Bioresource Technology 300 (2020) 122647

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Energy neutrality versus carbon footprint minimization in municipal wastewater treatment plants

Mojtaba Maktabifard[a,*], Ewa Zaborowska[a], Jacek Makinia[a]

[a] Faculty of Civil and Environmental Engineering, Gdansk University of Technology, Narutowicza Street 11/12, 80-233 Gdansk, Poland

ARTICLE INFO

Keywords:
Co-digestion
Emission factors
Energy recovery
Greenhouse gases
Nitrous oxide gas

ABSTRACT

This work aimed to compare the carbon footprint (CF) of six full-scale wastewater treatment plants (WWTPs). The CF was estimated in the range of 23–100 kg CO_{2e} per population equivalent. In the total CF, the direct emissions held the highest share (62–74%) for the plants with energy recovery from biogas. In the plants depending entirely on the power grid, the indirect emissions due to energy consumption dominated the total CF (69–72%). The estimated CF was found highly sensitive towards the choice of N_2O emission factors. A dual effect of external substrates co-digestion on the CF has been presented. After co-digestion, the overall CF decreased by 7% while increasing the biogas production by 17%. While applying the empirical model, the level of energy neutrality was strongly related to the ratio of the indirect to direct emissions.

1. Introduction

Traditionally, the operation of WWTPs was focused on pollutants removal from wastewater in order to meet water quality standards for public health and environmental protection. In the last decades, new objectives have been postulated and put into practice to move towards sustainability in WWTPs. The sustainability is a multi-dimensional concept targeting economic, environmental and social aspects of WWTPs (Sweetapple et al., 2015). Each of those aspects can be subdivided into a large number of elements. Energy is one of the key elements of sustainability and a shift from the negative energy balance (energy demand covered by external sources) to the energy neutral or even energy positive wastewater treatment has been postulated (Gao et al., 2014; Lopes et al., 2018; Maktabifard et al., 2018; Song et al., 2018).

Carbon footprint (CF) is a new measure of sustainability in wastewater sector to determine the overall impact of WWTPs on climate change (Delre et al., 2019) and as a consequence, the focus of discussion for the WWTPs performance has recently turned to the CF minimization (Ødegaard, 2016; Xu et al., 2017). All relevant forms of the energy demand (electricity, heat, chemicals, fossil fuels, transport) and GHG emissions (carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O)) are commonly accounted in the CF assessment. These GHGs are among the six GHGs to be mitigated under the Kyoto Protocol and reported in GHG inventories (IPCC, 2006). Fugitive CO_2, CH_4 and N_2O, originated from wastewater treatment processes, are anthropogenic GHGs and produced on-site in WWTPs. CO_2 of the fossil origin is mainly produced off-site due to energy and material used in WWTPs (Yoshida et al., 2014). All GHG emissions can be expressed as CO_2 equivalents (CO_{2e}) with respect to their global warming potential (GWP). CH_4 and N_2O have respectively 28 and 265 times greater GWP compared to CO_2 in a 100-year time horizon (IPCC, 2013). The wastewater sector produces 0.37% of the total national carbon emissions in the USA (Wang et al., 2016) and it has been reported up to 3% globally (Xu, 2013).

The CF analysis is an important tool to recognize GHG emissions of specific units within the WWTPs and discover the potential solutions to minimize those emissions. The GHG emissions incorporated in the CF analysis of WWTPs can be classified based on the Intergovernmental Panel on Climate Change approach (IPCC, 2014). According to this classification, direct GHG emissions are related to activities within well-defined boundaries, such as, within-the-fence of a WWTP. Indirect emissions are a consequence of the activities within these boundaries but occur outside the specified boundaries. Components of both direct and indirect emissions can be gathered in scopes as adopted by the IPCC Guidelines (IPCC, 2006).

Differences between the contributions of the indirect and direct GHG emissions reported in the literature revealed the importance of evaluating the impact of various parameters and operational strategies on the total CF of WWTPs (Maktabifard et al., 2019). Based on the literature data, the direct emissions hold a large share in the total CF attributed to wastewater treatment (Gustavsson and Tumlin, 2013). Specifically, the reported contributions of N_2O emissions exceeded in

* Corresponding author.
E-mail address: mojmakta@pg.edu.pl (M. Maktabifard).

https://doi.org/10.1016/j.biortech.2019.122647
Received 15 October 2019; Received in revised form 15 December 2019; Accepted 17 December 2019
Available online 20 December 2019
0960-8524/

some cases half of the total CF in the biological nutrient removal (BNR) WWTPs (Maktabifard et al., 2019). The increasing attention on the process emissions resulted from the high GWP of the N_2O gas (Xu, 2013). N_2O is mainly produced as a result of incomplete nitrification or denitrification, low dissolved oxygen concentration and short solid retention time (Thakur and Medhi, 2019). The direct N_2O emissions can make enormous differences in the final total CF results in WWTP.

There are numerous studies focused on reduction of CF by improving the energy balance from wastewater (Chen et al., 2019; Gu et al., 2016; Mamais et al., 2015; Sun et al., 2017; Sweetapple et al., 2015; Wang et al., 2016). However, different emission factors (EFs) and contributions of direct and indirect GHG emissions to the total CF reported in the literature (Delre et al., 2019; Mamais et al., 2015; Zhao et al., 2019) do not explain clearly a relationship between the energy neutrality and the CF in the WWTPs. To improve the energy balance in municipal WWTPs, the common trend is to increase co-digestion of sewage sludges with external substrates (Maktabifard et al., 2018). Nevertheless, the choice of co-substrates and feeding strategy is fundamental because mobilised nutrients are recirculated to the main stream with the digester supernatants. Moreover, by adding the carbon source to support the anaerobic digestion (AD) process, the direct CF of the plants can also increase. Therefore, the trade-off between the increase in the direct CF and decrease in the indirect CF should carefully be analysed. This analysis can support decision-making and give a rationale to implement a sustainable strategy meeting both energy neutrality and CF minimization targets.

One of the important parameters which can significantly influence the CF of WWTPs is the sludge disposal scenario. Therefore, it is necessary to achieve a sustainable sludge disposal strategy. The conventional strategies, such as landfilling, are progressively restricted (e.g., due to EUR-Lex Directive (2018)) and development of innovative strategies to reduce the CF and increase energy recovery are required. Kacprzak et al. (2017) recommended increasing energy recovery through AD while achieving good quality product for further processing and agricultural use. On the other hand, external factors such as, social acceptance and countries restrictions of the usage of sludge for food production should be considered. In general, there is no universal technological solution which will consider local issues and sustainable development (Barberio et al., 2013). Hao et al. (2020) recommended direct incineration of excess sludge (without AD) over other scenarios such as landfilling and agricultural use. Their findings suggest that this scenario has the lowest energy deficit and cost while decreasing the overall indirect CF. Gherghel et al. (2019) reported that from environmental point of view, agricultural use of excess sludge still remains one of the preferable options. Among other novel technologies, production of biofuels from excess sludge and electricity production by using microbial fuel cells were reviewed but still more research is required to reach full-scale application stage in WWTPs. Nakatsuka et al. (2020) proposed integrating wastewater treatment and municipal solid waste incineration plants to overcome significant energy consumption required for sludge drying. On the other hand, other sludge drying and management methods such as drying lagoons might have low energy consumption but substantially increase the overall CF of the WWTP due to high CH_4 emissions (Pan et al., 2016).

The objective of the present study is to determine the relations between the energy neutrality and the CF in municipal WWTPs. Six medium-scale and large-scale facilities located in Poland have been analysed based on routinely collected operational data. The tool applied to calculate the CF of WWTPs is based on the empirical models (CFCT, 2014). The most influential parameters within the CF components have been identified and subjected to more detailed analysis of their influence on the final results. An analytic hierarchy process (AHP) evaluation has been applied in order to select the most proper sludge disposal strategies. A relationship between the energy neutrality level and a ratio of the indirect to direct GHG emissions have been investigated in terms of energy recovery from biogas in WWTPs. For strategies involving co-digestion of sewage sludges with external substrates, possible trade-offs between the two discussed targets have been revealed. The present study aims at identifying parameters sensitive to assumptions and enhancing understanding of the complex relation between achieving energy neutrality and CF minimization.

2. Materials and methods

2.1. Study sites

The studied WWTPs represent BNR plants with different process configurations, levels of energy neutrality and options for sludge disposal. The plants are termed A–F and ordered based on the design capacity expressed as population equivalents (PE). In this study, the sludge disposal refers to the fate of sludge after AD (operations conducted both inside and outside the WWTPs) and can go beyond the plants boundaries. The WWTP A (73% energy neutral) benefits on-site electricity production through combined heat and power (CHP) system using biogas produced from mesophilic AD of primary sludge, waste activated sludge (WAS) and external organic material (fat) for co-digestion (4% of the total feedstock volume on average). The WWTPs B and D are both partially energy neutral (68% and 29%, respectively). The main difference is that the plant D employs A_2O configuration in the biological step, while the plant B is the only studied WWTP which uses a sequencing batch reactors (SBR) configuration. The plant C is the most energy efficient studied WWTP (98% energy neutral). Plant C has the highest biogas production among all the studied cases. The plants E and F are medium-size facilities which purchase all the required energy for wastewater treatment from the power grid since they do not have close mesophilic AD units. In the plant F, psychrophilic AD of WAS is performed in an open chamber. These six WWTPs apply different sludge disposal practices. Plants A and B convert the digested sludge to compost, while plants C, D and F distribute the stabilized sludge directly to farmlands. Plant E is the only WWTP which disposes the excess sludge to landfill. Basic characteristics of each plant is provided in Table 1. Fig. 1 shows the scheme of wastewater treatment and sludge lines in the studied WWTPs indicating different system boundaries for each plant.

2.2. Calculation tool and classification of GHG emissions

The Carbon Footprint Calculation Tool (CFCT) dedicated for WWTPs is MS Excel spreadsheet (CFCT, 2014), developed in the project entitled “Calculation of the CF from Swedish WWTPs” (SVU 12-120) (Gustavsson and Tumlin, 2013). The detailed results for full-scale

Table 1
Basic annual characteristics of the studied municipal WWTPs.

WWTP	Design size (PE)	Configuration	Average influent flow rate (m^3/d)	COD load (Mg/yr)	TN load (Mg/yr)	Energy Neutrality (%)	Sludge disposal method
A	200,000	A_2O	23,000	9578	664	73	Compost
B	130,000	SBR	7000	2971	287	68	Compost
C	100,000	JHB	6500	3747	257	98	Farmland
D	80,000	A_2O	8700	2168	187	29	Farmland
E	70,000	A_2O	9600	3666	250	0	Landfill
F	60,000	A_2O	8500	2431	250	0	Farmland

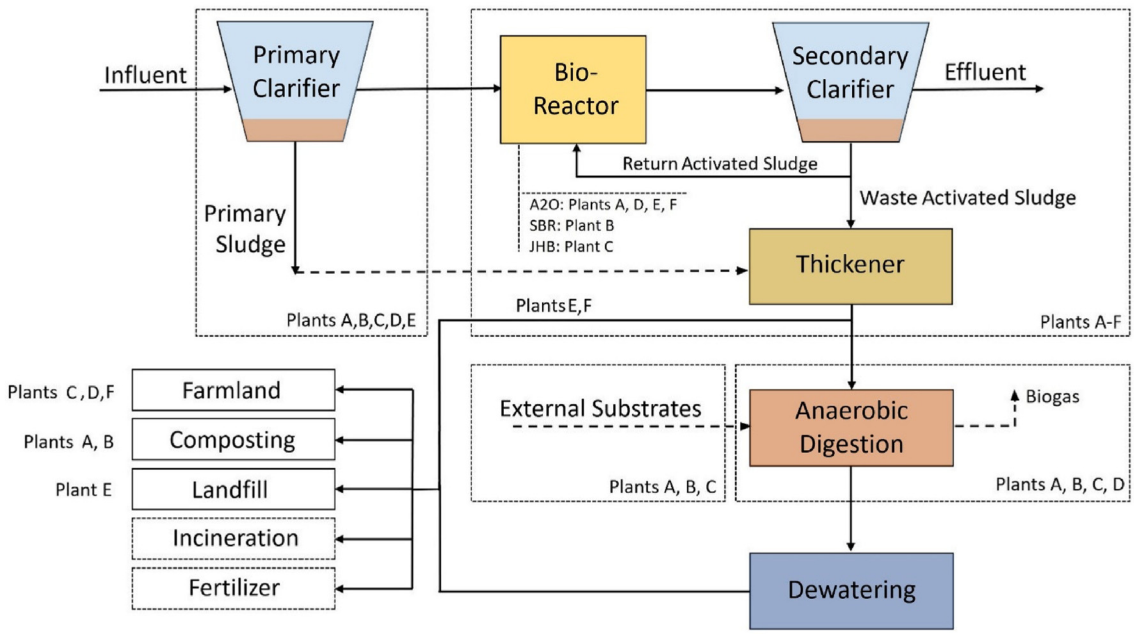

Fig. 1. General concept of wastewater and sludge lines in the studied WWTPs.

facilities are based on empirical models applied in the CFCT. GHG (CO_2, CH_4 and N_2O) emissions are expressed in CO_{2e} units with respect to their GWP (IPCC, 2013) and gathered by five categories (wastewater treatment, energy consumption, biogas production, sludge handling and miscellaneous). In the CFCT, the results can be analysed and evaluated to determine the most influential and sensitive parameters affecting the total CF. The approach based on the empirical models (the EFs) is advantageous by providing the feasibility of calculating the total CF of the whole WWTP including the three scopes of GHG emissions as proposed in (IPCC, 2006).

The AHP evaluation has been applied in order to select the most proper sludge disposal strategies with respect to the criteria, such as, energy and CF. For energy, both consumption (the lower the better) and production (the higher the better) has been considered while for the CF, CH_4 and N_2O emissions as the main GHGs emitted from WWTPs have been considered and different scenarios were rated based on these criteria.

2.3. Databases for CF calculations

The CF analysis was based on a wide range of annual historical operating data. Daily average samples of wastewater were collected and analysed for the pollutants concentrations in accredited laboratories meeting the Polish standards which are in accordance with the Standard Methods (APHA, 2005). One of the major reasons for choosing the aforementioned case studies was availability of the comprehensive data, specifically in the energy section. The wastewater flow rates, fuels (including biogas) production/consumption and electric energy production/consumption were measured and registered by water meters, gas meters and electricity meters, respectively. The amounts of sludge and wastes as well as the chemicals consumption were registered routinely by the plant operators. In the plants A, B and C, heat production and consumption were measured directly by heat meters. In other cases, the heat consumption was estimated based on the fuels consumption and the energy conversion devices (CHP units, boilers) thermal efficiency. The data for all six case studies drawn from the annual reports (2016 or 2017) were introduced to the CFCT (CFCT, 2014).

The complete necessary data related to co-digestion of external substrates were available for the plant A. The daily characteristics of the AD substrates and biogas production were collected on-site for different scenarios (mixed sludge with and without co-substrates).

2.4. Assumptions for CF calculations

Following the recommendations of the IPCC (IPCC, 2006), only CO_2 emissions due to external fossil carbon use were considered in the calculations while the CO_2 emissions of biogenic origin were not taken into account for the CF estimation.

Missing data (the ones not routinely measured) were estimated based on the values adopted from the literature. Among them, the direct emissions from wastewater and sludge treatment processes were calculated based on the EFs reported in the previous studies.

2.5. Uncertainty analysis

The accuracy of calculations depends on the availability and quality of routine data provided by the plants operators. Specifically, uncertainty of the assumed scope 1 emissions related to direct emissions from wastewater and sludge treatment processes results from scarcity of routine measurements. The results reported in the literature demonstrated considerable variability in the N_2O EFs and were site specific. For biological processes in the activated sludge (AS) bioreactors, the EF of 15.7 g N_2O/kg $N_{denitrified}$ (Foley et al., 2010) was used as a reference value, following the defaults implemented in the CFCT. In the present study, the total CF for each case was subsequently calculated based on various EFs reported in the literature and the uncertainty effect was demonstrated.

3. Results and discussion

3.1. Specific CF indicators – current state

The final results of the CF for each WWTP were compared with respect to the plant's capacity (expressed in PE), influent flow rate, removed loads of nitrogen (TN), phosphorus (TP) and chemical oxygen demand (COD). The results in Table 2 clearly explain that the total CF of the WWTPs is not correlated with the capacity of the plants. Many other parameters, such as, influent wastewater characteristics, energy efficiency of the plant and sludge disposal strategies may affect the CO_{2e} emissions.

Table 2
Total and specific annual CO_{2e} emissions for the studied plants.

Total/specific emission (Unit)	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F
CF (Mg CO_{2e}/year	4740	2336	1825	2055	6026	2350
CF_{PE} (kg CO_{2e}/PE)	25.8	56.5	23.7	42.1	99.7	38.8
CF_V (kg $CO_{2e}/m^3_{wastewater}$)	0.6	0.9	0.8	0.6	1.7	0.8
CF_{TN} (Mg CO_{2e}/Mg $TN_{removed}$)	7.9	8.9	7.6	13.1	25.9	11.5
CF_{TP} (Mg CO_{2e}/Mg $TP_{removed}$))	49	57	62	70.9	229	110
CF_{COD} (Mg CO_{2e}/ Mg $COD_{removed}$)	0.5	0.8	0.5	0.6	1.7	1.0

The total annual produced CO_{2e} per PE (CF_{PE}) has been selected to demonstrate a comparison within the WWTPs Plants A and C leave a relatively low CF_{PE} (25.8 and 23.7 kg CO_{2e} per PE, respectively) due to higher energy recovery from biogas in comparison with other plants. Plants B, D and F are considered medium in terms of the CF_{PE} (56.5, 42.1 and 38.8 kg CO_{2e} per PE, respectively). Finally, plant E has the highest CF_{PE} (99.7 kg CO_{2e} per PE) due to the synergistic effect of the off-site emissions in the energy category and landfilling of the sludge.

The comparative analysis with the literature data showed that the estimated specific CF indicators (Table 2) were within the reported ranges for all WWTPs considered in the present study. In other case studies (Gustavsson and Tumlin, 2013; Mamais et al., 2015), the CF_{PE} was reported in the extremely broad range of 7–161 kg CO_{2e}/PE. Another specific CF per m^3 of wastewater influent (CF_V) was reported in the range of 0.1–2.4 kg CO_{2e}/m^3 (Li et al., 2017; Wang et al., 2016). In the present study, the lowest value was observed in plants A and D (0.6 kg CO_{2e}/m^3) and the largest one belonged to plant E (1.7 kg CO_{2e}/m^3). Wang et al. (2016) reported the range 0.1 to 0.96 kg CO_{2e}/m^3 for the facilities located in different countries. The CF_V was affected by different discharge limits for the effluent concentrations of pollutants as well as different EFs for electricity. For instance, for the plant with the minimum CF_V reported by Wang et al. (2016) (0.1 kg CO_{2e}/m^3), which is significantly smaller than plants A to F, the EF used was 0.36 kg CO_{2e}/kWh. That value is less than half of the one assumed in the present study. Mannina et al. (2019) developed a new model for CF estimation in WWTPs. The range of the specific CF_V reported in that study was 0.18–1.18 kg CO_{2e}/m^3 considering both direct and indirect emissions. The results with the relatively high values for the CF_V, were attributed to the high indirect emissions (for scenarios with high electricity consumption). Moreover, high direct emissions attributed to the high TN concentrations in the influent increased the contribution of the N_2O emissions.

For more accurate estimation of the annual CF of WWTPs, it has been suggested to consider both operational and construction phases. Nevertheless, the calculation of CF related to construction phase is challenging due to the lack of data and thus this stage has often been neglected in the literature. Mo and Zhang (2012) calculated the annual CF of a large WWTP in Florida (USA) with the design capacity of 360,000 m^3/d. The results showed 53,000 Mg CO_2/year for the operational phase and 5700 Mg CO_2/year for the construction phase which makes about 10% of the total CF of this WWTP.

The carbon emissions due to energy consumption accounted for 38–50% of the GHG emissions in WWTPs investigated by Bao et al. (2016). In the present study, the estimated CFs, related to energy, were beyond that range. The studied WWTPs can be divided into two categories: (i) plants A to D with on-site energy recovery units and low energy-related CF (1–23% of the total CF), and (ii) plants E and F without any energy recovery and with a high energy-related CF (69–72%). These results demonstrate the significance of energy recovery from wastewater. Ødegaard (2016) emphasized that future WWTPs should be energy neutral while leaving a low CF. The highest values of CO_{2e} emissions were usually obtained from the indirect emissions caused by electricity consumption for aeration (Mamais et al., 2015). Thus, increasing the aeration efficiency at WWTPs has some potential for reducing emission of GHGs. However, the trade-off between the cost-efficient aeration and N_2O emission must be carefully monitored due to the high GWP of N_2O gas. Direct N_2O and CH_4 emissions are not easy to measure due to the complexity of gas measurements, especially at open WWTPs (Yoshida et al., 2014). In WWTPs built underground, measuring the N_2O gas emissions is feasible like in the Viikinmaki WWTP in Finland (Blomberg et al., 2018). For electricity, the EF can easier be estimated. If the electricity is produced from coal, the effect is large whereas the production by wind power or photovoltaic has a very small climate effect. If the incineration of WAS is considered, the additional CF must be taken into account in view of the emerging environmental regulations worldwide. The increased energy recovery does not necessarily correspond with a shift towards sustainability, particularly in terms of environmental sustainability as represented by sludge production.

A shift from classical, central systems of energy production and distribution, primarily based on fossil fuel power plants, towards decentralized energy system (DES) has been observed. Plant C can be a good example of decentralized local energy plants which only leaves 0.4 kg CO_{2e}/PE. Based on the online CF calculator (Carbon Footprint Calculator, 2019) considering an average family car which drives 15,000 km per year, the total CO_{2e} emissions would be 500 kg per person. Comparison of the car emission with the WWTP indirect emission shows that the latter one can be almost negligible while maximizing the renewable energy recovery.

3.2. Energy neutrality vs. CF of the studied plants

Fig. 2 shows the comparison between the level of energy neutrality and the CF_{PE}. The plants are ordered from the highest (98% energy neutral for plant C) to the lowest (0% energy neutral for plants E and F). The total CF_{PE} (including both direct and indirect emissions) is not directly correlated with the level of energy neutrality. By increasing the on-site energy recovery from biogas, the share of the indirect emissions in the CF_{PE} decreases. As shown in Fig. 2, the share of the indirect emissions for plant E is 69% (0% energy neutral) while for plant C, this share falls to 1% (98% energy neutral). For plants E and F, the indirect emissions are as high as 69.8 and 28.2 kg CO_{2e}/PE, respectively. This emission was estimated at 7.0 and 0.3 kg CO_{2e}/PE, for plants A and C, respectively. This trend shows that increasing the energy efficiency in WWTPs would readily affect the indirect CF and would help to decrease the total CF in comparison with the current state.

Fig. 3 shows the correlation between the level of energy neutrality and indirect to direct CF ratio, for the studied WWTPs in Poland as well as for the data reported in the literature (plants 1–7) from different regions, such as Australia (plants 1, 2 and 3), USA (plant 4), Scandinavia (plants 5 and 6) and Italy (plant 7). As it is demonstrated, the variables are strongly correlated (R^2 = 0.97). Following the previously introduced classification, the WWTPs can be divided into two categories. The first group comprises plants A–D and 1–7 which have the on-site energy recovery units and with the indirect to direct emission ratio lower than 50%. These group of WWTPs are located on the left side of Fig. 3. Between the ratios of 50–200% of the indirect to direct GHG emissions there is a gap of data and no WWTP was found to fit in that area. This lack of data is due to either the WWTPs (mostly medium or large size) had the AD unit on-site and were producing a portion of

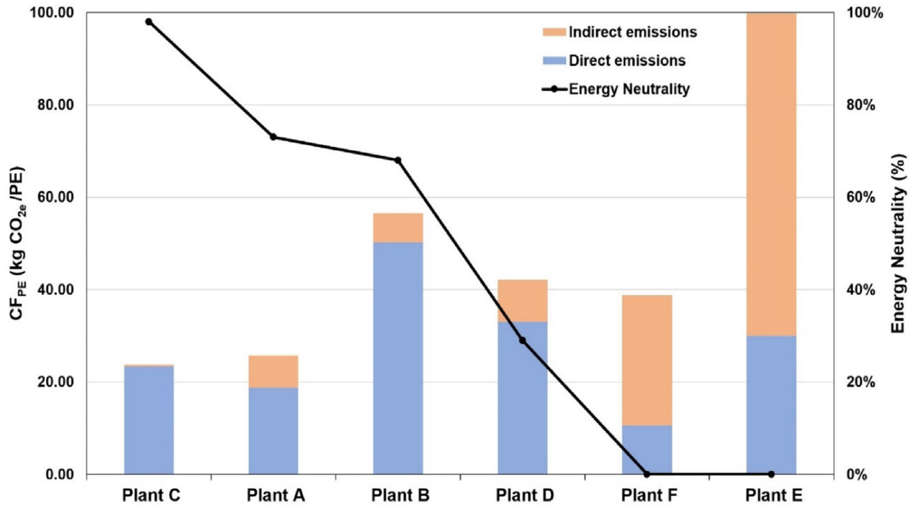

Fig. 2. Comparison between the level of energy neutrality and CF_{PE} in the studied WWTPs.

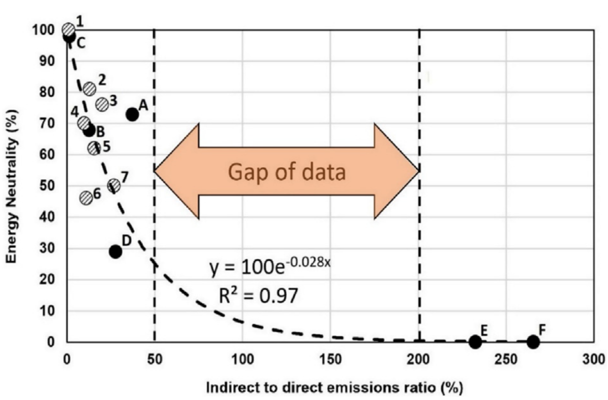

Fig. 3. Correlation between energy neutrality and indirect to direct CF ratio in the studied WWTPs, data from the literature were derived from de Haas (2018) for plants 1, 2 and 3; Gu et al. (2016) for plant 4; Gustavsson and Tumlin (2013) for plants 5 and 6; and Mannina et al. (2019) for plant 7.

their required energy (25–100%) or, on the other hand, they were missing the AD unit (mostly small size facilities) and had zero energy recovery. Plants E and F belong to the second group without any energy recovery and with the ratio over 200%. By decreasing the indirect emissions to zero the model indicates that the level of energy neutrality would go close to 100% (energy neutral) and when the indirect to direct emission ratio passes 200%, the level of energy neutrality becomes zero. In the case of plant A, the highest residual was observed because of the high value of indirect emissions not only those related to electricity but related to chemicals and transportation (scope 3). As shown in Fig. 4, a share as high as 6% of scope 3 in the total CF was calculated for plant A and only 1–2% for the other studied WWTPs. Based on the proposed model shown in Fig. 3, in order to fit into the gap of data zone, the on-site energy recovery must be lower than 25% while the share of indirect emissions decreases. An example could be a WWTP which recovers a relatively small share of the energy consumption by renewable energies (e.g., solar energy) in order to partially overcome the indirect emissions.

Aiming at the energy neutrality is reasonable in terms of the CF provided that the total CF is not increased. This condition can be satisfied while applying AD of sewage sludges. Apart from the environmental aspects, increasing the energy self-sufficiency contributes to lower costs of the electricity purchased from the grid. Such an option can be considered safe, since the energy prices might increase as the fossil fuel resources become more limited and in view of the prospective regulations regarding CF mitigation.

3.3. Components of the CF

Results of the CF calculations for six full-scale case studies are

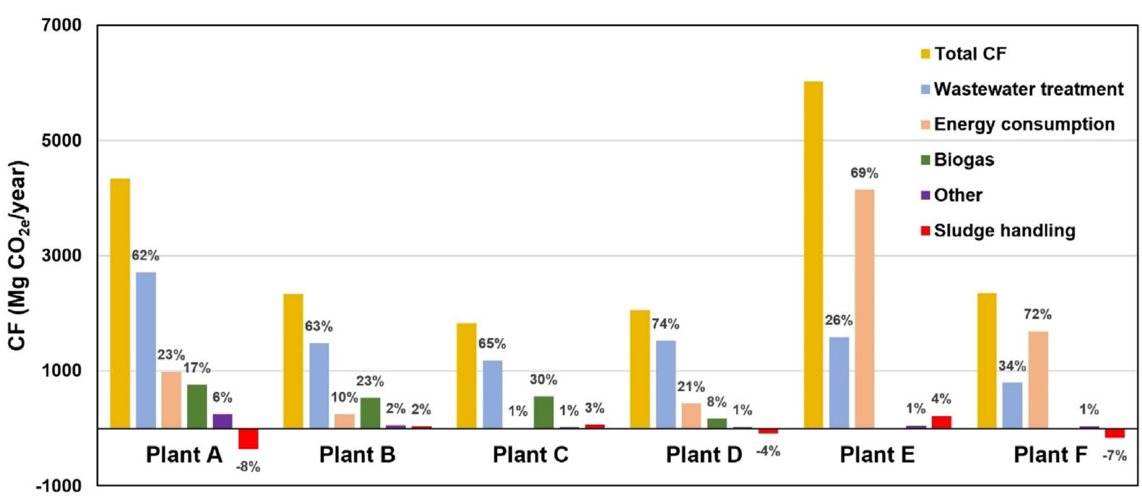

Fig. 4. The shares of different CF components in the studied WWTPs.

shown in Fig. 4. The plants are ordered A to F based on the size (PE) and the first bar represents the absolute value of the total annual CF of the plants. For plants A–D, the direct emissions from wastewater treatment had the highest share in the total CO_{2e} emissions (62–74%), followed by the energy consumption (1–23%) and biogas production (8–30%). The biogas production had both positive and negative effects on the CF. The positive influence was considered in the energy consumption category and resulted from the energy recovery and reduction in the off-site emissions related to the electricity supplied from the power grid. The negative influence resulted from biogas slip during its production, CH_4 and N_2O release during the incomplete biogas combustion in the CHP engines and boilers. Plant C would be a good example in this case. While it holds the highest share of CF related to biogas (30%), it has the lowest CF related to energy consumption (only 1%). Plant C is in 98% energy neutral, therefore, the share of the energy consumption category is almost negligible. For this advanced WWTP, 65% of the total CF originated from wastewater treatment. Since this WWTP had the highest biogas production among the six studied WWTPs, the share of the biogas category in CF was thus considerably higher than other WWTPs. Similar to plant C, the contribution of the biogas category in the total CF was relatively high (23%) for plant B. This large CF from biogas was due to the higher biogas production in these plants and thus relatively higher biogas loss. WWTPs A to D have the on-site energy recovery units, therefore, CF shares related to energy consumption and biogas production interact with each other. Considering the sum of these two components, the share of energy related CF, would be 30–40%.

For WWTPs A, D and F, the proper sludge disposal practice (farmland distribution of anaerobically stabilized or composted sludge) reduced the total CF by 8, 4 and 7%, respectively. These reductions (in the sludge disposal category) resulted from the avoided emissions of GHGs due to substituting production of chemical fertilizers. In contrast, for plants B, C and E, the sludge disposal increased the total CF. For plant C, it was due to a relatively long sludge storage time span before further handling within the plant that caused additional CH_4 emissions. In the case of plant B, the increase was due to the additional energy usage by the composting process, and for plant E, the sludge disposal did not contribute to reduction of the total CF due to the emissions released while landfilling the excess sludge.

Plants E and F produced most of the GHG emissions through the indirect way (69 and 72%, respectively in the energy consumption category) due to supplying all the energy demand from the power grid. Other significant CF contributors in those two plants belonged to the wastewater section (26–34%).

Plant E had the highest CF even though it was one of the smallest studied WWTPs in size. The high absolute CF value of the plant resulted from the energy consumption (69%) followed by the direct emissions from wastewater treatment (26%). In terms of CF reduction, this case study showed the importance of implementation of the AD process with biogas production and energy recovery.

3.4. *Effect of the N_2O EFs on the CF*

As described in Section 2.5, the N_2O EFs (EF_{N2O}) for full-scale BNR WWTPs have been reported in a wide range. Only the sole EF_{N2O} can make enormous differences in the final total CF_{PE}. For the plants with on-site energy recovery (A-D) higher uncertainty of the calculated CF was revealed. The result for the total CF could increase by as high as 105% (after re-calculating based on the highest EF_{N2O}) for plant B. On the other hand, the level of uncertainty decreases to 18 and 25% for plant E and F, respectively. In the sensitivity analysis, the direct emissions from wastewater treatment were found as one of the most influential parameters affecting the total annual CF_{PE}.

The significant share of the direct GHG emissions from wastewater treatment in the total CF (as high as 74% in plant D) confirmed the observation of Koutsou et al. (2018) about domination of the direct emissions in the CF of partially energy neutral WWTPs. Koutsou et al. (2018) analysed data from 220 WWTPs in Greece. The authors concluded that 68.8% of the total GHG emissions were associated with the direct emissions. The contribution of the N_2O emission in the total CF was reported as high as 86% (Kosonen et al., 2016). Results of the study by Gruber et al. (2020) confirmed the high N_2O emissions for different AS processes and highlighted N_2O as the most important GHG produced by WWTPs.

A broad range of N_2O emissions for different types of wastewater treatment configurations and process conditions can be found in the literature. Based on the study of Nguyen et al. (2019), the level of direct N_2O from WWTPs using A2O configuration accounted for 0.97 g $N_2O/m^3_{wastewater}$. In the present study, the average value calculated for plants A, D, E and F with the same configuration (A2O) was 1.01 g $N_2O/m^3_{wastewater}$. Wang et al. (2016) analysed N_2O EFs in the full-scale WWTPs by online measurements and reported the produced gaseous N_2O for A2O process in the range of 0.095–3.44% of the total nitrogen load. Sun et al. (2017) emphasized that N_2O EF could not be precisely determined based on the experimental methods and thus model simulation method could be a better choice. According to the experimental results, the N_2O EF varied significantly from 0.2 to 1.6% of the total nitrogen load (Sun et al., 2017). Gruber et al. (2020) performed a long-term monitoring campaigns in 3 WWTPs with different biological configurations in Switzerland and assessed the N_2O EF, 1.7% of the total nitrogen load. They also reported that considering all the published long-term campaigns, the N_2O EF would be 1.9%.

Such a significant different reported values for the N_2O EF can be attributed to differences in WWTPs technologies and processes conditions as well as various methodologies used for the monitoring strategies. To demonstrate the substantial effect of the N_2O EF on the total CF, uncertainty level of the total CF for each studied plant has been shown in Fig. 5. The lowest uncertainty, belongs to plants E and F because the share of energy in these plants dominates the total CF and direct emissions are 26 and 34% respectively. In contrast, in the plants dominated by direct emissions, the uncertainty of the total CF estimation could exceed 100%. In the case of plant B, the highest level of uncertainty was observed possibly due to the unique configuration of the bioreactor (SBR). The choice of the EF_{N2O} may significantly affect the final results. This explains the need for individual full-scale measurements at WWTPs and further studies to calculate the direct emissions by more accurate modelling tools instead of using the assumed EFs from the literature.

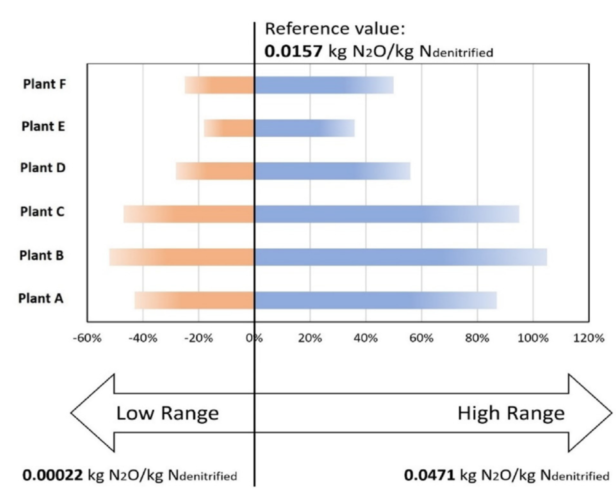

Fig. 5. Uncertainty level of CF for each studied WWTP based on different N_2O EFs.

Table 3
Effect of different sludge disposal scenarios on the total CF.

Sludge disposal scenario	Total CF (Mg CO_{2e}/year) and the relative effect of each scenario on total CF (%)					
	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F
Farmland	4478 (−5%)	2304 (−1%)	**1825 (0%)**	**2055 (0%)**	5823 (−4%)	**2350 (0%)**
Composting and farmland	**4740 (0%)**	**2336 (0%)**	2102 (15%)	2237 (8%)	5923 (−2%)	2794 (19%)
Incineration	5004 (5%)	3332 (42%)	1997 (9%)	2169 (5%)	5814 (−4%)	2612 (11%)
Landfill	5278 (11%)	2555 (9%)	2219 (21%)	2176 (6%)	**6026 (0%)**	2748 (17%)
Fertilizer after drying	7119 (50%)	2552 (9%)	2241 (22%)	2323 (27%)	7096 (16%)	3677 (56%)

Note: The highlighted numbers represent the actual sludge disposal scenarios applied at the plants.

3.5. Effect of sludge disposal practices on the CF

One of the most important ways which can help reduce the CF of WWTPs is the proper disposal of dewatered sludge. Sludge usage scenarios could make significant changes in the total CF. Five different scenarios for sludge disposal were considered and their influence on the CF have been shown in Table 3. Each possible scenario was assumed in the CFCT by using different EFs for each type of the sludge disposal strategy. Distribution on farmlands, composting (with subsequent distribution on farmlands), landfilling, incineration, and conversion to fertilizer after drying were taken into consideration. The results shown in bold in Table 3 are based on the current sludge disposal practice while the remaining results are based on the assumed scenarios.

Plants C, D and F distribute the dewatered sludge on farmlands and such an approach was found to be the best scenario in terms of the CF. The other possible methodology for sludge disposal is converting the anaerobically stabilized sludge to compost as it is at the plants A and B. Distribution of sludge on landfills was found as one of the most unfavourable strategies. Plant E and A would save around 200 and 250 Mg CO_{2e}/year of emissions, respectively, by applying the farmland scenario. The percentages in Table 3 show the significant effect of the sludge disposal scenarios on the total GHG emissions produced by each WWTP. Consequently, due to the possible "minus" effect on the total CF (as demonstrated in Fig. 4), it is crucial to consider the environmental effects of sludge disposal.

The scenarios selected for the detailed analysis represent well-established technologies commonly applied for sludge disposal. Current research trends are focused on technologies to reduce sludge production, emerging pollutants content and fate, thermal pre-treatment, as well as recovery of carbon, nutrients and energy (Zhang et al., 2017a). Sludge pre-treatment technologies, such as thermal hydrolysis can improve both biogas production and sludge dewaterability. The process self-sufficiency is a prerequisite in full-scale applications to become neutral for both the energy balance and indirect GHG emissions. Supercritical water treatment and torrefaction for treating biomass can immobilize most metals in the bio-char residue but are cost-intensive processes. Thermal processes offering carbon and nutrients recovery have been proposed as the options for sludge treatment to satisfy the concept of sustainability and circular economy. The novel technologies need further development, reducing costs and evaluation with respect to the overall environmental impact as considered in the life-cycle assessment approach.

In the AHP evaluation, five different sludge disposal scenarios were compared and ranked based on the energy consumption and production (energy criteria) and CH_4 and N_2O gas emissions (CF criteria). Farmland disposal of sewage sludge for agricultural usage was found as the best selected scenario in terms of both energy and the CF followed by the incineration scenario. For the fertilizer scenario, the better results were observed when the CF reduction was priority. On the other hand, landfilling of sludge would be preferred when primarily low energy consumption was expected.

While farmland disposal of the sludge was found as a better scenario in terms of the CF reduction, on the other hand, the standards concerning land application of the sludge should be considered. EUR-Lex Directive (2018) provides a waste hierarchy that shall apply as a priority order in the waste management legislation and policy. The major problems related to sludge disposal comprise the presence of heavy metals and organic contaminants (Islam et al., 2013). Some European countries introduced restrictions based on the European directives such as limit values for pathogens and organic micropollutants. Consequently, it is crucial that sludge reuse for farmland disposal has to meet those qualitative requirements, as in case of Denmark (Collivignarelli et al., 2019).

3.6. Impact of carbon intensity of the consumed electricity

Zhao et al. (2019) noticed that the EFs attributed to the power grid (scope 2) significantly affect the overall CF. In the present study, results shown in Fig. 4 proved that the share of the electricity consumption category in the total CF of the WWTPs could be as high as 72%.

The EFs for the off-site electricity production are country specific. A recent study by Vuarnoz and Jusselme (2018), reported the electricity EFs for Switzerland, Austria, France and Germany 203, 352, 80 and 534 g CO_{2e}/kWh, respectively. The variations of the EFs for electricity in different countries are attributed to significant differences in the fuel mix of each region. The countries which rely heavily on fossil fuels report higher electricity EFs. Based on the fact that all the case studies are located in the same country, the marginal Polish EF (810 g CO_{2e}/kWh) was chosen (NCEM, 2017). For plants E and F, the total CF was more sensitive (in comparison with plants A–D) to changes in the EF for electricity. By applying the Nordic standard EF of 58 g CO_{2e}/kWh, the overall CF would decrease by considerable amounts of 59% and 65% for plants E and F, respectively. This proves that by moving towards less carbon intensive sources in the energy mix, such as biomass, wind and solar, the total CF of WWTPs can be enormously decreased without the need of modifying the technological processes within the WWTPs.

3.7. Impact of co-digestion on the CF

Co-digestion of sewage sludges with external substrates was applied in four of the studied WWTPs (A–D). This process enhanced both the biogas production and the on-site energy production.

On-site biogas production is one of the best solutions to achieve energy neutral conditions in WWTPs (Gao et al., 2014; Maktabifard et al., 2018). Energy recovery via biogas driven CHP units or boilers helps decrease the indirect GHG emissions (scope 2) related to energy consumption from the power grid. However, biogas production may adversely affect the direct GHG emissions (scope 1) due to biogas loss from the AD as well as due to CH_4 and N_2O release during incomplete biogas combustion by engines or burners.

The effect of co-digestion on the total CF has not been addressed in the literature. Therefore, this section is dedicated to compare two possible scenarios: (i) AD of sewage sludges, and (ii) AD of sewage sludges and external biomass (co-AD). This issue is further shown based on the data from plant A. The results showed that the average biogas yield was 500 and 540 m^3/Mg volatile suspended solids (VSS) for sewage sludges and a mixture of the sewage sludges and fat, respectively. The biogas yield for fat was roughly calculated based on the difference in the biogas yield between the two mixtures and the weight fraction of fat in the mixture. For comparison, the biogas yield for WAS and fat was reported 250–350 m^3/Mg VSS and > 1000 m^3/Mg VSS, respectively (Weiland, 2010). Zhang et al. (2017b) investigated co-digestion of food waste and fat. Based on their findings, for the optimum fat content of 30%, the methane yield was as high as 630 ml/gVSS.

In the present study, it was estimated that by co-digesting 7% additional (in addition to the mixed sewage sludges) organic matter of fat, the biogas production increased by 17%. This additional biogas production increased the on-site electricity production and the level of energy neutrality of the WWTP from 63% to 73%. However, the addition of fat simultaneously increased the direct CF (scope 1) in the WWTP while decreasing the indirect CF (scope 2). It resulted in the overall CF decrease by 7%. Therefore, the increased direct emission (235 Mg CO_{2e}/year) was balanced by the enhanced biogas production and energy recovery. At the current state of the EFs for electricity from the grid, increasing the biogas production by co-digestion remains reasonable in terms of the total CF. However, in the future, this judgment may alter due to the ascending trend of the direct CF in the overall WWTP emission.

4. Conclusions

The analysis of the energy neutrality and carbon footprint minimization targets in municipal WWTPs revealed that:

- The total specific CF_{PE} is not directly related to the level of energy neutrality in WWTPs. However, strong correlation was found between the level of energy neutrality and the indirect to direct GHG emissions ratio.
- WWTPs applying energy recovery demonstrate the indirect to direct GHG emissions ratio lower than 50%.
- A shift towards less carbon intensive energy sources is an alternative to CF mitigation via technological upgrades.
- Co-AD remains a reasonable option in terms of both energy neutrality and the total CF.

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.

Acknowledgement

The project was financially supported by Regional Fund for Environmental Protection and Water Management in Gdansk/Poland in the “Pomeranian R & D projects” competition (2017 edition), grant No. WFOS/D/201/3/2018.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biortech.2019.122647.

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