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Energetic Potential for Biological Methanation in Anaerobic Sewage Sludge Digesters in Austria

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Abstract: Biological methanation as a method of sector coupling between electric and gas grids is expected to be an integral part of the green energy change. Wastewater treatment plants (WWTPs) involving anaerobic digestion (AD) allow existing infrastructure to operate as energy conversion plants, to close carbon cycles and to generate long-term storable energy in the form of biomethane. Therefore, municipal raw sludge and additional organic residuals (co-substrates) are converted into biogas. Hydrogen is added to convert the carbon dioxide in the biogas into methane via biological methanation (BM). In this study, the energy amount that is convertible via BM in municipal digesters in Austria was calculated. The amount of energy, which can be transformed from electric surplus energy into biomethane, was assessed. Operational data from lab-scale digesters were combined with data from 28 Austrian full-scale wastewater treatment plants with AD. They represent 9.2 Mio population equivalents (PE), or 68% of Austria's municipal AD capacity for WWTPs > 50,000 PE (in sum, 13.6 Mio PE). Energy flows for BM including water electrolysis and anaerobic digestion were created on a countrywide basis. It was found that 2.9–4.4% (220–327 GWh·y^{-1}) of Austria's yearly renewable electricity production (7470 GWh·y^{-1}) can be transformed into biomethane via BM in municipal digesters.

Keywords: anaerobic digestion; biological methanation; carbon dioxide; energy conversion; hydrogen; methane; renewable energy storage

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1. Introduction

The share of fluctuating renewable energy sources, such as wind and photovoltaics, is increasing worldwide. In Austria, the number of wind turbines doubled within 7 years (from 2011 to 2018) from 662 to 1313, while the installed electrical power increased from 1099 to 3045 MW [1]. Especially in the eastern part of Austria, large numbers of wind turbines have been installed (Lower Austria 729 and Burgenland 429). To reach a fully renewable electricity supply, further systems are needed. With an increasing share of fluctuating renewable energy production, network stabilization and long-term energy storage are urgently needed.

Countrywide, 54 storage and pump storage hydropower plants with a total installed power of 8.8 GW and a storage capacity of 9.3 TWh (3 TWh in pump storage reservoirs) are in operation [2,3]. Resch et al. [4] reported that the storage capacity in hydropower plants in Austria is almost fully exploited, and this capacity is needed for medium-term energy storage from days up to weeks. The existing natural gas infrastructure could be utilized for long-term or seasonal storage ranging from weeks up to months.

Due to its location in central Europe, Austria is a transit land for natural gas. Approximately 49 × 10^9 m^3 of natural gas is transported through the country per year. An energy amount of 551 TWh of natural gas was imported, 10 TWh was produced in Austria, 430 TWh was exported, 69 TWh was stored, 36 TWh was discharged from the storage and 91 TWh was given to end customers/consumed in the year 2020 [5]. From 10 TWh of natural gas produced in Austria, 0.152 TWh (1.5%) is derived from renewable biogas. To achieve the goals of Austria's national energy strategy, biomethane production must increase dramatically from approximately 0.16% (152 GWh$\cdot$y^{-1}) of the natural gas demand in the year 2020 to 10% in the year 2030 [6].

Austria's natural gas storage capacity with a volume of 7794 $\times$ 10^6 m^3 (87.2 TWh) is the fourth largest in Europe [7]. The countrywide gas demand of approximately one year is storable in this gas storage. The maximum storage rate is 2.7 $\times$ 10^6 m^3$\cdot$h^{-1} (30.6 GW), and the maximum discharge rate is 3.3 $\times$ 10^6 m^3$\cdot$h^{-1} (37.4 GW) [8]. For dimension conversion between energy content and natural gas volume, a factor of 11.19 kWh/m^3 was applied according to e-control (2021) [5]. When comparing energy storage in pump storage plants and gas storage facilities, the different efficiencies in the conversion into electricity must be considered. While pump storage plants reach efficiencies between 70 and 85% for electricity storage and discharge, for the energy that is stored in gas storage facilities, a recovery efficiency between 35 and 40% for electrical usage and approximately 80% for electrical and thermal usage, e.g., in combined heat and power plants, must be considered.

Table 1 provides an overview of Austria's countrywide energy storage capacities in hydropower plants and gas storage facilities.

Table 1. Austria's energy storage capacities in hydropower plants and gas storage facilities.

			Hydropower (114 Plants)		**Gas Grid (9 Storage Facilities)**	**Sum**
			Storage Plants (60)	**Pump Storage Plants (54)**		
power	storage	(GW)	-	3.4 (2)	30.6 (4)	34
	discharge	(GW)	5.4 (1)	3.4 (2)	37.4 (4)	46.2
capacity	storage	(GWh)	3000 (1)		87,200 (3)	90,200

(1) e-control (2020) [3], (2) Austria Forum (2020) [2], (3) European Union (2014) [7], (4) Gas Infrastructure Europe (2012) [8].

In times of high renewable electricity production in Austria, a maximum of 3 GW can be exported to neighboring states via the high-voltage network [4], and 3.4–4.9 GW can be stored using pump storage plants [2,4]. The plan is to expand the electricity long-distance transport lines to 11 GW within the next few years.

Excess electricity production, which cannot be exported, during times of high production in the neighboring countries and due to limited transport capacities, can be used to produce hydrogen and long term-storable biomethane using biological methanation. Thus, a seasonal load shift of excess energy from summer to wintertime with high energy demand is feasible.

The centerpiece of the so-called Power-to-Gas (PtG) concept is the production of hydrogen and oxygen via water electrolysis (Equation (1)) using renewable surplus energy from wind turbines and photovoltaic systems. Schäfer et al. [9] reported that the produced oxygen can be used in other processes, such as aeration in the biological wastewater treatment or ozone production to remove organic trace substances via ozonation.

$$H_2O \rightarrow H_2 + 0.5O_2 \; (\Delta H^0 = + \, 285.9 \text{ kJ}\cdot\text{mol}^{-1}) \tag{1}$$

Fu et al. [10] noted that hydrogen gas is an attractive option to decarbonize the present energy system and to extend the usage of the existing gas infrastructure.

Due to the legal framework in Austria, direct injection of pure hydrogen into the natural gas grid is not possible, but the injection of methane gas containing up to 10% hydrogen has been allowed since June 2021. Due to technical reasons such as the low calorific value and hydrogen diffusion through pipelines, a direct injection also does not appear useful. Therefore, using hydrogen to upgrade CO_2 contained in biogas to biomethane can be an attractive alternative.

To reach climate and energy goals determined in Austria's national climate and energy strategy, hydropower pump storage plants should be used for grid stabilization and shortterm storage in phases of excess electricity production. Technologies such as Power-to-Gas should be used for seasonal energy storage, by utilizing existing infrastructure, such as the natural gas grid and gas storage facilities [6].

2. Biogas Quality and State of the Art in Biogas Upgrading

2.1. Biogas Composition

The anaerobic digestion (AD) of sewage sludge is a proven and worldwide established technique with which to generate biogas for energetic use with an enormous potential worldwide [11]. Biogas and biomethane are believed to be building blocks for achieving climate and energy goals, as a replacement for natural gas for use in high-temperature processes and as a possibility for long-term energy storage. In general, the composition of biogas depends on the substrate used in anaerobic digestion and the mean oxidation state of the carbon in the substrate [12]. A typical composition of biogas from AD contains CH_4 (53–70%), CO_2 (30–47%), H_2O (5–10%), N_2 (0–3%), O_2 (0–1%) and traces of H_2S, NH_3, hydrocarbons, total chlorine (HCl and CH_3Cl explained as Cl) and siloxanes (0–10,000 ppm, 0–100 ppm, 0–200 mg·m^{-3}, 0–5 mg·m^{-3} and 0–40 mg·m^{-3}) [13,14]. Biogas from AD can contain traces of H_2; when the digestion process is disturbed, unbalanced, or co-substrates are digested, the percentage of H_2 can rise to 3% [13].

Quality standards for biogas injection into the natural gas grid vary between different countries. Muñoz et al. [15] report that in the Netherlands, injected biogas must contain 80% CH_4. In most other European countries, stricter quality standards must be fulfilled. The costs for a biogas upgrade strongly depend on these quality standards.

2.2. Quality Standards for Biogas Grid-Injection in Austria

In Austria, the biogas that is injected into the natural gas grid must fulfill natural gas quality standards at the injection point, which are regulated by two guidelines. Guideline G31 regulates the natural gas quality [16], and Guideline G33 regulates the biogas injection into the gas grid [17]. In Table 2, typical biogas composition and quality standards for natural gas grid injection in Austria are compared.

Table 2. Typical biogas composition and Austrian quality standards for biogas injection.

Parameter		Unit	Biogas Composition	Quality According Austrian Standards ÖVGW G31 [16] and ÖVGW G33 [17]
methane	CH_4	(%)	53–70	>97
carbon dioxide	CO_2	(%)	30–47	$\leq$2
oxygen	O_2	(%)	0–1	0.5
nitrogen	N_2	(%)	0–3	$\leq$5
ammonia	NH_4	(mg·m^{-3})	<100	technical free
hydrogen sulfide	H_2S	(mg·m^{-3})	<1000	$\leq$5
water—dewpoint	H_2O	(°C)	$\leq$37 °C, 1 bar	$\leq$−8 °C, 40 bar
calorific value	Hi	(kWh·m^{-3})	6.7–8.4	10.7–12.8
Wobbe Index	Wi	(kWh·m^{-3})	6.9–9.5	13.3–15.7

Injected biogas must meet methane concentrations of >96% v/v (or >97% v/v if the rest are inflammable gas components) and a minimum calorific value of 10.7 kWh·m^{-3} at the point of injection [16]. To reach the required methane concentration value, biogas must be upgraded. To reach the required minimum calorific value and Wobbe Index, additional gas with a higher calorific value than methane must be added before grid injection. Often, ethane (Hi = 16.37 kWh·m^{-3}) or propane (23.22 kWh·m^{-3}), so-called mix-gases, are used to raise biogas' lower energy content (9.17 kWh·m^{-3}) to the minimum of 10.7 kWh·m^{-3} required for injection. In June 2021, a new guideline (Guideline G B210 regenerative gas—biogas) for biogas injection was validated. This guideline allows higher hydrogen concentrations of up to 10% H_2 and lower methane concentrations in injected biogas, which will simplify the requirements for biogas production to promote biologically upgraded technologies, using added hydrogen [18]. Nevertheless, the calorific value and Wobbe Index will have to meet former requirements. Due to hydrogen's low calorific value of 2.66 $kWh \cdot m^{-3}$, depending on the concentration in the upgraded biogas, consequently, more high caloric mix-gases will need to be added.

Further legal simplifications for biomethane grid injection are currently being discussed to reach the goal of Austria's national energy strategy, which claims a share of 10% (approximately 800 Mio $m^3 \cdot y^{-1}$) of the biogas in the natural gas grid, by the year 2030. In addition to increased biogas production and upgrading, long-term storage in the natural gas network is also promoted [18].

2.3. State of the Art in Biogas Upgrading to Quality Standards Required for Grid Injection

Muñoz et al. [15] reported that there are different techniques for upgrading biogas to reach the required quality for injection into the natural gas grid. Two principles can be distinguished, which are compared in Figure 1: first, CO_2 separation and removal, and second, CO_2 conversion through methanation, by adding additional hydrogen to reach quality standards in natural gas grids.

2.3.1. Biogas Upgrading via CO_2 Separation and Removal

The separation and removal of the CO_2 contained in the biogas with thousands of applications worldwide is very common. Pressure swing adsorption (PSA), water scrubbing, chemical scrubbing and membrane separation techniques are used to remove the contained CO_2 to reach methane concentrations >97%. By using cryogenic CO_2 separation, other impurities such as H_2O and H_2S, can also be removed in a single step. Biological CO_2 fixation using microalgae is also a possibility to remove CO_2 from biogas, but it requires sunlight for biomass growth. During these biogas upgrade processes, the CO_2 removed from the biogas is usually released into the atmosphere and lost for further reactions. Specific upgrading costs strongly depend on the upgrade capacity and the technology used and vary between 0.02 and 0.1 $EUR \cdot m^{-3}$ biomethane [15]. Additionally, 0.5–5% of the energy contained in the biogas is needed for the upgrade process. Furthermore, all the CO_2 separation and removal processes come with a methane slip, which has to be considered in the carbon footprint.

2.3.2. Biogas Upgrading with CO_2 Conversion through Methanation

The use of additional hydrogen (H_2) for a conversion of the contained CO_2 into CH_4 via methanation (Equation (2)) offers the opportunity to close carbon cycles and to produce storable biomethane in one step. The Sabatier process is an approved thermo-chemical high-temperature–high-pressure method for CO_2 conversion operated at 300–400 °C and 30 bar using nickel, ruthenium-alumina catalysts. With its high energy consumption and high-demand reaction conditions, it is only feasible on a large scale, for example, in petrochemical refineries.

An alternative method is a biological methanation using hydrogenotrophic archaea. Kim et al. [19] consider biological methane production as a more environmentally friendly method for carbon dioxide reduction than physical and chemical methods. To benefit from the CO_2 content in the biogas, additional hydrogen is used to produce additional methane.

There are two common techniques for the biological methanation of carbon dioxide (CO_2) to methane (CH_4) providing externally produced hydrogen (H_2) in situ and ex situ. During in situ methanation, the biogas reactor (digester) is used for BM, while during ex situ methanation, an additional methanation reactor is operated. BM is used to upgrade biogas to biomethane or synthetic natural gas (SNG) and to reduce CO_2 emissions [15]. Schäfer [20] supposed that as part of a green energy transition and to increase the share of flexibly produced renewable energy, a transformation of the energy system is necessary. WWTPs, including anaerobic digesters and gas infrastructure, can serve as a building block for the green energy transition. Existing digesters can be operated as in situ methanation reactors to increase the methane content of the biogas by adding CO_2- neutral produced hydrogen. Equation (2) shows the stoichiometric equation of the methanation reaction.

$$4\,H_2 + CO_2 \rightarrow CH_4 + 2\,H_2O\ (\Delta G^0 = -130.7\ kJ\cdot mol^{-1}) \quad (2)$$

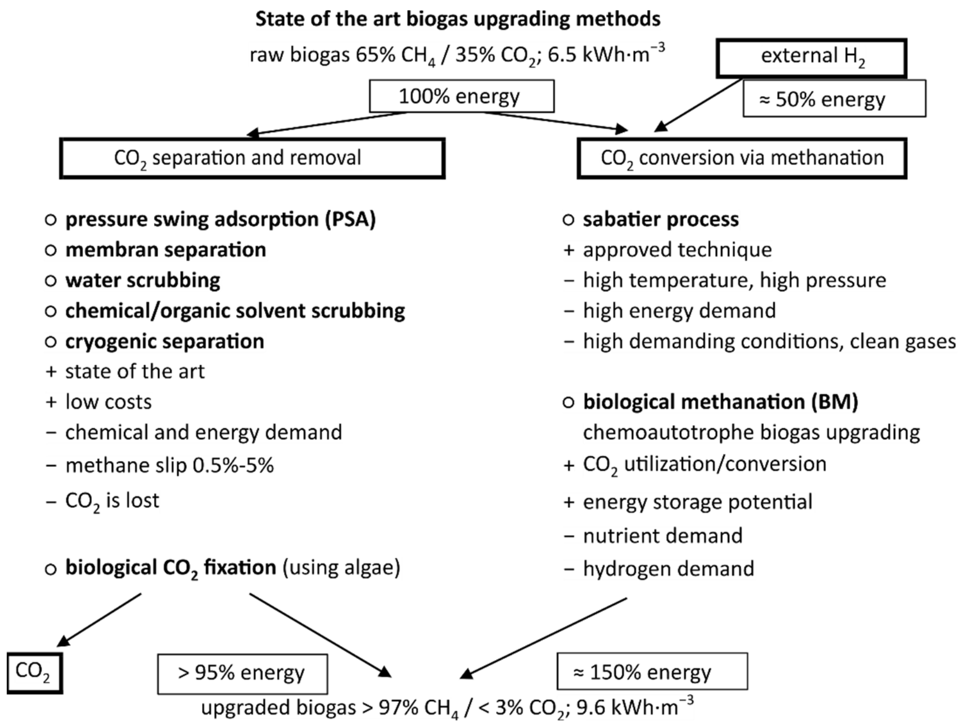

Figure 1. Different biogas upgrading methods according to Muñoz et al. 2015 [15].

Different reactor types for BM, such as continuously stirred tank reactors (CSTRs) [21], bubble columns [19], up-flow anaerobic sludge blanket (UASB) reactors [22,23], packed bed columns, fixed bed reactors [24], trickle-bed reactors [25–27] and stirred tank reactors (STRs) with gas sparging via membranes [28,29], have been examined in recent years.

Based on biogas containing 65% CH_4 and 35% CO_2, biomethane with >96% CH_4 (the rest being H_2 and CO_2) can be produced via both methanation principles. The calorific value is thereby increased by 50% from 6.5 to 9.6 kWh·m^{-3}. Especially large WWTPs provide the framework necessary for BM. They offer trained and experienced employees, and safety and infrastructure equipment, such as explosion protection, robust electricity and gas–grid connections. Additionally, large quantities of CO_2 are available in high concentrations, which can be transformed into methane.

Currently, the biogas produced in Austria is usually not upgraded due to the high effort and low market price and rather is burnt in combined heat and power plants (CHPs) for direct electricity and heat production. Currently, only two biogas upgrade plants are operated at Austrian WWTPs.

2.4. Biogas Production in Agricultural Biogas Plants and Sewage Sludge Digesters in Austria

Biogas production from organic waste, wastewater and energy crops is a worldwide established technique with an increasing amount of biogas and biogas plants each year. Data for the biogas production on a countrywide basis are available for the 14 member states of IEA Bioenergy Task 37 (Energy from Biogas). In Table 3, the total biogas production, the number of agricultural biogas plants and sewage sludge digesters and the share of biogas produced in ADs at WWTPs are displayed with data reported from the International Energy Agency (IEA 2020) [30]. Data for Norway were added from the IEA (2015) [31]. In total, 102,559 GWh·y^{-1} of energy is generated as biogas, and 12,885 GWh·y^{-1} (13%) is the share of biogas generated from sewage sludge digestion.

Table 3. Total biogas production and biogas production in WWTPs, data from IEA, 2020 [30], and data for Norway IEA, 2015 [31].

Country	Reference	Total Biogas Production from Sewage Sludge, Industrial Wastewater, Biowaste, Aggricultural Residuals and Landfills	Number of Biogas Plants Agricultur and Sewage Sludge AD's	Biogas Production in WWTPs Sewage Sludge Digestion only		Number of WWTPs Including AD
	Year	$GWh \cdot y^{-1}$		$GWh \cdot y^{-1}$	% of Total	#
Australia	2017	1587 (3)	242	381 (3)	24%	52
Austria	2020	1613 (1)	287	n.a.	n.a.	n.a.
Brazil	2016	5219	165	210	4%	10
Canada	2019	n.a.	150	n.a.	n.a.	31
Denmark	2018	3723	172	308	8%	51
Finland	2017	692	96	126	18%	16
France	2017	3527 (2)	687	442 (2)	8%	88
Germany	2019	52,158 (2)	10,551	3657 (2)	7%	1274
Norway	2010	500	129	164	33%	129
South Korea	2017	2815	119	630	38%	36
Sweden	2018	2044	280	715	40%	138
Switzerland	2018	1454	634	633	49%	473
The Netherlands	2018	3465	262	640	20%	80
United Kingdom	2018	23,762 (1)	994	4266 (1)	11%	163
		sum	sum	sum	mean	sum
		102,559	14,640	12,885	23%	2678

(1) calculated from electricity production with $\eta_{EL} = 0.35$; (2) electricity + heat production; (3) calculated from installed capacity.

For Austria, no detailed data about the number of sewage sludge digesters and their biogas production are available in this dataset. Additionally, no data for the total biogas and energy production from sewage sludge are available to calculate the share of the total biogas production. To overcome this lack of information, a detailed assessment was necessary to obtain the number of WWTPs using AD from different additional sources.

3. Materials and Methods

3.1. Survey on Anaerobic Sewage Sludge Digestion in Austria

In 2019, 136 WWTPs including AD with a design capacity > 20,000 PE and 58 with a design capacity > 50,000 PE were operating in Austria [32,33]. In 2020, Austria's largest AD at the Vienna main WWTP (4 Mio PE design capacity) started its operation. Since then, countrywide, a total of 164 ADs with a capacity of approx. 15.1 Mio PE or 70% of the Austrian WWTP design capacity (total 21.49 Mio PE capacity) have been in operation to treat sewage sludge. The 59 WWTP > 50,000 PE-operating ADs (design capacity of 13.6 Mio PE) were identified as candidates for implementing BM in municipal digesters.

Before this study, no detailed data for Austria's entire biogas production from sewage sludge digestion at WWTPs were available. Only the total number of agricultural and sewage sludge digestors and their total energy production were available (Table 3). To overcome this lack of data, a survey was conducted using an email questionnaire and telephone interviews at 30 Austrian WWTPs including AD. The survey data were returned and available for 28 of the 30 interviewed WWTPs. Thirteen questions were asked about the operational data, digester's design details and infrastructure connection of the plants. The plant's name and catchment area, design capacity based on chemical oxygen demand (COD), yearly average COD load, number of digesters in operation, digester volume, daily average gas production, raw sludge and co-substrate loads, co-substrate types and the existing gas infrastructure (connection to the natural gas grid) were evaluated. Informal additional information was also considered, e.g., the usage of flotation instead of conventional primary treatment.

By using representative survey data from 28 of the 137 WWTPs including AD, it was possible to calculate PE-specific biogas production rates and the total biogas production for all anaerobic sewage sludge digesters in Austria. These data were used to calculate the yearly energy production. Using the PE-specific digester volume, the maximum BM capacity for Austria was calculated. Efficiency factors for water electrolysis were obtained from the literature. Efficiency factors for biological methanation were obtained from the literature [34] and own lab-scale experiments [35]. Using these efficiency factors and the yearly produced CO_2 load from biogas production, the producible amount of energy through BM was calculated. A sensitivity analysis was performed for the following three different scenarios: Scenario 1: all WWTPs > 50,000 PE (n_1 = 59); Scenario 2: all WWTPs > 20,000 PE (n_2 = 137); and Scenario 3: all WWTPs including AD in Austria (n_3 = 164) are operating BM.

Additionally, 151 agricultural biogas plants with a total of 565 $GWh \cdot y^{-1}$ of electricity and 350 $GWh \cdot y^{-1}$ of heat production were operated in Austria in 2019 [36]. These agricultural biogas plants were not considered in the following calculations, because they are optimized to electricity production and most of them are located in rural areas without a gas grid connection in their surroundings.

3.2. Energy Flows for Electricity, and Bio- and Natural Gas, at a National Level

To create an energy flow diagram (SANKEY-diagram) at a national level for Austria for electricity, and natural and biogas, the Software STAN (substance flow analysis) [37] was used. Different sources were applied as input data. National statistical data about energy production and the transport of electricity, and natural and biogas [36,38,39], as well as international statistical data [30,31] for sectoral energy production, energy import and export, were combined.

As described above, three different scenarios were considered. For these three scenarios, transferable energy loads via BM were calculated using transformation efficiency from the literature and lab-scale tests. Efficiency for water electrolysis was set to η_{EL} = 0.9 according to Muñoz et al. [15]. According to the observed methane production in continuous lab-scale tests using in situ BM, the efficiency for biological methanation was set to η_{BM} = 0.7 (0.7 mol CH_4 is produced, consuming 4 mol H_2 according to Equation (2)).

3.3. Maximum Volume-Specific Biological Methanation Rate

To calculate the maximum volume-specific capacity of an AD, a volume-specific biological methanation rate (BMR) of 0.3 L $CH_4 \cdot L^{-1} \cdot d^{-1}$ was considered according to values from the literature for in situ BM in CSTRs. Luo and Angelidaki (2013b) [40] reported a specific BMR of 0.34–0.39 L $CH_4 \cdot L^{-1} \cdot d^{-1}$ and 96.1% CH_4 at thermophilic conditions (55 °C), while Tauber et al. (2020) [35] reported a BMR of 0.29–0.43 L $CH_4 \cdot L^{-1} \cdot d^{-1}$ for in situ BM in a lab-scale sewage sludge digestor at mesophilic temperatures (38 °C) and a hydraulic retention time of 25 d. The maximum BM capacity (BM_c) was calculated, using the BMR, the PE-specific digester volume (sV_{ADvol}) and the design capacity of the AD based on COD (PE_{design}) (Equation (3)).

$$BM_c = BMR \cdot sV_{ADvol} \cdot PE_{design}\ [W] \tag{3}$$

4. Results

4.1. Survey Data

Survey data from 28 Austrian WWTPs including AD with a design capacity between 24,500 PE (based on COD_{120}; 120 g $COD \cdot PE^{-1} \cdot d^{-1}$) and 4 Mio PE was evaluated (Table 4). Additional data for WWTP #28 was added from ebswien (2021) [40]. The yearly average inflow COD load varied between 1272 (10,600 PE) and 374,400 kg $COD \cdot d^{-1}$ (3,120,000 PE).

This indicates relative COD inflow loads between 43 and 125% of the design capacity (70% on average). Most of the plants, especially >100,000 PE, are operated between 60 and 85% of their design capacity.

Table 4. Organic capacity, loading and data from anaerobic digestion, from 28 Austrian WWTPs in the year 2020.

WWTP	Design	Inflow	Relative	Digesters	Digester	Co-Substrate	Specific	Specific	Gas Grid	Yearly
	Capacity	Load	Load	Number	Volume	Share	Gas Production	AD Volume	Connection	Gas Production
#	(PE_{COD})	(PE_{COD})	(%)	(-)	(m^3)	(% COD)	($L \cdot PE^{-1} \cdot d^{-1}$)	($L \cdot PE^{-1}$)	(Yes / No)	($m^3 \cdot y^{-1}$)
1	24,500	10,600	43	1	1690	0	24.91	68.98	No	96,360
2	45,000	22,200	49	1	1300	36	38.91	28.89	Yes	315,279
3	62,500	30,012	48	1	1600	5	31.29	25.60	Yes	342,768
4	65,000	48,690	75	1	1800	0	14.82	27.69	Yes	263,394
5	71,670	63,070	88	2	3200	0	26.11	44.65	Yes	601,088
6	90,000	38,850	43	2	5000	5	38.92	55.56	Yes	551,953
7	100,000	60,000	60	2	7000	0	21.67	70.00	Yes	474,500
8	100,000	-	-	2	4600	-		46.00	-	730,000
9	100,000	91,663	92	1	2500	0	12.69	25.00	-	424,680
10	110,000	75,000	68	2	5500	0	22.32	50.00	Yes	610,946
11	120,000	150,000	125	3	8250	43	56.32	68.75	Yes	3,083,520
12	120,000	75,309	63	2	5400	47	75.53	45.00	Yes	2,076,263
13	130,000	55,000	42	2	4800	5	21.82	36.92	No	438,000
14	130,000	78,300	60	2	3000	0	23.51	23.08	Yes	672,000
15	135,500	108,300	80	2	6000	6	31.39	44.28	Yes	1,241,000
16	150,000	-	-	3	7200	-		48.00	-	1,095,000
17	160,000	150,000	94	2	9000	0	23.74	56.25	Yes	1,300,000
18	167,000	138,000	83	2	5000	23	36.72	29.94	Yes	1,849,384
19	170,000	110,000	65	2	5000	0	14.55	29.41	No [(1)]	584,000
20	200,000	170,000	85	2	11,000	0	29.01	55.00	n.a.	1,800,000
21	255,000	110,000	43	2	6000	0	24.91	23.53	Yes	1,000,000
22	260,000	102,000	39	2	10,500	45	49.02	40.38	Yes [(2)]	1,825,000
23	280,000	219,000	78	3	11,000	0	24.13	39.29	No	1,928,832
24	370,000	-	-	2	8000	-	-	21.62	-	-
25	400,000	260,000	65	2	9200	20	37.93	23.00	Yes	3,600,000
26	457,579	507,913	111	5	12,000	5	28.23	26.22	Yes	5,232,742
27	950,000	745,625	78	3	31,200	0	24.14	32.84	Yes [(2)]	6,570,000
28 [(3)]	4,000,000	3,120,000	78	6	75,000	0	21.60	18.75	No	24,600,000
sum	9,223,749	6,539,532		62	261,740		22.01 [(4)] /40.72 [(5)]		18 Yes/5 No	63,306,709
mean			70	4		10	30.17 [(6)]	39.45		≈411 $GWh \cdot y^{-1}$

[(1)] Gas grid connection is currently under construction; [(2)] Biogas upgrading is already installed; [(3)] data from Project EOS (ebswien, 2021) [40]; [(4)] average specific gas production for plants without co-substrate; [(5)] for plants dosing co-substrate; [(6)] all plants.

Two of the observed WWTPs were operated above their organic design capacities—WWTP #11, with a design capacity of 120,000 PE, an average load of 150,000 PE (125%), an organic loading from the municipal discharge of 85,000 PE and 65,000 PE from the milk industry, and WWTP #26, with 457,579 PE COD design capacity and 507,913 PE average daily inflow load (111%)—while the hydraulic utilization was 85%. It should be noted that all the observed WWTPs, including these two, reached the required effluent limits. In Figure 2, the COD design capacity, the COD inflow load and the relative COD load are shown for the 28 WWTPs participating in the survey.

The number of digesters operated per WWTP varies between one and six reactors, with an overall digester volume between 1300 and 75,000 m^3 (Table 4). Five of the observed WWTPs operate one digester, 16 operate two digesters, five operate three digesters, one WWTP operates five digesters and one operates six digesters. The PE-specific digester volume varies between 21.62 and 70.00 $L \cdot PE^{-1}$ with an average of 39.11 $L \cdot PE^{-1}$ (Figure 3). At 11 of the 28 observed plants, in addition to primary and waste-activated sludge from biological wastewater treatment, between 5 and 47% of the COD-input load is added as co-substrates (10% on average).

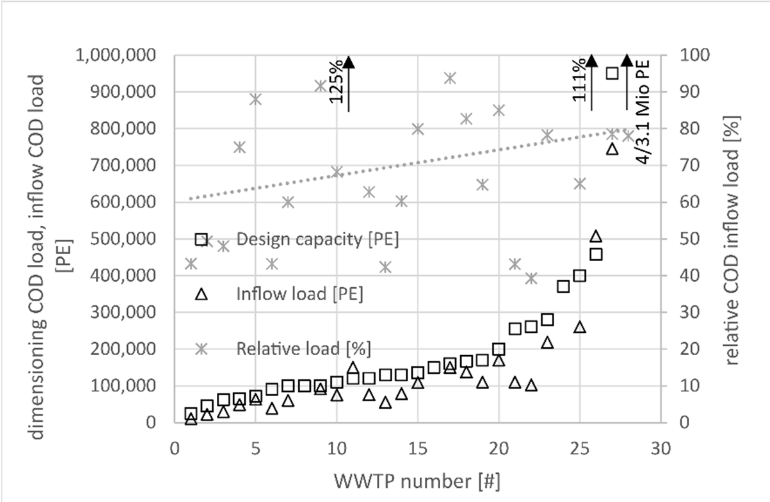

Figure 2. Design capacity PE_{COD} (squares), inflow load PE_{COD} (triangles) and relative COD load (stars) of 28 Austrian WWTPs.

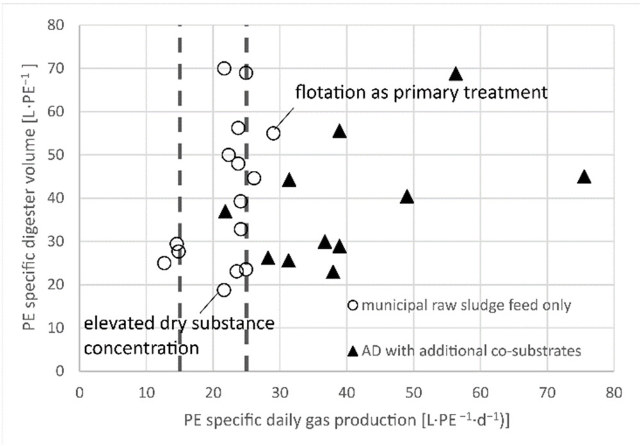

Figure 3. PE-specific digester volume ($L{\cdot}PE^{-1}$) and PE-specific daily gas production ($L{\cdot}PE^{-1}{\cdot}d^{-1}$) for 28 Austrian municipal digesters: ADs including co-substrate dosing are marked with triangles; the expected range for the specific gas production from the literature (15–25 $L{\cdot}PE^{-1}{\cdot}d^{-1}$) is marked with dashed lines.

In total, 11 of the 28 observed plants (40%) treated co-substrates, and the share of co-substrate ranged from 5 to 47% of the COD input of the AD (Figure 3). The PE-specific daily gas production varies between 12.7 and 29.0 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ for plants without co-substrates and between 21.8 and 75.5 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ for plants with co-substrate dosing. The average specific gas production was 22.0 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ for plants without co-substrates and 40.7 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ for plants including co-substrates. The average specific gas production for all the examined anaerobic sewage sludge digesters was 30.2 $L{\cdot}PE^{-1}{\cdot}d^{-1}$, which is within the range of values found in the literature. Lindtner (2008) [41] reported a PE-specific gas production without co-substrates of 15–24 $L{\cdot}PE^{-1}{\cdot}d^{-1}$; Haberkern et al. (2017) [42], 17 $L{\cdot}PE^{-1}{\cdot}d^{-1}$; and VSA (2010) [43] reported 25 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ without and 31 $L{\cdot}PE^{-1}{\cdot}d^{-1}$ with co-substrates, which also matches the numbers calculated in this work.

AD #20 has the highest observed specific gas production without co-substrate dosing (29.01 $L{\cdot}PE^{-1}{\cdot}d^{-1}$). This can be explained by the high share of wastewater from the milk industry and the usage of flotation as primary wastewater treatment instead of conventional primary clarifiers. AD #28 is operated with an elevated dry substance concentration in the digester sludge of approx. 7 $g{\cdot}L^{-1}$, which leads to a relatively low specific AD volume of 18.75 $L{\cdot}PE^{-1}$.

Two of the examined WWTPs operate biomethane upgrading facilities—WWTP #22, a membrane upgrade plant with a 120 m^3 $CH_4 \cdot h^{-1}$ capacity, and WWTP #27, a water scrubbing plant with a 450 m^3 $CH_4 \cdot h^{-1}$ capacity.

To put these numbers into context, in Table 5, data about all the 164 operating Austrian sewage sludge digesters, sorted by the design capacity, are summarized (data from ÖWAV, 2019) [32]. The 59 largest plants (36%) with a design capacity > 50,000 PE have a share of 90% of the overall capacity. At the same time, the smallest 27 (16%) plants > 20,000 PE provide a share of 2% of the overall treatment capacity.

Table 5. Austrian WWTPs including AD, their number, design capacity, sum and share of treatment capacity data (ÖWAV, 2019) [32] combined with own calculations.

Category	Number of WWTPs	Sum	Share	Biogas	Energy
Design Capacity	Including AD			Production	Production
PE_{COD}	#	PE_{COD}	[-]	Mio $m^3 \cdot y^{-1}$	$GWh \cdot y^{-1}$
>50,000	59	13.6 Mio	90%	76.45	734
>20,000	137	14.8 Mio	98%	83.19	799
All	164	15.1 Mio	100%	84.88	815

Considering a PE-specific gas production of 22 $L \cdot PE^{-1} \cdot d^{-1}$, a design capacity of 13.6 Mio PE and a utilization rate of 70%, this equals a biogas production of 76.45 Mio $m^3 \cdot y^{-1}$ (734 $GWh \cdot y^{-1}$) for Austria's 59 WWTPs > 50,000 PE design capacity including AD. Note that seven WWTPs with a design capacity > 50,000 PE (in sum, 475,000 PE) in Austria have no AD.

For plants > 20,000 PE, 83.19 Mio $m^3 \cdot y^{-1}$ (799 $GWh \cdot y^{-1}$) of biogas and for all plants, 84.88 Mio $m^3 \cdot y^{-1}$ (815 $GWh \cdot y^{-1}$) of biogas can be produced per year.

In Table 6, the biogas production of Austria's agricultural and sewage sludge biogas plants is displayed. In total, 315 biogas plants were operating in Austria in the year 2020, 164 of which are sewage sludge digesters.

Table 6. Total biogas production and biogas production in WWTPs in Austria (data for Austria's biogas production in 164 sewage sludge ADs from own calculations).

Country	Reference	Total Biogas Production from Sewage Sludge, Industrial Wastewater, Biowaste, Aggricultural Residuals and Landfills	Number of Biogas Plants Agricultur and Sewage Sludge AD's	Biogas Production in WWTPs Sewage Sludge Digestion Only		Number of WWTPs Including AD
	Year	$GWh \cdot y^{-1}$		$GWh \cdot y^{-1}$	% of Total	#
Austria	2020	1715 (1)	315	815 (2)	44% (2)	164 (3)

(1) calculated from electricity production with η_{EL} = 0.35; (2) calculated from survey data, using 15.1 Mio PE and 44.6 $kWh \cdot PE^{-1} \cdot d^{-1}$; (3) ÖWAV, 2019.

With 44% of the total biogas production, biogas from sewage sludge digestion has a relatively large share in Austria (Table 6), compared to the mean share of 23% of all IEA bioenergy in the 14 countries displayed in Table 3.

4.2. Potential for Long-Term Energy Storage Using BM Considering Different Efficiencies

To calculate BM's overall efficiency between electric input and biomethane output, a model sewage sludge AD with 100,000 PE capacity was used. Figure 4 shows the energy and mass flow chart for the model AD including electrolysis and in situ BM. This digester's design capacity is 12,000 kg COD input per day. If 50% of the input COD is degraded (η_{AD} = 0.5), the gas production without BM is 3500 $m^3 \cdot d^{-1}$ (2100 $m^3 \cdot d^{-1}$ of CH_4 and 1400 $m^3 \cdot d^{-1}$ of CO_2). If the CO_2 content in the biogas is reduced from 40 to 10%, and 1050 $m^3 \cdot d^{-1}$ of additional CH_4 is produced via BM, the biogas composition is then 3150 $m^3 \cdot d^{-1}$ of CH_4, 350 $m^3 \cdot d^{-1}$ of CO_2 and 1800 $m^3 \cdot d^{-1}$ of H_2. Therefore, 6000 m^3·d^{-1} of H_2 is needed. Taking into consideration that the efficiency of biological methanation is η_{BM} = 0.7 and the efficiency of the water electrolysis is η_{el} = 0.9, an amount of 23.63 MWh·d^{-1} (85 GJ·d^{-1}) of electrical energy is needed. This results in a specific energy consumption of 22.5 kWh$_{el}$·m^{-3} of CH_4 produced from BM.

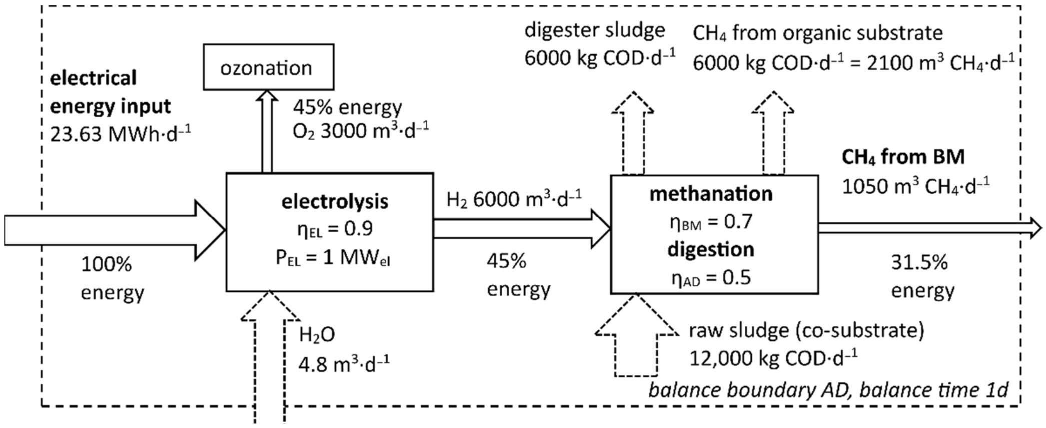

Figure 4. Energy and mass flow chart for in situ methanation in municipal digesters: values calculated for a 100,000 PE model WWTP with AD.

In addition to the hydrogen, 3000 m^3·d^{-1} of oxygen is produced and can be used in the biological wastewater treatment, or for ozone production to operate a fourth treatment step for the removal of trace substances.

Excluding the oxygen production, the overall energy efficiency between electricity input and CH_4 including water electrolysis and BM can be calculated to 31.5%. An additional 1050 m^3·d^{-1} of CH_4 is produced with an input of 23.63 MWh·d^{-1} of electricity (22.5 kWh·m^{-3} of CH_4). Including oxygen production, the overall efficiency is increased to 76.5% when pure oxygen is efficiently used in the treatment process. For comparison, the efficiency of pump storage hydropower plants is 70–85%.

4.3. Extrapolation of the Efficiency of Biological Methanation Transforming Electricity into Biomethane

The efficient use of the provided hydrogen is important for the overall efficiency of the PtG concept. In Table 7, the biogas composition and produced gas volumes are compared for different hydrogen conversion factors and an AD without H_2 addition. The daily gas production for all the main components and their shares for five different BM efficiencies, as well as a classic AD without BM for comparison, are displayed. At a BM efficiency of 93% (η_{BM} = 0.93), gas concentrations required for direct grid injection in Austria can be reached in one step. At lower efficiencies, an upgrade step must be connected downstream. η_{BM} is thereby defined as H_2 used for CO_2 conversion divided by H_2 input to BM.

Table 7. Daily produced gas volumes and shares in the product gas for CH_4, CO_2 and H_2 for a model AD including biological methanation for five different BM efficiencies and classic AD without BM as a reference for the model WWTP.

Gas	Classic AD		Anaerobic Digestion Including In Situ Methanation							
Components	without BM		η_{BM} = 0.7		η_{BM} = 0.8		η_{BM} = 0.93		η_{BM} = 1.0	
	Daily Volume	Share	Daily Volume	Share	Daily Volume	Share	Daily Volume	Share	Daily Volume	Share
	(m^3·d^{-1})	(%)	(m^3·d^{-1})	(%)	(m^3·d^{-1})	(%)	(m^3·d^{-1})	(%)	(m^3·d^{-1})	(%)
H_2 input	0	-	5600	-	5600	-	5600	-	5600	-
CH_4	2100	60%	3150	64%	3220	70%	3500	88%	3500	100%
CO_2	1400	40%	350	7%	280	6%	108	2%	0	0%
H_2	0	0%	1400	29%	1120	24%	392	10%	0	0%
sum output	3500	100%	5300	100%	4620	100%	4000	100%	3500	100%

Austria's 59 WWTPs with a design capacity > 50,000 PE produced 76.45 Mio $m^3 \cdot y^{-1}$ (855 $GWh \cdot y^{-1}$) of biogas in 2019. If the CO_2 content of the biogas is reduced from 40 to 10% via BM (22.9 Mio $m^3 \cdot y^{-1}$), this leads to an additional energetic potential of 220 $GWh \cdot y^{-1}$ (792 $TJ \cdot y^{-1}$) due to BM for Austria (Scenario one). In comparison, this is in the same range as that of the biogas injected from agricultural biogas plants (152 $GWh \cdot y^{-1}$) in 2020 [5]. According to the calculations above, a total efficiency of 31.5% can be assumed for BM. To produce this additional 22.9 Mio $m^3 \cdot y^{-1}$ of CH_4 via BM, 698 $GWh \cdot y^{-1}$ of electrical energy is needed.

For Scenario two (all WWTPs > 20,000 PE), an additional 25 Mio $m^3 \cdot y^{-1}$ of methane can be produced through BM, which equals 279 $GWh \cdot y^{-1}$.

For Scenario three (all WWTPs including AD), an additional 25.5 Mio $m^3 \cdot y^{-1}$ of methane can be produced, which equals 285 $GWh \cdot y^{-1}$. Therefore, 904 $GWh \cdot y^{-1}$ of electrical energy can be stored. As the transferable amount of energy depends on the available digester volume and the available CO_2, BM should be implemented in the large plants first.

4.4. Maximum BM Capacity in Austria's Sewage Sludge Digesters

Considering the BMR of 0.3 L $CH_4 \cdot L^{-1} \cdot d^{-1}$, the PE-specific AD volume of 40 $L \cdot PE^{-1}$ and the efficiency of 22.5 $Wh \cdot L^{-1}$ of CH_4, a maximum PE-specific BM capacity based on electricity input (sBM_{c_el}) can be calculated by dividing Equation (3) by the PE-specific AD volume, which results in 11.25 $W \cdot PE^{-1}$.

$$sBM_{c\_el} = 0.3\frac{L\,CH_4}{L_{AD} \cdot d} \cdot 40\frac{L_{AD}}{PE} \cdot 22.5\frac{Wh}{L\,CH_4} = 270\frac{Wh}{PE \cdot d} = 11.25\frac{W}{PE}$$

With this PE-specific electric input, approximately 8.8 L $CH_4 \cdot PE^{-1} \cdot d^{-1}$ can be additionally produced from CO_2 via BM, based on the PE-specific biogas production of 22 L $CH_4 \cdot PE^{-1} \cdot d^{-1}$. Using a countrywide design capacity of 15.1 Mio PE, the BM capacity based on electricity input (BM_{c_el}) for Austria can be calculated as follows:

$$BM_{c\_el} = sBM_c \cdot design_{capacity} = 11.25\frac{W}{PE} \cdot 15.1 \text{Mio PE} = 169.9 \text{ MW}$$

In this regard, approximately 170 MW electrical input out of the 177,000 m^3 $CH_4 \cdot d^{-1}$ (64.5 Mio $m^3 \cdot y^{-1}$) of methane gas can be produced through in situ BM, which equals 625 $GWh \cdot y^{-1}$. This maximum capacity is limited by the BMR. The available AD reactor volume would allow an approximately three times higher energy throughput than the 220 $GWh \cdot y^{-1}$, which can be transformed into biomethane using all the CO_2 contained in the biogas. Therefore, the maximum capacity of biological methanation is limited by the amount of CO_2 available in the biogas. However, as shown in Figure 3 the volume-specific amount of biogas and, thus, of CO_2 can be increased by up to three times, by dosing additional co-substrates.

4.5. SANKEY Diagram for Energy Flows through Austrian including BM at ADs

Energy flows and storage capacities for electricity and methane gas (natural gas and biomethane) for Austria are displayed in an energy flow diagram in Figure 5. All amounts of energy flows are displayed in $GWh \cdot y^{-1}$, energy stocks are displayed in GWh, respectively. The spatial balance boundary is Austria's political limit, and the balance time is one year (2020).

The efficiency factor for electricity storage in pump storage hydropower plants was considered as 75%. The efficiency factor for the conversion of electricity to methane through biological methanation was considered as 31.5% (excluding the oxygen production), as shown above. For comparison, Sterner and Stadler [44] indicated an efficiency factor in the range of 49–79% for PtG via biological methanation, depending on the used storage type, pressure level and withdrawal technology. Thema et al. [45] analyzed 36 methanation projects worldwide and reported an efficiency factor of 41% on average. The analyzed methanation plants have an average capacity of 380 kW_{el}, which is about half of the assumed electrical power needed for the BM at the model WWTP in Figure 4.

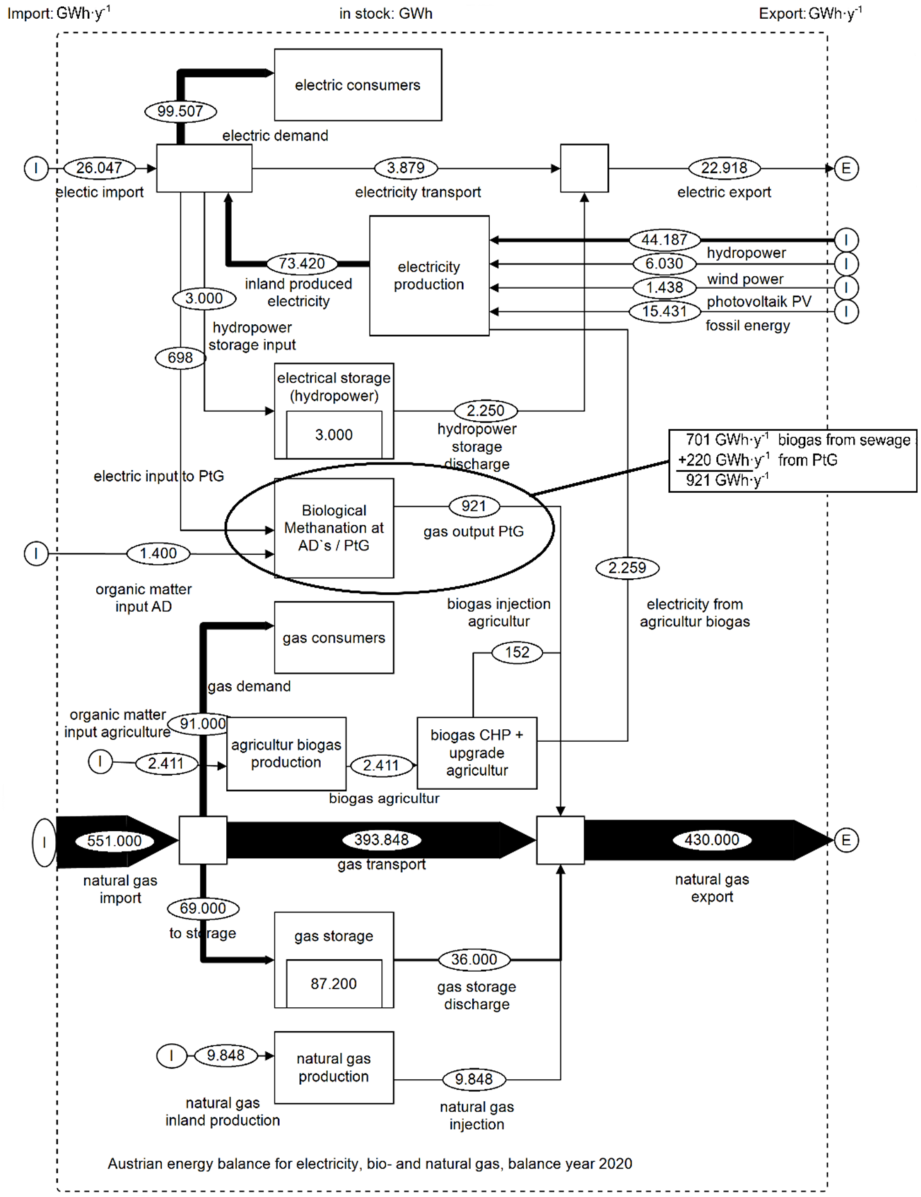

Figure 5. Austrian energy balance for electricity, and bio- and natural gas, for the balance year 2020, with energy flows for PtG through BM in sewage sludge digesters added.

The calculated electric input into the PtG electrolysis was 698 GWh·y^{-1}, which is approximately 23% of the electrical input into hydropower pump storage plants (3000 GWh·y^{-1}). In total, 701 GWh·y^{-1} of biogas from sewage sludge is produced per year. If all WWTPs > 50,000 PE are utilized and all of the produced CO_2 is converted into CH_4, an additional 220 GWh·y^{-1} can be produced through BM. In total, 921 GWh·y^{-1} of biomethane is produced, which equals 1% of Austria's natural gas consumption. A share of 16% (15,431 GWh·y^{-1}) of Austria's electricity production is fossil-based (mainly natural gas), and 26% (26,047 GWh·y^{-1}) is imported electricity [46].

As Thema et al. [45] supposed, biological methanation is nowadays more expensive than chemical methanation using the Sabatier process, but costs are expected to drop by 75% to below EUR 500·kW_{el}^{-1} for both methods in the next few years. It is also expected that methanation systems with a capacity of 50–250 m^3·h^{-1} will be realized.

With the additional long-term storage capacity generated through BM, wind and PV systems do not have to be shut down, when long-distance electricity transportation and hydropower storage are fully occupied. At the same time, the existing infrastructure is used, and no new hydropower reservoirs have to be created, which is becoming more difficult for environmental reasons.

5. Summary

With an increasing share of fluctuating energy from renewable sources, the need for network stabilization and long-term storage technologies will increase. Power-to-Gas is an opportunity to transfer electricity to the gas grid. Large wastewater treatment plants with anaerobic sewage sludge digestion provide ideal conditions and existing infrastructure for Power-to-Gas through biological methanation.

To obtain the detailed data necessary for the calculation of the energetic potential of biological methanation in Austria, a survey was conducted. Data from 28 Austrian WWTPs showed that ADs at plants > 50,000 PE are operated at 70% of their COD design capacity on average. The PE-specific digester volume was 40 $L \cdot PE^{-1}$, while the PE-specific gas production was 22 $L \cdot PE^{-1} \cdot d^{-1}$ without co-substrate dosing and 30.5 $L \cdot PE^{-1} \cdot d^{-1}$ including co-substrates, which lies in the range of the values found in the literature.

By supplying hydrogen to the digester, the CO_2 concentration in the biogas can be reduced by biological methanation and, thus, the gas quality that is required to feed into the gas network can be achieved. For Austria, this is now possible due to significant changes in the relevant guidelines, which now allow up to 10% hydrogen in the gas that is fed into the natural gas grid. In addition, compared to the thermochemical Sabatier process, biological methanation is a relatively simple method for producing biomethane, which can also be implemented in small units with a capacity of 50–250 $m^3 \cdot h^{-1}$ of biogas.

It was shown that the maximum amount of energy, convertible through methanation in sewage sludge digesters, is limited by the per time unit available quantity of CO_2 produced in the digesters, which could be increased by increasing the biogas production by digesting co-substrates. Considering volume-specific methanation rates, the existing digester volume would allow an energy throughput that is approximately three times higher.

Anaerobic sewage sludge digesters with a total capacity of 15.1 Mio PE are currently in operation. This capacity allows the production of approximately 109 Mio $m^3 \cdot y^{-1}$ of biogas from municipal sludge (at 100% utilization, without co-substrates). By implementing BM at all of these WWTPs > 50,000 PE, a maximum of 32.7 Mio $m^3 \cdot y^{-1}$ of methane can be additionally produced and stored. The maximum potential to store electricity in methane gas was calculated to 698 $GWh \cdot y^{-1}$, which is 9.3% of Austria's electricity from wind and PV facilities. At 70% utilization, the potential is 22.9 Mio $m^3 \cdot y^{-1}$ of methane, which equals 220 $GWh \cdot y^{-1}$ or 2.9% of Austria's yearly green energy production. At the same time, the carbon dioxide emission would be reduced by 65,400 t $CO_2 \cdot y^{-1}$. Considering that CO_2 from biological processes is climate neutral by definition, a positive carbon footprint could be reached.

Approximately 1% of Austria's natural gas demand could be replaced by the additional biomethane from BM at anaerobic sewage sludge digesters. Considering that a large share of Austria's electricity production is still fossil-based (16%), and 26% is imported electricity including fossil-based shares, there is still work to do to reach zero-emission targets for electricity production. Considering that the hydropower storage capacity is fully occupied, storing energy in the natural gas grid and its storing facilities is a practicable alternative, especially with a storage capacity of 87.2 TWh in Austria and widespread infrastructure, including a long-distance transporting network through Europe.

For the 14 counties in the IEA bioenergy task group 37 as shown in Table 3, the energetic potential of BM at municipal ADs is approximately 6000 $GWh \cdot y^{-1}$, which lies in the range of Austria's yearly renewable electricity production.

Considering benefits such as oxygen production for the fourth treatment stage, the overall efficiency of PtG is 76.5%. This is comparable with efficiency factors for hydropower pump storage plants (70–85%). When long-distance electricity transport is fully occupied, alternatives for network stabilization, such as PtG, are the only solution, if installed renewable power plants such as windmills and PV are to be fully utilized. Biological methanation in anaerobic sewage sludge digestors is, therefore, a good opportunity.

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