. Expected methane yield is difficult to predict, especially on a commercial scale, where feedstock composition is constantly changing, mainly depending on the characteristics of the delivered raw materials [69]. Nevertheless, biogas yields may differ, yet laboratory experiment results can be used as a guide for large-scale implementation.

1.4. *Aim of the study*

Most of the research papers dealing with biogas plant scaling, site selection and resource management focus on only one substrate, e.g. manure, without taking other feedstocks into account. Therefore, some planning and operation problems may remain hidden. Co-digestion of LM with common, yet unutilised raw materials like CR as feedstock is an obvious way to enhance biogas production; however, the latter has strict sustainability constraints. Therefore, sustainable removal and fair accessibility of CR may become endangered by the promotion and increased utilisation of such resources.

Feedstock distribution conflicts, especially in the case of CR and the digestate, receive disproportionately little attention in the field of bioenergy utilisation, compared to their role and importance. By applying a GIS-based resource allocation analysis, this research examines the geographical availability of some agricultural waste types and their effect on spatial patterns of supply areas, feedstock logistics and AD plant site selection and sizing in a Hungarian study area. The aim of the study is to examine the possible threats regarding resource allocation in a regional biogas concept with several actors, with the main assumptions of minimised manure transport and increased demand for CR.

2. Materials and methods

2.1. *Data preparation and resource potential analysis*

2.1.1. *Livestock manure*

A database of 2018 provided by the Hungarian Food Chain Safety Office (NÉBIH), with high spatial resolution was used to calculate the livestock units (LU), considering pig and cattle types and age groups. The database also contains precise spatial information for every pig and cattle breeding location in the study area, ranging from households to large-scale farms. Due to the large number of locations, and because of the practical problems of collecting and transporting small quantities of LM, only the sites with 10 LUs or higher were considered for further calculations. Under this threshold, manure production rate is low, and the collection often faces difficulties due to extensive or semi-extensive breeding; therefore, its utilisation would be inefficient.

The Bioenergy and Food Security Rapid Appraisal (BEFS AR) Tool from FAO was used to calculate yearly LM yield on the farms [70]. Animal breeding technology for each farm is uncertain; therefore, dry collection of CM with 100% collection efficiency was supposed, which was determined empirically. Despite that extensive breeding in large-scale farms is not typical in the study area, visual inspections using satellite images and orthophotos were made for every location in order to determine whether the cows have access to pastures or not. The possibility of grazing can reduce manure collection significantly [71]. Based on experiences of local experts and farm owners, 10% reduction from the amount of collectable CM was assumed at farms with attached pastures. In the case of pigs, deep litter husbandry is less typical, thus flushing of PS was considered (Table 1.).

2.1.2. *Crop residues*

Information regarding cultivated area coverage in 2018 in the study area was obtained from the Hungarian State Treasury. The database contains a 100 ha resolution grid, and the size of the cultivated area for different crop species in each quadrat in the grid. Wheat, triticale, oat, barley and rye were selected and handled as a group (referred to as WS), based on their similar characteristics. Corn was considered as another group (referred to as CS), excluding its volume used for silage purposes, since the latter refers to the utilisation of the whole crop, e.g. for livestock feeding [72]. The amount of gross CR (considering straw of cereals and stalk of corn) for each group was estimated by multiplying the areal values by the annual crop yield (t/ha), using the local agricultural statistics from Ref. [73]. Values regarding residue to crop ratios are diverse, mainly due to the different geographic and climatic conditions, as well the applied cultivation methods [74–76]. Since consistent studies regarding this ratio are not available for the local conditions, values of BEFS AR Tool were applied [70].

Respecting strict sustainability constraints, maximum removal rates were determined for the two CR groups in order to preserve soil resource supply [77–79]. However, the literature is not consistent about these removal rates, which are also affected by several factors. Therefore, average values have been applied from Refs. [80–84]. Other needs for competitive uses of CR were defined as 1%, i.e. for mushroom cultivation, surface mulching, utilisation in biomass power plants and for other industrial uses [85]. Demand for livestock bedding, which represents the highest share of competitive use of WS, was not taken into account, since information about husbandry technologies at livestock farms regarding bedding methods are not available. A 2% loss of the gross amount was assumed during harvesting, transportation and storing the CR [86]. Amounts of competitive use and losses were applied equally for the agricultural grid of the whole study area. Characteristics of WS and CS, applied from the above-mentioned literature, are presented in Table 1.

2.2. *Applied feedstock compositions and scenarios*

In order to make the amount of feedstock comparable, DM and VS contents were defined, using average values of laboratory measurements for both LM and CR types. Applied values are presented in Table 1. These characteristics depend on, i.e. livestock feeding, breeding technology, manure collection and storage practice as well as harvesting method of crops [98].

Since available studies regarding co-digestion of the considered raw materials are limited, four different feedstock composition groups were made, each with three different substrate mixing ratios on VS basis: one with high LM and low CR content (~2:1), one with equal raw material content (1:1), and a third mix with low LM and high CR content (~1:2). It was assumed that only one type of LM could be co-digested with only one type of CR, e.g. CM or PS with either WS or CS. Mixture characteristics were selected and applied based on the relevant articles, which approaches meet the following three criteria:

- Feedstock composition uses the raw materials assumed in this research (i.e. WS, CS, PS and CM);

Table 1

Properties of considered raw materials. LM = livestock manure; CR = crop residue; DM = dry matter content; VS = volatile solid content; CM = cow manure; PS = pig slurry; WS = wheat straw; CS = corn stalk.

LM type	Yield (t/LU/yr)		Collection rate (%)	DM (%)	VS (% of DM)	Reference (DM and VS)
CM	13.66		90–100	12.16	77.53	[54,59,87–92]
PS	15.26		100	8.47	79.46	[60,87,90,91,93,94]
CR type	Yield (t/ha/yr)	Residue to crop ratio	Removal rate (%)	DM (%)	VS (% of DM)	Reference (DM and VS)
WS	4.89	1.39	41	84.56	92.57	[50,64,92,93]
CS	6.77	1.96	38	90.02	90.25	[95–97]

- Feedstock composition ratios of the mixtures correspond to other mixtures to be comparable with each other;
- It is indicated in the related studies whether the feedstock was pre-treated or not.

Considering the aspects mentioned above, the applied mixing ratios and ultimate methane yields (UMY) were collected and presented in Table 2. For originally untreated feedstock mixtures, pre-treatment (mechanical, thermal or chemical) was assumed to increase the hypothetical biogas yield, based on earlier findings [99,100]. Regarding PSWS mixtures, according to the knowledge of the authors, no appropriate studies are available which would fulfil all the above mentioned three criteria; therefore, a feedstock composition of PS mixed with rice straw was chosen, because of its similar characteristics to other cereal straws [60].

Accordingly, 12 scenarios were created, and their names reflect the composition and ratio of the corresponding mixtures. Considering constant LM weights, the amount of required CR on VS basis can be expressed by the crop residue multiplier (CRM_{VS}) factor, while the crop residue share (CRS, %), considering the net weight of the feedstock mixture, varies depending on the different composition of the scenarios (Table 2).

2.3. GIS analysis of feedstock management

2.3.1. Manure source merging and site selection

Other biogas related studies usually perform multi-criteria site selection in order to determine optimal locations. In contrast, the sole criteria for AD plant site selection in the present method is the location of livestock farms as reference points, where other substrates, i.e. crop residues are transported to. Availability of such feedstocks was not considered as a criteria initially, which allows the method to uncover a potential local overdemand on them.

Every farm was considered as a candidate, and to be hosting an AD plant on site. However, construction of new biogas plants in each livestock breeding location is not realistic because of economies of scale and technical equipment constraints on the market [24]. Therefore, merging the sources via transporting the generated LM is recommended.

Farms that would not be able to support a biogas engine of 100 kW$_e$ as a limit, have been merged to sites already above this limit. This lower threshold is therefore only technical, without taking economical constraints into account. Engine capacity specifications are presented later, in Section 2.4. The possibility to exceed the threshold for a given candidate site varies in each scenario, according to the associated potential methane yield (PMY). All candidate sites that exceed the capacity limit were proposed to operate a biogas plant, without taking any other constraints into account. Separation of CM apart from PS was assumed in cases of farms where both cows and pigs are kept, supposing there were two AD plants at the same location. It was possible to take just one or both types of LM to other locations, depending on which is sufficient to utilise on-site.

Merging was implemented by locating the closest site above the threshold with the same utilised LM type, based on the actual road network. This ensures the lowest energy and cost requirements, and emission mitigation of the transport as well. Defining the closest location was implemented by the Closest Facility Tool from ArcMap 10.2, using the road network of the study area from OpenStreetMap. Every farm under the proposed 100 kW$_e$ threshold was allowed to transport their manure to only one (the closest) receiving site. The AD plants at these sites could accept LM without any quantitative limitation. During LM transportation, no travel distance restrictions were assumed, noting that manure transportation itself is controversial [101,102]. In order to reduce load weight and consequently transportation cost of both PS and CM, moisture content reduction to 40% was assumed, i.e. using centrifugation at the farms before delivery to the AD plants [103]. The total transportation route length (T_{LM}, km) to the facilities has been calculated as follows:

$$T_{LM} = LM/25*d$$

where LM stands for the amount of livestock manure (PS or CM, depending on the actual scenario based on Table 2) (t), 25 is the capacity of the assumed transporting vehicle (t) and d is the geodetic distance between a given manure source and the AD plant (km), measured on the actual road network of the study area by basic GIS operations.

Following the merging process, transported amounts of LM were added to the LM generated at the sites of the receiving facilities. This procedure practically corresponds to a simple site selection, where only basic technical aspects were considered, e.g. minimal facility size and transportation, since determining the optimal location for AD pants was

Table 2

General properties of the applied scenarios. VS = volatile solid; CRM = crop residue multiplier; CRS = crop residue share; MC = methane content; UMY = ultimate methane yield.

Feedstock composition	Composition (VS ratio)	Scenario name	CRM_{VS}	CRS (%)	MC (%)	UMY (Nm^3/tVS)	Reference
Cow Manure and Cereal Straw	7:3	CM7WS3	0.43	5.18	58	350	[55]
	1:1	CM1WS1	1.00	12.05	58	420	[55]
	3:7	CM3WS7	2.33	28.07	50	396	[55]
Cow Manure and Corn Stalk	2:1	CM2CS1	0.50	5.81	57	588	[56]
	1:1	CM1CS1	1.00	11.61	62	614	[56]
	1:2	CM1CS2	2.00	23.23	58	603	[56]
Pig Slurry and Cereal Straw	2:1	PS2WS1	0.50	4.30	56	536	[60]
	1:1	PS1WS1	1.00	8.60	56	534	[60]
	1:2	PS1WS2	2.00	17.20	54	482	[60]
Pig Slurry and Corn Stalk	7:3	PS7CS3	0.43	3.57	55	421	[62]
	1:1	PS1CS1	1.00	8.29	55	337	[62]
	3:7	PS3CS7	2.33	19.33	55	294	[62]

not the main purpose of this research.

2.3.2. Allocation of crop residues and digestate

After merging the LM sources and designating the AD plant sites, it became possible to allocate CR to them, knowing the aggregated, total amount of manure and the required amount of CR at each AD plant, according to the scenarios. The Location-allocation Tool and its Maximize Capacitated Coverage option from ArcMap has been applied to find the proper source of CR in the study area. The tool fulfils CR requirements (capacities) of the possible AD plants by allocating WS or CS from the data points (quadrats) of the agricultural grid, considering the road network of the study area. Original CR values of each agricultural quadrat were divided into 5 equal parts, from which 2 were retained (2 * 1/5). 2 of the 3 remainder parts were subdivided into 2-2 equal shares (4 * 1/10), then the last one fifth into 5 equal parts (5 * 1/25). This operation allowed the tool to allocate the CR from the same quadrat to different biogas facilities. By applying this method, such facilities could share the amount of CR of a given quadrat by overlapping their supply areas, thus, making the biomass distribution more realistic. AD plants prefer agricultural CR sources closest to them, and are extending their supply areas until their previously defined demand is fulfilled. Maximum distance for CR transportation was set to 40 km as a limit recommended in other studies [23,32,42].

The transportation route length of CRs (T_{CR}, km) between the AD plants and the centre of the agriculture quadrats as crop fields has been calculated by the following formula:

$$T_{CR} = CR/20^{*}d$$

where *CR* is the amount of crop residues (WS or CS, depending on the actual scenario based on Table 2), 20 is the capacity of the assumed bale transporting vehicle (t), and *d* is the distance between a given crop residue source and the AD plant (km).

The amount of annual production of digestate corresponds to 90% of the feedstocks [86]. Similarly to LM transport, separation of the liquid and solid phase of digestate was assumed. It was also assumed that the solid digestate is transported to the fields where the CR originate from. The liquid fraction can be used for dilution of the mixture, while solid fraction is utilised as fertiliser on croplands. The transport route length of the digestate (T_{Dig}, km) has been calculated by:

$$T_{Dig} = Dig/25^{*}d$$

where *Dig* stands for the amount of solid digestate (t), 25 is the capacity of the assumed transporting vehicle (t), and *d* is the distance between a given AD plant and the origin of the delivered CR (km).

2.4. Technical and economic assessment

2.4.1. Energy potentials and plant sizing

Calculations regarding raw material utilisation in each scenario were implemented on VS basis, by defining the amounts of the required CR, considering constant amounts of manure and changing CRM_{VS}. Annual potential methane yield (*PMY*, Nm^3/yr) has been calculated for each AD plant in terms of the scenarios with the following formula, assuming that the required CR amount is fully obtainable as follows:

$$PMY = (LM * DM_{LM} * VS_{LM} + LM * DM_{LM} * VS_{LM} * CRM_{VS})^{*}UMY$$

where *LM* is the amount of livestock manure available at a given site (PS or CM) (t), DM_{LM} and VS_{LM} stand for the dry matter content (%) and the volatile solid content (%) of LM, respectively, CRM_{VS} is the volatile solid multiplier of crop residues on VS basis according to the given feedstock composition, and *UMY* is the ultimate methane yield (Nm^3/tVS). In the case of insufficient amount of CR, lower *PMY* can be achieved.

Mesophilic conditions and wet fermentation were assumed in continuously stirred tank reactors (CSTR) during single-stage digestion, as the most common practice for biogas production [86]. 1.5% of biogas loss from leaking was considered during the whole process according to Ref. [104]. In determining the capacity of the required gas engines, optimisation of engines for power system regulation purposes and covering daily peak electricity demands were assumed, with 5000 h/yr operation time [105]. Properties of the proposed biogas engines are presented in Table 3. The size of the gas engine that can be installed on each site is determined by which category it can reach based on *PMY*.

Annual electricity production (E_e, MWh/yr) of a given facility has been calculated by:

$$E_e = PMY^{*}36^{*}\eta/3600$$

where *PMY* stands for potential methane yield (Nm^3/yr), 36 is the lower heating value of methane (MJ/Nm^3), η is the electrical efficiency of the gas engine (%) and 3600 is the conversion factor of MJ to MWh.

2.4.2. Economic assessment

Favourability comparison between scenarios has been made by basic techno-economic calculations. The same method was applied as in Ref. [42], except for transportation costs. Capital (CapEx) and operation expenditures (OpEx) and their components have been carried out with revenue calculations. In order to take annual depreciation of CapEx into account, the Modified Accelerated Cost Recovery System (MACRS) has been applied with annual depreciation rates of 0.75, 5.95, 5.59, 5.26, 4.94, 4.65, 4.37, 4.10, 3.82, 3.63 and 3.58 from the 10th year, for a total of 25 years operation period. The economic indicators were proposed based on the current Hungarian economic characteristics. The annual inflation rate of 4% has been applied for OpEx and revenue from electricity ($Revenue_e$). Also, a 6% interest rate has been assumed for the discounted net cash flow to calculate the discounted payback period for each individual AD plant.

Regarding feedstock expenditures, CM and PS prices have been set to be zero, as recently manure management is rather a problem than a valuable raw material, reported by local farm owners. However, both WS and CS prices cost an average of 25 €/t in the study area [106].

The total transportation cost ($OpEx_T$, €) for a given AD plant has been calculated by the following formula:

$$OpEx_T = (LM * 2.6 + T_{LM} * 0.85 * 2) + (CR * 2.6 + T_{CR} * 0.85 * 2) + (Dig * 2.6 + T_{Dig} * 0.85 * 2)$$

Transportation of feedstock types and digestate was assumed by commonly used semi-trailers and tipper trucks, with a fixed transport cost of 2.6 €/t load for *LM*, *CR* and *Dig* (t), as average local tariffs. Furthermore, 0.85 €/km as a varying cost was assumed for T_{LM}, T_{CR} and T_{Dig}, according to Ref. [107]. 2 was used as a multiplication factor in order to take the routes on the way back to the biogas plants without load into account, affecting only the varying transport cost part.

The only potential income of an AD plant considered in this study is electricity. Thermal energy utilisation from gas engines, e.g. for district heating purposes was out of the scope of this study, despite the favourable conditions of some settlements in the region [108].

9% of self-consumption from electricity was applied to calculate net electricity that could be sold on the market [42,86]. $Revenue_e$ was

Table 3
Applied biogas engine properties and fuel demands.

Engine capacity (kWe)	η (%)	Required fuel (1000 Nm^3 CH_4)
100	28	179
200	35	263
350	40	438
500	42	595
1000	43	1163
2000	43	2326
3500	43	4070
5000	43	5814

calculated by multiplying the net amount of electricity (MWh) by 107.06 (€/MWh) and 96.1 (€/MWh) for AD plants with engine capacity under 1000 kW_e and over 1000 kW_e, respectively. These subsidised, capacity dependent electricity prices can be provided over a 25-year period in Hungary, according to the Hungarian Renewable Energy Support System, METÁR [109]. However, this support scheme currently does not promote flexible power production, which could help the integration of intermittent energy sources.

3. Results

3.1. Quantity and spatial distribution of feedstock, theoretical feasibility

Spatial distribution of livestock farms is heterogeneous in the study area; however, the range of their size is very diverse. There are 237 farms in the region that exceed the 10 LU limit, of which medium and large scale are dominant (Fig. 1b). A total 20,259 LU of pigs are distributed among 76 farms, while 42,290 LU of cows are being held at 197 farms, both distributed unequally within the area. There are 36 farms where both pigs and cows are kept, but typically farms are specialised; therefore, one type of animal is dominant on each farm. There are overlaps between these classifications. Generated PS and CM weights 309,150 t/yr and 577,595 t/yr, respectively. Site specific results were validated by real life experiences in two farms in the study area (Harsány and Onga), where AD plants are in operation already.

Intensive arable agriculture is dominant in the river valleys and plains in the centre and the north-eastern parts of the 7250 km^2 study area, while the land use of the hilly northern parts is more heterogeneous (Fig. 1c and d). According to the statistics of the year 2018, cultivated area of cereals is 81,288 ha and 40,175 ha of corn, which result in residues of 513,068 t/yr WS and 532,853 t/yr CS. The total amount of CR available for energy recovery of WS and CS are 194,965 t/yr and 186,499 t/yr, respectively, considering sustainable removal rates, competitive uses and losses.

Table 4
Raw material requirements for co-digestion on volatile solid (VS) basis), and potential methane yield (PMY) values for each scenario. LM = livestock manure; CR = crop residues.

Scenario name	LM_{VS} (t)	Required CR_{VS} (t)	Demand rate from total CR (%)	PMY (1000 m^3)
CM7WS3	55,862	23,941	15.67	27,931
CM1WS1	55,862	55,862	36.57	46,924
CM3WS7	55,862	1,30,345	85.34	73,738
CM2CS1	55,862	27,931	18.43	49,253
CM1CS1	55,862	55,862	36.87	68,576
CM1CS2	55,862	1,11,724	73.73	1,01,121
PS2WS1	20,328	10,164	6.65	16,349
PS1WS1	20,328	20,328	13.31	21,726
PS1WS2	20,328	40,656	26.62	29,418
PS7CS3	20,328	8712	5.75	12,221
PS1CS1	20,328	20,328	13.42	13,688
PS3CS7	20,328	47,432	31.30	19,936

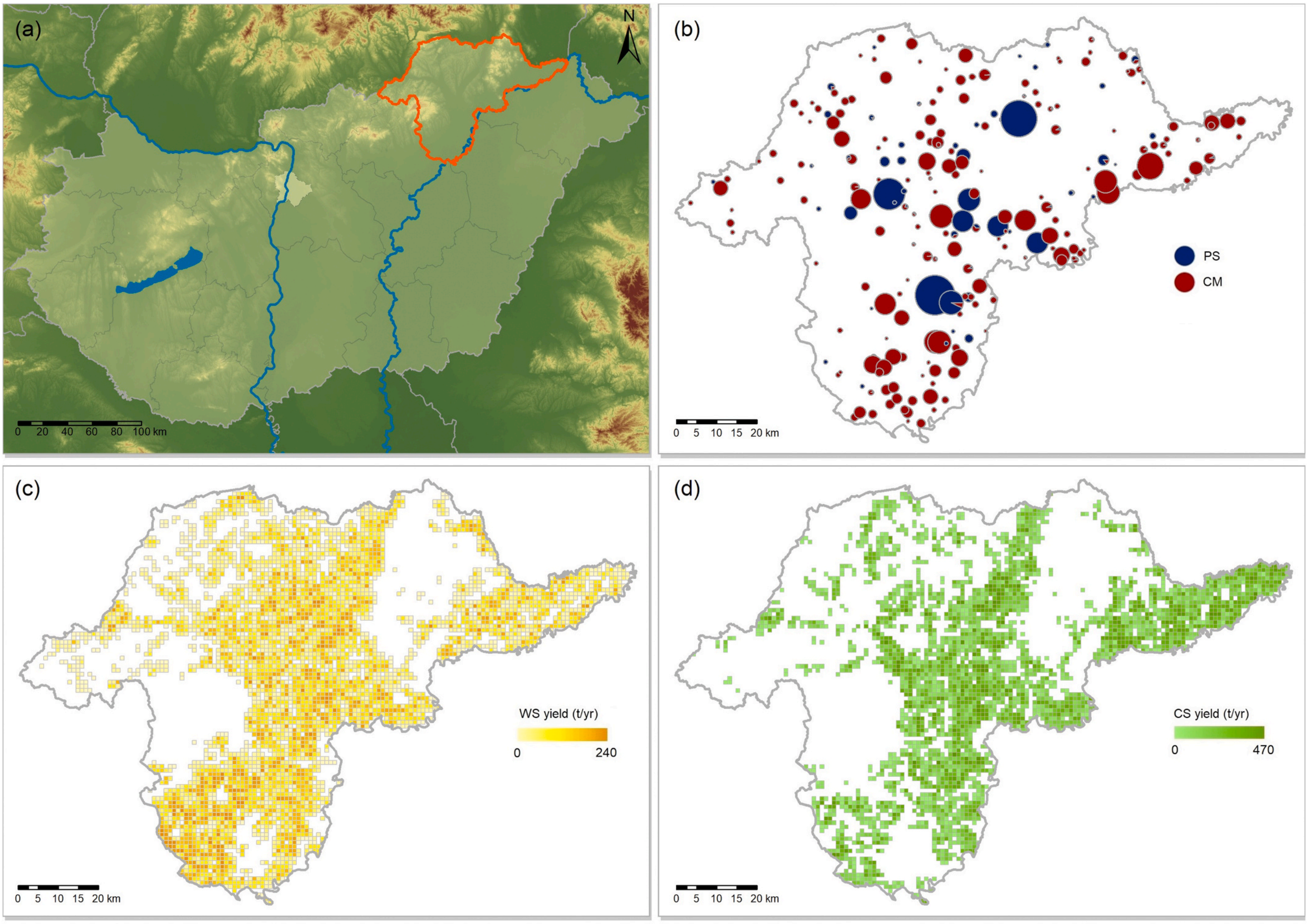

Fig. 1. (a) Geographical location of the study area in Hungary; (b) Distribution and relative quantities of pig slurry (PS) and cow manure (CM). In this frame, categories in the legend are not presented because of data protection reasons; (c) Distribution and quantities of cereal straw (WS); (d) Distribution and quantities of corn stalk (CS).

Constant LM and varying CR amounts (on VS basis) are presented in Table 4, by assuming that the total amount of PS and CM is utilised for biogas production. It can be stated that despite the large quantities of LM, CR needs for co-digestion can be fully supplied theoretically, even in cases of scenarios with the largest WS or CS demand. Despite UMY in each feedstock composition being different, PMY is increasing with a higher share of CR for all feedstocks, since more raw material is utilised for energy recovery. From an energy recovery point of view, co-digesting PS with WS and co-digesting CM with CS are more favourable scenarios according to the results in Table 4. Therefore, only these two composition types and their 6 scenarios were chosen for further analysis and presented in detail in the subsequent sections.

3.2. Feedstock logistic patterns

3.2.1. Site selection and collection of manure

Since site specific PMY values are changing according to feedstock characteristics, the possibility of exceeding the fuel demand limit of a

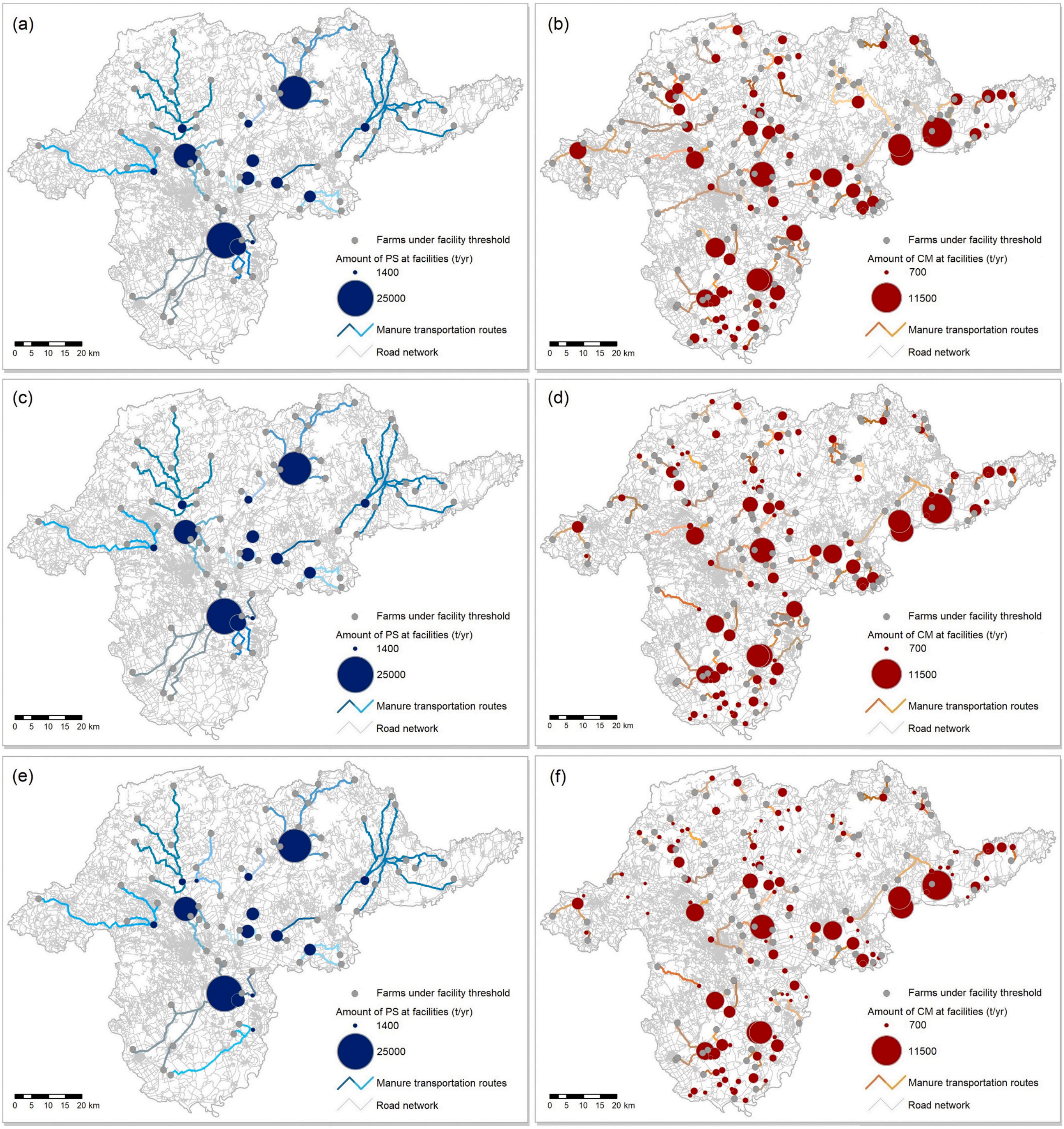

Fig. 2. Computed optimal (shortest) routes of manure transportation to the selected AD plant sites, for scenarios (a) PS2WS1; (b) CM2CS1; (c) PS1WS1; (d) CM1CS1; (e) PS1WS2 and (f) CM1CS2. Markings of farms under the plant threshold are presented in the same size because of data protection reasons. PS and CM stand for pig slurry and cow manure, respectively.

100 kW$_e$ gas engine can affect more candidate sites, thus they would be able to host AD plants. PS and CM from farms under the limit are assumed to be transported to the closest AD plant; however, transportation route length and direction also vary according to the number and spatial pattern of the receiving facilities in each scenario. For this reason, a given candidate AD plant site in another scenario could lose its delivered manure, if other competitors appear in their vicinity, resulting in a distraction effect (Fig. 2).

According to Fig. 1b, there are less possible AD plants using PS than CM, due to the more centralised character and fewer number of pig farms. Moreover, this rigid structure of PS utilising plants is well illustrated by the fact that the number of facilities is the same in the case of PS2WS1 and PS1WS1 scenarios. PS utilising AD plants in the central part of the study area collect all additional LM, while no other candidate site near the borders exceeds the threshold to become a receiving facility. It is also associated with much longer mean transportation routes, compared to hauling of CM (Table 5). Maximum CM hauling distances are 23.3, 16.6 and 16.6 km for CM2CS1, CM1CS1 and CM1CS2, respectively, and 43.8 km for all the three PSWS scenarios. Nevertheless, only a small percentage of the routes exceed the 10 km transportation distance which is an economic limit according to Ref. [32].

The value of T_{LM} is decreasing by the growing number of AD plants for two reasons. On the one hand, a high number of farms offer more possibilities to find closer receiving locations for sites under the 100 kW$_e$ engine threshold. On the other hand, livestock farms that are exceeding the threshold, utilise their own LM locally in the model; therefore they do not need to transport the manure to another location. Due to the large number and sensitive data of individual farms in each scenario, results hereafter are presented only in a summarised form in tables and diagrams.

3.2.2. Allocation efficiency of crop residues

The GIS analysis proved that potential AD plant sites firstly should gather the required CR from their vicinity, if such materials are even available. If the CR were homogeneously distributed around the AD plants, the size of supply areas would be in direct proportion to the amount of required WS and CS. Nevertheless, except for sites in the centre of the study area, this linear relation cannot be realised due to the heterogeneity of the cultivated area. Uncertainty of allocation efficiency increases towards the borders, as the analysis considers the study area as it was isolated from the surroundings. Nevertheless, continuity of cereal and corn cultivated areas is ensured beyond the borders mainly to the South and East, therefore CR acquisition would be less challenging in reality for AD plants close to the margins.

As the demand for WS and CS increases with their growing CRS according to the scenarios, supply areas are extended further (Fig. 3.). This can lead to difficulties to obtain CR, where the supply areas of neighbouring AD plants overlap, and the facilities would be forced to meet their demand from the same cultivated areas. For the PS2WS1, PS1WS1, PS1WS2 and CM2CS1 scenarios, the allocation succeeded without such complications; therefore, the supply of CR would be ensured. For CM1CS1, in the southern parts of the study area some overlaps occur because of the dense distribution of proposed AD plants and insufficient amount of CS. Yet, their demand can be fulfilled by hauling the required amount of CS from longer distances. For CM1CS2, however, significant overlaps can be observed.

Note that transportation of CR, similarly to LM transport, was also applied on the actual road network. Nonetheless, maps of Fig. 3 show CR allocations as an origin-destination connectivity between the CR source quadrats and the AD plants as straight lines. This visualisation allows simpler display by avoiding route overlaps on the actual road network. For this reason, some agriculture quadrats connected to farther AD plants rather to closer ones may seem illogical, whilst their computed transportation routes are still correct. This is due to the characteristics of the local road network, which is affected by several geographic factors, e.g. terrain or limited possibilities to cross rivers on bridges.

For the scenario CM1CS2, the location-allocation analysis resulted in that CS demands could not be fulfilled completely, but only by 95.3% if sustainability limits regarding CR removal are respected. The shortage could affect 9 AD plants out of 114: these 9 sites are highlighted in Fig. 3 (f). Their CR supply deficit ranges from 5% to 100% and concerns smaller and even larger facilities, thus affecting the techno-economic results of the CM1CS2 scenario. Therefore, a correction was made by substituting these problematic AD plants with their corresponding sites from the CM1CS1 scenario, including gas engine capacities, transportation and economic attributes, in order to make CM1CS2 scenario feasible without shortages. One out of the 9 AD plants could not be substituted, because in the CM1CS1 scenario it was under the engine capacity threshold and merged to the nearest AD plant; therefore, it was neglected.

Unlike LM transport, defining maximal distances is less relevant, since the longest allocation routes concern only very small amounts of CR acquisition. It is negligible compared to most of the CS or WS demand that can be collected close to the AD plants. The average distances that trucks need to drive between an AD plant and the CR source are 8.9, 13.3 and 15.3 km for CMCS scenarios, and 7.2, 10.7 and 13.8 km for the PSWS scenarios, respectively, which is in proportion to the growing crop demands.

Due to possible overlaps, exact CR supply areas are difficult to define and visualise. Fig. 4 shows the relative changes and spatial distribution of the remaining CR for the highest CRS scenarios, after performing the allocation operations. The two maps clearly represent the difference between the strict supply areas of PS1WS2 and the diverse patterns of overlapping supply areas of CM1CS2 in Fig. 3.

Expanding CR supply areas can also be defined by presenting average specific round trip distances on the required mass of feedstock basis (Fig. 5). Specific WS transport is characterised by a linear growth since such CR is distributed homogeneously in the study area, and only minor overlaps of supply areas occur, even for the PS1WS2 scenario. In contrast, a rather parabolic growth can be observed for specific CS transport, especially for the CM1CS2 scenario, indicating extended supply areas due to higher total demands and the large number of AD plants.

Also, the geographical extension of cornfields was smaller than that of cereal fields in the year of observation. In the case of CM1CS2, site specific transport needs range from 0.07 to 3.40 km/t. The largest values

Table 5

Feedstock and digestate transportation route lengths of the 6 modelled scenarios. T_{LM} = transportation of livestock manure; T_{CR} = transportation of crop residues; T_{Dig} = transportation of digestate.

Scenario name	Farm merging		T_{LM}		T_{CR}		T_{Dig}		Total transport (km)
	Sites hosting AD plants	Sites under treshold	Sum (km)	Mean (km)	Sum (km)	Mean (km)	Sum (km)	Mean (km)	
CM2CS1	70	126	14,921	213	7867	112	86,880	1241	1,09,668
CM1CS1	91	106	8457	93	20,436	225	1,46,206	1607	1,75,099
CM1CS2	114	81	4510	40	67,841	595	1,83,557	1610	2,55,908
PS2WS1	13	63	10,477	806	2458	189	34,637	2664	47,572
PS1WS1	13	63	10,477	806	6808	524	56,472	4344	73,757
PS1WS2	15	61	9568	638	19,247	1283	85,724	5715	1,14,539

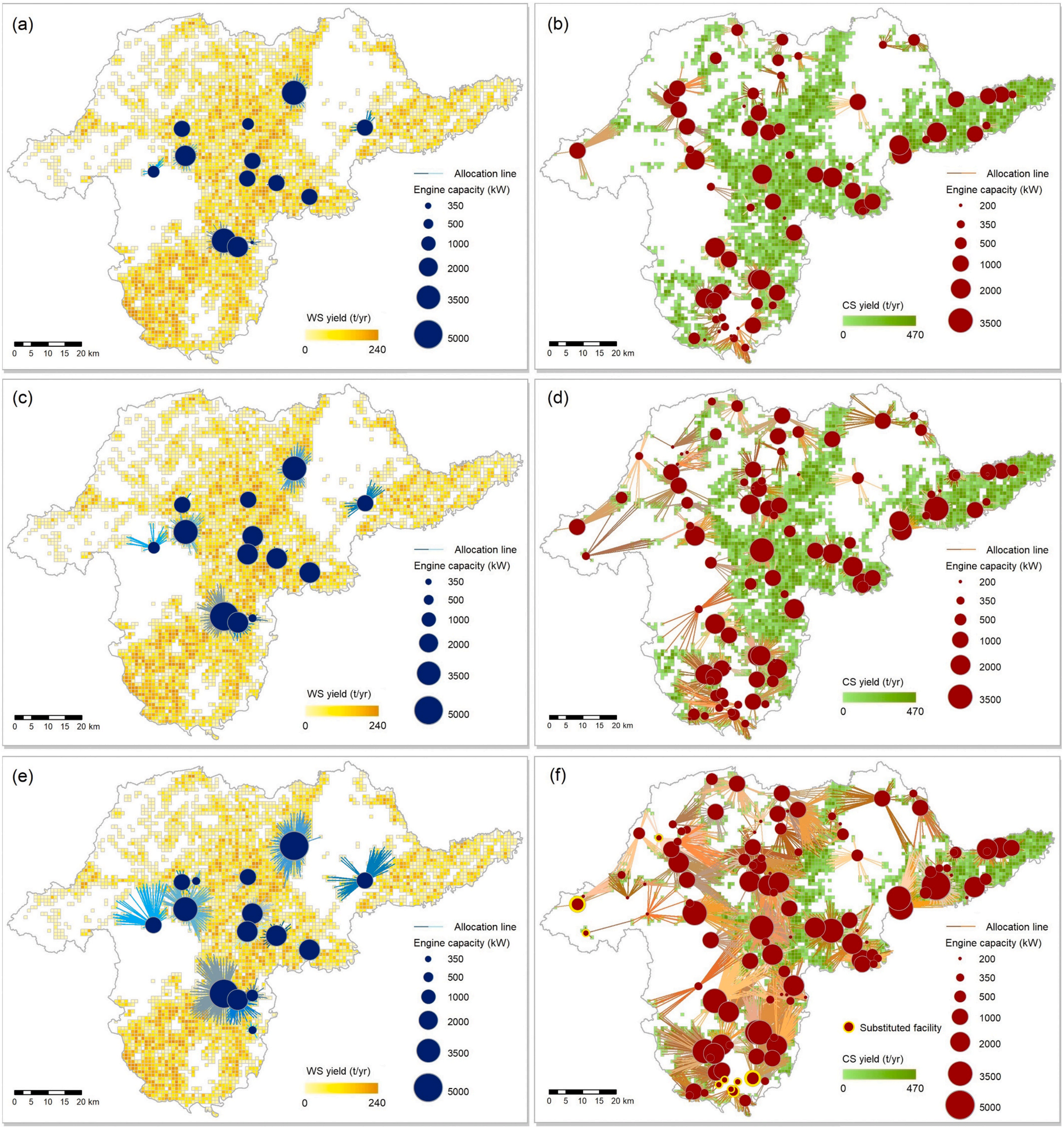

Fig. 3. Origin of crop residues and their allocation to facilities for scenarios (a) PS2WS1; (b) CM2CS1; (c) PS1WS1; (d) CM1CS1; (e) PS1WS2 and (f) CM1CS2. For allocation, actual transportation via the road network was considered. WS and CS stand for wheat straw and corn stalk, respectively.

appear in the case of AD plants near the border of the study area, located in low CR density areas or in high AD plant density areas. High specific transport needs can influence expenditures of feedstock transportation of AD plants, particularly in Scenario C1CS2, to different degrees according to the location of the facilities and their individual CS demand.

Transportation distances are high for the digestate in all scenarios, since the co-digested material contains not only the CR but also the manure. Digestate was assumed to be transported back to the fields where CS and WS were originated from in proportion to the amount of CR collected, according to the corresponding scenario (Table 5).

3.3. Energy potentials and economic feasibility

Results of the technical and economic assessment are presented in Table 6 by summarising the value of each AD plant in each scenario. Since the modelling resulted in that 9 sites could not be fully supplied with CR for CM1CS2, replaced facilities affect the PMY of the scenario. In that case, expected methane yield is 2% lower than the theoretical PMY.

Total installed capacity was computed to increase through the scenarios, according to the PMY. Even though gas engine size arrangement

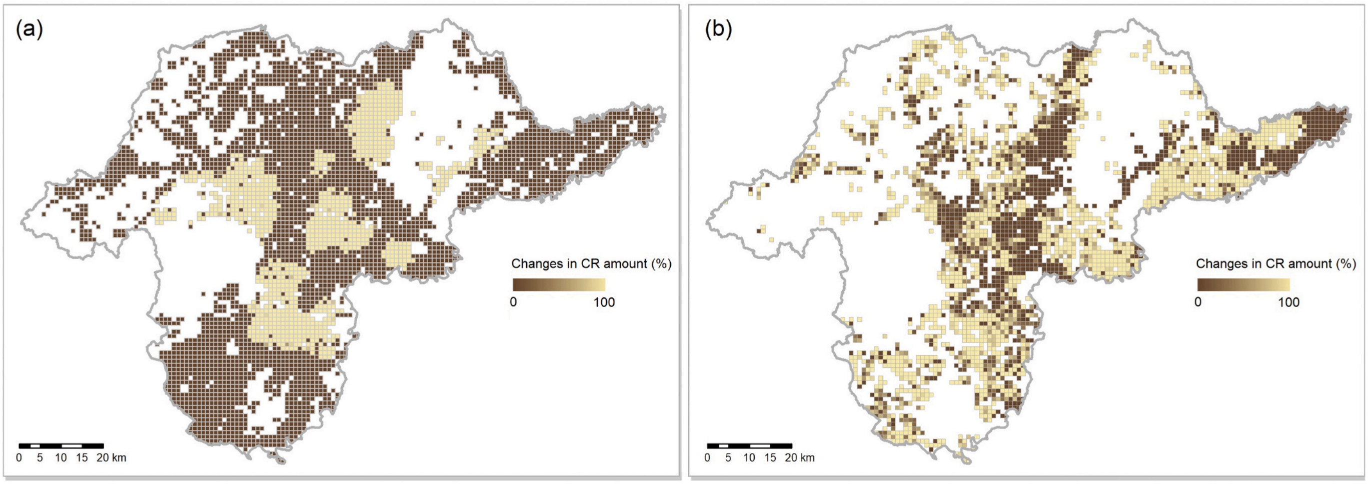

Fig. 4. Relative changes in available amounts of crop residues (CR) after the allocation process of (a) PS1WS2 and (b) CM1CS2 scenarios.

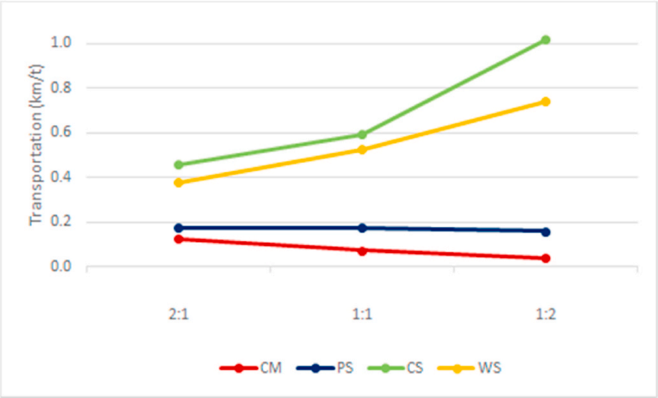

Fig. 5. Specific mean transport needs per one ton of raw material for each feedstock composition ratio. CM = cow manure; PS = pig slurry; CS = corn stalk; WS = wheat straw

is diverse, scenarios can be characterised by a tendency towards larger capacities as site specific methane yield increases with higher CRS.

The total electricity demand of the study area was 3.84 TWh in the year of observation [110]. Combining two different feedstock composition scenarios from the same CRS could supply 6.4%, 8.8% and 12.5% of the total electricity demand for 2:1, 1:1 and 1:2 ratios, respectively.

Average values of CapEx and OpEx are increasing as growing amounts of feedstock are utilised in the scenarios. Although the lower CRS scenarios result in rather smaller installed gas engine capacity AD plants and thus higher, subsidised revenues, economies of scale affects negatively these small facilities. The economic return of small biogas plants is uncertain, and most of these investments would not pay off even in a 25-years-long operation period. An exact economic feasibility threshold cannot be specified, but all sites with 100 kW_e and 200 kW_e, as well as most of the 350 kW_e engine capacity plants would be unprofitable, which concerns all scenarios. The share of non-recoverable AD plants ranges from 7.7% to 21.0% (over a 25-year-long period). The average discounted payback period was calculated by taking all AD plants in each scenario into account, resulting in high values because of the relatively high share of non-recoverable biogas facilities.

Fig. 6 shows the annual average specified cost distribution together with the annual share of CapEx, per MWh net electricity. CapEx share, $OpEx_{CHP}$ (the gas engine) and $OpEx_{Digester}$ costs decrease in line with economies of scale, which trend is well presented by the PS scenarios. For the CM1CS2 scenario, these components are higher than in CM1CS1, which can be explained by the higher share of non-recoverable facilities, in contrast with the obvious cost reduction of PSWS scenarios.

The share of transportation cost from $OpEx_{Total}$ decreases slightly, while feedstock costs increase significantly towards scenarios with higher CR demand. Total cost of T_{LM} would be less as more receiving sites and shorter routes are available (Fig. 7). Regarding $OpEx_T$, T_{CR} and T_{Dig} costs together account for 64%, 74% and 83% for CM2CS1, CM1CS1 and CM1CS2, respectively. These proportions are slightly lower in cases of PSWS scenarios, namely 57%, 62% and 71% for PS2WS1, PS1WS1 and PS1WS2, respectively. Proportional differences between the two feedstock composition types can mainly be attributed to the cumbersome CS supply.

4. Discussion

4.1. Result evaluation

The results show that large quantities of CR and LM could be utilised for energy recovery purposes in the study area, whilst their spatial distribution is diverse. Despite Scenarios PS2WS1 and CM1CS1 having the most favourable UMY values within their corresponding feedstock

Table 6
Technical and economic characteristics of the selected scenarios. PMY = potential methane yield.

Scenario name	PMY (1000 m^3)	Capacity (MWe)	Net electricity (MWh)	Average values				Non-recoverable biogas plants
				$CapEx_{Total}$ (1000 €)	$OpEx_{Total}$ (1000 €/yr)	$Revenue_e$ (1000 €/yr)	Discounted payback period (yr)	
CM2CS1	49,253	56.6	1,83,847	1526	107	211	19.3	14 (20.0%)
CM1CS1	68,534	74.8	2,55,974	1509	110	226	17.9	15 (16.5%)
CM1CS2	99,063	114.5	3,70,320	1707	129	254	20.8	24 (21.0%)
PS2WS1	16,350	18.2	61,935	2335	189	372	17.4	1 (7.7%)
PS1WS1	21,726	25.9	82,510	2924	247	482	15.7	1 (7.7%)
PS1WS2	29,418	28.7	1,11,673	3192	293	566	16.3	2 (13.3%)

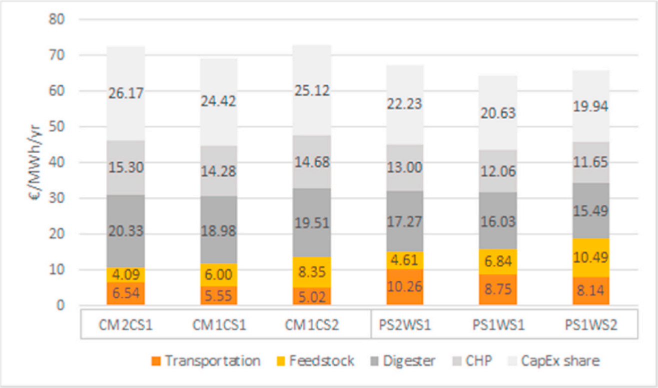

Fig. 6. Average specified cost distributions with the annual share of capital expenditures (CapEx).

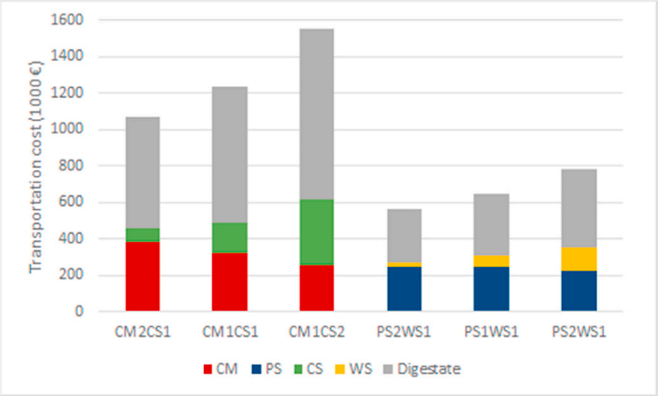

Fig. 7. Total transportation cost of the feedstock and the digestate. CM = cow manure; PS = pig slurry; CS = corn stalk; WS = wheat straw

compositions, scenarios with the highest CRS have higher PMY for the simple reason that more raw materials could be utilised. Therefore, these scenarios are preferred; however, it may cause difficulties regarding resource acquisition. Considering low CRS scenarios, there may be supply difficulties, i.e. supply areas overlap in two cases: in areas where CR availability is insufficient near a given AD plant; and in areas where large quantities of CR are available, although there are a large number of AD plants operating in the area. These problems appear in several parts of the study area in the CM2CS1 scenario. In line with growing CR demand, e.g. in the case of CM1CS2 scenario, tensions increase in these two types of areas, but are not limited to them. Expanding and overlapping supply areas have a further impact and create conflicts in otherwise trouble-free locations as well.

Average CR transport distances are found to be of the same magnitude as the findings of [111]. WS and CS demands can be covered in most scenarios, but as the number of AD plants and their CR demand increase, the struggle for resources unfolds. In the case of the CM1CS2 scenario, CS needs can only be met theoretically. Yet, the allocation process resulted in that in practice it is impossible to fulfil the demands for certain facilities due to the insufficient amount of CS in their vicinity. It indicates that under certain allocation conditions, the study area can reach its sustainable capacity limit from a resource management viewpoint. This is similar to the findings of [42], where CRS was high in the proposed feedstock mixture. Despite the less favourable UMY values for mixing CM with WS, it may still be worthwhile to attach more importance to this feedstock composition in order to balance the demand between WS and CS. Furthermore, it is likely that each site would use different feedstock mixing ratios, which may reduce the supply conflict to some extent.

The size of the facilities, their location, and the concept in which their LM delivery areas were merged in the present methodology have decisive influence on the results of the WS and CS allocation efficiency. Together with the spatial availability of the CR, these factors are associated with higher transportation costs and the conflict of supply area overlaps as CRS increases. Although many farm owners would have the possibility to order CR from a close range, in practice owners rather transport it from their own fields from larger distances, as it is the case e.g. for livestock bedding purposes in the study area. It means, theoretically, the digestate should be taken back to the origin of both CR and livestock feedstuff, making the process environmentally and economically inefficient [112,113]. This mostly affects scenarios in which CRS is high. In these scenarios, costs of T_{CR} and T_{Dig} exceed the cost of T_{LM} so much that it can influence the initial AD plant site selection. Meanwhile, many agricultural quadrats are not allocated to any proposed biogas facility; therefore, abundant amounts of CR are still available for biogas production or other utilisation purposes.

4.2. *Indirect effects of co-digestion on planning*

According to the results, initial AD plant site selection by LM source merging had a crucial role for further raw material allocation. Transporting manure to the nearest appropriate receiving site results in the lowest costs and emission. Nevertheless, without knowledge of the CR allocation possibilities and the competitor facilities in the vicinity of the proposed receiving AD plant, this type of logistic method leads to unfavourable results. In the case of AD plants where adequate CR supply is inherently difficult, LM transportation from other locations would exacerbate the problem. This especially concerns scenarios with high WS or CS demand, where transport costs of CR and digestate outweigh transport costs of LM. In such a situation, LM transport has a subordinate role; therefore, its allocation towards areas properly resourced with CR is recommended. Results showed that in certain regions in the study area, significant amounts of cereal straws would remain unutilised. Proposed biogas facilities that would apply high CRS in the feedstock composition should prefer these regions during site selection in order to reduce T_{CR} and T_{Dig}, thus the $OpEx_T$. Moreover, it would result not only in more homogeneous AD plant distribution, but also in less conflict of overlapping supply areas.

Several studies emphasize the importance of manure transport reduction as an important efficiency factor in the biogas production chain [101,102]. These research papers, however, focus on manure or other mono-digested feedstock solely, which makes planning and site selection easier, since they are point sources [114–116]. The present study points out that this strategy is inappropriate when large quantities of co-substrates with different spatial availability properties are also considered. Nonetheless, transportation of LM in order to apply the positive effects of economies of scale may be still favourable. Appropriate site selection efforts may justify the transport of manure over longer distances.

Regarding CR supply, occurrence of overlaps and shortages are prevented in similar studies because their supply areas have been determined initially and demands are constant, i.e. they have not been allowed to exceed local raw material potentials [39,117]. In contrast, the current methodology allows more flexible feedstock acquisition, while also taking land application of the digestate into account. Surprisingly, digestate management, especially transportation, is a neglected issue in most studies [118], or digestate is considered only to be transported back to point sources, e.g. processing facilities [119]. Yet, Skovsgaard and Jacobsen [69] confirm that it has a crucial role in OpEx reduction and therefore, AD plant site selection and scaling.

The unfavourable economic results of each scenario can clearly be attributed to the high number of small AD plants, but additional financial support can help make the small-scale plants competitive. Decentralised facility distribution is favourable, but in terms of $OpEx_T$, the advantage of lower T_{LM} costs is not as significant as the cost increment due to the growing CR demand.

If certain AD plants could only meet their CR demands by delivering over longer distances, there is a risk of dedicated energy crop cultivation or CR removal beyond sustainability limits. Note that straw used for animal bedding can reduce the amount of CR demand in each feedstock mixture scenario, but it was not taken into account due to uncertainties. In order to avoid resource shortage, mapping other competitive uses of CR is also important by taking other digestible organic materials into account. Besides agricultural waste, it is important to consider other feedstock sources, such as organic share of municipal waste or organic industrial by-products, which can reduce CR-dependence [120].

5. Conclusions

Biogas-related studies often deal with livestock manure and its logistics in GIS-based analyzes, since the spatial distribution of manure has a crucial role in AD plant site selection and scaling. However, its co-digestion with other substrates, such as crop residues, and its possible effect on spatial feedstock allocation and energy planning is less investigated.

The methodology presented in this paper designates AD plant sites from a least manure transport intensive viewpoint, in line with similar studies. The methodology also considers the beneficial effects of co-digestion for different feedstock compositions, and allocates crop residues to the proposed facilities in varying proportion, which is a novel approach in the field of biogas feedstock allocation modelling.

The results showed that a high share of crop residue utilisation should be promoted for planned biogas facilities in order to achieve higher gas yield. Yet, high crop residue demand may result in overlapping biomass supply areas of the competing AD plants, which can lead to feedstock shortages. Many facilities have to satisfy their crop demand from greater distances, which results in increased transportation costs, especially if the land application of the digestate at its origins is also considered. By analysing the geographical patterns of the proposed AD plants and the feedstock transportation routes together with the results of technical properties and economic feasibility, it can be concluded that these issues are clearly attributed to the inappropriate manure-focused site selection approach, which neglects the crop residue supply during the planning phase. The results draw attention to the importance of regional resource management regulation and spatial energy planning, in order to avoid possible tensions and preserve strict sustainability limits of biomass utilisation.

Author statement

Tamás Soha: Conceptualization, Methodology, Software, Formal analysis, Investigation, Resources, Writing – original draft, Writing – review & editing, Visualisation, Project administration. Luca Papp: Conceptualization, Investigation, Resources, Visualisation. Csaba Csontos: Formal analysis, Writing – review & editing. Béla Munkácsy: Writing – review & editing, Project administration, Supervision.

Declaration of competing interest

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

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