Science of the Total Environment 806 (2022) 150436

Contents lists available at ScienceDirect

Science of the Total Environment

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

# Comprehensive evaluation of the carbon footprint components of wastewater treatment plants located in the Baltic Sea region

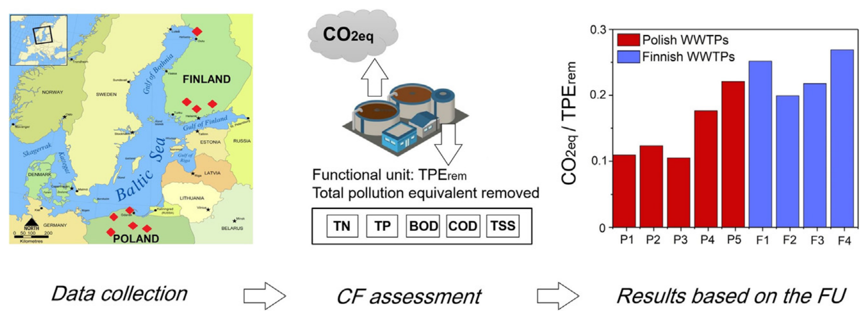
Mojtaba Maktabifard [a,b,*], Alexis Awaitey [b], Elina Merta [c], Henri Haimi [c], Ewa Zaborowska [a], Anna Mikola [b], Jacek Mąkinia [a]

[a] Faculty of Civil and Environmental Engineering, Gdańsk University of Technology, Narutowicza Street 11/12, 80-233 Gdańsk, Poland
[b] Department of Built Environment, Aalto University, PO Box 15200, FI-00076 AALTO, Finland
[c] FCG Finnish Consulting Group, PO Box 950, 00601 Helsinki, Finland

## HIGHLIGHTS

- CF of WWTPs mainly depends (70%) on direct emissions from the processes.
- Energy consumption is the main component (>30%) of the WWTPs indirect emissions.
- Functional unit based on collective pollutant removal enabled to better compare WWTPs
- WWTPs could offset up to 27% of CF by applying new practices, e.g. selling biofuel.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 6 August 2021
Received in revised form 14 September 2021
Accepted 15 September 2021
Available online 21 September 2021

Editor: Qilin Wang

*Keywords:*
Emission offset
Functional unit
Greenhouse gas emissions
Low-carbon operations
Nutrient removal

## ABSTRACT

Finland and Poland share similar environmental interests with regard to their wastewater effluents eventually being discharged to the Baltic Sea. However, differences in the influent wastewater characteristics, treatment processes, operational conditions, and carbon intensities of energy mixes in both countries make these two countries interesting for carbon footprint (CF) comparison. This study aimed at proposing a functional unit (FU) which enables a comprehensive comparison of wastewater treatment plants (WWTPs) in terms of their CF. Direct emissions had the highest contribution (70%) to the total CF. Energy consumption dominated the total indirect emissions in both countries by over 30%. Polish WWTPs benefitted more from energy self-sufficiency than Finnish plants as a result of higher electricity emission factors in Poland. The main difference between indirect emissions of both countries were attributed to higher chemical consumption of the Finnish WWTPs. Total pollution equivalent removed ($TPE_{rem}$) FU proposed enabled a better comparison of WWTPs located in different countries in terms of their total CF. High correlations of $TPE_{rem}$ with other FUs were found since $TPE_{rem}$ could balance out the differences in the removal efficiencies of various pollutants. Offsetting CF was found a proper strategy for the studied WWTPs to move towards low-carbon operation. The studied WWTPs could reduce their CF from up to 27% by different practices, such as selling biofuel, electricity and fertilizers. These findings are applicable widely since the selected WWTPs represent the typical treatment solutions in Poland, Finland and in the Baltic Sea region.



* Corresponding author at: Faculty of Civil and Environmental Engineering, Gdańsk University of Technology, Narutowicza Street 11/12, 80-233 Gdańsk, Poland.
*E-mail address:* mojmakta@pg.edu.pl (M. Maktabifard).

https://doi.org/10.1016/j.scitotenv.2021.150436

## 1. Introduction

Municipalities require resources to build infrastructure and provide services for society. The consumption of resources and provision of services to society lead to an inevitable footprint on the environment. Although municipal operations, such as waste and wastewater management, are beneficial for environmental protection, they still have negative environmental impacts. For example, the operation of wastewater treatment plants (WWTPs) reduces the environmental impact of the sewage effluent on the receiving waterbodies. On the other hand, substantial amounts of greenhouse gases (GHGs) are released from WWTPs (Delre et al., 2017).

According to the Intergovernmental Panel on Climate Change (IPCC), waste and wastewater treatment is responsible for approximately 3% of the global GHG emissions (IPCC, 2014). GHG can be emitted from WWTPs directly or indirectly. The direct GHG emissions occur during wastewater and sludge treatment processes (IPCC, 2007). The consumption of resources, such as energy and chemicals, which are required for wastewater treatment, indirectly contributes to the GHG emissions (Fitzsimons et al., 2016; INCOPA, 2014).

Carbon footprint (CF) is a tool to measure the total set of GHG emissions caused by an activity, product or a defined system such as WWTPs (Flores et al., 2020). Most literature studies use carbon dioxide ($CO_2$) equivalent as the CF, which is defined as direct and indirect GHG emissions released from the WWTPs (Chen et al., 2020). The reported CF studies of WWTPs could be varied based on CF assessment methodologies, type of treatment processes for sludge and wastewater, the country-specific carbon intensity of the energy mix and etc. Despite these differences, the major contributors to CF for most studies are similar, including fugitive GHGs, especially nitrous oxide ($N_2O$) from wastewater treatment (Delre et al., 2017; Gustavsson and Tumlin, 2013; Mölsä, 2020; Maktabifard et al., 2020). This is due to the large amount of $N_2O$ emissions and its global warming potential (GWP), which is 265 times higher than $CO_2$ (IPCC, 2014).

The energy mixes of different regions and countries may have a variable effect on the CF of WWTPs, depending on the carbon intensities of their energy production. In the countries which energy mixes are heavily reliant on fossil fuels, WWTPs tend to have a high share of their CF attributed to energy consumption. In contrast, the CF shares for energy consumption are lower in the countries with a higher reliance on renewable fuels (Gustavsson and Tumlin, 2013; Maktabifard et al., 2020).

One common methodological difference between the CF assessment studies for WWTPs is the choice of a functional unit (FU). The ISO 14040:2006 standard, which is a framework for life cycle assessment (LCA), defines a FU as "*quantified performance of a product system for use as a reference point*" (ISO, 2006). The most commonly reported FU for the CF of WWTPs is the population equivalent (PE), followed by a unit volume of treated wastewater (1 $m^3_{wastewater}$) (Flores et al., 2020; Gustavsson and Tumlin, 2013; Mölsä, 2020; Maktabifard et al., 2020; Parravicini et al., 2016). In a few CF studies, the FUs were based on the removed load of a specific pollutant (e.g., kg nitrogen or kg phosphorus removed) (Delre et al., 2017; Parravicini et al., 2016). The aforementioned FUs solely focus on either one water quantity or quality parameter and do not reflect the overall treatment efficiency of the plant. As a consequence, CF results with different FUs cannot accurately be compared and a FU accounting for various pollutant loads and removal performances in a single unit is lacking.

Finland and Poland follow the European environmental legislation (UWWTD, 2017). Furthermore, both countries share similar environmental interests with regard to their wastewater effluents, eventually being discharged to the Baltic Sea. However, differences in the influent wastewater characteristics, treatment processes, operational conditions, carbon intensities of energy mixes, and local effluent limits of both countries make these two countries interesting for a CF comparative study. This comparison would highlight the different carbon intensities, and consequently would be a step forward for implementing the net zero carbon strategy in wastewater management in different countries.

The main aim of this study is to propose a single FU that relates the total CF to collective pollutant removal. Such a FU enables a more consistent comparison of WWTPs located in different countries in terms of their CF. Therefore, the CF was initially assessed for the selected plants located in Finland and Poland. The effect of key differences in both direct and indirect emissions was investigated. Then the correlations between the single FU and other commonly used FUs were analyzed to determine their effect on the CF of WWTPs. Finally, specific recommendations were given for the studied WWTPs to reduce their CF. These recommendations could be applicable widely since the selected WWTPs represent the typical treatment solutions in the Baltic Sea region.

## 2. Material and methods

### 2.1. Site descriptions

The sites selected for CF assessment (five Polish (P1-P5) and four Finnish (F1-F4) WWTPs) were medium or large scale municipal facilities. The basic annual characteristics, including the size, biological process configuration, and sludge handling and disposal processes can be found in the supplementary material (see Appendix A, Table A1). The average annual values (based on data from the year 2017) of the influent flow rates, pollutant loads, effluent permits and 7 days Biochemical Oxygen Demand ($BOD_7$) to nitrogen and phosphorus ratios of the plants are listed in Table A2 in the supplementary material. The sampling method used for the plants in both countries was the 24-hour flow proportional composite sampling.

#### 2.1.1. Polish wastewater treatment plants

The five Polish municipal WWTPs were located in northern Poland and discharged their effluent (directly or indirectly) to the Baltic Sea. All the studied plants implemented enhanced biological phosphorous removal (EBPR), which is a common practice in Poland. Coagulants, polymers and other chemicals were only used for accidental supporting phosphorus removal, sludge dewatering and odour control. External carbon in the form of methanol was used in one plant (P2). Due to sufficient alkalinity of the influent wastewater, no additional alkaline chemicals were used among the studied plants.

The studied WWTPs were electricity self-sufficient to some degree (29-98%) except for plant P5, which totally relied on the grid's electricity. Plant P3 was most energy efficient with the highest biogas production among all the studied cases. Plants P1, P2 and P3 used external substrates for co-digestion with sewage sludges. Aside from plant P5, the Polish plants produced their required heat primarily from the combined heat and power (CHP) generators. Plant P4 combusted additional biogas in boilers to cover the heat consumption (a quarter of the heat consumption). In the case of plant P5, the required heat was partially produced by burning natural gas in on-site boilers, while the remaining portion of heat was provided through a heat pump.

#### 2.1.2. Finnish wastewater treatment plants

The four Finnish municipal WWTPs were located in the southern and central Finland and implemented chemical precipitation for phosphorous removal, which is a commonly used process in Finland. Aside coagulants, polymers and alkaline chemicals were used for enhancing sludge dewatering, sedimentation, and pH adjustment. Polymers were also used in the tertiary flotation process in plant F1. Methanol was used by plants F1 and F4 to enhance denitrification. During the studied period, additional chemicals were consumed by plant F4, which implemented the KemiCond (Cornel et al., 2005; Schaum et al., 2014) and DesinFix (effluent disinfection) processes by Kemira. That plant was the only site in this study with a requirement for effluent disinfection.

The studied Finnish WWTPs relied on the grid for >90% of their electricity consumption. For heat consumption, plant F4 relied fully on the district heat, while the other plants supplemented the district heat with heat from biogas (produced from the anaerobic digestion (AD)) or natural gas combustion. During the studied period, plant F3 sold biogas and electricity to the grid, while plant F4 sold heat from biogas combustion to the district heating network.

### 2.2. Data collection for carbon footprint analysis

The CF analysis was based on the annual operating data for the year 2017. Data collection forms in a spreadsheet format were given to the case study plants to be filled. The forms were separated into different categories as shown in Table A3 in the supplementary material.

The emission factors (EFs) for chemicals were adopted from chemical manufacturers and literature studies. The electricity and district heat EFs were obtained from the energy suppliers of the WWTPs and literature. The direct emissions were calculated based on literature EFs since on-site measurement data were not available for most of the studied WWTPs. Additional information on the treatment efficiencies was derived from environmental permits and official monitoring reports of WWTPs. More details on the EFs used in this study are provided in Table A4 in the supplementary material.

### 2.3. Carbon footprint calculation

The CF calculation was based on the routine plant activity data, combined with the chosen EFs from literature, and energy or chemical suppliers (Table A4 in the supplementary material). The schematic diagram, shown in Fig. A1 in the supplementary material, summarizes the guidelines used for the CF assessment in this study. The boundary used for the CF study was the gate to gate which comprises all the treatment processes, including sludge and screenings, chemical use, transport of screenings and sludge, and energy consumption. Furthermore, the emissions from the disposal of sludge were also included. The sludge disposal scenarios refer to the fate of sludge after AD (operations conducted both inside and outside the WWTPs) and can go beyond the plants boundaries. The emissions from the sewer network, infrastructure and equipment were less important and excluded from the analysis as suggested by (Gustavsson and Tumlin, 2013).

CF is divided to direct and indirect emissions. Direct emissions comprise $N_2O$ and methane ($CH_4$) released from different sections of the plants. The main empirical EFs used for calculating the direct emissions comprise $EF_{N2O} = 0.016$ kg $N_2O$/kg $N_{influent}$ for calculating the direct $N_2O$ emissions from wastewater treatment processes (IPCC, 2019), $EF_{CH4} = 0.0025$ for calculating the direct $CH_4$ emissions in the activated sludge process (Gustavsson and Tumlin, 2013), $CH_{4\ slippage} = 1\%$ of $CH_4$ combusted (Schaum et al., 2016) and 0.4% leakage of the total produced biogas from AD (Tauber et al., 2019). Indirect emissions include GHG emissions from energy, chemical and transportation sectors. The EFs include both general factors and plant-specific factors. The general factors are kept constant for all the sites in each country. These factors refer to transportation, chemical manufacturing and fugitive emissions of $CH_4$ and $N_2O$. The plant-specific factors are unique and variable between WWTPs. These vary between sites depending on the energy supplier and source of energy or fuels used in their electricity and heat generation processes. For the Polish WWTPs, the EF for electricity production was a nationwide average, whereas the Finnish WWTPs used regional EFs. Eventually, the calculated fugitive emissions of $CH_4$ and $N_2O$ were converted to $CO_2$ equivalents ($CO_{2eq}$) based on the conversion factors from the IPCC report (IPCC, 2014).

### 2.4. Carbon footprint offsets

In the present study, the offsets are defined as products (e.g., energy, biogas or fertilizer) recovered within the WWTP's boundary, which can subsequently be sold to replace more carbon-intensive conventional products (e.g., natural gas, synthetic fertilizer). The net emission reduction by replacing the conventional product, constitute offsets. The considered offsets were based on the recovered electricity or heat used on-site or sold to the grid, biogas sold as vehicle fuel, and the amount of synthetic fertilizer replaced by composted (or non-composted) sludge. The offsets were calculated based on EFs from the literature, the nationwide EF for electricity production (Polish plants) and the EFs from local energy suppliers for the Finnish WWTPs (Table A4 in the supplementary material).

### 2.5. Proposed functional unit

In addition to the PE, total phosphorus (TP) removed, total nitrogen (TN) removed and unit volume of treated wastewater used as the conventional FUs, this study proposes the modified FU previously introduced by Longo et al. (2016) to account for the total pollutant load removed. The pollutants considered comprise BOD, chemical oxygen demand (COD), total suspended solids (TSS), TN and TP concentrations. The total pollution equivalent removed ($TPE_{rem}$) has been already utilized for energy efficiency (Longo et al., 2016) and operational cost monitoring (Haimi et al., 2020) but it has not been applied to CF assessment in the literature so far. In this method, the $TPE_{rem}$ is calculated by a weighted sum of the compounds that have a major influence on the quality of receiving water body from the following formula:

$$TPE_{rem}(t) = \sum_{i=1}^{n} W_i * POL_{rem,i}(t) \quad (1)$$

where n is the number of different pollutants, $W_i$ is the weight for the specific pollutant, $POL_{rem}$ is the daily amount of the single pollutant removed (kg/day) and *t* is the calculation period (365 days).

The weights assigned in this study are $W_{BOD} = 1$, $W_{COD} = 1$, $W_{TSS} = 2$, $W_{TN} = 18$ and $W_{TP} = 100$. These weights were originally adopted from Benedetti et al. (2008) and were modified based on the oxygen consumption potential index used by Finnish Water Utilities Association (FWUA, 2013).

## 3. Results and discussion

### 3.1. Overall carbon footprint of the wastewater treatment plants

Table 1 shows the annual amounts of GHG emitted by each plant, along with the CF expressed in different FUs.

The CF of the studied plants ranged from 18 to 77 kg $CO_{2eq}$/PE. The average annual CF of the Polish and the Finnish WWTPs was 31 and 63 $CO_{2eq}$/PE, respectively. These fall within the wide range of 7 to

**Table 1**
Total annual CF of the studied WWTPs based on various functional units.

| WWTP | ton $CO_{2eq}$ | kg $CO_{2eq}$/kg $TPE_{rem}$ | kg $CO_{2eq}$/kg $TN_{rem}$ | kg $CO_{2eq}$/kg $TP_{rem}$ | kg $CO_{2eq}/m^3$ treated wastewater | kg $CO_{2eq}$/PE /year |
|---|---|---|---|---|---|---|
| P1 | 4894 | 0.11 | 8 | 47 | 0.6 | 20 |
| P2 | 1846 | 0.13 | 7 | 45 | 0.7 | 38 |
| P3 | 1575 | 0.11 | 7 | 53 | 0.7 | 18 |
| P4 | 2042 | 0.18 | 13 | 73 | 0.6 | 36 |
| P5 | 2619 | 0.22 | 13 | 125 | 0.8 | 43 |
| F1 | 5874 | 0.25 | 20 | 92 | 0.7 | 72 |
| F2 | 3345 | 0.20 | 16 | 91 | 0.6 | 44 |
| F3 | 6157 | 0.22 | 18 | 100 | 0.6 | 57 |
| F4 | 12910 | 0.27 | 32 | 93 | 0.7 | 77 |

100 kg $CO_{2eq}$/PE reported in the literature studies (Gustavsson and Tumlin, 2013; Maktabifard et al., 2020; Mölsä, 2020; Parravicini et al., 2016).

The CF represented in terms of wastewater volume showed more similarities for both countries, having the same average emission of approximately 0.7 kg $CO_{2eq}/m^3_{wastewater}$. This suggests that variations in the influent volume and pollutants concentration affect the CF result in terms of unit of treated wastewater. The average shares of GHG emissions by the category for the Polish and Finnish WWTPs are shown in Fig. 1.

### 3.2. Direct emissions

The direct emissions had the highest share in the total CF (67% Finnish and 72% Polish plants, respectively). Furthermore, those emissions primarily originated from $N_2O$ released from the bioreactors. For both countries, over a half of the total CF was attributed to the $N_2O$ emissions, i.e., 52% and 58% for the Finnish and Polish plants, respectively. High contributions (>50%) of $N_2O$ emissions in the CF of WWTPs have been also reported in the literature (Daelman et al., 2013; Desloover et al., 2011).

Due to the EF for $N_2O$ being based on TN concentration in the influent wastewater, higher N-load resulted in higher $N_2O$ emissions per PE. Mannina et al. (2019) highlighted that increasing N-load, leads to the growth of ammonia oxidizing bacteria (AOB) favouring $N_2O$ formation in the aerobic reactors. Based on their results, the most influencing factor on GHG emission was related to the influent COD to TN ratio. Fig. 2a shows the correlation between the influent TN load to the studied plants and the daily CF per PE. The average daily N-load between studied WWTPs varies from 7 to 18 g N/PE/d. The increasing specific N-load had a direct effect on the CF. On average, the Finnish plants had a higher daily N-load per PE than the Polish plants (~14 vs. ~11 g N/PE/d). One of the factors, which could result in higher influent N-loads, is the amount of protein consumed per capita. The Food and Agriculture Organization of the United Nations (FAO) reported the specific protein consumption of 39 and 36 kg/PE/year in Finland and Poland, respectively (FAO, 2010). For comparison, a recent CF assessment on the Iranian WWTPs by Nayeb et al. (2019) reported consumption of protein at 31 kg/PE/year and assumed a lower specific N-load (10 g N/PE/d). The research by Ramírez-Melgarejo et al. (2020) emphasized that TN should be the parameter used to know the variable amount of nitrogen in the system and thus calculate the $N_2O$ emissions.

All the studied plants fall within 90% confidence limit in Fig. 2a besides plant P2. Plant P2 is located on the coastline of the Gulf of Gdańsk and collects wastewater in a partially combined sewer system from the adjacent towns (touristic region). As a consequence, there was a significant difference between the N load in summer (1090 kg N/d) and winter (700 kg N/d).

GHG emissions from the sludge treatment processes, such as, sludge digestion and composting, were the second largest contributor to the direct CF. These were primarily the direct $CH_4$ emissions but also $N_2O$ emissions from composting were included. The total CF from sludge disposal refers to the fate of sludge after AD and was responsible for approximately 10% of the total CF in both Finnish and Polish WWTPs. Comparing to the direct $N_2O$ emissions, less information is available in the literature on contribution of direct $CH_4$ emissions from bioreactors to the total CF of WWTPs. Ribera-Guardia et al. (2019) emphasized a potentially important role of the direct $CH_4$ emissions. Based on long-term monitoring campaigns, the reported share of $CH_4$ emissions from the aerated zones was as large as 45% of the total CF.

### 3.3. Indirect emissions

The indirect emissions constituted 28% and 33% of the CF for the Polish and Finnish WWTPs, respectively. Energy consumption was the dominant component in the total indirect emissions with 18% and 26% shares of the total CF for the Finnish and Polish WWTPs, respectively. The main difference between the indirect emissions of both countries was attributed to chemical consumption (1% and 8% of the total CF for Polish and Finnish WWTPs, respectively). Higher chemical consumption of the Finnish plants was mostly due to the coagulants consumed for chemical precipitation and alkaline chemicals for pH adjustment. The indirect emissions from transportation in both countries had a marginal share of 1%.

#### 3.3.1. Electricity consumption

To determine the energy efficiency of the WWTPs, the electricity consumption was normalized to the $TPE_{rem}$ FU (kWh/ton $TPE_{rem}$) to account for the actual treatment efficiency of the plants. The electrical efficiencies were higher for larger WWTPs, which consumed less electricity per $TPE_{rem}$ (Fig. 2b). Plants P1, F3 and F4 were the three studied WWTPs with over 100,000 PE in size and had the lowest specific electrical consumptions (78, 112 and 123 kWh/ton $TPE_{rem}$, respectively) among all the plants. A study by Tukiainen (2009) assessed the specific energy consumption of WWTPs in Finland which also resulted in larger plants having the least consumption per 1 $m^3$ wastewater treated. Another study by Haslinger et al. (2016) presented operating energy consumption of Austrian WWTPs and reported the average specific energy consumption of large plants (>100,000 PE) 28 kWh/PE-$COD_{120}$, while for medium sized plants (50,000 to 100,000 PE) it was 20% higher (34 kWh/PE-$COD_{120}$). The COD-population equivalents (PE-$COD_{120}$) were calculated from the mean annual COD-load of the plant (120 g COD per person and day corresponding to 60 g $BOD_5$ per person and day).

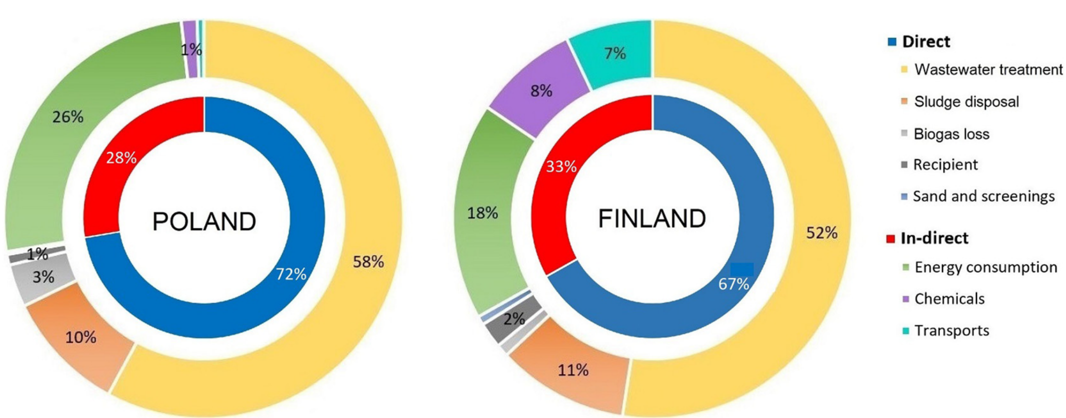



**Fig. 1.** Average annual share of the different emissions in Polish and Finnish WWTPs.

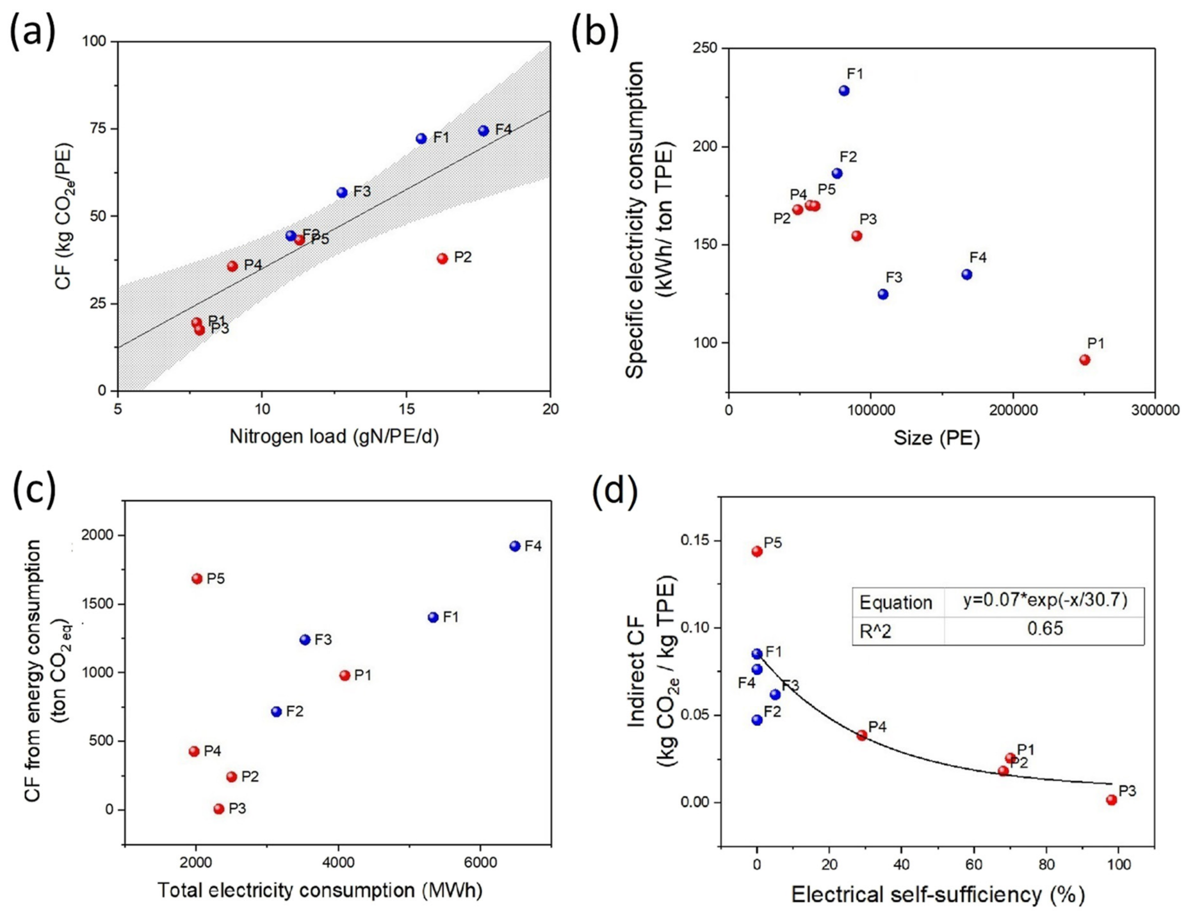



**Fig. 2.** Comparison of the relationships in the studied plants in terms of (a) Daily CF of the studied WWTPs vs. N-load per PE correlation (±90% confidence region); (b) specific electricity consumption vs. size of the WWTP; (c) energy related CF of the studied WWTPs vs. total electricity consumption; (d) correlation between indirect CF and the degree of electricity self-sufficiency of the plants.

Four out of the five Polish WWTPs partially covered their required electricity with the electricity produced on-site from biogas combustion. This reduced the Polish plants reliance on the grid for electricity and the associated emissions in external power plants. Only one Finnish WWTP (plant F3) was partially electrical self-sufficient (~5%) with all other plants relying fully on the grid. Although the Finnish WWTPs consumed more electricity on average (Fig. 2c), they were still in the same emission range as those in Poland. This is due to the lower electricity production EFs in Finland compared to Poland.

As shown in Fig. 2d, there was a negative correlation between the degree of energy self-sufficiency of the plants (mostly Polish WWTPs) and their indirect CF. Higher on-site electricity production of the Polish WWTPs resulted in a greater save of $CO_{2eq}$ off-site emissions. This was based on the high electricity EF (0.81 kg $CO_{2e}$/kWh) in Poland due to electricity being produced mainly in the coal power plants (NCEM, 2017). Zaborowska et al. (2021) emphasized that by increasing the share of renewable energies instead of coal, the focus shifts from the indirect to direct emissions.

On the other hand, since the EF for electricity production is much lower in Finland, increasing the energy self-sufficiency of the Finnish WWTPs would lead to relatively lower emissions savings compared to Poland. From a CF perspective, the Polish WWTPs would benefit more from energy self-sufficiency than Finnish WWTPs.

### 3.3.2. Chemicals

The studied Finnish WWTPs used chemical precipitation for phosphorous removal, while EBPR was applied in Polish WWTPs. The implementation of EBPR greatly reduced the amount of used chemicals and the respective emissions. Furthermore, in comparison with the Finnish sites, the Polish WWTPs had higher TP effluent standard limits (1 or 2 mg P/l, depending on the plant size) which could be achieved biologically.

For chemical precipitation, the amount of phosphorous removed is mostly based on the molar ratio of the metal ion dosed to the initial phosphorous concentration ($Me_{dose}/TP_{initial}$). The relationship between phosphorous removal efficiency and the $Me_{dose}/TP_{initial}$ ratio is not fully linear. According to literature, for very low target phosphorous concentrations, higher $Me_{dose}/TP_{initial}$ ratios are required than the theoretical demand (Szabó et al., 2008). An experimental study by Luk (1999) showed that the $Me_{dose}/TP_{initial}$ ratio ranged from 3 to 5 when achieving phosphorous removal rates >90% for municipal wastewater, with the higher ratios applied in practice. To achieve the residual TP concentrations between 0.3 and 1.0 mg P/l, ratios of 1.2-4 for aluminium or iron are suggested (Minnesota Pollution Control Agency, 2006). Since the effluent phosphorous requirements for the Finnish WWTPs are stricter (>50% lower) than the Polish plants, it is logical that Finnish WWTPs would require a high dosage of coagulants. Based on the site data, the $Me_{dose}/TP_{initial}$ ratios ranged from 5.1 to 5.8, which is close to the range from the literature. GHG emissions related to the chemical precipitation method are anticipated to increase in the future if more stringent TP requirements are enforced. This is based on the fact that much higher Me/P ratios are needed for higher removal rates.

Plant F4, implemented the KemiCond process for treating sludge, which is a chemical intensive process accounting for approximately 28% of the total indirect chemical emissions of the plant. Additionally, since plant F4 was the only site with an effluent disinfection

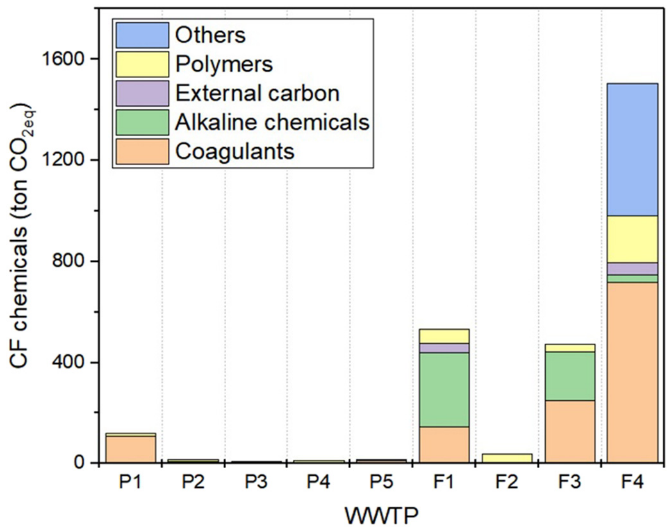



**Fig. 3.** Chemical consumption indirect emissions for WWTPs in this study.

requirement, chemicals which were used for this purpose accounted for ~7% of the total indirect chemical emissions. The chemicals used for the aforementioned processes are categorized under "others" in Fig. 3.

### 3.4. Offsets

The CF offsets in this study originated from selling the composted sludge as fertilizer, energy in the form of heat or electricity and biogas which was replacing natural gas for vehicular use. Plant F3 achieved the highest offsets, partly due to the sale of high amounts of biogas derived from co-digestion of sludge and municipal/industrial bio-waste. The total offsets for plant F3 accounted for over 10% of its total CF. For the other Finnish WWTPs (F1, F2 and F4), the majority of offsets were attributed to the replacement of conventional synthetic fertilizers with composted sludge. The offsets from the sale of heat and electricity did not contribute significantly due to the low EF for electricity and heat production in Finland. Fig. 4a shows the amount of CF that each WWTP could offset on the annual basis in the year of the study.

The Polish WWTPs benefitted more from replacing the grid's electricity with on-site produced electricity due to the high national EF for electricity production. Plant P2 managed to offset 505 ton $CO_{2e}$/year accounting for 27% of its total CF (Fig. 4b).

Offsetting the CF is a proper approach for WWTPs to move towards carbon neutrality. On the other hand, biogas utilization or implementing CHP units require complex control strategies, which increases management complexity as highlighted by Chen et al. (2020). Moreover, Pahunang et al. (2021) emphasized that more research is required to assess the cost-effectiveness of different technologies which could increase the CF offsets of WWTPs. Fig. 4b compares the CF of the studied plants with and without consideration of offsets.

### 3.5. Correlation between influencing parameters and CF

Although a high correlation does not necessarily indicate a major contributor to the CF, it can help identify any trends between the contributing factor and the CF. These trends were analyzed with the Pearson's correlation co-efficient (Fig. 5). The PE and $TPE_{rem}$ FUs were chosen for comparison, and positive correlations between all parameters and the CF were observed, except for the biogas production and degree of energy self-sufficiency.

Although $N_2O$ from bioreactors was the highest contributor to GHG emissions, the influent N-load had the highest positive correlation with CF when using PE as the FU. On the other hand, while using $TPE_{rem}$ as the FU, it had the second lowest positive correlation with the CF.

As shown in Fig. 5, there was a moderate negative correlation between the biogas loss emissions and the overall CF of the plants. This was logical since more biogas production would lead to more energy recovered to be consumed on-site (increased energy self-sufficiency). This will lower the CF through the emission saved from replacing the grid energy consumption. This is further supported by all plants having strong positive correlations between energy consumed from the grid and the overall CF. The negative correlation between the biogas produced and the CF will only be applicable if the emission saved by replacing the grid energy balances the emissions duo to biogas loss, as in the case of the Polish WWTPs. Therefore, in the countries with very low electricity production EFs, it might be more beneficial to consume grid energy rather than energy consumed through biogas production on-site.

### 3.6. Assessment of the total pollution equivalent as a functional unit

The suitability of $TPE_{rem}$ as a FU was assessed by calculating the correlation of each FU vs. $TPE_{rem}$ which is presented in Fig. 6. Plant P5 was the only Polish WWTP without on-site biogas production. It was found as the only studied plant with higher indirect emissions than the direct $N_2O$ emissions. Furthermore, it had the lowest phosphorus removal

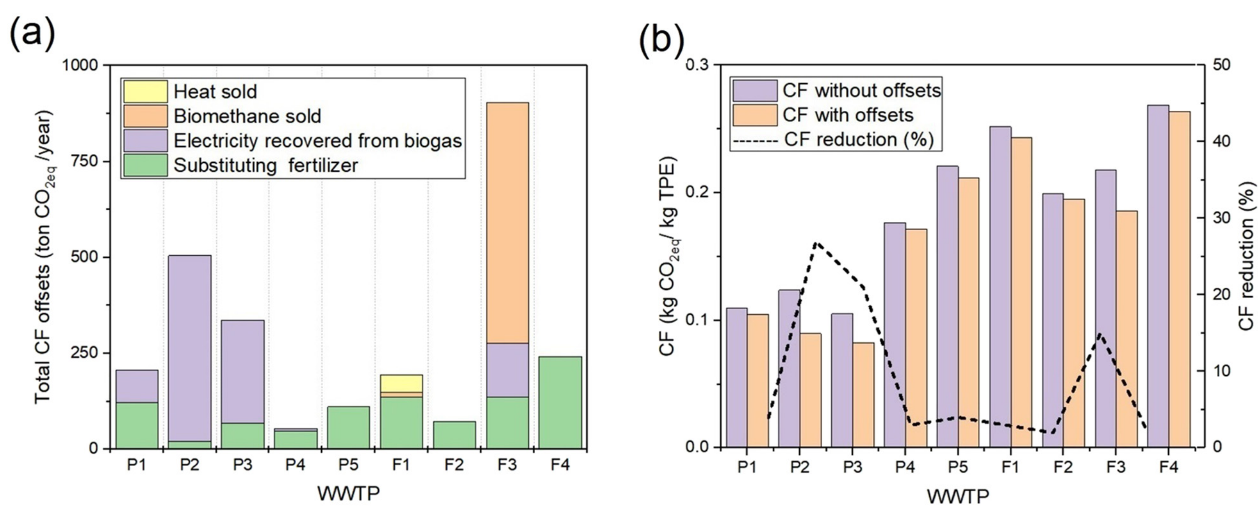



**Fig. 4.** Comparison of the offsets in the studied plants in terms of (a) total CF offsets; (b) CF without and with offsets.

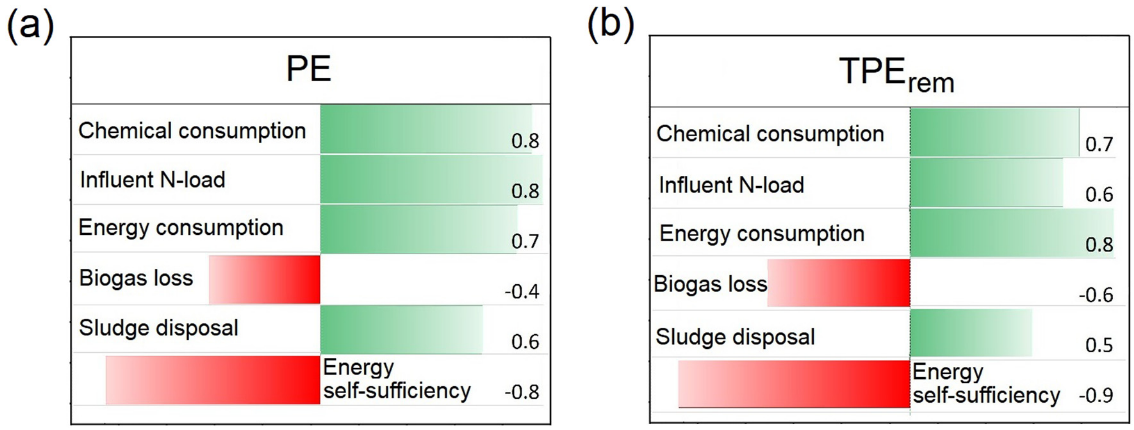



**Fig. 5.** Pearson's correlation co-efficient between contributing factors and overall CF when using (a) PE as FU; (b) $TPE_{rem}$ as FU.

efficiency (90%) due to higher regional standards for effluent TP (2 mg P/l).

The Pearson's correlation coefficients matrix is shown in Fig. 7. Since $TN_{rem}$, $TP_{rem}$, and BOD were incorporated in the $TPE_{rem}$ FU, $TPE_{rem}$ would correlate best with most of the FUs in this study. However, the unit volume of treated wastewater FU had the lowest positive correlation between all the FUs (including $TPE_{rem}$).

To illustrate the differences between the FUs, plants P5 and F3 were compared. In terms of $TPE_{rem}$, both plants had similar CF results. However, different CFs were observed between the plants in terms of unit volume of treated wastewater, $TN_{rem}$, $TP_{rem}$ and PE FUs (see Fig. 6). Plant P5 had higher emissions per kg $TP_{rem}$ and unit volume of treated wastewater. However, plant F3 had higher emissions per kg $TN_{rem}$ and PE served. Plant P5 had a lower TP removal efficiency than plant F3 as shown in Table A2 in the supplementary material. On the other hand, the TN and BOD removal efficiencies were lower for plant F3. Lower pollutant treatment efficiencies lead to higher CF values for the respective FUs ($TN_{rem}$, $TP_{rem}$, etc.). This explains the differences

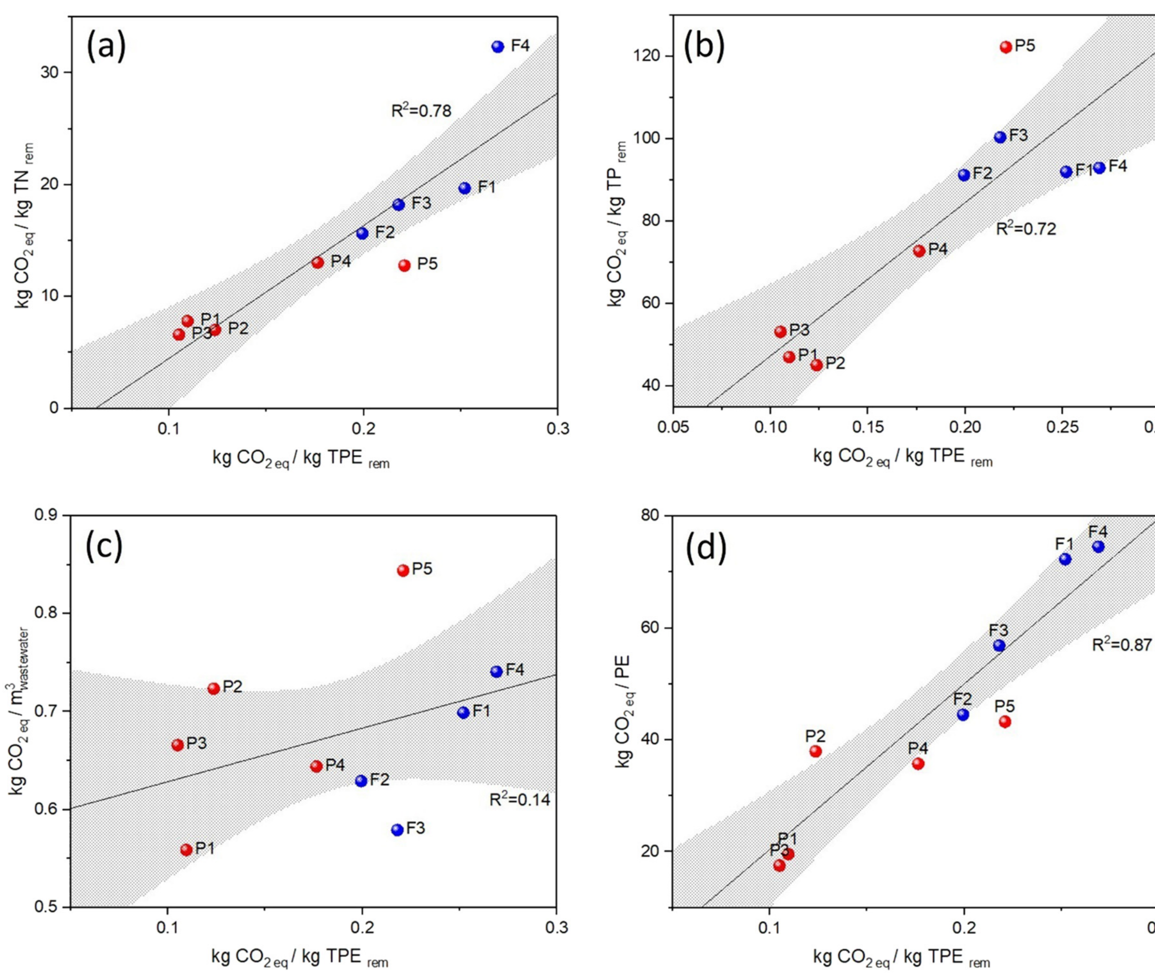



**Fig. 6.** Correlations between $CO_{2eq}$ per $TPE_{rem}$ vs. different FUs used in this study (trend lines with ±90% confidence regions) (a) TN, (b) TP, (c) $m^3_{wastewater}$ and (d) PE.

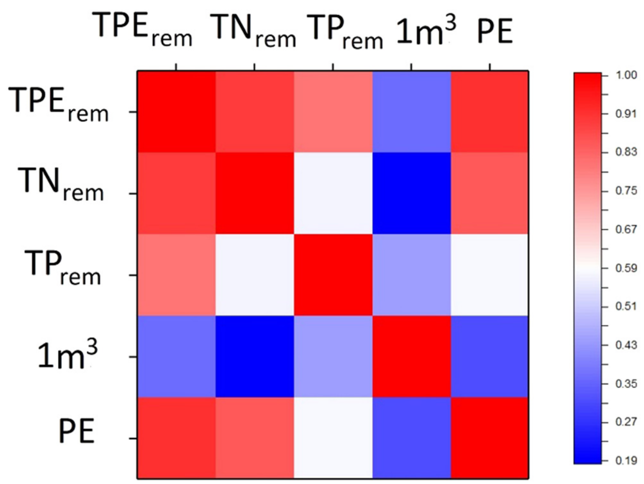



**Fig. 7.** Correlation matrices of the FUs applied in this study.

between the results for $TN_{rem}$, $TP_{rem}$ and PE FUs between plant P5 and plant F3.

A low correlation with the unit volume of treated wastewater FU is due to differences in the influent characteristics between the Polish and Finnish WWTPs. The average COD to volume ratio was approximately 1 kg COD/$m^3_{influent}$ and 0.7 kg COD/$m^3_{influent}$ for the Polish and Finnish plants, respectively. This difference is most likely due to the sewer network system of both countries. The Finnish sewer networks are typically separate but in old city centres, these are still combined with storm water run-off. In addition, the sewer networks in Finland are ageing with possible higher infiltration rates into the sewer networks. Moreover, the snow-melting period during spring would increase the volume of influent wastewater in the areas with combined networks. Hence, even though the $TPE_{rem}$ might be the same for a Polish and Finnish plant, the volume of wastewater treated would vary, leading to different results for unit volume of treated wastewater as a FU. Additionally, more influent wastewater would require more pumping, further increasing the energy consumption and CF of the plants.

The specific weights for the $TPE_{rem}$ FU can be modified based on the importance of the pollutant to the effluent receiving waterbody. For this study, TP was ranked the most important due to its eutrophication impact on the Baltic Sea (HELCOM, 2018). If other pollutants are identified as having more negative impact on the receiving waterbody, the weights can be modified accordingly.

### *3.7. Recommendations for WWTPs to reduce carbon footprint*

In plant F4, the use of polyaluminium chloride (PAC) alone constituted ~5% of the total CF. While the use of ferric sulphate accounted for just <1% of the total CF, even though the plant used ~500 ton/year more ferric sulphate than PAC. This illustrates the potential benefits for WWTPs while selecting chemicals with lower EFs. Based on the chemical production EFs, ferric sulphate and ferrous sulphate had the lowest EFs and could be alternative options to reduce the CF.

Approximately 4% of the CF in plant F4 was associated with the KemiCond process (chemical consumption and transport emissions). The main benefits of the KemiCond process were related to the improved dewatering of sludge (reducing the amount of sludge), disinfection of sludge, and reduction of bad odours. The plant should assess if the reduction of sludge transport emissions (due to more efficient dewatering from the KemiCond process) balances the emissions from the consumption and transport of KemiCond process chemicals. An alternative solution could be replacing the KemiCond process with on/off-site AD of the primary and waste activated sludge produced.

Due to the relatively high EF for electricity production in Poland, the Polish WWTPs can benefit from high offsets if more electricity from biogas is consumed on-site or sold to the grid. This would require optimizing the AD process to increase biogas production (Jenicek et al., 2012). A suitable option could be co-digestion of sewage sludge and bio-waste (Zhao et al., 2019).

The EBPR process with low chemical consumption could be an alternative for Finnish WWTPs. However, EBPR requires an optimal BOD to TP ratio. Based on Table A2 (supplementary material), the average BOD to TP ratio for the Polish plants is ~51 while that of Finnish plants is only ~31. The lower average ratio for the Finnish WWTPs would indicate less suitability for EBPR in Finland, unless an additional carbon source is added. The Finnish site with the highest BOD to TP ratio was plant F2 (the high ratio of 51 resulted from industrial influent) and thus, EBPR could be applicable to that plant.

Around half of the reviewed CF studies (available in the supplementary material, Table A5) reported the CF based on PE and therefore it was found as the most frequent FU followed by unit of influent wastewater which was adopted in 25% of the studies. Other studies reported the CF based on TN or TP removed, with Delre et al. (2017) being the only reviewed study which reported the CF based on kg C removed. There were studies in the literature such as, Chai et al. (2015) which did not report the CF based on any of the aforementioned FUs and instead used the absolute value of $CO_{2eq}$ emissions. These differences in expressing the CF of different WWTPs make the CF comparison challenging. This highlights the need for a comprehensive FU such as $TPE_{rem}$ that could be widely adopted for CF comparison of WWTPs located in different countries.

## 4. Conclusions

Direct emissions from the treatment processes had approximately 70% share in the total CF of the studied WWTPs in Finland and Poland. The main portion of the total CF (55%) was attributed to $N_2O$ released from the bioreactors. Energy consumption was the main component (over 30%) of the total indirect emissions. From the CF perspective, WWTPs located in the countries with a high EF for grid electricity benefitted more from energy self-sufficiency. The main difference between the indirect emissions of both compared countries was attributed to CF of chemicals. In the Finnish plants, that component was important and mainly resulted from coagulants consumed for chemical phosphorus precipitation. Furthermore, the FU ($TPE_{rem}$), which relates total CF to collective pollutant removal, enabled to better compare WWTPs in terms of the total CF. High correlations of $TPE_{rem}$ with other FUs were found albeit $TPE_{rem}$ could balance differences in the removal efficiencies of various pollutants. Offsetting CF appears a viable strategy to move towards carbon neutrality. WWTPs could reduce their CF up to 27% by new operational practices, such as selling biofuel, electricity and fertilizers.

## CRediT authorship contribution statement

**Mojtaba Maktabifard:** Formal analysis, Methodology, Visualization, Writing – original draft. **Alexis Awaitey:** Formal analysis, Methodology, Writing – original draft. **Elina Merta:** Data curation, Writing – review & editing. **Henri Haimi:** Conceptualization, Data curation, Writing – review & editing. **Ewa Zaborowska:** Data curation, Supervision, Writing – review & editing. **Anna Mikola:** Conceptualization, Supervision, Writing – review & editing. **Jacek Mąkinia:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing.

## 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.

## Acknowledgements

The authors acknowledge the support provided by InterPhD2 Program funded by European Union/European Social Fund (Project No. POWR.03.02.00-IP.08-00-DOK/16). The authors would like to further thank "Finnish Water Utilities Development Fund" that provided funding for the project "Carbon Footprint of Finnish Wastewater Treatment Plants".


## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.150436.

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