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Carbon Footprint of Finnish Wastewater Treatment Plants

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Abstract of master's thesis

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Abstract

Waste and wastewater treatment currently constitute approximately five percent of global greenhouse gas (GHG) emissions. Mitigating nation-wide GHG emissions requires collaboration between municipalities in reducing their carbon footprints. Since wastewater treatment plants (WWTPs) are essential municipal infrastructure, knowledge of their GHG emissions is important for assessing part of the total carbon footprint of municipalities. The aim of this thesis was to evaluate the carbon footprint of four WWTPs in Finland and provide recommendations for the sites in this study to reduce their GHG emissions.

The processes included in the carbon footprint boundary were the treatment of wastewater, treatment of sludge and screenings, transport of chemicals, transport of screenings and sludge, as well as the energy and chemical consumption of the plant. The emissions from the wastewater network and disposal of sludge were excluded from the boundaries in this study.

Carbon footprint results ranged from 45 – 82 kg CO_2eq PE^{-1} $year^{-1}$. Total emissions were dominated by fugitive emissions from wastewater and sludge treatment, with an average of 72 % of total carbon dioxide equivalent emissions (CO_2eq). Energy consumption emissions were the second highest with an average of 19 %, followed by chemical consumption emissions with 7 % of total CO_2eq emissions. Transport emissions constituted a minor share of 1 % of total CO_2eq emissions. N_2O emissions from wastewater treatment constituted an average of 59 % of total GHG emissions. Due to the high global warming potential of N_2O, results showed high sensitivity of the carbon footprint to the N_2O emission factor used. Based on the results, it was concluded that WWTPs with higher energy efficiencies or low indirect emissions would have higher shares and sensitivities to N_2O emission factors.

Due to the high share of direct emissions (particularly N_2O), it is recommended that WWTPs in Finland evaluating their carbon footprint should invest in on-site measurement campaigns of fugitive N_2O emissions of at least one year. This would reduce the under/overestimation of on-site emissions. In addition, it is also recommended that WWTPs monitor process-specific electricity consumptions which would help them identify energy-intensive processes on-site.

Keywords Carbon footprint, wastewater treatment, greenhouse gases

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Tiivistelmä

Jätteiden ja jäteveden käsittely muodostaa tällä hetkellä noin viisi prosenttia maailman kasvihuonekaasupäästöistä. Kasvihuonekaasupäästöjen alentaminen ja hiilijalanjäljen pienentäminen valtakunnallisesti vaativat kuntien välistä yhteistyötä. Koska jätevedenpuhdistamot ovat olennainen osa kuntien infrastruktuuria, niiden kasvihuonekaasupäästöjen tuntemus on tärkeää kuntien kokonaishiilijalanjäljen arvioimiseksi. Tässä työssä arvioitiin neljän Suomessa sijaitsevan jätevedenpuhdistamon hiilijalanjälki ja annettiin suosituksia tarkasteltujen jätevedenpuhdistmoiden kasvihuonekaasupäästöjen vähentämiseksi.

Hiilijalanjäljen arviointiin sisällytettiin jäteveden käsittely, lietteen ja välppäjätteen käsittely, kemikaalien kuljetus, välppäjätteen ja lietteen kuljetus, laitoksen energiankulutus sekä kemikaalien kulutus. Viemäriverkoston ja lietteen loppusijoituksen päästöt jätettiin tämän tutkimuksen rajauksen ulkopuolelle.

Hiilijalanjälkitulokset vaihtelivat välillä 45 – 82 kg CO_2ekv AVL^{-1} a^{-1}. Suorat päästöt aiheuttivat merkittävimmän osan kokonaispäästöistä. Niiden osuus oli keskimäärin 72 % kaikista CO_2ekv-päästöistä. Toiseksi suurin päästölähde oli energiankulutus, jonka päästöjen osuus oli keskimäärin 19 %. Kemikaalien kulutuksen osuus oli 7 % ja liikenteen osuus 1 % kaikista CO_2ekv-päästöistä. Jätevedenpuhdistuksen N_2O-päästöt vastasivat keskimäärin ~ 59 % kaikista kasvihuonekaasupäästöistä. Tulokset osoittivat, että hiilijalanjäljen tulos on herkkä laskennassa käytettävälle N_2O:n päästökertoimelle, mikä johtuu N_2O:n suuresta ilmaston lämmityspotentiaalista. Tulosten perusteella N_2O-päästöjen osuus hiilijalanjäljestä ja tulosten herkkyys käytettävälle N_2O-päästökertoimelle on suurempi jätevedenpuhdistamoilla, jotka ovat energiatehokkaita tai joiden epäsuorat päästöt muutoin ovat alhaiset.

Suorien päästöjen (erityisesti N_2O) suuren osuuden vuoksi on suositeltavaa, että suomalaisilla jätevedenpuhdistamoilla, joiden hiilijalanjälkeä arvioidaan, investoitaisiin ajallisesti riittävän pitkäkestoisiin N_2O-haihduntapäästöjen mittauskampanjoihin. Tämä vähentäisi puhdistamolla tapahtuvien päästöjen ali- tai yliarviointia. Lisäksi on suositeltavaa, että jätevedenpuhdistamot mittaavat ja seuraavat prosessikohtaisia sähkönkulutuksia, mikä auttaa tunnistamaan puhdistamon energiaintensiiviset prosessiyksiköt.

Avainsanat Hiilijalanjälki, jätevedenkäsittely, kasvihuonekaasut

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Alexis Awaitey

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Abbreviations

AMO	Ammonia Monooxygenase
AOB	Ammonia Oxidizing Bacteria
BNR	Biological Nutrient Removal
BOD	Biochemical Oxygen Demand
CAS	Conventional Activated Sludge
CF	Carbon Footprint
COD	Chemical Oxygen Demand
DO	Dissolved Oxygen
DS	Dry solids
EF	Emission Factor
EU	European Union
FU	Functional Unit
GHG	Greenhouse Gases
GWP	Global Warming Potential
HAO	Hydroxylamine Oxidoreductase
IPCC	Intergovernmental Panel on Climate Change
LCA	Life Cycle Assessment
LCIA	Life Cycle Impact Assessment
MBBR	Moving Bed Biofilm Reactor
MBR	Membrane Bioreactor
NaR	Nitrate reductase
NiR	Nitrite reductase
NOB	Nitrite Oxidizing Bacteria
NOR	Nitric oxide reductase
PAC	Polyaluminium chloride
PAO	Phosphorous-Accumulating Organisms
PE	Person Equivalent
SBR	Sequencing Batch Reactor
SS	Suspended Solids
TN	Total Nitrogen
TOC	Total Organic Carbon
TP	Total Phosphorous
VS	Volatile solids
WWTP	Wastewater Treatment Plants

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1 Introduction

1.1 GHG emission monitoring and trends in Finland

Anthropogenic greenhouse gas (GHG) emissions have long been identified as a driving factor for global warming (IPCC, 2014). Efforts have been made, both on a global and national scale, to reduce these GHG emissions. Globally, the Kyoto protocol and Paris agreement are examples of international agreements in mitigating GHG emissions of which Finland is a participant.

Depending on the international agreement countries are involved in, they might be obliged to report their yearly GHG emissions as part of the GHG monitoring process. For example, the Kyoto protocol obliges participating countries to establish a national system for monitoring GHG emissions (Statistics Finland, 2018). The Inter-Governmental Panel on Climate Change (IPCC) also developed guidelines for countries to estimate their greenhouse gas emissions (IPCC, 2006). Finland established its own national Greenhouse Gas Inventory System in 2005 and its actors and networks are illustrated in Figure 1 (Statistics Finland, 2018).

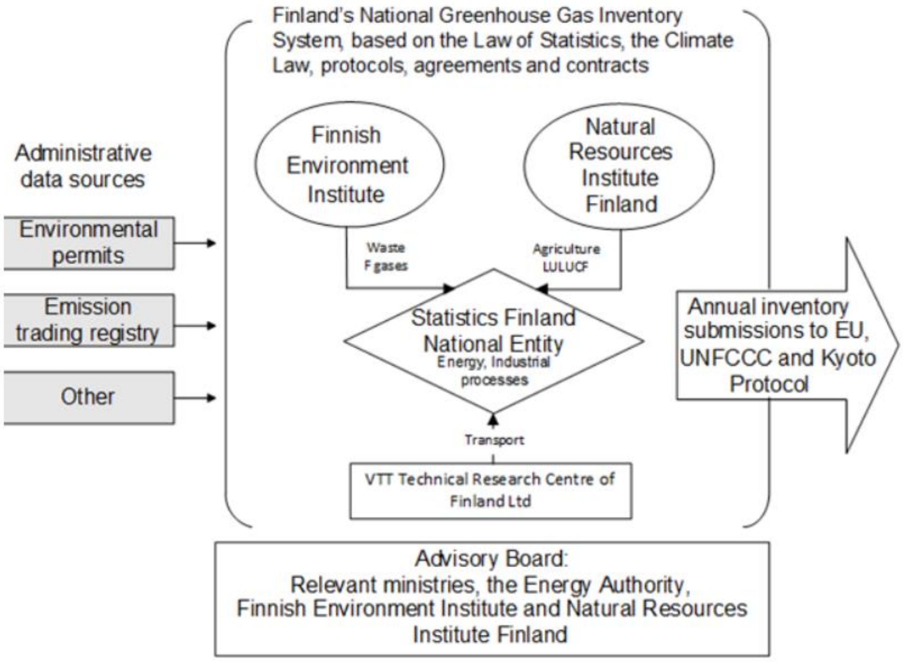
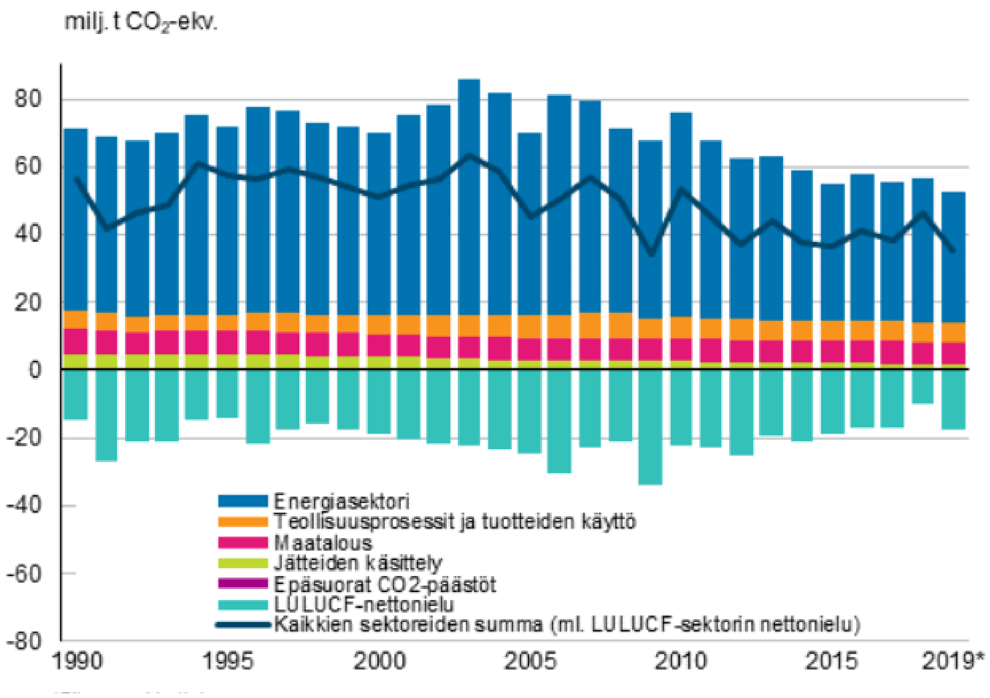


Figure 1 - National GHG inventory system in Finland (Statistics Finland, 2018)

Finland's GHG emissions have steadily declined compared to 1990 levels as shown in Figure 2. This reduction is mainly attributed to the energy sector, whose emissions have declined ~19%, relative to 1990 emissions (Statistics Finland, 2019). The energy sector is still the largest GHG emitter accounting for over 70% of total emissions as of 2019 (Statistics Finland, 2019). Although Finland's success in GHG mitigation is evident, its goal of becoming carbon neutral by 2035 would require mitigation in other sectors as well.



Blue= energy sector, Orange = Industrial processes, Pink = Agriculture, Green = waste treatment, Teal = carbon offsets

Figure 2 - Greenhouse gas emissions by sectors from 1990 to 2018 in million-ton CO_2eq (Statistics Finland, 2019)

1.2 Climate change mitigation in Finnish municipalities

On a regional scale, the Finnish Ministry of Environment implemented the "Municipal Climate Change Solutions Programme (2018–2023)" to boost greenhouse gas mitigation in municipalities. The Ministry of Environment emphasized the importance of municipalities as key players in facilitating and improving regional climate work (Finnish Ministry of Environment, 2020). Thus, the collective monitoring and quantification of GHG emissions in municipalities are important for the country to meet its national GHG mitigation targets.

The city of Oulu, for example, is a signatory of the Covenant of Mayors. This is an EU cooperation between regions and municipalities to reduce their GHG emissions through increased energy efficiency and use of renewable energy sources. This is to further support the EU's goal of reducing CO_2 emissions by 20% by 2020 compared to 1990 levels. Since national goals depend on collective regional and municipality efforts, the city of Oulu and other signatories must commit to a 40% GHG reduction from 1990 levels by 2030. This would translate to a GHG emission per capita of 4.7 t CO_2eq by the year 2030 (Liljeström and Monni, 2018).

A regional calculation model known as "ALas" was developed to calculate the emissions from municipalities and regions in Finland (Finnish Environment Institute, 2020). This model is in accordance with the IPCC (2006) guidelines, of which Finnish GHG reporting is based on. The results of the calculation based on the ALas model showed an average GHG decrease of 15% for municipalities in 2018 compared to 2005 (Finnish Environment Institute, 2020).

1.3 GHG trends in the Finnish Wastewater sector

According to IPCC, waste and wastewater treatment constitutes less than 5% of global GHG emissions (IPCC, 2007). The wastewater sector in Finland has experienced a 16% decrease in GHG emissions since 1990 as of 2017 (Statistics Finland, 2019). This reduction has been attributed to a decrease in uncollected domestic wastewaters and its associated GHG emissions (refer to Figure 3). This suggests a constant increase in sewer network connections since 1990. The emissions reported for the wastewater sector comprise of both methane (CH_4) and nitrous oxide (N_2O) emissions, converted to carbon dioxide (CO_2) equivalents.

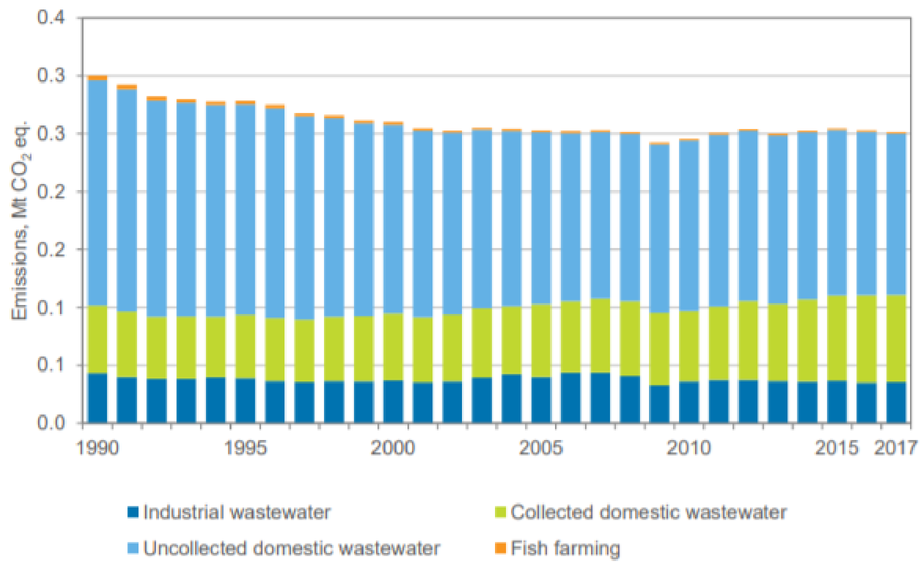


Figure 3 - Emissions from wastewater sector from 1990 to 2017 in Mt CO2eq (Statistics Finland, 2019)

Based on data gathered from the report "Greenhouse Gas Emissions in Finland 1990-2017" by Statistics Finland (2019), CH_4 emissions from the wastewater sector have gradually been decreasing overtime (refer to Figure 4). However, N_2O emissions have shown a steady, overall increase since 1990. According to the data, majority of N_2O emissions is attributed to collected domestic wastewater and hence, directly associated with the wastewater treatment process (Statistics Finland, 2019). Gradual CH_4 reduction was attributed to an increase in wastewater collection.

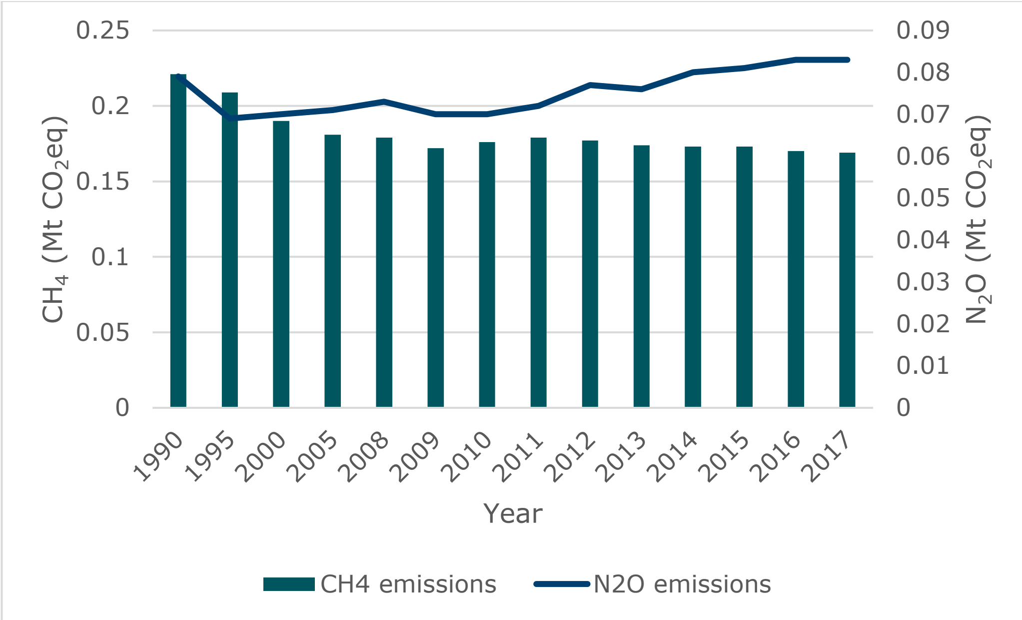


Figure 4 - Total CH_4 and N_2O emissions (in megatons) from the wastewater sector. Data taken from Statistics Finland, 2019

Opportunities exist to leverage the wastewater treatment process through resource recovery and optimized operation of biological processes, with GHG emission reduction as a goal. Wastewater treatment generates excess sludge that can be anaerobically digested to produce energy in the form of biogas. This reduces reliance on fossil fuel and results in lower emissions in the energy sector, which constitutes the highest share of emissions in Finland. Additionally, sludge can be transformed into fertilizers which further prevent their respective production emissions (Singh and Agrawal, 2008). Additionally, processes such as RAVITA™, that recovers soluble phosphorous, further demonstrates the resource recovery potential of wastewater treatment (Rossi *et al.* 2018). Thus, the wastewater sector can be regarded as an opportunity to reduce emissions in other sectors such as energy production.

However, challenges exist in the wastewater industry with regards to simultaneously balancing operational costs and meeting multiple environmental objectives. This was illustrated in a study by Arnell *et al.* (2017) which evaluated the effects of changing the operational strategy of a Swedish Wastewater Treatment Plant (WWTP) to enhanced chemical primary treatment. The results showed that the biogas production increased by 14% while the global warming potential decreased by 28%. On the other hand, due to the increased consumption of chemicals, the operational cost increased by 87%. This illustrates the importance of a wholistic, life cycle perspective when setting targets for WWTPs.

1.4 Research aim and objectives

In Finland, carbon footprint studies for WWTPs are few at the time of this study (these are reviewed in Chapter 5). The National GHG inventories rely on rough estimates that are not plant specific enough and might be under or overestimated. Considering the limited studies available, comparing and benchmarking the carbon footprint of WWTPs in Finland would be difficult without additional studies done. This thesis is part of a project involving four municipal WWTPs of varying capacities and aims to quantify their carbon footprint using as accurate plant-data as possible. This thesis aims to contribute more information regarding carbon footprint of WWTPs in Finland and highlight areas that will require further future research. The research objectives of this thesis are:

- To quantify the total carbon footprint of four case study WWTPs in Finland
- To identify major GHG emission hotspots in these WWTPs
- Propose possible measures these sites could take to reduce their carbon footprint
- Provide guidelines for WWTPs on how to assess their carbon footprints.

The thesis begins with a literature review on Life Cycle Assessment (LCA) and carbon footprint (CF) procedures to familiarize the reader with the CF process (Chapter 2.1 – 2.2). The conventional wastewater treatment process is then explained in Chapter 2.3, followed by a review of GHG emissions in WWTPs (Chapter 2.4). Literature studies regarding carbon footprint studies in WWTPs is reviewed in Chapter 2.5. The methods used in estimating their carbon footprint is discussed in Chapter 3, followed by the results and discussion in Chapter 4 – 5. Finally, the conclusion of the thesis will be given in Chapter 6.

2 Literature review

2.1 Concept of LCA

In order to assess the environmental performance of an organization, product or service, a structured methodology known as the Life Cycle Assessment is often used. According to the ISO 14040:2006 standard, LCA is defined as the "compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product or service throughout its life cycle" (ISO, 2006) In an LCA, the environmental impacts are categorized. Each impact category indicates the performance of an organization, product or service with regards to the environmental aspect of the studied entity. For example, the LCA category for assessing the climate change impact of an entity could be the global warming potential (GWP). Figure 5 illustrates the different steps in an LCA. The phases of an LCA include: i) Goal and scope definition ii) Inventory analysis iii) impact assessment and iv) Interpretation (ISO, 2006).

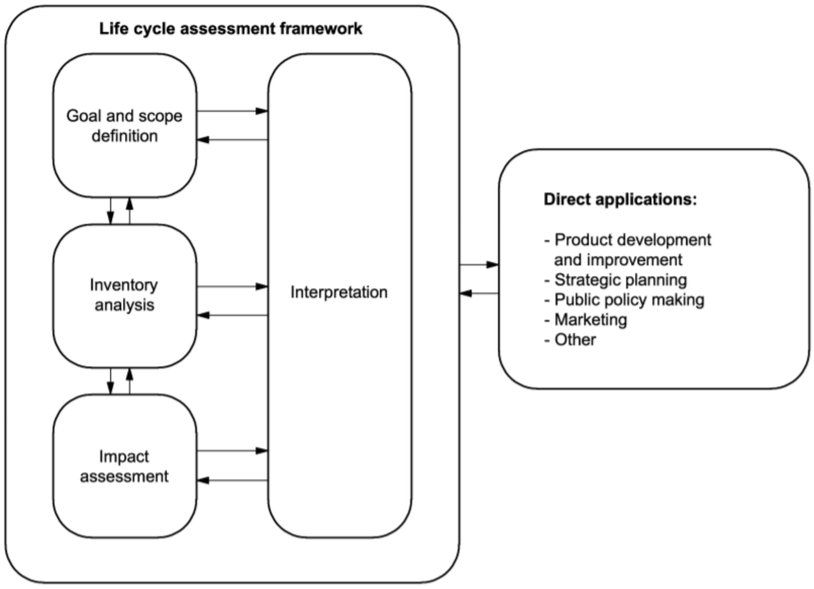


Figure 5 - Phases of an LCA taken from ISO 14040:2006 (ISO, 2006)

During the goal and scope definition phase, the entity decides the aspects of its operation to be assessed (scope/boundary). It then compiles all the processes that will be included in the scope. It also compiles the environmental indicators and calculation procedures to be used in the LCA study (ISO, 2006).

The inventory analysis involves data collection regarding the processes included in the scope. An example of such data could be amount of electricity consumed by the facility (ISO, 2006).

The impact assessment step involves the main calculation of the results which is subsequently analyzed in the interpretation phase (ISO, 2006)

2.2 Carbon footprint assessment

When the LCA assessment solely focuses on the global warming potential of a product or service, this is known as the carbon footprint assessment (ISO, 2006). The aim of the carbon footprint is to quantify the GHG emissions that are associated with the entity being analyzed.

Greenhouse gases comprise of methane (CH_4), carbon dioxide (CO_2), nitrous oxide (N_2O) and fluorinated gases, such as hydro fluorocarbons. Carbon dioxide of biogenic origin is not included in carbon footprint assessments as it is assumed to be part of the short-term natural cycle and regarded as carbon neutral (IPCC, 2014). Greenhouse gases have varying global warming potentials, represented in terms of carbon dioxide equivalents (CO_2eq). The potencies of the common GHG over a 100-year period, taken from the 5th IPCC assessment report are shown in Table 1 (IPCC, 2014).

Table 1 - GHG potencies in CO_2eq (IPCC, 2014)

Greenhouse gas	**CO_2eq**
CO_2	1
CH_4	28
N_2O	265

The Greenhouse Gas Protocol is a standard that provides guidelines for companies to assess their carbon footprint (Greenhouse gas protocol, 2011). This assessment is based on activity data of the organization. Activity data refers to data regarding the daily processes of the facility/organization. Examples of operational data could be quantity of fuel consumed by the facility, or on-line measurements of fugitive emissions on-site. This data is subsequently used in conjunction with emission factors derived from either literature or on-site measurements to calculate the GHG emissions (Greenhouse gas protocol, 2011).

2.2.1 Categorization of emissions

According to the GHG protocol, greenhouse gas emissions from an organization can be divided into 3 scopes (Greenhouse gas protocol, 2011).

Scope 1 emissions: These comprise of emissions that originate on-site. This is either fugitive gases from the site's processes or the combustion of fuels on-site.

Scope 2 emissions: These emissions originate from the generation of electricity used on-site

Scope 3 emissions: These are emissions that are out of the control of the facility. These include transport emissions, outsourced services, etc.

2.2.2 Boundary of carbon footprint assessment

The extent of carbon footprint assessments depends on the boundaries (or scope) defined. Typical boundaries used in footprint calculations are the "cradle to grave" and "cradle to gate". Cradle to grave involves all carbon dioxide equivalent emissions throughout the life of the product. This includes the extraction of raw materials, manufacturing of the product, operation, maintenance, transport and finally the disposal of the product (Bhatt *et al.* 2019). The "cradle to gate" approach encompasses all the impacts from the extraction of raw materials to the departure of the finished product from the facility (Laurent *et al.* 2016). The boundary of the carbon footprint assessment needs to be established before the carbon footprint calculation can begin. Depending on the scale of the company, organizational boundaries could also exist, as illustrated in the example shown in Figure 6.

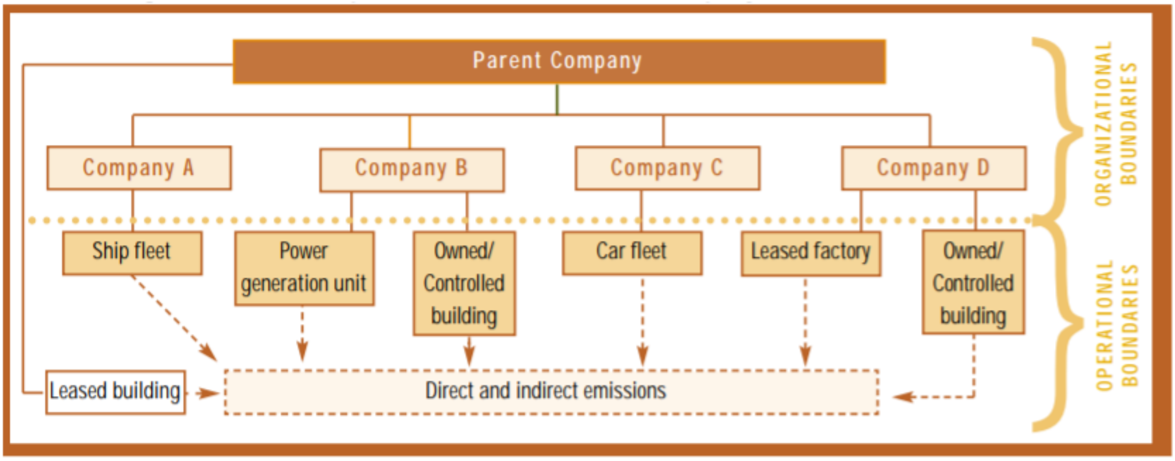


Figure 6 - Example of boundaries in a carbon footprint study (Greenhouse Gas Protocol, 2011)

2.2.3 Functional unit

Due to varying scales of facilities or organizations, benchmarking or comparing performance efficiencies between facilities would be difficult without a common denominator. This is when a functional unit needs to be defined. The ISO 14040:2006 standard defines a functional unit as "quantified performance of a product system for use as a reference point" (ISO, 2006). This could be in terms of the unit product produced or unit service provided.

For example, in carbon footprint assessment of WWTPs, most literature studies might express the emissions of the plant in terms of CO_2eq/ m^3 of wastewater treated (Xu, 2011; Maktabifard *et al*. 2019). If the CF is represented in CO_2eq/ m^3, the functional unit is one m^3 of wastewater treated and the results can be compared between various WWTPs provided they all share the same functional unit.

2.2.4 Carbon offsets

In order to be carbon neutral, a facility would need carbon offsets to balance out its GHG emissions. A carbon offset is defined as a unit of CO_2eq that is avoided or reduced in order to compensate for emissions created elsewhere (Goodward and Kelly, 2010). The carbon offsets are usually expressed with a negative value while the CO_2 equivalent emissions are reported as positive values (Greenhouse gas protocol, 2011).

2.3 Wastewater treatment process

In a centralized system, wastewater is collected in sewer networks that lead to the wastewater treatment plants. Additional wastewater is brought externally to the site from septic tanks of customers not connected to the sewer network. Once the wastewater reaches the facility, it undergoes preliminary treatment, primary treatment, biological nutrient removal (BNR) or secondary treatment, and finally tertiary treatment (Hopcroft, 2015). A simple illustration of a typical wastewater treatment process with anaerobic digestion of sludge can be seen in Figure 7.

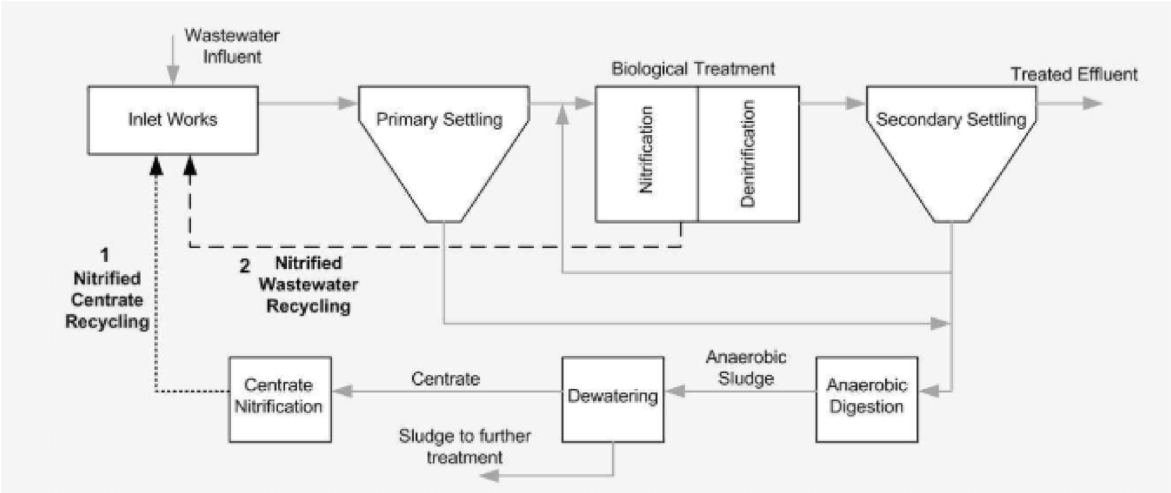


Figure 7 - Illustration of a typical wastewater treatment process with digestion of sludge (Estrada et al.*2015)*

The first stage of preliminary treatment is screening. This is done to filter out large debris in the wastewater that could damage equipment downstream of operations. This is followed by grit removal, which is done to remove gravel and other larger inorganic constituents of the wastewater (Hopcroft, 2015).

Primary treatment follows the preliminary treatment phase. This stage consists of primary sedimentation tanks which retain the wastewater for a specific period to allow heavier objects to settle (Hopcroft, 2015).

The effluent from the primary-treatment step is then led to the activated sludge process, where microbes break down the contaminants in the wastewater and biological nutrient removal takes place. Nitrogen removal in WWTP comprises of both nitrification and denitrification steps. Nitrification occurs in oxic environments (aerated zones) and denitrification occurs in anoxic environments (oxygen-deficient zones). The equations for nitrification and denitrification can be seen below:

Equation 1 - nitrification reaction (e^{-1} = electron)

$$NH_3 + O_2 = NO_2^- + 3H^+ + 2e^-$$

Equation 2 - Conversion of nitrite to nitrate (e^{-1} = electron)

$$NO_2^- + H_2O = NO_3^- + 2H^+ + 2e^-$$

Equation 3 - Denitrification reaction (e^{-1} = electron)

$$2NO_3^- + 10e^- + 12H^+ = N_2 + 6H_2O$$

Ammonia (NH_3) is first oxidized to nitrite (NO_2^-) by ammonia oxidizing bacteria (AOB). The nitrite is then oxidized to nitrate (NO_3^-) by nitrite oxidizing bacteria (NOBs). During denitrification, the NO_3^- is reduced to NO_2^- (nitrite) which is in turn reduced to NO (nitric oxide). This nitric oxide is then converted to N_2O before finally being converted to N_2, by heterotrophic bacteria (Hopcroft, 2015). Phosphorous is removed either by chemical precipitation or biologically through phosphorous-accumulating organisms (PAOs) with the former being the most common in Finland (Finnish Water Utilities Association, 2018). Depending on the WWTP, sidestream processes might be implemented. Sidestream processes consist of separate treatment units for the supernatant from biosolids treatment, due to its rich nutrient concentration and low organic content (Rodriguez-Garcia *et al.* 2014). This rich nutrient concentration increases the loading rate in the mainstream bioreactors and sidestream treatment leads to more stable conditions in the activated sludge process.

Depending on the WWTP, tertiary treatment might be a final step in the treatment process. In this stage, impurities such as suspended solids (SS) and phosphorous are further removed (Hopcroft, 2015). An example of tertiary treatment is the flotation process. During flotation, bubbles are injected, unto the which oils and grease attach and rise to the surface (Wang, 2010).

During the wastewater treatment process, excess sludge builds up in the clarifiers and bioreactors which is removed and treated either on-site or off-site before being disposed of. Treatment methods are usually anaerobic digestion or composting of the sludge (Hopcroft, 2015).

Other biological processes used in Finland for wastewater treatment are the Membrane Bioreactor (MBR) and Moving Bed Biofilm Reactor (MBBR). The MBR system is a combination of the ultrafiltration and Conventional Activated Sludge (CAS) systems. The MBR system filters out the solids and biomass associated with the activated sludge process and results in a clearer effluent (Judd, 2010). The MBBR system consists of carriers, which serve as surfaces for biomass to grow on. These carriers are suspended and move freely in the bioreactor, with the current caused by tank aeration. The benefits of this system include lower suspended solids in the reactor (Mannina *et al.* 2018).

2.4 Greenhouse gases in wastewater treatment

The main fugitive GHGs emitted in WWTPs are CH_4 and N_2O, which can be released along various points on the treatment plant if conditions are favorable (Kosse *et al.* 2016). CO_2 is also emitted during microbial degradation of organic matter in the aeration tanks during respiration (Kosse *et al.* 2016). Although CO_2 from wastewater treatment is not included in the scope of carbon footprints due to its biogenic origin, few studies suggest fossil fuel carbon can constitute up to 20% of the total carbon in wastewater (Griffith *et al.* 2009). Law *et al.* (2013) studied fossil carbon in wastewater with isotopic mass balances and concluded that between 4-14% of influent Total Organic Carbon (TOC) was of fossil origin with 88-98% being removed during treatment. The main production points for fugitive gases in WWTPs can be seen in Figure 8.

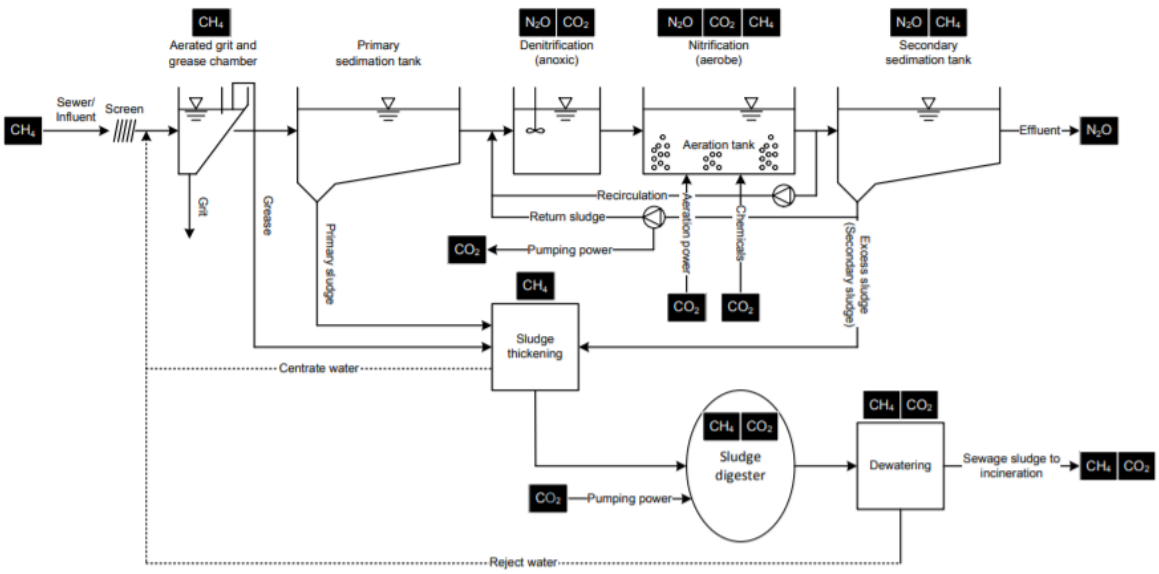


Figure 8 - Production sites of fugitive gas emissions in a conventional activated sludge plant (Kosse et al. *2016)*

Indirect emissions result from the use of resources during the treatment process. These resources could be in the form of energy (electricity and heat) or chemical use. Emissions from transportation of chemicals and sludge are also included in this category (Kosse *et al.* 2016).

The following chapters will outline the formation of fugitive gases and sources of indirect greenhouse gases in conventional wastewater treatment plants. It will also discuss findings from literature studies regarding N_2O and CH_4 emissions in WWTPs.

2.4.1 Nitrous oxide

Nitrous oxide is a potent greenhouse gas having 265 times more global warming potential than carbon dioxide (IPCC, 2014). WWTPs have been known to release nitrous oxide during operations, although the dynamics of its formation is yet to be fully understood. This chapter will give a brief description of N_2O formation pathways in WWTPs based on literature studies, followed by a summary of main findings in N_2O emission studies in WWTPs.

2.4.1.1 Formation of nitrous oxide in WWTPs

In this section, the various pathways of N_2O production in WWTPs will be described. Based on existing knowledge, there are three N_2O production pathways: hydroxylamine oxidation, nitrifier denitrification and heterotrophic denitrification pathways (Wunderlin *et al.* 2013, Ni and Yuan, 2015). The illustration in Figure 9, taken from Massara *et al.* (2017), sums up the formation of N_2O from the three different pathways.

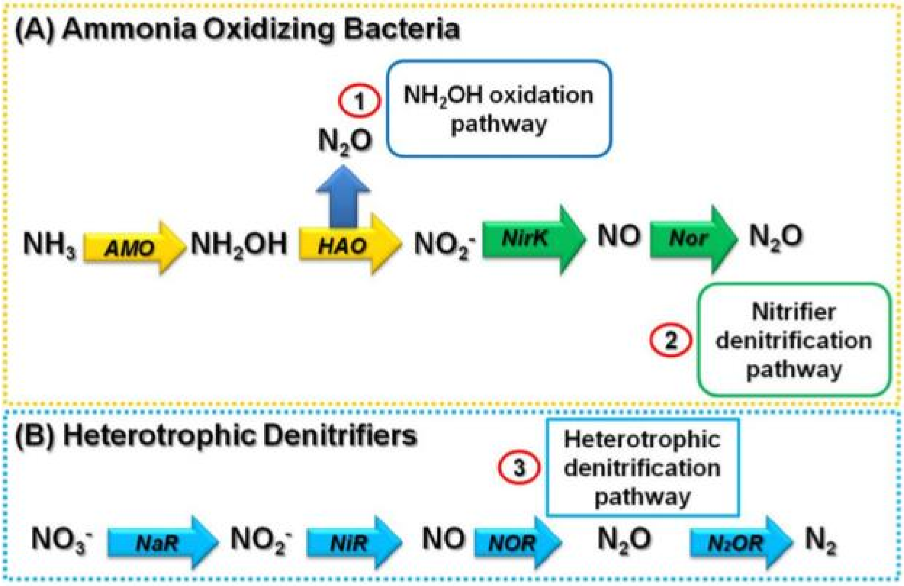


Figure 9 - Formation pathways of N_2O (Massara et al. *2017)*

N_2O emissions from hydroxylamine oxidation occur when AOBs oxidize ammonia (NH_3) to hydroxylamine (NH_2OH) with the ammonia monooxygenase (AMO) enzyme as a catalyst (Massara *et al.* 2017). NH_2OH is then oxidized to NO_2^- with hydroxylamine oxidoreductase (HAO) as the catalyst and oxygen as the electron acceptor. N_2O can be produced during this pathway if the oxidation of hydroxylamine (NH_2OH) to NO_2^- is incomplete (Massara *et al.* 2017). According to a review article by Vasilaki *et al* (2019), majority of N_2O emission studies in WWTPs suggest the hydroxylamine oxidation pathway to be of low contribution to WWTPs' overall N_2O emissions.

The second pathway for N_2O production occurs from the continuation of the AOB oxidation process described in the previous paragraph. In addition to the AMO and HAO enzymes, AOBs have nitrite reductase (NiR) and nitric oxide reductase (NOR) enzymes. Studies have shown the AOBs express the enzymes (NiR and NOR) during anaerobic periods to break down NO_2^- to NO, and subsequently, NO to form N_2O (Beaumont *et al.* 2004). NiR acts as a catalyst for reducing NO_2^- to NO and NOR acts a catalyst for reducing NO to N_2O. This is known as the nitrifier denitrification pathway (Hooper *et al.* 1997, Law *et al.* 2012a, Ni and Yuan, 2015).

The third pathway of N_2O emissions is the heterotrophic denitrification pathway (Massara *et al.* 2017). The first step is the conversion of nitrate (NO_3^-) to nitrite (NO_2^-), which is catalyzed by the nitrate reductase enzyme (NaR). NO_2^- is then converted to nitric oxide (NO), which is catalyzed by nitrite reductase. The nitric oxide is then converted to N_2O with the help of the nitric oxide reductase (NOR). The nitrous oxide is then converted to diatomic nitrogen catalyzed by the nitrous oxide reductase. However, if the heterotrophic denitrification is disrupted at the nitrous oxide production stage and does not reach completion, accumulation of N_2O occurs. Thus, the heterotrophic denitrification pathway serves as a mechanism for N_2O consumption if the process is not disturbed or stopped at the N_2O formation stage (Massara *et al.* 2017, von Schulthess and Gujer, 1996; Pan *et al.*, 2012, 2013a; Ni and Yuan, 2015). However, the two AOB pathways mentioned previously are considered the major production mechanisms since they can occur simultaneously (Massara *et al.* 2017).

2.4.1.2 N_2O from effluent in receiving water bodies

The IPCC stated that there was limited data available regarding the nitrification and denitrification dynamics in the receiving water bodies. The IPCC's default emission factor based on limited data for wastewater effluent in receiving bodies is 0.005 kg N_2O-N/ kg $TN_{effluent.}$ One assumption for the emission factor given was that the N_2O production is directly linked to nitrification and denitrification in rivers and estuaries and to the nitrogen load to the river (IPCC, 2006).

2.4.1.3 Main findings from N_2O emission studies in WWTPs

Previous carbon footprint studies have attributed majority (in some cases, over 70%) of the total GHG emissions to fugitive emissions, i.e. methane and nitrous oxide emissions (Daelman *et al*. 2013). Due to the high GWP of N_2O, WWTPs carbon footprint could heavily depend on N_2O emissions, especially in sites having low energy consumption emissions (Maktabifard *et al.* 2019). This sub-chapter sums up findings from different studies related to N_2O emissions in WWTPs followed by a short summary table of the studies done with their respective emission factors.

In conventional activated sludge plants, nitrification is expected to be the main process responsible for N_2O emissions (Shen *et al.* 2013, Sun *et al.* 2014 & Li *et al.* 2015). Incomplete nitrification leads to accumulation of nitrite (NO_2^-) which can be subsequently converted to N_2O through the nitrifier-denitrification pathway. For this reason, it is important that dissolved oxygen is sufficient for the completion of nitrification and thereby reducing the NO_2^- accumulation (Massara *et al.* 2017). Denitrification has also been suggested to be a N_2O sink, due to N_2O being an intermediate compound in the conversion of NO_3^- to N_2 (Massara *et al.* 2017). Studies further support the nitrifier-denitrification pathway as being dominant based on: i) N_2O peak formation when transitioning from anoxic to aerobic zones, ii) simultaneous increase in N_2O emissions when NO_2^- and NH_4^- concentrations increase under sub-optimal aerobic conditions and iii) N_2O peaks coinciding with increased NO_2^- concentrations in aerobic zones (Vasilaki *et al.*2019; Rodriguez-Caballero *et al.* 2014).

The existence of multiple, inter-linked process parameters in the wastewater treatment process proves difficult to establish a clear dependence of N_2O emissions on a specific process parameter (Kosonen *et al.* 2016). Currently, the consensus as to which factors influence the formation of nitrous oxide emissions is yet to be fully reached. However, strong evidence from literature suggests that increasing the nitrogen loading rate of the treatment plant close to, or beyond its capacity, will lead to an increase in the nitrous oxide emissions (Quan *et al.* 2012; Frison *et al.* 2015; Zheng *et al.* 2015). Also, the nitrous oxide emissions could occur in other non-nitrifying process units such as the secondary clarifiers, although it is assumed part of these emissions originate from the aerated units (Mikola *et al.* 2014). Finally, sub-optimal aeration, leading to incomplete nitrification, could favor the nitrifier-denitrification pathway due to the accumulation of nitrites (NO_2^-) (Vasilaki *et al.* 2019). Vasilaki *et al.* (2019) summarized the contributing factors to N_2O emissions found in literature studies which are, but not restricted to the following: i) low DO, ii) NO_2^- accumulation in both aerated and denitrifying zones, iii) limited COD:N ratio, and iv) sudden changes in processes or shocks to the systems.

In addition to emissions being process-dependent, the variability in methodologies used for sampling and measurement could also play a role in the accuracy of these results (Kosonen *et al.* 2016). For example, some studies measured N_2O emissions from grab-samples from specific points in the reactor whereas other studies covered the whole reactor with hoods to capture the N_2O gases which were then connected to an online measurement system. The study done on the Viikinmäki WWTP measured N_2O emissions from exhaust gas of the whole plant and also used sensors submerged in the aeration basin (Kosonen *et al.* 2016). The variability in measurement methodologies makes comparison of different results difficult and thus, a standardized method for measuring and reporting N_2O emissions is needed (Massara *et al.* 2017; Delre *et al.* 2017b).

An extensive review on N_2O measurement studies by Vasilaki *et al* (2019), showed that more than 80% of existing articles represent the EF in terms of percentage nitrogen load to the WWTP. Depending on the N_2O production pathway, EFs might not be fully representative. For example, an EF represented in terms of "% nitrogen denitrified emitted as N_2O" would give low emission values for a WWTP having low denitrification rates even though N_2O emissions could occur through the nitrifier-denitrification pathway during nitrification. If indeed the nitrifier-denitrification pathway is more dominant, as suggested in literature, the EF in terms of NH_4^+ removed would be more suitable. This will account for both the nitrifier-denitrifier and heterotrophic denitrification pathway since all the pathways originate from ammonia reactions, as seen in Figure 9 (Bellandi *et al.* 2018). It is therefore important to understand the N_2O generation pathways and its triggering conditions in order to make better EF representations (Vasilaki et. al., 2019).

N_2O emission factors for WWTPs differ significantly in literature (Vasilaki *et al.* 2019). In their 2019 refinement, the IPCC updated their default emission factor to 1.6% of total nitrogen (TN) in influent wastewater emitted as N_2O-N (IPCC, 2019). IPCC's selection of a new EF was based on nitrogen loading rates and respective N_2O emissions from 30 scientific literature studies. The respective loading rates and emissions were plotted and the trendline equation resulted in 1.6% of the total nitrogen load as shown in Figure 10.

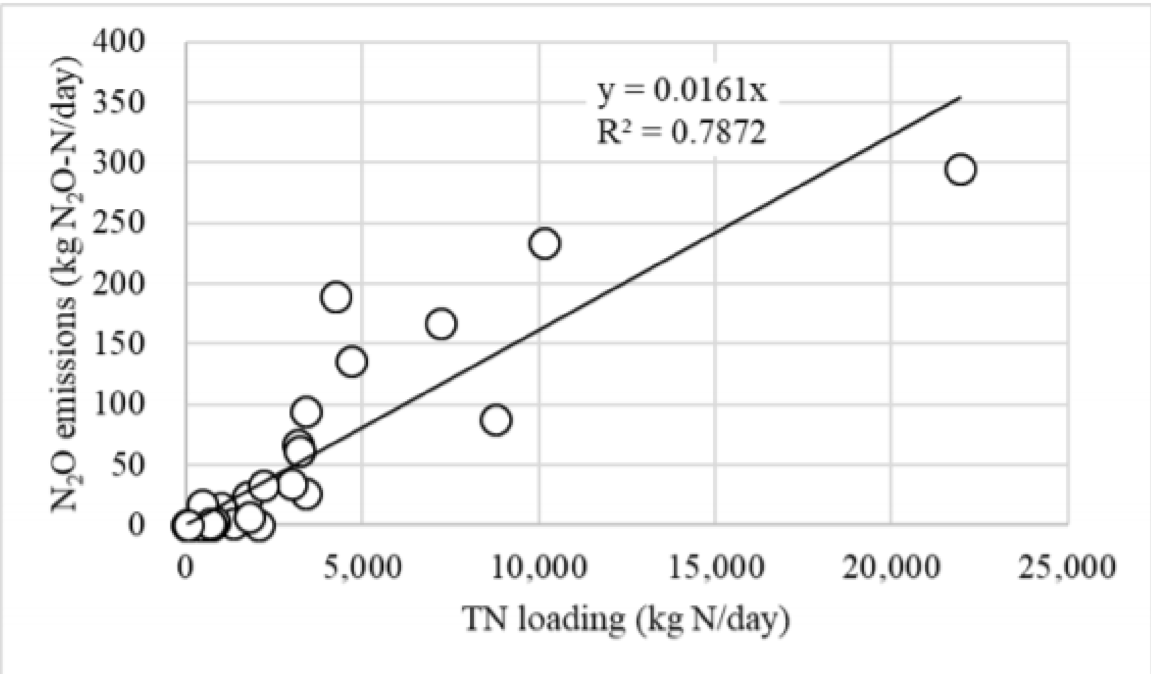


Figure 10 - IPCC's resulting N_2O EF in their 2019 refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC,2019)

A review of existing, long-term N_2O measurement studies (>70 days measurement periods) resulted in an average EF of 1.7% $TN_{influent}$ while short-term N_2O measurement studies have an average EF of 0.8% $TN_{influent}$ (Vasilaki *et al.* 2019). This suggests that short-term measurement studies tend to underestimate N_2O emissions while long-term measurements might more accurately account for seasonal changes in process conditions (e.g. changes in wastewater temperature, flow rates, influent concentrations etc.).

The average of EFs obtained from literature regarding WWTPs having sidestream processes was found to be 2,1% of TN in influent wastewater emitted as N_2O-N (Vasilaki *et al.* 2019). In a WWTP in Austria, the sidestream DEMON® process (which utilizes anaerobic AOBs to reduce ammonium concentrations in reject waters) accounted for ~90% of the total N_2O emissions on-site (Schaubroeck *et al*., 2015). The reason for the higher EF for side-stream processes is due to the high ammonia oxidation rate and build-up of NO_2^- which favors the nitrifier-denitrification pathway. However, there are existing studies for WWTPs with Anammox side-stream processes that resulted in EFs of <1% influent TN (Joss *et al*., 2009; Weissenbacher *et al*., 2010). In conclusion, NO_2^- accumulation has been identified as one of the key scenarios for N_2O formation along with low DO, limited organic substrate, abrupt system shocks and low COD:N ratios (Duan *et al.* 2017; Guo *et al.* 2017; Massara *et al.* 2017).

A list of N_2O emission studies found in literature along with their country of study, emission factors, and treatment configuration can be seen in Table 2.

Table 2 - WWTP N_2O emission studies found in literature with their respective emission factors (N/A = not available)

Reference	**Country of study**	**Configuration**	**EF-N_2O**	**Source studied**
Yoshida *et al*. (2014)	Denmark	Bio Denitro	0.1% - 2.72% N_2O-N/ TN influent	Whole plant
Thomsen (2016)	Denmark	N/A	0.318% N_2O-N / TN influent	BNR reactors
Czepiel *et al*. (1995)	U.S.A	Activated sludge process	0.014%-0.055% N_2O-N/ TN influent	Wastewater line
Aboobakar *et al*. (2013a)	U.K.	Anoxic.oxic	0.04% N_2O-N/ TN influent	BNR reactors
Daelman *et al*. (2013)	Netherlands	Anoxic-oxic	2.8% N_2O-N/ TN influent	Whole plant
Oshita *et al*. (2014)	Japan	N/A	0.013% - 0.07% N_2O-N/ TN influent	Sludge treatment units
Masuda *et al*. (2015)	Japan	Anoxic-oxic	0.37% - 1.24% N_2O-N/ TN influent	BNR reactors
Ahn *et al*. (2010)	U.S.A	Activated sludge process	0.006% - 1.546% N_2O-N/ TN influent	BNR reactor

Sommer *et al*. (1995)	Germany	Activated sludge plant	0.02% N_2O-N/ TN influent	BNR reactor
Rodriguez-Caballero *et al*. (2014)	Spain	Anoxic-Oxic	6.8% N_2O-N/ $NH^{+}_{4\text{-}N}$ load	BNR reactor
Brotto *et al*. (2015)	Brazil	Activated sludge process	0.013% - 0.197% N_2O-N/ TN influent	BNR reactor
Kimochi *et al* (1998)	Japan	Anoxic-oxic	0.01% - 0.08% N_2O-N/ TN influent	BNR reactor
Kosonen *et al* (2016)	Finland	Anoxic-Oxic	1.9% N_2O-N/ TN influent	Whole plant

2.4.2 Methane

Methane is a greenhouse gas with a global warming potential 28 times higher than carbon dioxide (IPCC, 2014). Although methane is a threat to the climate, it can also serve as a resource in the form of biogas derived from anaerobic digestion in WWTPs (Maktabifard *et al.* 2019). This chapter will describe the formation of methane in WWTPs and the various sources on-site, followed by a table summarizing measurement studies from WWTPs and their respective emission factors.

2.4.2.1 Formation and sources of methane in WWTPs

Methane is formed in WWTPs both deliberately (through anaerobic digestion), and unintentionally when favorable conditions in the treatment process facilitate its production (IPCC,2014). Methane is produced by the breakdown of organics by microbial organisms in the absence of oxygen. Fermentative bacteria degrade large organic material into fatty acids and alcohols. These fatty acids and alcohols are then further broken down by another set of microbes into methane and carbon dioxide (Smith *et al.* 2015). Methane formation in WWTPs are usually slow due to the conditions not always being favorable. According to the IPCC background report "CH_4 And N_2O Emissions from Wastewater Handling", very little methane is released in the short-term processing and storage of sludge with larger emissions occurring in the ultimate, long-term disposal of raw sewage sludge (Penman, 2000).

According to IPCC, methane could possibly form in sewer networks if the sewer is poorly graded and mostly runs at a full capacity or even flooded. This results in lack of air space in the sewer resulting in anaerobic zones. In temperate climates however, this formation pathway is considered insignificant, due to the lower temperature conditions in the sewers (IPCC,2014). Additionally, a study by Daelman *et al* (2012) suggested that 80% of the dissolved methane in wastewater influent was oxidized in the activated sludge reactor indicating that the activated sludge process acts as a sink for dissolved methane entering the WWTPs (Daelman *et al.* 2012).

According to the IPCC, leaks from digesters constitute a major source of methane emissions in a WWTP (Penman, 2000). Daelman *et al*. (2012) studied the methane emissions from a WWTP in Netherlands. It was discovered that 72% of the fugitive methane emissions were attributed to the digestion process and sludge processing units, namely: the sludge thickener, centrifuge, storage tanks for dewatered sludge and the methane emissions from gas engines (Daelman *et al*. 2012). Schaum *et al*. (2015) performed a detailed COD balance for the anaerobic digestion process based on experimental data and concluded that the main sources of methane regarding the digestion process are: i) methane slips in Combined Heat and Power (CHP) generators/ biogas combustion units, ii) residual biogas emissions from digestate storage/thickening process and iii) dissolved methane in digestate.

Methane can also be emitted during composting of sludge (IPCC,2014). A study by Maulini-Duran *et al*. (2013) showed higher methane emissions during the composting of digested sludge compared to raw sludge. It was suggested that the dissolved methane in the digestate may be stripped off into the atmosphere during composting. Hence, it is expected that WWTPs which compost anaerobically digested sludge would have higher methane emissions than composting raw sludge (Maulini-Duran *et al*. 2013).

According to IPCC, methane formation during sludge storage is expected to be slow due to fermentative reactions resulting in a pH decrease (IPCC, 2014). In such acidic environments, methane forming bacteria cannot thrive and methane production would be reduced (IPCC, 2014). Additionally, IPCC stated that at temperatures below 15°C, significant methane production is not likely to occur due to the inhibition of methanogens (IPCC, 2014). This is of interest for this study since Finland is located in the temperate climate zone. By this logic, WWTPs in Finland that store sludge outdoors should experience lower yearly average methane emissions than other WWTPs in warmer geographical areas. A study done in Sweden by Willén *et al*. (2016) measured the methane emissions from silos storing dewatered, digested sludge in a WWTP. The results of the study showed that methane emission fluxes were low and even close to zero during winter but began increasing after spring. Since Sweden's climate could be considered close to that of Finland, it can be assumed that the yearly average methane emissions from outdoor sludge storage is expected to also be low in Finland.

Based on the set of conditions required for methane formation, it is assumed that majority of the methane emissions from wastewater treatment originate from the sludge treatment and handling processes.

A list of some previous studies done, and their corresponding emission factors can be found in Table 3.

Table 3 - List of CH_4 measurement studies in WWTPs with their respective emission factors (N/A= Not applicable)

Reference	Country of study	Configuration	EF-CH_4	Source studied
Gustavsson & Tumlin (2013)	Sweden, Norway, Finland, Denmark	Activated sludge	0.25% CH_4/ COD influent	Whole plant
Delre *et al.* 2017b	Sweden & Denmark	Bio Denipho Bio Denitro SBR & deammonification Activated sludge	1.1% - 21.3% CH_4/ CH_4 produced 0.2% to 3.2% CH_4/ COD influent	Whole plant

Yoshida *et al*. (2014)	Denmark	Bio Denitro	0.61% – 4.35% CH_4/ COD influent 2.1% – 4.4 % CH_4/ CH_4 produced	Whole plant
Petersson (2012)	Sweden	N/A	0.15% - 2.6% CH_4/kg CH_4 produced	Sludge treatment units
			0.3% - 5.25% CH_4/kg CH_4 produced	Whole biogas plant
Thomsen (2016)	Denmark	N/A	1.3% CH_4/kg CH_4 produced	Biogas plant
Flesch *et al*. (2011)	Canada	N/A	0.8% - 3.2% CH_4/kg CH_4 produced	Biogas plant
Czepiel *et al*. (1993)	U.S.A	Activated sludge process	0.29% CH_4 / COD influent	BNR Reactors
Aboobakar *et al*. (2013b)	U.K	Anoxic.oxic	0.04%-0.1% CH_4/ COD influent	BNR reactors
Daelman *et al*. (2012)	Netherlands	Anoxic-oxic	1.13% CH_4/ COD influent	Whole plant
Wang *et al*. (2011)	China	A-A-O	0.06%-0.1% CH_4/ COD influent	Whole plant
Oshita *et al*. (2014)	Japan	N/A	0.16% - 0.6% CH_4/ COD influent	Sludge treatment units
Masuda *et al*. (2015)	Japan	Anoxic-oxic	0.15% - 0.69% CH_4/ COD influent	BNR reactors

2.4.3 Indirect emissions

Wastewater treatment plants consume resources during their operations. These products in turn have a specific manufacturing carbon footprint. Thus, indirectly, wastewater treatment emits carbon dioxide equivalents of the products and resources they consume (Campos *et al.* 2016). This chapter will briefly discuss the various sources of indirect emissions in a conventional wastewater treatment plant.

2.4.3.1 Energy consumption

WWTPs consume energy in the form of electricity and heat. Unless a WWTP is self-sufficient, it will have to depend on the grid or other external energy source for electricity, heat, or both. The emissions associated with the use of electricity from the grid will depend on the fuel source used by the producer to generate the electricity/heat energy. National average emission factors for electricity/heat generation are typically used in lieu of specific supplier data in carbon footprint studies (Delre *et al.* 2017a; Gustavsson & Tumlin, 2013).

The energy mix in Finland has experienced an overall decrease in fossil fuel shares since 2003, while simultaneously reducing the carbon dioxide emissions associated with energy generation, as shown in Figure 11 (Official Statistics of Finland, 2019). As of 2018, the share of fossil fuels in Finland's energy mix was ~35% (Official Statistics of Finland, 2019). Due to the current trend of fossil fuel reduction in the energy mix, it is expected that the overall share of the CF for WWTPs regarding energy consumption will gradually reduce in the future.

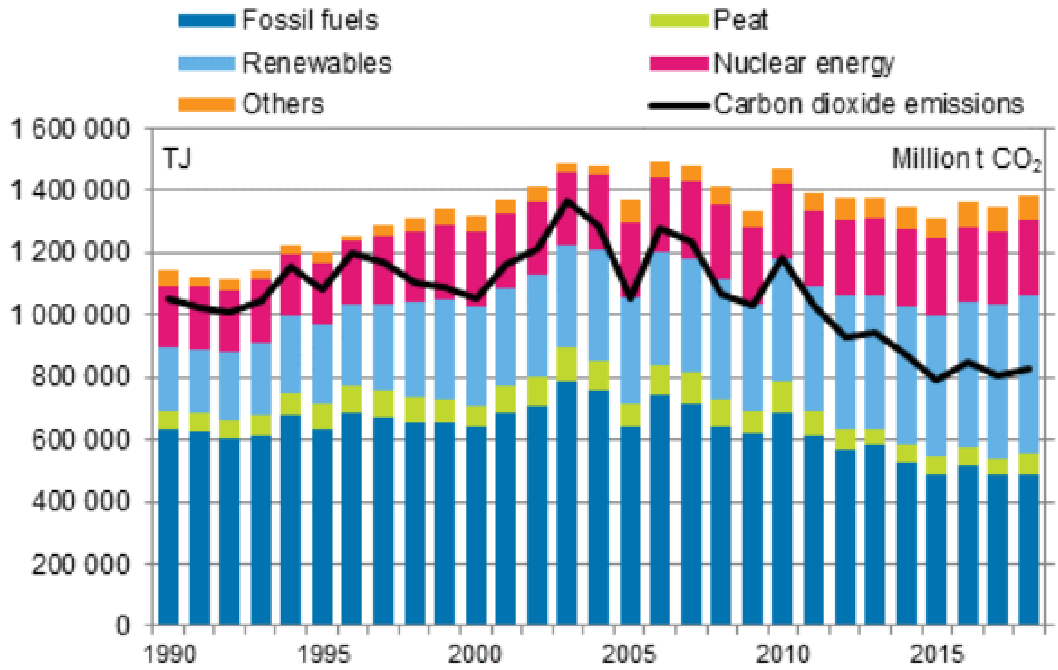


Figure 11 - Shares of fuel sources in Finland's primary energy source mix since 1990 (Official Statistics of Finland, 2019)

A study by Delre *et al.* (2017a) illustrated the differences in carbon footprint shares related to energy consumption for plants situated in Denmark and Sweden. The WWTPs situated in Denmark had between 16% and 29% of the total footprint attributed to energy consumption while those in Sweden had between 3% and 4% of the total footprint related to energy consumption. This was due to the different energy mix between the countries. Sweden had a higher share of lower emitting fuels (mostly hydro and nuclear) in their energy mix than Denmark and therefore had lower emissions per unit electricity consumed. From a carbon footprint perspective, Gustavsson & Tumlin, (2013) recommended that WWTPs consuming a high renewable energy mix should sell more energy produced on-site to the grid. This could be in the form of biogas, electricity or heat to the grid. This could reduce the overall carbon footprint of the site.

In terms of WWTP scale, literature suggests that larger treatment plants benefit from a lower specific electricity consumption than smaller treatment plants as shown in Figure 12 (Tukiainen, 2009). Electricity consumption is also linked to the degree of effluent quality set out in the site's environmental permit. For example, a site having stricter nitrogen removal requirements will tend to consume more electricity than a site will lower requirements. This is due to more aeration needed for the nitrification process, more pumping for external carbon sources, as well as increased tank volume which require more energy (Tukiainen, 2009). Moreover, studies show that approximately 50% of the WWTP's electricity consumption is used for the activated sludge process suggesting how energy intensive aeration is in a WWTP process (Gurung *et al.* 2018).

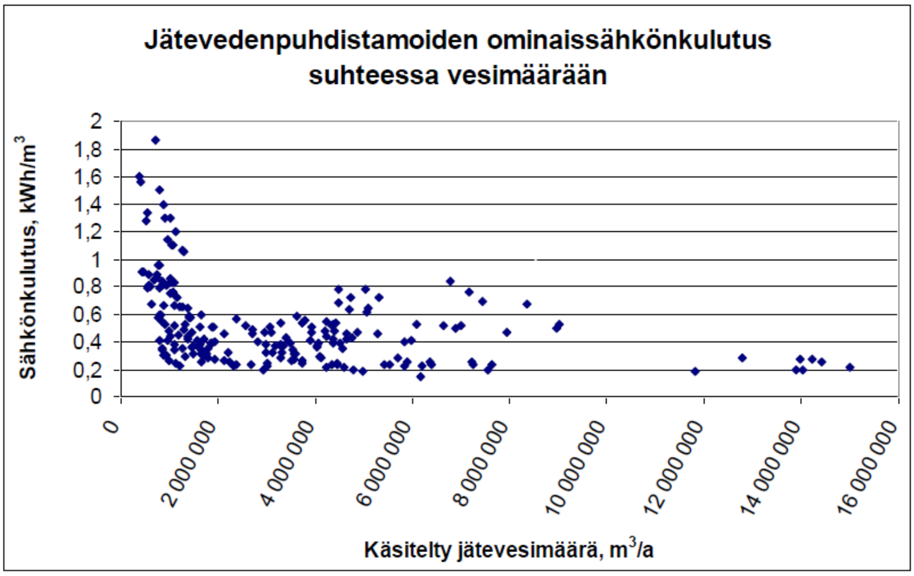


Figure 12 - Energy consumption per m³ of WWTP treated (vertical axis) and cubic metres of wastewater treated per year (horizontal axis) of WWTPs in Finland (Tukiainen, 2009)

2.4.3.2 Chemical usage

The main chemicals used in a wastewater treatment process are coagulants, flocculants/polymers, external carbon sources such as methanol, and alkaline chemicals for pH adjustment (Hopcroft, 2015). These chemicals require energy input and have direct emissions during their manufacturing. Therefore, they constitute indirect emissions when consumed in wastewater treatment processes. From the emission factors derived from literature and chemical manufacturers, the most carbon intensive chemicals used in WWTPs are polymers which average ~4 kg CO_2eq/ kg polymer produced while coagulants averaged ~ 0.3 kg CO_2eq/ kg (Kettunen, 2020).

2.4.3.3 Transport

Operations at the WWTP require supplies and produce waste activated sludge, digestate, sand and screening waste that need to be delivered and disposed of. This requires some form of transportation. The indirect emissions from transport are typically presented as gCO_2eq/ton km (VTT Technical Research Centre of Finland Ltd., 2017). The factors affecting the indirect transport emissions are therefore the mass of the substance being transported and the distance being transported. Different modes of transport have different carbon emission factors. Previous carbon footprint studies on WWTPs indicate that transport emissions constitute a minor share of the total carbon footprint of the WWTP (Maktabifard, *et al*. 2019).

In northern Finland, transport distances are relatively longer as compared to the southern areas, due to areas being more sparsely populated (Lauronen, 2017). It is therefore expected that WWTPs in more sparsely populated areas to have slightly higher transport emissions.

2.4.3.4 Construction

Depending on the boundary defined for the carbon footprint study, the indirect emissions from the construction of the WWTP can be included in the carbon footprint calculation. A study by Chai *et al.* (2015), included the emissions from the construction phase when calculating the carbon footprint of a WWTP in China, assuming a 20-year life span of the plant. The results showed that construction of a WWTP accounted for 4% - 6% of the total carbon footprint of the treatment site over the lifespan of the plant. Genzowsky *et al*. (2011) also concluded that the construction phase of two WWTPs accounted between 10 -20% of the total GHG emissions over an assumed lifespan of 30 years. Gustavsson & Tumlin (2013) who analyzed 16 WWTPs in Scandinavia also concluded that the carbon footprint of the operational phase was far more significant for the carbon footprint than the construction phase (Gustavsson & Tumlin, 2013). Although the choice of building material would impact the overall footprint, it is regarded as much lower than the operational emissions of the treatment plant in the long run.

2.5 Carbon footprint studies of WWTPs

This chapter covers previous WWTP carbon footprint studies done in Nordic countries as well as other geographic areas of the world. At the end of the review, main studies cited in literature along with their findings are provided in Table 4.

2.5.1 Studies done in the Nordic region

In the Nordic region, few studies exist that cover the whole wastewater treatment plant. One of the earliest full-scale studies was done by Gustavsson & Tumlin (2013). The study was done on 16 WWTPs situated in Norway, Sweden, Denmark and Finland. The assessment's boundaries comprised of the wastewater treatment process, the sludge treatment process and emissions from the effluent in the receiving waterbodies. Indirect emissions included transport, energy consumption and chemicals. Calculations were based on emission factors from literature, although few sites had short-term measurement data of fugitive emissions that were included in the calculation. The sites studied had a carbon footprint in the range of 7 – 108 kg CO_2eq PE^{-1} $year^{-1}$ with an average of 46 kg CO_2eq PE^{-1} $year^{-1}$. Results indicated that direct GHG emissions and energy consumption emissions were the most important contributors to the carbon footprint of the sites studied. The highest emitter among the 16 sites attributed most of its emissions to nitrous oxide. Chemical and transport related emissions did not significantly affect the final carbon footprint result.

Another study done in the Nordic region was by Delre *et al*. (2017) who quantified the carbon footprint of seven WWTPs (four Danish and three Swedish). The boundary for the study included the wastewater and sludge treatment processes, (along with chemical and energy consumption), transport emissions and emissions from the receiving body of the effluent. It excluded the collection of wastewaters to the plant and the construction and demolition phase. Quantification of direct emissions was done using data of on-site GHG measurements while other emissions were model based. In line with the results of Gustavsson & Tumlin (2013), between 44% and 71% of the total carbon footprint of the plants was attributed to direct GHG emissions with N_2O having a higher share than CH_4. The remaining 29% – 56% was attributed to energy consumption and chemical use with energy consumption emissions being dependent on the energy mix of the country (Delre *et al.,*2017). At the end of the study, the authors gave a list of suggestions regarding carbon footprint assessment in WWTPs. One suggestion was that system boundary cut-offs should only be done if the carbon emissions of respective processes being cut-off constitute less than 1% of the total environmental burden (Delre *et al.,*2017). This will prevent the cut-off of processes that could account for a significant share of the total carbon footprint of the WWTP.

The third study discussed is the life-cycle analysis performed on the Viikinmäki WWTP in Finland (Mölsä, 2019). This study mainly focused on the broader environmental burdens with numerous environmental indicators. There was a section of the LCA that included the carbon footprint of the plant. The calculation was done with an MS-excel tool developed by the Swedish Water Development for the project done by Gustavsson & Tumlin (2013) (VA-teknin Södra). It was then compared with the results from another tool used, known as GaBi (GaBi solutions). A few emission factors in the excel tool were modified to better suit the Viikinmäki plant. These modifications were related to the soil additives for composting process which were not initially available in the excel tool. Also, ferrous sulphate was considered to have zero impact in the tool due to it being a by-product. The transport emissions were modified to include return trips of empty trucks, a ferry transport emission factor was introduced, and a train transport emission factor was added. The boundary used for the carbon footprint study was the gate to gate boundary which includes all processes involved in the treatment of wastewater and sludge, chemical use, transport and energy consumption. Infrastructure and equipment were excluded as it was assumed to have little effect on the results based on a previous research done by Lundin *et al*. (2000). The functional unit used was the wastewater inflow to Viikinmäki in the year 2018. Results of the CF was 41 kg CO_2eq PE^{-1} $year^{-1}$ for the wastewater treatment process and 9 kg CO_2eq PE^{-1} $year^{-1}$ for the sludge treatment. The total carbon footprint result from the Swedish excel file was 14% higher than the result obtained from the GaBi 6 model. As with previous carbon footprint studies, nitrous oxide constituted majority of the total footprint share with 64% followed by methane with 17%, when using the Swedish tool. Moreover, the Life Cycle Impact Assessment (LCIA) results indicated that the most influencing factors in the result were linked with nitrogen removal (Mölsä, 2019).

2.5.2 Studies in other geographic areas

Chai *et al.* (2015) studied the carbon footprint of three wastewater treatment configurations in China along with different sludge treatment scenarios. The treatment technologies for wastewater studied were A-A-O (Anaerobic-Anoxic-Oxic), SBR and the oxygen ditch configurations. The sludge treatment options were anaerobic digestion and aerobic fermentation. This study's carbon footprint calculation was solely based on emission factors with no measurement of fugitive emissions on-site. Results showed that the SBR treatment option combined with anaerobic digestion of sludge had the lowest carbon footprint. Combined with the anaerobic digestion of sludge, the oxygen ditch wastewater treatment method had the highest carbon footprint scenario. The authors attributed this to the amount of sludge produced for digestion. Among the 3 treatment technologies studied, SBR produces the most sludge, followed by A-A-O and finally, the oxidation ditch method produced the least sludge due to its prolonged aeration and Sludge Retention Time (SRT) (Chai *et al.* 2015). However, studies have shown that the oxygen ditch treatment method is at "low risk" for nitrous oxide emissions due to its relatively more stable process conditions and uniform dissolved oxygen concentrations (Yu *et al.* 2010). For each wastewater treatment technology scenario, the disposal of sludge to landfill as a sludge treatment method had the highest carbon footprint due to the amount of CH_4 emitted. Each treatment technology combined with anaerobic digestion on-site had the lowest carbon footprints due to emission offsets from biogas produced (Chai *et al.* 2015). It should be noted that this study was based on process design data of WWTPs treating 20 000 m^3 of wastewater and not actual case studies. Since process conditions might fluctuate occasionally in the WWTP, the GHG and hence the overall carbon footprint would be dynamic.

Rodriguez-Caballero *et al*. (2015) studied the formation of N_2O emissions and combined it with the electricity consumption of the plant to determine the carbon footprint of the plant with an SBR configuration. Different aeration cycles were implemented during the measurement period. Results showed that approximately 61% of the overall carbon footprint of the plant was attributed to N_2O emissions with electricity consumption accounting for the remaining 39%. The plant's total nitrogen removal was high (89% TN removal). Some studies associate a high TN removal with low N_2O emissions (Parravicini *et al.* 2016). Interestingly, the calculated N_2O emission factor of the plant, based on online measurement, was 6.8% of the influent NH_4^+-N. This is much higher than reported EFs from previous studies. Results showed that intermittent aeration with short oxic and anoxic phases mitigated N_2O emissions in the reactor and subsequently led to a considerably lower carbon footprint of the plant. It should be noted that this study excluded emissions from sludge treatment, as well as indirect emissions from transport and other resources (e.g. chemicals) used on-site.

Xu (2011), studied the carbon footprint of two treatment plants in Spain. The first plant had a treatment capacity of 1 000 000 PE while the second had a PE of 70 000 PE. The first plant had a Pre-Anoxic-Anaerobic-Anoxic-Oxic configuration with tertiary (disinfection) and pre-treatment of wastewater. Anaerobic digestion was used as sludge treatment with cogeneration of heat and electricity from biogas. The second plant had the same treatment configuration excluding the pre-anoxic tank. Also, the sludge treatment option for the second plant was anaerobic digestion with the gas being torched instead of generating heat or electricity. The first and second plants had nitrogen removal capacities of 85% and 75% respectively. The carbon footprint was assessed using the GaBi 6 software (GaBi solutions). The nitrous oxide emissions were calculated based on the emission factor of 3.2 g N_2O/person/year proposed by Czepiel *et al* (1995). The dissolved N_2O measurements were carried out on-site but were not used in the calculation. This could be due to inconsistencies and occasional power failures affecting measurements, as stated by the author. Results of the first plant showed that approximately 62% of the total CF was attributed to electricity consumption while the second highest share was associated with direct emissions from the activated sludge process (17.2%). The second plant's results attributed 77% of the CF to electricity consumption, with the second highest source being attributed to the emissions in the effluent receiving body (15%). The high share of emissions from electricity consumption could be associated with the high emission factor of electricity generation in Spain. Emissions from the secondary treatment (mainly N_2O) were surprisingly low compared to other studies (Delre *et al.*2017; Mölsä, 2019). However, recent studies have discredited the EFs proposed by Czepiel *et al* (1995), which was used for this study, due to its outdated methodology and lack of representativeness (Foley et. al., 2015).

Parravicini *et al*. (2016), assessed the carbon footprint of two model WWTPs with PEs of 50 000 each. One plant was modelled with anaerobic digestion for sludge treatment while the other incorporated aerobic sludge stabilization. Emissions from biosolids disposal, sewer system and construction phase were excluded from the scope. For N_2O emissions, the authors used a regression model based on data derived from a measurement campaign of eight municipal WWTPs in Austria. In the regression model, the emission factor was dependent on the degree of TN removal (higher TN removal, lower emission factor). For fugitive gas emission factors used, refer to table 4. The carbon footprint of the plant with anaerobic digestion was 36 kg CO_2eq PE^{-1} $year^{-1}$ with N_2O emissions accounting for 43% of total emissions. For the WWTP with aerobic sludge stabilization, the total footprint was 24 kg CO_2eq/PE/a with 60% of emissions attributed to electricity consumption. The CH_4 emissions from the long-term storage of the dewatered sludge contributed 19% of the total emissions. The author stated that plants with higher TN removal efficiencies would have lower emissions in the effluent receiving body while those with lower TN removal efficiency, higher emissions in the receiving body. Thus, plants with low TN removal should consider including the receiving body in the scope of their footprint calculations as excluding this can lead to underestimated results. Aside the significant impact of direct emissions, the authors concluded that optimized plant design for higher TN removal, combined with controlled sludge treatment, are prerequisites for a low carbon footprint (Parravicini *et al*. 2016).

Maktabifard *et al* (2019) evaluated the carbon footprint of two municipal WWTPs in northern Poland, both of which were biological nutrient removal plants. The first plant, located in Slupsk, had a PE of 250 000 while the second, located in Starogard, had a PE of 60 000. This is the second study found in literature that utilized the carbon footprint excel tool developed by Gustavsson & Tumlin (2013). The Slupsk WWTP co-digested the sludge and other organics to produce biogas which was utilized on-site for heat and electricity. The digestate was then composted and distributed to farmlands. The WWTP in Starogard relied 100% on energy from the grid and chose psychrophilic digestion as a sludge treatment method. The stabilized sludge was then distributed to farmlands. Results showed that the Slupsk WWTP had 62% of its footprint associated with fugitive gas emissions with 23% attributed to energy consumption. The energy from biogas used on-site was already factored in since the site did not have to buy extra energy from the grid. Additionally, the spread of the compost, derived from digestate, on the farmlands resulted in emission savings of 8%. The Starogard WWTP on the other hand, attributed 71% of its total footprint to energy consumption since it relied 100% in the energy from the grid. This high contribution stems from Poland's energy mix being heavily dependent on coal and hence, has a higher footprint for electricity generation. The psychrophilic digestion of sludge and the subsequent distribution on farmlands led to emission savings of 7%. In conclusion, WWTPs operating in countries heavily relying on fossil fuel for electricity generation will gain more benefits in CF reduction by utilizing biogas produced on-site as was also suggested by Delre *et al*. (2017). Also, due to high uncertainty in N_2O emission factors, the authors recommended plant-specific, full-scale measurements as the best method for quantification. The author also illustrated the increasing dependency of CF on direct emissions with increasing energy neutrality in WWTPs. In the study, a sensitivity analysis was carried out to determine which sites were more sensitive to changes in N_2O EFs. According to the study, the sites which had energy recovery (use/sale of biogas) had their CF more dependent on the direct emissions resulting in an increase in uncertainty when EFs from literature were used. Selling & utilization of biogas on-site reduces dependency on energy from the grid and overall reduction in the CF. Therefore, the indirect emissions regarding energy consumption are reduced and hence, more dependency on direct emissions (Maktabifard *et al.* 2019)

Table 4 - Previous studies on plant-scale carbon footprint assessment in WWTPs (N/A = not available)

Reference	Country of study	treatment system	FU	EF-CH_4	EF-N_2O	Boundary	Results
Gustavsson & Tumlin (2013)	Sweden, Norway, Finland, Denmark	N/A	Swedish PE 70 g BOD/person/day	0.25% CH_4/ COD influent	1.57% N denitrified	Direct emissions, Transport, chemical use, emissions from receiving waterbodies, energy consumption and production, sludge treatment and application on land	7 – 108 kg CO_2eq PE^{-1} $year^{-1}$, average value was 46 kg CO_2eq PE^{-1} $year^{-1}$ Majority of emissions attributed to N_2O from wastewater treatment
Delre *et al.* 2017	Sweden & Denmark	Bio Denipho Bio Denitro SBR & deammonification Activated sludge	kg C removed kg N removed kg P removed	1.1% - 21.3% kg CH_4/kg CH_4 produced	0.1% - 5.2% N_2O-N/kg TN influent	Direct emissions, Chemical use, Transportation, Energy consumption/production, emissions from receiving waterbodies, sludge treatment & application on land	0.9 – 2.2 kg CO_2eq (kg C removed)$^{-1}$, 6.5 – 12.6 kg CO_2eq (kg TN removed)$^{-1}$, 31.3 – 94.8 kg CO_2eq Direct GHG accounted for between 44% - 71%.
Mölsä, (2019)	Finland	Activated Sludge process	total inflow for the year (m^3)	0.25% CH_4/ COD influent	1.57% N denitrified	Wastewater & sludge treatment, chemical use, production and use of energy, transportation, recipient water body emissions	41 kg CO_2eq PE^{-1} $year^{-1}$ for wastewater treatment, 9.7 kg CO_2eq PE^{-1} $year^{-1}$ for composting process Nitrous oxide constituted majority of the total footprint share with 64% followed by methane with 17%
Xu, (2011)	Spain	Anaerobic-Anoxic-Oxic Pre-anoxic-Anaerobic-Anoxic-Oxic	Total inflow for the year (m^3)	not stated, GaBi model used	3.2 g N_2O/person-year 4.6% total nitrogen load as N_2O	Wastewater treatment, sludge treatment, energy consumption, sludge transportation, recipient water body emissions	Electricity consumption constituted between 60% and 77% of total footprint.

Chai *et al.* (2015)	China	Anaerobic-Anoxic-Oxic	N/A	13.4 g CH_4/ kg sludge	0.035 kg N_2O-N / kg N denitrified	Construction, Electricity consumption, chemical use, transport, sludge treatment, wastewater treatment,	5817 – 9928 tons CO_2eq / year SBR combined with anaerobic digestion of sludge had the lowest carbon footprint. Oxidation ditch combined with anaerobic digestion had the highest carbon footprint.
Parravicini *et al*. (2016)	Modelled plant	Anoxic- Oxic	PE	8.7 g CH_4/ kg $COD_{influent}$	0.75% N_2O-N / $TN_{influent}$ for a site having 77% TN removal	All processes except disposal of biosolids, emissions from sewer networks, and construction phase of WWTPs	24- 36 kg CO_2eq PE^{-1} $year^{-1}$ Plant with anaerobic digestion had a higher carbon footprint than a plant with aerobic sludge stabilization.
Maktabifard *et al*. (2019)	Poland	Anaerobic-Anoxic-Oxic SBR	m^3 of wastewater treated	0.25% CH_4/ COD influent	1.57% N denitrified	N/A	24- 100 kg CO_2eq PE^{-1} $year^{-1}$ Slupsk WWTP had 62% of its footprint associated with fugitive gas emissions with 23% attributed to energy consumption. Starogard WWTP attributed 71% of its CF to energy consumption.

3 Methods

The WWTPs in this study were selected based on their differences in treatment processes, and differences in the scale of PEs treated. This was done to diversify the case study plants and investigate how their differences affected the carbon footprint results.

The Methods section begins with a general guideline to assist WWTPs in assessing their carbon footprint. Each case study WWTP's process layout is then described and finally, the methodology used for data collection and calculating their carbon footprints will be given.

3.1 General guidelines for CF assessment in WWTPs

This chapter presents a general set of guidelines to assist WWTPs in assessing their carbon footprint. The schematic shown in Figure 13 summarizes the list of guidelines presented.

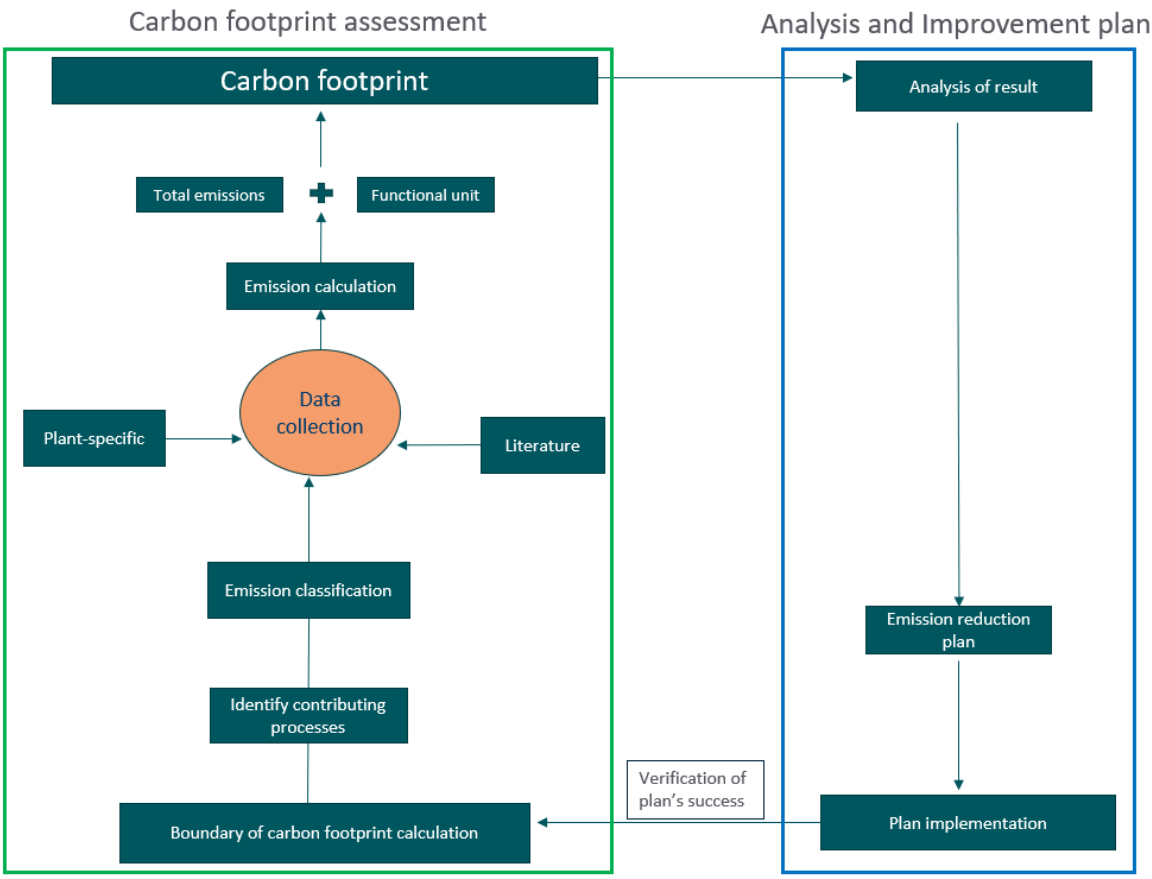


Figure 13 - Schematic for WWTP carbon footprint assessment

Step 1 - Boundaries: The WWTP must first decide what aspects of the plant's operations and outsourced processes should be included in the CF assessment. In general, the treatment of wastewater and excess sludge is always included in the boundary for most literature studies. Processes that are occasionally left out of the scope are the collection of wastewater and treatment of screenings due to the minor shares they contribute, according to literature.

Step 2 - Contributing processes: All processes that fall within the defined boundary should be identified. The main processes to consider are the electricity and heat consumption, chemical use, transportation emissions, and fugitive emissions from wastewater and sludge treatment processes.

Step 3 - Emission classification: Once the contributing processes have been identified, the site could classify the emissions sources either as direct or indirect emissions. Indirect emissions originate from resource consumption (e.g. chemical, purchased electricity, etc.) and transportation emissions. Direct emissions are the fugitive emissions from the site's treatment processes. Classification of emissions enables easier identification of key CO_2 contributors and the degree of control the WWTP has over these emissions.

Step 4 - Frequency of data collection: Before commencing the data collection, the WWTP should decide the frequency of data collected (monthly, quarterly or yearly values etc.). If the WWTP wants to analyse their CF seasonal variation, monthly or quarterly values could be used. If seasonal variation in CF is not of interest, then the site can rely on yearly values.

Step 5 - Operational data collection: The operational data for the processes identified in Step 3 are then collected. Operational data are the quantifiable aspects of the WWTP's operation. For example, the operational data for electricity and chemical consumption are the amounts of chemicals and electricity consumed. Depending on the WWTP, energy, chemical consumption and sludge production data are usually readily available. If data is not readily available, estimations based on site's discretion or literature values would need to be performed.

Step 6 - Fugitive emission data collection: Regarding fugitive emissions (N_2O and CH_4), the site has two options. It can either perform measurement campaigns on-site or it can rely on estimated/modelled results. It is recommended for sites to perform measurement campaigns of at least one year, in order to capture the seasonal changes in emissions. Measurement campaigns might provide more site-specific emission values with lower uncertainty than estimated or modelled results. Information regarding N_2O measurement techniques in WWTPs can be found in the study by Marques *et al.*, (2016) titled "Assessment of online monitoring strategies for measuring N2O emissions from full-scale wastewater treatment systems".

Step 7 - Gathering emission factors: EFs are used in conjunction with the site's data to calculate its total CO_2 equivalent emissions. EFs are predominantly used for indirect emissions, although default EFs for direct emissions have been used in literature studies as well. EFs for energy and chemical consumption can be obtained from the site's energy and chemical suppliers. Transportation EFs can be obtained from VTT Research Institute of Finland Ltd (lipasto.vtt.fi).

Step 8 - Calculation phase: Once all the plant-specific data and emission factors are collected, the emissions can then be calculated. If the EF is represented in terms of N_2O or CH_4 emitted, it is important to multiply the result with the respective GWPs of N_2O and CH_4. These GWPs can be obtained from the IPCC 2014 report (IPCC, 2014).

Step 9 - Functional unit (FU): The total emissions calculated in Step 8 could be represented in terms of the intended comparison purpose. If the site's goal is to analyse the nutrient removal process, the FU could be a kg of nitrogen/phosphorous removed or other elements of choice. If the site wants to relate the emissions to the population served, the functional unit could be population equivalent (PE). It should be noted that the results with different FUs lack comparability. For example, results given in CO_2eq/ PE cannot be compared with a result given in CO_2eq / m^3. Thus, if the site aims to compare results from different CF assessment campaigns, it should ensure that the FUs are the same. The use of multiple FUs are recommended for easier analysis and benchmarking of results.

Step 10 - Analysis of results: The key contributors of the sites CO_2 emissions should be identified along with their shares of the total emissions. Once the WWTP identifies these contributors, they can begin strategizing ways to reduce their emissions.

Step 11 - Improvement plan: The degree of control the site has over reducing emissions will depend on the key contributing process. For example, if electricity consumption has a major contribution to the CF, the site can either change to a more renewable energy supplier or reduce its electricity consumption. Control of other aspects, such as fugitive emissions, might require process optimization but may not be fully controlled if the formation of these gases are not fully understood (e.g. N_2O). The control of the contributing factors should therefore be assessed accordingly.

3.2 Site descriptions

3.2.1 Taskila WWTP, Oulu

The Taskila WWTP, located in Oulu, is the largest among the case studies treating municipal wastewater for ~ 160 000 PEs. Primary treatment involves screenings, grit removal, flocculation and pre-sedimentation. The plant's biological treatment process consists of both MBR and CAS reactors. The MBR process came into effect from Autumn 2018. Approximately 35% of the wastewater is diverted to the MBR line with the remaining being treated in the old CAS line. The effluent from the CAS treatment line undergoes post-sedimentation, biological filtration and finally chemical disinfection (DesinFix™ by Kemira Oy).

The chemicals used in the wastewater and sludge treatment during the study period include the following:

- Polyaluminium chloride
- Polymers
- Ferric sulphate ($Fe_2(SO_4)_3$)
- Sodium carbonate (Na_2CO_3)
- Calcium hydroxide ($Ca(OH)_2$)
- Methanol (CH_3OH)
- Sulphuric acid (H_2SO_4)
- Hydrogen peroxide (H_2O_2)
- Sodium hydroxide (NaOH)
- Peracetic acid ($C_2H_4O_3$)
- Formic acid (CH_2O_2)
- Sodium hypochlorite (NaClO)
- Citric acid ($C_6H_8O_7$)

The excess sludge generated undergoes a process called KemiCond (developed by Kemira). In this process, sludge is mixed with sulphuric acid and hydrogen peroxide in a reactor before being forwarded for mechanical dewatering. Benefits associated with the process include reductions in sludge amount, polymer consumption and odours. During the studied period, the treated sludge had three possible management options. These were:

- part of the sludge was spread on fields without any prior composting
- part of the sludge was composted
- part of the sludge was given to Gasum Oy biogas plant in Oulu
-

Due to lack of information of the treatment process that occurs at Gasum, it was assumed that the sludge was anaerobically digested to produce biogas intended for vehicular use. This is due to Gasum being a natural gas and vehicular biogas distributer (Gasum, 2020).

The schematic of the Taskila WWTP can be seen in Figure 14.

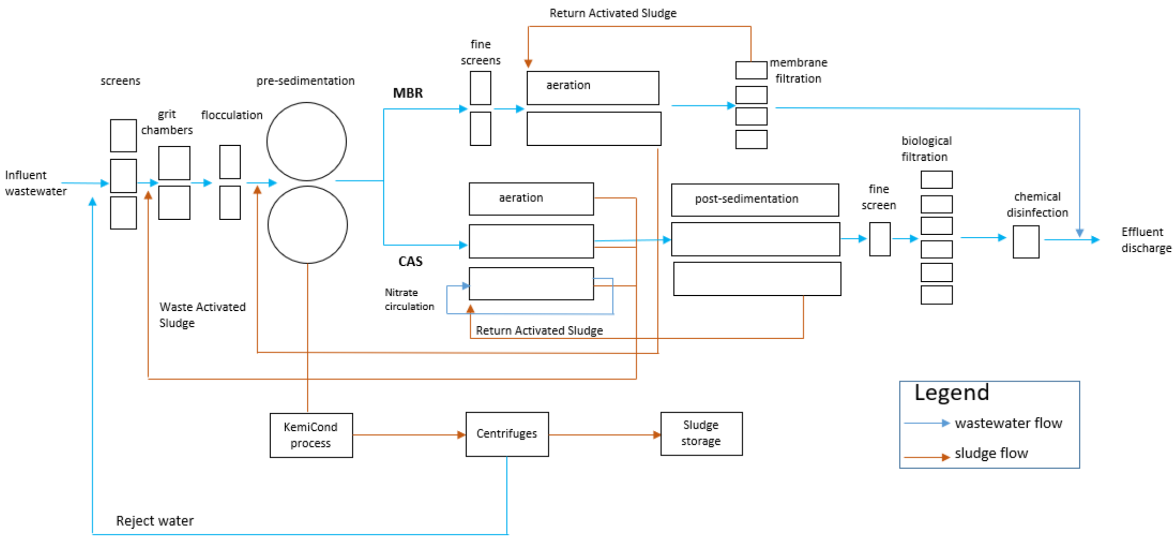


Figure 14 - Schematic of Taskila WWTP

3.2.2 Riihimäki WWTP

The Riihimäki WWTP treats municipal wastewater for ~75 000 PEs, as well as dairy industrial wastewater. It is an activated sludge plant with anaerobic digestion as a sludge treatment process. The wastewater treatment processes include screening, sand removal, primary sedimentation, anoxic-oxic bioreactors and secondary clarifiers. Sand filtration is the tertiary treatment process of the plant. The dairy industrial wastewater received at the plant undergoes aeration as a pre-treatment before being introduced into the anoxic-oxic bioreactor.

The chemicals used by the plant include:

- Ferrous sulphate ($FeSO_4$)
- Polymers

The excess sludge produced is gravity thickened, digested and centrifuged before being stored for a few days. It is then transported to Humuspehtoori Oy (located 90 km away) for further composting.

The biogas generated is burnt on-site in a gas boiler to produce heat. Since the generated heat from biogas is insufficient for the whole site, natural gas is purchased and combusted as supplementary fuel for heating. In cases of excess biogas generated, the gas is flared with a torch.

The layout of the site can be seen in the Figure 15.

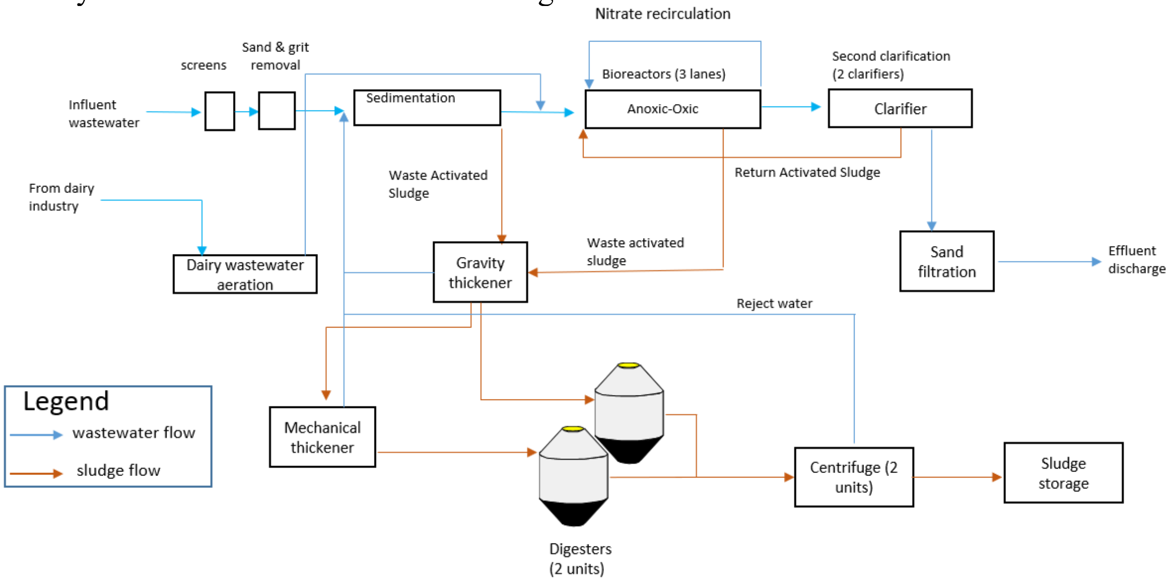


Figure 15 - Schematic of Riihimäki WWTP.

3.2.3 Paroinen WWTP, Hämeenlinna

The Paroinen WWTP, located in Hämeenlinna, treats municipal wastewater for ~ 90 000 PEs. The plant's preliminary treatment steps include screening, sand and grit removal. There is no primary treatment and the wastewater is led to the first activated sludge bioreactor (anoxic-oxic). It is then clarified before being led to the second bioreactor whose zones can be modified to either oxic or anoxic. There is nitrate circulation back to the first activated sludge process for denitrification in the anoxic zone. Flotation process comes after the second activated sludge process before the effluent is discharged from the site.

The chemicals used by the site are:

- Ferrous sulphate ($FeSO_4$)
- Methanol (CH_3OH)
- Calcium carbonate ($CaCO_3$)
- Polyaluminium chloride (PAC)
- Polymers

75% of the excess sludge produced is gravity-thickened before being anaerobically digested on-site. The remaining 25% of the sludge is digested off-site by ENVOR Oy (third party). The digestate is then centrifuged and composted on-site. The site's environmental permit sets a limit to how much sludge can be composted on-site and hence any excess sludge must be sent to a third party for further treatment.

The supernatant from sludge dewatering undergoes a separate aeration process before being led to the first bioreactor. This is done to reduce the ammonium load to the bioreactors.

All the biogas generated on-site, from anaerobic digestion of sludge, is combusted with a gas boiler to produce heat which supplements district heating purchased. During periods of excess heat produced, the site sells its surplus heat to the grid.

A simplified layout of the processes on-site can be seen in Figure 16.

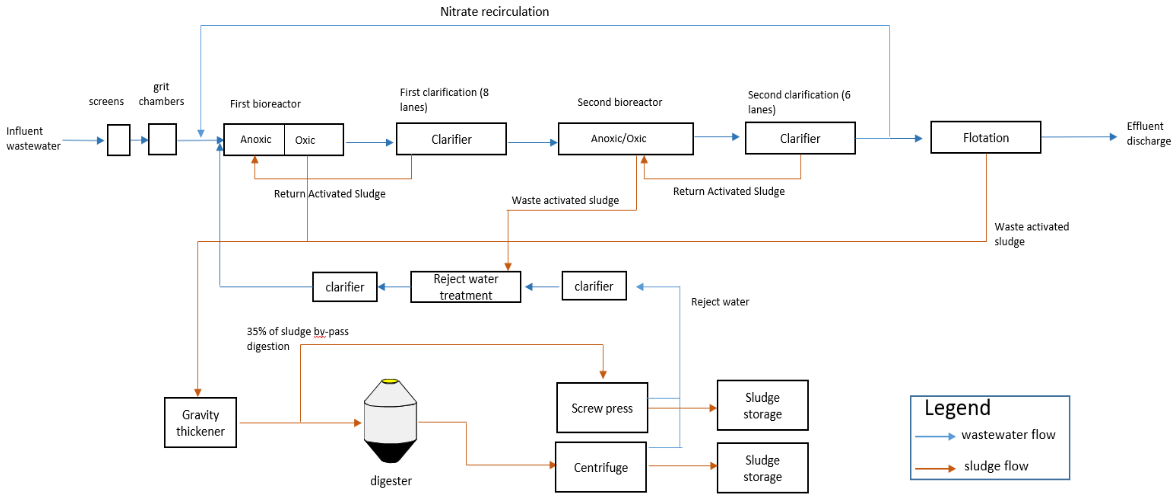


Figure 16 - Schematic of Paroinen WWTP processes

3.2.4 Mäkikylä WWTP, Kouvola

The Mäkikylä WWTP, located in Kouvola, treats the wastewater for ~100 000 PEs. Its pre-treatment steps consist of screening, sand and grit removal and primary sedimentation. The biological treatment stage consists of four treatment lines with modifiable anoxic-oxic zones, which include nitrate recirculation. The effluent from the biological treatment is then clarified before leaving the site.

Chemicals used by the site include:
- Ferric sulphate ($Fe_2(SO_4)_3$)
- Calcium carbonate ($CaCO_3$)
- Lime (CaO)
- Polyaluminum chloride
- Polymers

Between 2017 and June 2019 the excess sludge was centrifuged and co-digested on-site with municipal and industrial biowaste. In June 2019, the site sold its biogas production plant and began transporting the sludge to ENVOR Oy for digestion.

The schematic of the plant can be seen in Figure 17.

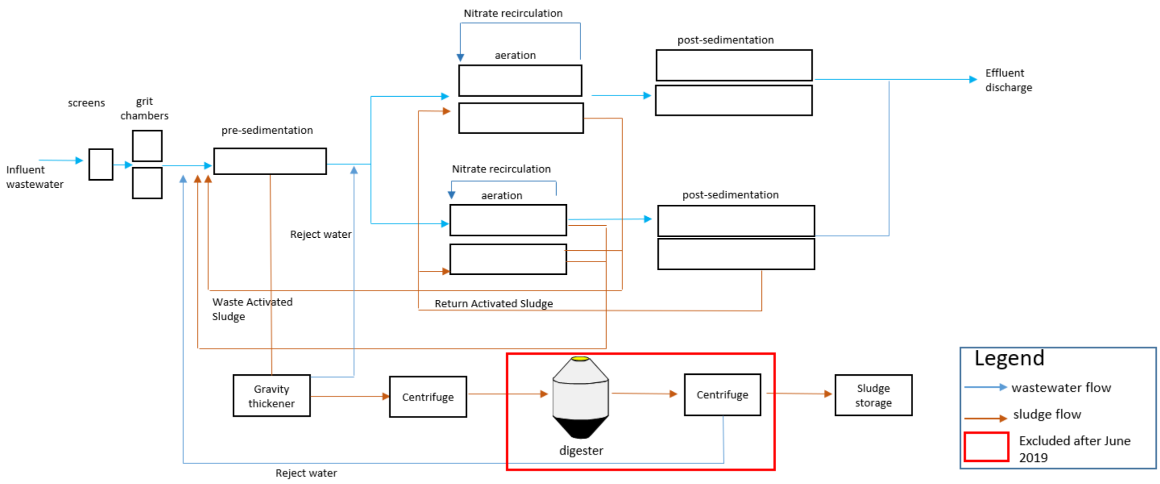


Figure 17 - Schematic of the Mäkikylä wastewater treatment process

3.3 Process parameters of WWTPs

The treatment parameters of the WWTPs can be seen in Table 5. The environmental permit of the sites in this study require nitrogen removal when the temperature of the influent is above 12°C and it requires sites to have a year-round average nitrogen removal of 70% (or 60% for Paroinen during the study period). Person Equivalent (PE) in this study refers to the inhabitants served according to the Government Decree on Urban Wastewater Treatment 888/2006.

Table 5 - Average wastewater treatment parameters of case study plants between 2017 and 2019

Parameter	Taskila	Paroinen	Riihimäki	Mäkikylä
daily treated water (m^3/day)	48 000	22 000	13 500	26 000
PE served**	163 521	90 000	75 238	100 219
Incoming COD_{Cr} (kg. day^{-1})	32 000	15 000	10 000	16 000
Incoming BOD_{7ATU} (kg. day^{-1})	11 500	6 300	5 300	6 900
Incoming Total N (kg. day^{-1})	3000	1300	900	1300
Incoming Total P (kg. day^{-1})	454	190	110	160
Average COD removal %	89 %	93 %	96 %	93 %
Average Total Nitrogen removal (%)	45 %	67 %	75 %	67 %
Average Total Phosphorous removal (%)	94 %	97 %	97 %	95 %
Permit requirements for nitrogen removal	≥70 %*	≥60 %*	≥70 %*	≥70 %*
Permit requirements for phosphorous removal	≥90 %	≥95 %	≥95 %	≥97 %

*= yearly average removal ** = PE calculated with assumption of 70 g BOD_7 PE^{-1} day^{-1}

3.4 Boundary of study

The carbon footprint analysis was done for the years 2017, 2018 and 2019. The boundaries chosen for this study can be seen in Figure 18. All related processes in the red box were included and processes outside the red box were excluded.

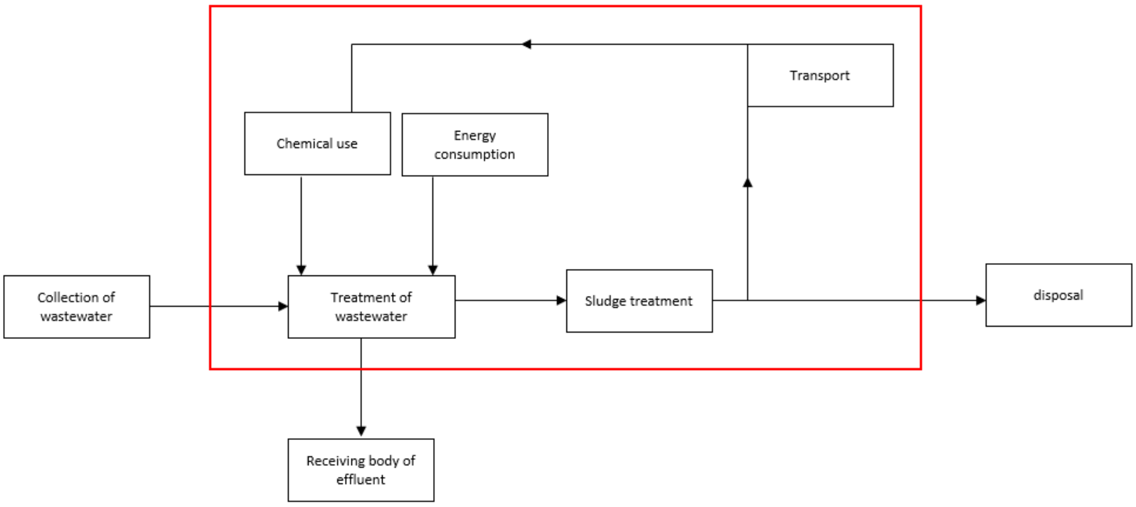


Figure 18 - Boundaries chosen for the carbon footprint calculation shown in the red box

The CO_2 emissions were separated into direct and indirect emissions with a third category for emission offsets. The list of processes included in the red box (shown in Figure 18) can be seen in Table 6.

Table 6 – Categorization of emission sources and in this study

Direct emissions	**Indirect emissions**	**Emission offsets**
N_2O emissions from wastewater treatment	Emissions from chemical manufacturing	Emission savings from the sale of electricity, heat and biogas produced on-site
CH_4 and N_2O emissions from composting of waste sludge	Emissions from transport of chemicals, sludge and screenings	
CH_4 emissions from anaerobic digestion of waste sludge	Emissions from generation of consumed electricity and heating	
CH_4 emissions from storage of waste sludge		
Emissions from biogas and natural gas combustion on-site		
CO_2eq emissions from screenings treatment		

The construction of WWTPs and sludge treatment facilities were excluded due to its low share of the total carbon footprint based on the review of previous studies (refer to Chapter 2.4).

The emissions from the receiving body were excluded based on limited data regarding the topic (refer to Chapter 2.4.1.2).

The wastewater collection phase was decided to be excluded from the scope. Based on reports from IPCC discussed in Chapter 2.4.2.1, methane formation in sewers of temperate climates would be insignificant due to colder temperatures in sewers. Since Finland is partly in the temperate zone, it was regarded as insignificant. The disposal of sludge was decided to be excluded since it was deemed to be outside the responsibilities of the WWTPs.

3.5 Data collection

Data collection forms in spreadsheet format were given to the case study plants to fill for each month of the years: 2017 -2019. The data collection form was separated into four different criteria, which were:

- Energy
- Chemicals
- Transport
- Sludge and screenings

The WWTPs in this study did not have any measurement system or data for fugitive gases during the period studied.

The information enquired for these categories can be seen in Table 7.

Table 7 - Activity data requested from the WWTPs in the data collection form

	Parameter	Unit
	Electricity consumption	kWh
Energy	District heating	kWh
	Natural gas sold/consumed	Nm^3 or kWh
	Amount of electricity sold from WWTP	kWh
	Amount of district heating sold from WWTP	kWh
	Electricity and District heat supplier	N/A
Chemicals	Each chemical used in the wastewater treatment process	tons
	Distance between chemical suppliers and WWTPs	km
Transport	Distance between outsourced sludge treatment and WWTP	km
	Distances between co-composting organics supplier and WWTP	km
	load capacity of delivery trucks	tons
	Amount of waste sludge produced	tons
Sludge	Amount of waste sludge anaerobically digested	tons
	Amount of waste sludge composted	tons
	Co-composting organics used for composting	tons
	Sludge storage time	days
	Dry matter of waste sludge produced	%
	Volatile solids of waste sludge produced	%

N/A = not applicable

Additional information regarding emission factors for chemicals were taken from chemical manufacturers and literature studies. Electricity and district heat EFs were taken from the energy suppliers of the WWTPs and literature (for sites using natural gas for heating). Due to lack of literature EFs regarding the use of peracetic acid, the EF for hydrogen peroxide consumption was used for peracetic acid. Since hydrogen peroxide is used in the manufacturing of peracetic acid, it is expected that the EF for peracetic acid would be higher than that of hydrogen peroxide.

Regarding the fate of biogas produced from outsourced digestion, data was only available for Paroinen WWTP but not for Mäkikylä WWTP. Since both plants are clients for the same outsourced company, it was assumed that the fate of the biogas produced was the same for both sites. According to ENVOR Oy, 50% of the biogas is used to power the ENVOR Oy facility, 35% of the biogas is given to industries for heat, and the remaining 15% is refined to biomethane to be used in transport and traffic (Laine, 2020).

3.6 Functional Unit

The main functional unit used in this study was the Population equivalent (PE). The PE of the sites in this study were calculated in accordance with the Government Decree on Urban Wastewater Treatment 888/2006. The decree gives an average value of 70 g BOD_7/person/day. Since the average daily BOD_7 loading to the sites in this study were known, the PE could be calculated. The average PEs of the sites over the study period can be found in Table 5.

In addition to the PE being the main FU, additional FUs were included for performance comparison among the plants. These FUs were 1 kg nitrogen removed, 1 kg of phosphorous removed and 1 m^3 treated wastewater.

3.7 Emission calculation phase

The emission calculation was based on plant activity data combined with chosen EFs from literature, and energy/chemical suppliers. The list of EFs used were divided into two categories, which were:

- common EFs (refer to Appendix 1)
- Plant-specific EFs (refer to Appendix 2 and Appendix 3)

Common EFs refer to emission factors that are constant for all sites. Examples are transportation EFs, chemical manufacturing EFs and fugitive emission factors for CH_4 and N_2O.

Plant-specific EFs refer to EFs that are unique and variable between WWTPs. These include the EFs for electricity and heat consumption. These vary between sites depending on the energy supplier and source of fuels used in their electricity and heat generation process.

The calculated fugitive emissions of CH_4 and N_2O were then converted to CO_2 equivalents based on the conversion factors from IPCC shown in Table 1.

This chapter will first talk about the calculation of indirect emissions from energy, chemical use, and transportation emissions, before discussing the methodology for direct CH_4 and N_2O emissions.

3.7.1 Energy consumption & sales

The emissions from electricity and district heat consumption for the case study plants were calculated based on emission factors gathered from each of the site's suppliers. For the Riihimäki and Mäkikylä WWTPs, heating was derived from natural gas and an EF for natural gas combustion in boilers was used for these plants.

The energy savings from selling biogas, heat or electricity were calculated based on how much CO_2eq emissions were saved from substituting the conventional fuel used. For example, the sale of biogas intended to be used as vehicular fuel will save all the emissions associated with the production and use of natural gas, of equal energy value, in vehicles. For EFs used regarding electricity and heating, refer to Appendices 2 and 3.

3.7.2 Chemical use

The emissions from chemical manufacturing was obtained partly from the suppliers of the plants and partly from literature values. A compiled list of all chemicals used by the case study plants along with their emission factors from manufacturing can be seen in Appendix 1. In this study, the use of ferrous sulphate was assumed to have zero emissions. This was based on communication with Kemira who stated that ferrous sulphate was produced from the company's ferrous sulphate landfill waste (Kettunen, 2020). It was also stated that little energy is consumed in the process of converting ferrous sulphate waste to a chemical product. Based on this information, ferrous sulphate was assumed to have zero emissions.

3.7.3 Transport

The transport emissions were calculated based on the mass of substance being transported, the distance transported, and the type of vehicle being used. The main mode of transportation was a loading truck with varying capacity. The emission factors for transportation were taken from VTT Research Institute of Finland's "Lipasto" database (VTT Technical Research Centre of Finland Ltd, 2017). In addition to inland transportation, transportation by ships were included due to some chemicals being manufactured outside Finland and it was assumed that the chemicals were shipped. Train transportation EFs were also included due to the use of methanol which was assumed to be imported from Russia. In this study, 2-way travel was included i.e. empty return trips to supplier. Transport EFs can be found in Appendix 1.

3.7.4 CH_4 emission calculation

Methane emissions in this study were assumed to be exclusively emitted from the sludge treatment processes and not from the wastewater treatment process. This is based on the literature review findings discussed in Chapter 2.4.3, which attributes methane formation to the sludge handling process and sewer networks. Since the collection phase is out of the scope of this carbon footprint study, it was excluded. Moreover, the report from the IPCC Task Force on National Greenhouse Gas Inventories stated that methane formation in sewers of temperate countries is very low due to the lower temperature of the wastewater (Penman, 2000).

Taskila WWTP composted KemiCond treated sludge while other plants compost digested sludge. No EFs regarding composting of KemiCond treated sludge existed at the time of this study. In order to maintain consistency between the sites, it was decided to use the

IPCC default EF for composting for all sites in this study. The EF can be found in Appendix 1.

EFs regarding methane emissions from WWTPs vary significantly in literature which could range from 0.06% to even 21% of the influent wastewater COD emitted as CH_4 (refer to Table 3). It was decided that the use of a single EF for the anaerobic digestion process was not representative due to different sludge storage times in the case studies plants. Also, a single EF might assume that all biogas is combusted which was not the case for the Mäkikylä plant in this study (they sold biogas). Thus, for the digestion process, three different EFs were used for the following emissions: i) leaks from digesters, ii) emissions from sludge storage and iii) methane slips from combustion units. These EFs can be found in Table 8

The Paroinen and Riihimäki WWTP did not measure how much biogas was produced since they did not sell it and all the biogas was subsequently combusted for heat. Due to this, an estimation of how much methane was produced had to be made in order to further estimate fugitive gases from the anaerobic digestion process. Additional EFs used to estimate this amount of produced biogas are also shown in Table 8.

The emissions from the digestion process at ENVOR Oy were based on leakages from anaerobic digestion and methane slip from combustion units. This was due to limited data regarding sludge storage times. Indirect emissions of the facility at ENVOR Oy were not included.

The outsourced composting emission were based on the amount of sludge sent off-site and did not include the indirect emissions of the facility handling the composting process.

The full list of methane EFs used can be seen in Table 8.

Table 8 - EFs used for estimating methane emissions

Source of methane emissions	**Value**	**Unit**	**literature source**
Composting of sludge	10	g CH_4. kg DS^{-1}	IPCC (2006)
Leakages from anaerobic digestion	0.4%	produced biogas	Tauber *et al* (2019)
Methane slip from combustion units	1%	CH_4 combusted	Schaum *et al*. (2015)
Sludge storage	8.9	gCH_4-C.ton DS^{-1}.day^{-1}	Willén *et al* (2016)
Biogas potential of sludge	308	Nm^3 biogas. ton VS^{-1}	Kiselev *et al* (2019)
Density of CH_4 at standard temperature and pressure	0.72	kg. Nm^{-3}	Suomen kaasuyhdistys (2016)
Methane content of biogas	60	% biogas produced	Kiselev *et al* (2019)

DS= Dry Solids, Nm^3 = volume at standard temperature and pressure, VS= volatile solids

3.7.5 N_2O emission calculation

Literature studies suggest that N_2O emissions are site specific and hence the only accurate way to quantify these are through on-line measurements on-site (refer to Chapter 2.4.1).

Since none of the sites had measurement data for the period of study, an EF had to be chosen, based on literature studies.

Due to numerous studies existing regarding N_2O emissions from the wastewater treatment process, criteria needed to be set regarding the choice of EF used. The sampling methodology and duration of measurement campaigns were decided as the most important criteria for selecting an EF for this study. N_2O studies with just a few days measurement campaigns were excluded from the potential EF list. This was done to eliminate the possibility of temporal anomalies arising from short-term changes in process conditions. Additionally, studies with short-term measurement campaigns have lower EF values on average (0,8%) compared to long term studies (1,7%), which suggests a degree of underestimation (Vasilaki *et al.* 2019). Secondly, studies which did not incorporate continuous monitoring (e.g studies using grab samples) were not considered. This is due to grab samples only representing the emission value of a specific point and time. Thus, continuous temporal variations are not adequately analyzed.

Taking all these criteria into account, the potential EFs were narrowed down to three studies by Daelman *et al* (2015), Kosonen *et al*. (2016) and Rodriguez-Caballero *et al.* 2014 which took place in Netherlands, Finland and Spain respectively. These studies were found to have the longest measurement periods (at the time of this study) and were based on continuous on-line measurements. The plants involved in these studies employed activated sludge processes consisting of plug-flow reactors, with anoxic-oxic configurations, which matched the sites in this study. These three studies represented their EFs in terms of % influent TN and the average of these three studies was found to be 1.61% TN. This was close to the average given by Vasilaki *et al.* 2019, who stated the average EFs of long-term studies to be 1.7%. This was almost the same as the IPCC emission factor of 1.6% of influent nitrogen emitted as N_2O-N.

It was finally decided to use the official IPCC default EF of 1.6% nitrogen load (released as N_2O-N) as the main EF in this study, since it was relatively closer to the average EF value for long-term studies (1.7%). In order to compare the sensitivity of the plants CF to the N_2O EF, two additional EFs were tested:

- 2.1% TN (average for studies involving side-stream processes)
- 0.8% TN (average for short-term studies)

4 Results

The results section first shows the monthly PEs served by the WWTPs based on their monthly BOD_7 loading gathered from official yearly reports. The overall CF results is then given for all WWTPs before going into more detail for each WWTP. At the end of the chapter, a comparison of the GHG emissions based on process performance among sites will be given.

4.1 Loading rates of WWTPs

Since the main functional unit used in this study is kg CO_2eq PE^{-1} $year^{-1}$, the monthly PEs for each site were first determined. Taskila and Mäkikylä WWTP both experienced the most fluctuations in PEs served during the spring of 2017 and 2018. Paroinen and Riihimäki WWTP had relatively more stable PEs during the study period than Taskila and Mäkikylä WWTP. The three-month average PEs served for each site and further used for the carbon footprint calculation can be seen in Figure 19.

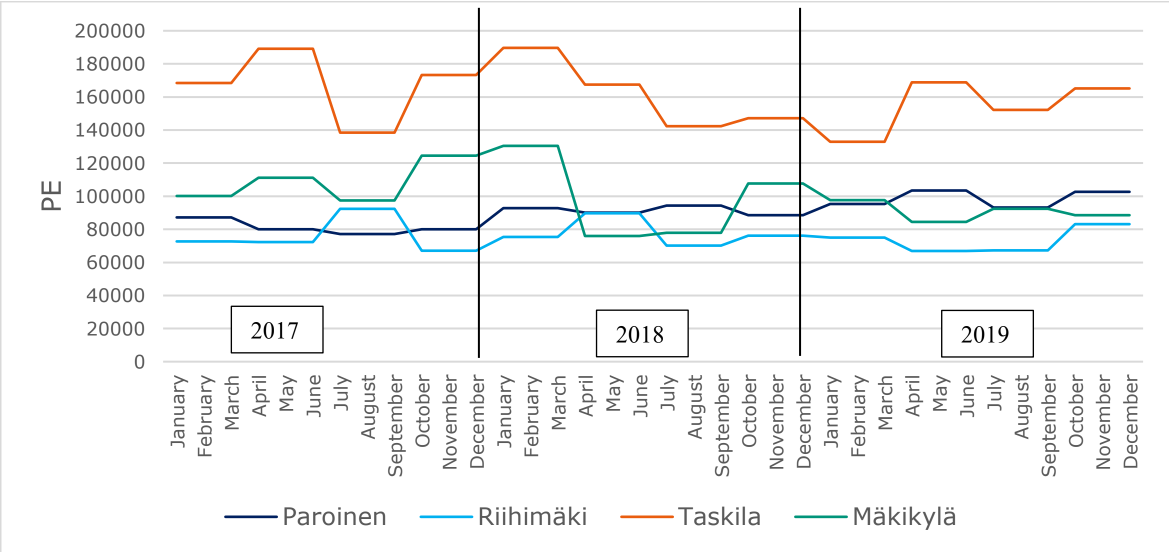


Figure 19 - Monthly PEs served by the WWTPs over the study period (3-month average)

Since the EF for N_2O emissions from wastewater treatment was based on total nitrogen loading to the sites, the three-month average total nitrogen loading per day were obtained and the normalized loads in terms of PE over the three-year period were calculated. The daily nitrogen load shown in Figure 20 appeared to be lower during the summer on average, for all WWTPs.

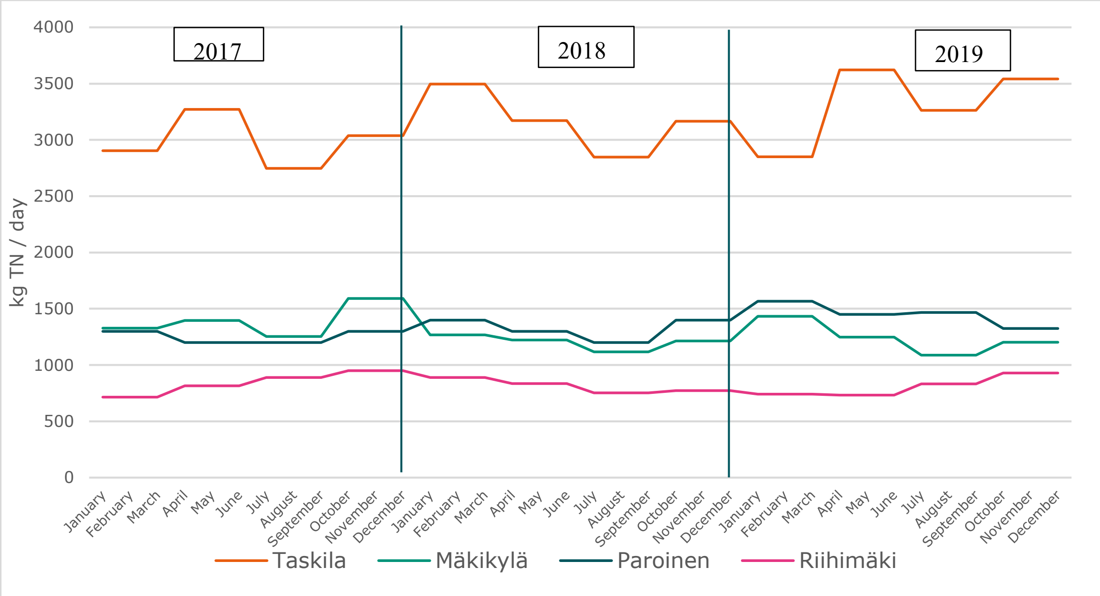


Figure 20 - Daily total nitrogen loading to studied WWTPs (3-month average)

Figure 21 shows the average daily nitrogen load and its normalized value, in terms of PE. Taskila WWTP had the highest daily load per PE, while the Riihimäki WWTP had the lowest.

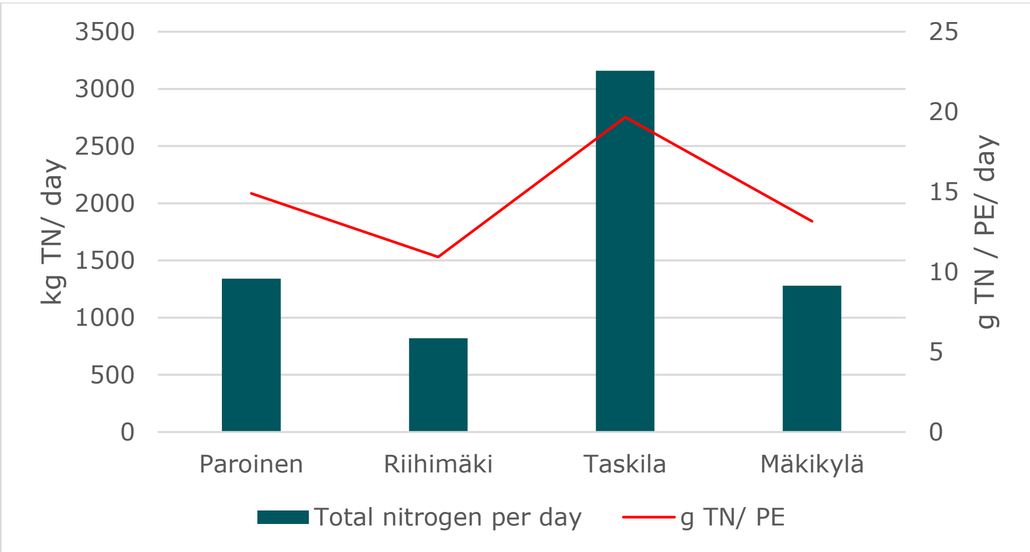


Figure 21 - Average total nitrogen loading per day and normalized load per PE for the WWTPs over the three-year period

4.2 Carbon footprint results of all WWTPs

The average CF of the studied plants ranged from 45 to 82 kg CO_2eq PE $^{-1}$ year $^{-1}$. The overall average CF between the sites was 63 kg CO_2eq PE $^{-1}$ year $^{-1}$. The monthly carbon footprint of the WWTPs can be seen in Figure 22. The summary of the average carbon footprint per year expressed in other FUs, for each of the WWTPs, is shown in Table 9.

The comparison of other FUs will be discussed in Chapter 4.6.

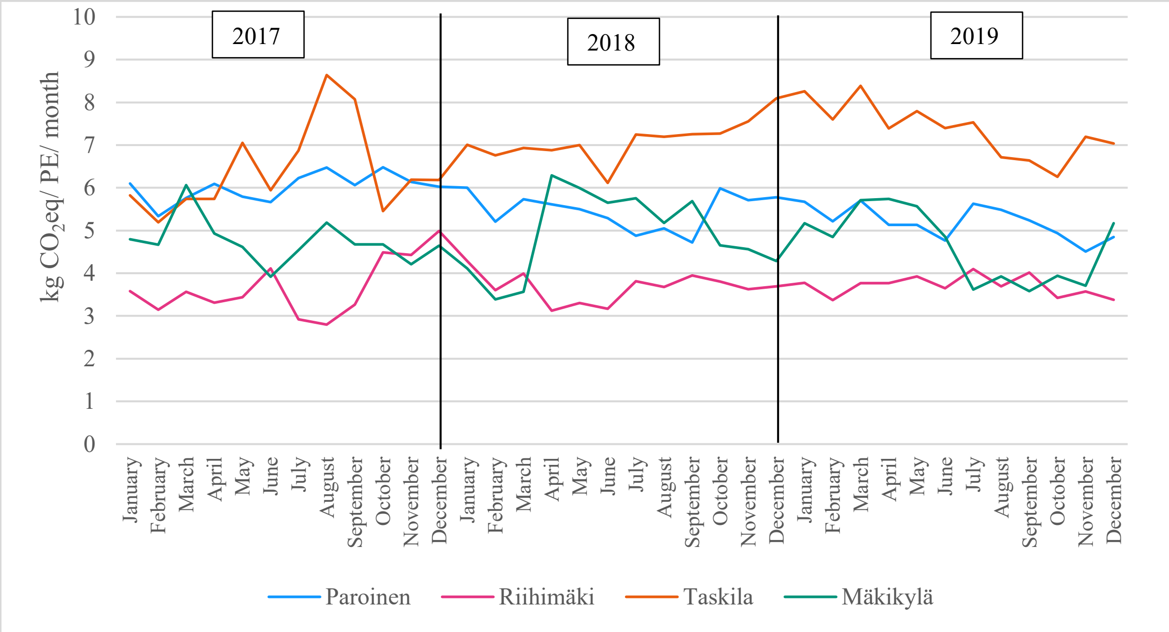


Figure 22 - Monthly carbon footprint of WWTPs in this study over the three-year study period.

In order to determine the variability of the monthly CF and PEs served, the standard deviation was calculated for all WWTPs. A site having a lower standard deviation indicates that the magnitude of monthly emissions for that site would be closer to the three-year average with less fluctuations. The results in Table 9 showed that Taskila WWTP had the highest variable monthly CF, while Riihimäki WWTP had the lowest. Monthly PEs served were the most stable for Paroinen WWTP and least stable for Taskila WWTP. Taskila WWTP's TN loading rate had the most fluctuations while Riihimäki WWTP's TN loading rate was the most stable.

Table 9 - Standard deviations of monthly CF, PEs served, and TN loading over the three-year period for the WWTPs.

WWTP	monthly CF	Monthly PEs served	Monthly TN loading
Mäkikylä	0.79	16 600	136
Paroinen	0.51	8 200	114
Taskila	0.84	18 200	284
Riihimäki	0.45	8 300	78

A summary of the total emissions, offsets and CF results for each WWTP and the respective years can be seen in Table 10. The CF expressed in other functional units are also shown.

Table 10 - Summary of CF results for all WWTPs in this study

Year	Functional unit	Paroinen	Riihimäki	Mäkikylä	Taskila
	Total emissions (ton CO_2eq)	5854	3333	6136	12335
	Total carbon offsets (tons CO_2eq)	-77	0	-767	0
	Total net emissions (tons CO2eq)	5777	3333	5370	12335
2017	kg CO_2eq/m^3 treated wastewater	0.7	0.6	0.6	0.7
	kg CO_2eq / kg TN removed	20	16	18	30

	kg CO_2eq / kg TP removed	92	91	100	88
	kg CO_2eq / PE/ year	72	45	57	75
	Total emissions (ton CO_2eq)	5979	3449	5603	13360
	Total carbon offsets (tons CO_2eq)	-155	0	-714	0
	Total net emissions (tons CO2eq)	5825	3449	4889	13360
2018	kg CO_2eq /m3 treated wastewater	0.8	0.7	0.6	0.8
	kg CO_2eq / kg TN removed	18	15	19	26
	kg CO_2eq / kg TP removed	86	89	112	83
	kg CO_2eq / PE/ year	65	45	60	83
	Total emissions (ton CO_2eq)	6124	3269	5103	13602
	Total carbon offsets (tons CO_2eq)	-150	0	-451	0
	Total net emissions (tons CO2eq)	5974	3269	4652	13602
2019	kg CO_2eq /m3 treated wastewater	0.8	0.7	0.5	0.8
	kg CO_2eq / kg TN removed	17	15	17	21
	kg CO_2eq / kg TP removed	82	95	95	84
	kg CO_2eq / PE/ year	62	45	56	88

The correlation coefficients between the monthly CF values, direct or indirect emissions and PEs served can be seen in Table 11. Generally, there were no strong positive correlations between the monthly CF and either indirect or direct emissions among the sites. All the WWTPs had moderate to strong negative correlations between the monthly PEs served and the monthly CF.

Table 11 - Correlation coefficients between indirect/direct emissions, monthly PEs served and the monthly CF of WWTPs in this study.

	Correlation coefficients between monthly CF and indirect emissions	**Correlation coefficients between monthly CF and direct emissions**	**Correlation coefficients between monthly CF and PEs served**
Paroinen WWTP	0.34	0.20	-0.83
Mäkikylä WWTP	-0.09	0.46	-0.58
Riihimäki WWTP	0.28	0.52	-0.71
Taskila WWTP	0.35	0.35	-0.64

4.2.1 Taskila WWTP

Taskila had an average of 82 kg CO_2eq PE^{-1} $year^{-1}$ over the three-year period (refer to Table 10). This was the highest carbon footprint in terms of population equivalent among the WWTPs for each year studied. The total monthly direct/indirect emissions and the monthly CF can be seen in Figure 23.

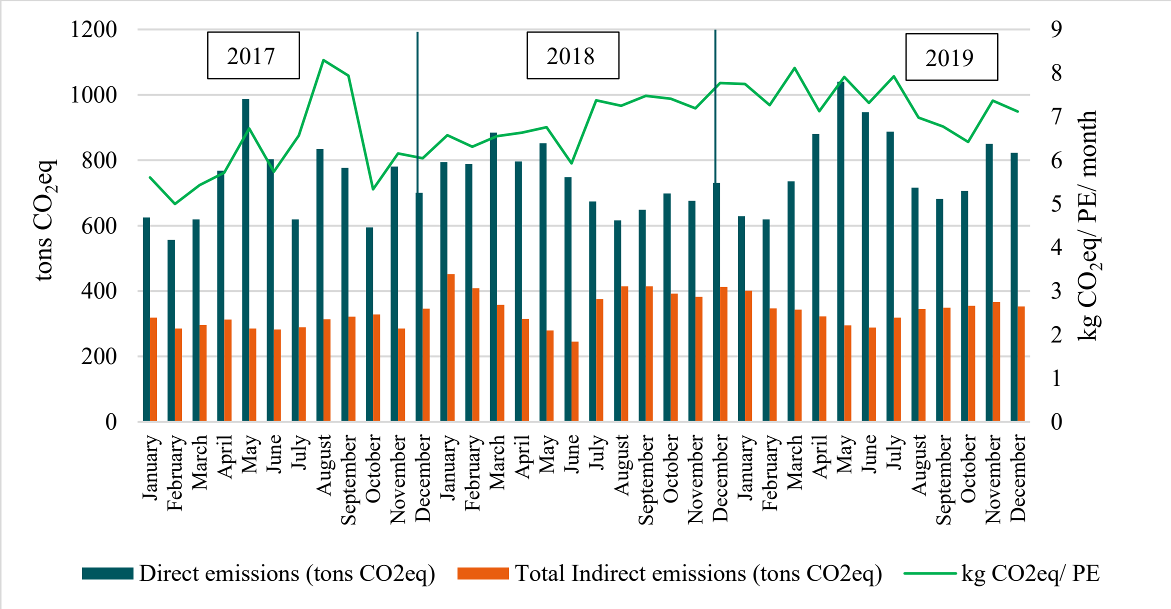


Figure 23 - Monthly carbon footprint, direct and indirect emissions of Taskila WWTP over the studied period.

The shares of the total emissions by category can be seen in Figure 24. Approximately 69% of the total CF was attributed to direct emissions from wastewater, sludge, and screenings treatment. Among the WWTPs, Taskila had the highest share for chemical consumption (14%) but had the lowest share for energy consumption (15%). For energy consumption emission shares, ~13% was attributed to electricity, and ~2% for heat consumption. Transport emissions contributed a low share of 2% of the CF.

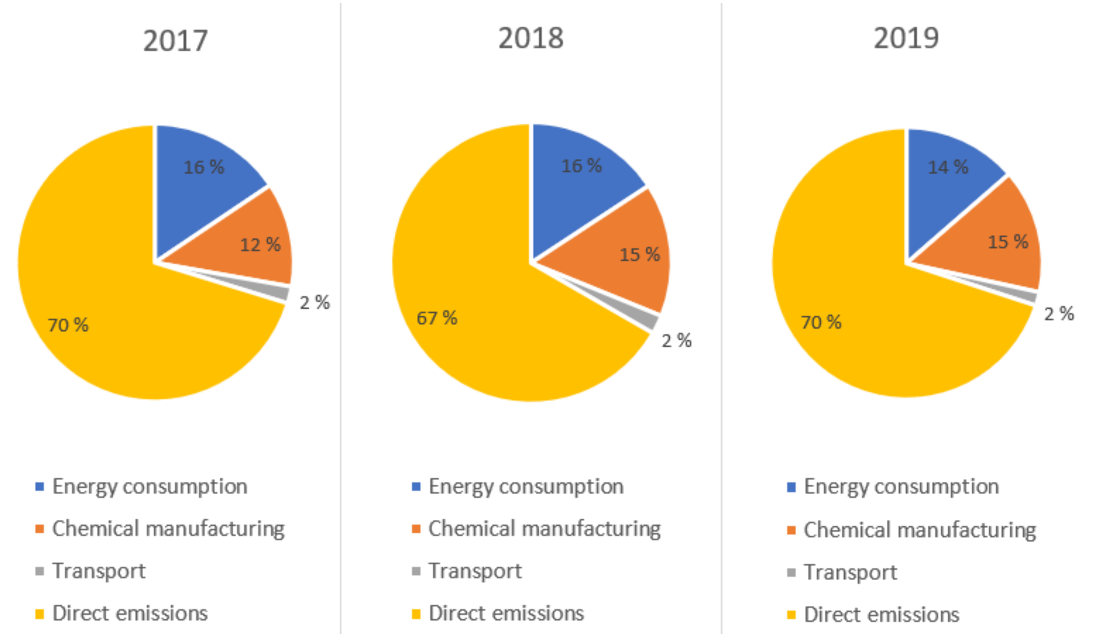


Figure 24 - Shares of Taskila WWTP's total emissions by category

Taskila's indirect emissions were dominated by chemical consumption emissions. The energy consumption emissions were highest during the winter period, with chemical consumption emissions showing similar peaks. The total indirect emissions and its contributors over the study period can be seen in Figure 25.

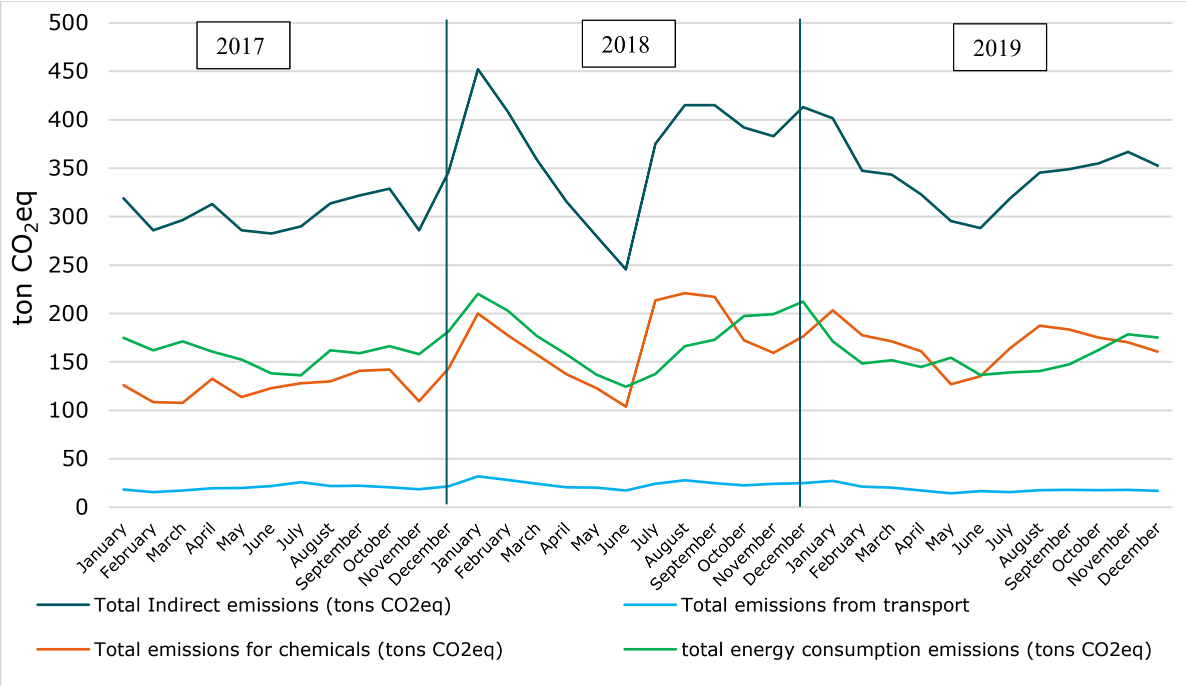


Figure 25 - Monthly indirect emissions of Taskila WWTP and contributing categories

The direct emissions for Taskila were heavily dependent on N_2O emissions from wastewater treatment. The peaks of direct emissions were mostly dictated by CH_4 and N_2O emissions from composting, due to its higher fluctuation and variability along the study period. The correlation coefficient between the total direct emissions and total composting emissions was 0.8, which suggests strong positive correlation. The correlation coefficient between the total direct emissions and N_2O emissions from wastewater treatment was 0.65, which also suggests moderate positive correlation. Other sources of direct emissions from Taskila WWTP constituted negligible shares of ~0.5% of total emissions (refer to Appendix 5).

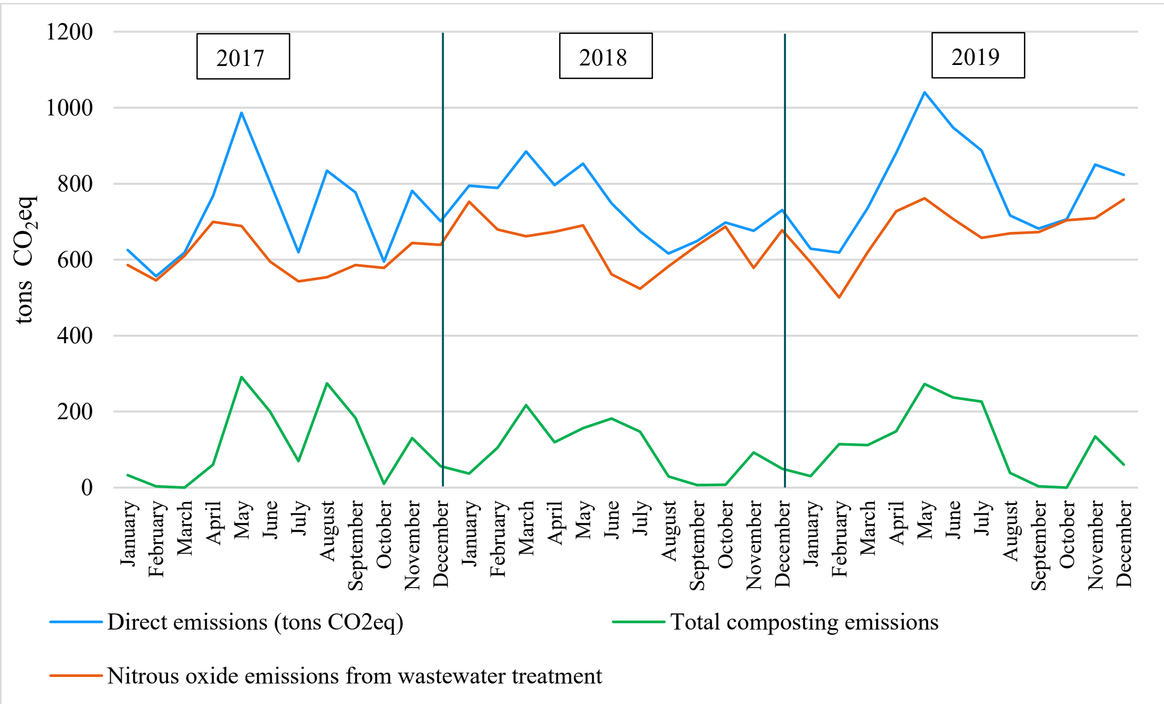


Figure 26 - Total direct emissions and contributing categories of Taskila WWTP.

The detailed emission sources and their respective shares for Taskila WWTP can be seen in Appendix 5.

4.2.2 Riihimäki WWTP

The Riihimäki WWTP had the lowest carbon footprint among the studied sites with an average of 45 kg CO_2eq PE $^{-1}$ year $^{-1}$. The dynamic of the CF over the whole 3-year period can be seen in Figure 27.

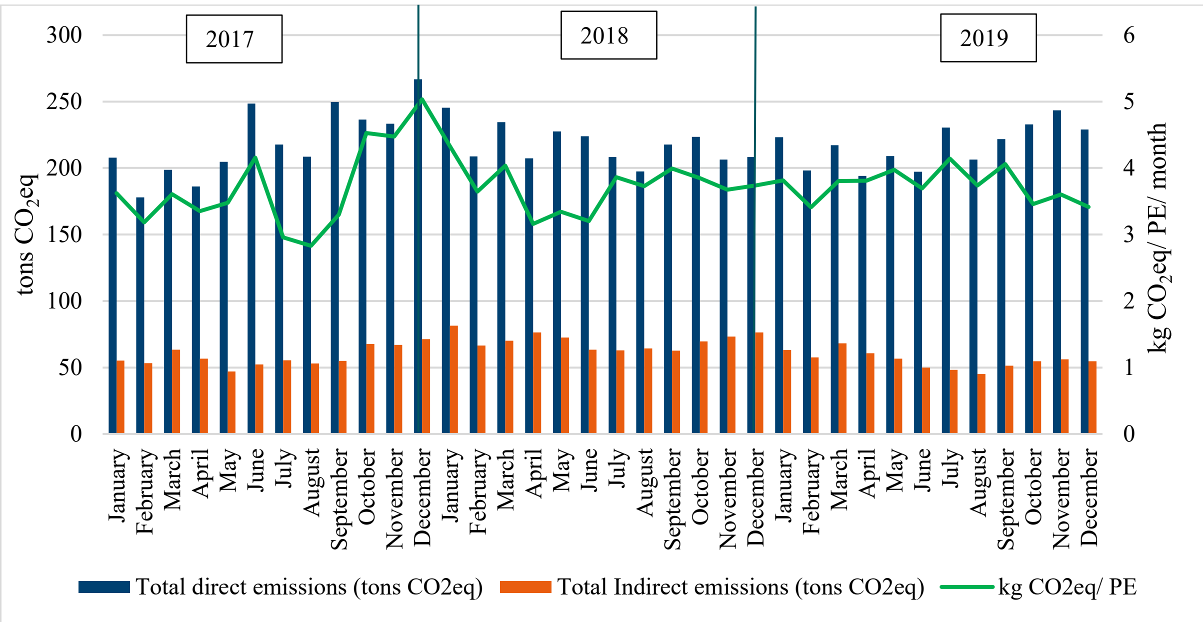


Figure 27 - Riihimäki WWTP's monthly carbon footprint along with direct and indirect emissions over the study period.

Among the sites, Riihimäki WWTP had the highest share for direct emissions, with an average of 78% of total GHG emissions. The second highest was electricity consumption with 20%, while transport and chemical consumption emissions had minor shares (~1% each). The site's share for chemical consumption emissions were the lowest among the WWTPs in this study. Since Riihimäki WWTP combusted natural gas on-site for heating, these emissions were added to the direct emissions of the site. The shares of emissions for the respective years can be seen in Figure 28.

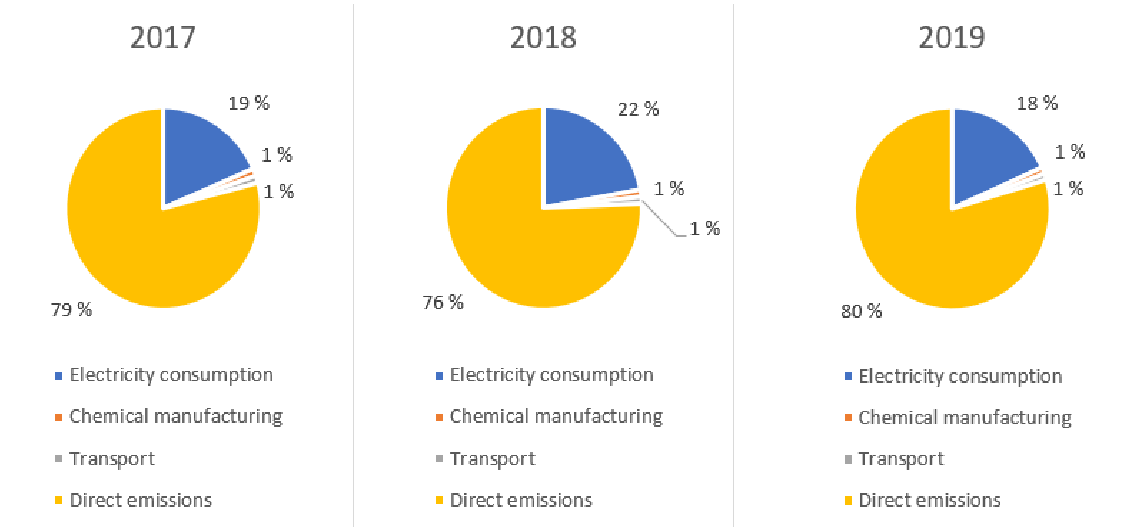


Figure 28 - Total emission shares by category of Riihimäki WWTP

Riihimäki WWTP's indirect emissions were heavily dependent on the electricity consumption emissions, as shown in Figure 29.

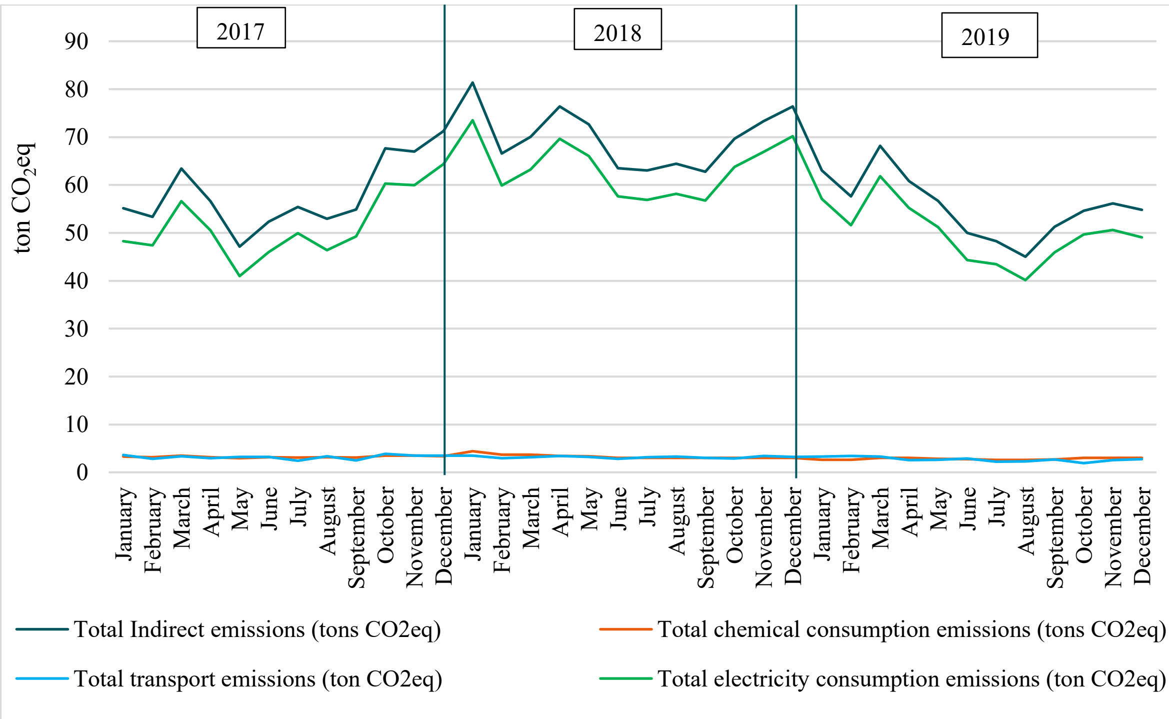


Figure 29 - Contributing categories for indirect emissions and their shares for Riihimäki WWTP

Direct emissions for Riihimäki WWTP were dominantly based on N_2O emissions from the wastewater treatment process. Methane emissions from anaerobic digestion constituted the minimum shares for direct emissions. The direct emissions over the studied period can be seen in Figure 30.

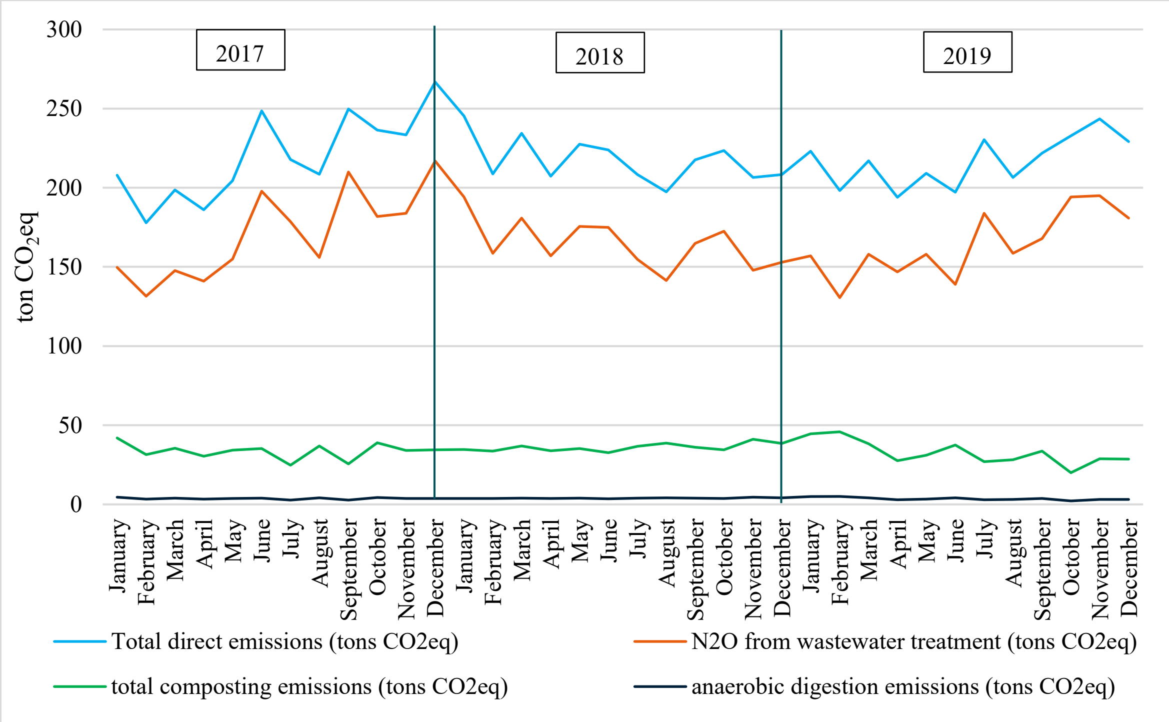


Figure 30 – Riihimäki WWTP's direct emissions and their contributing categories

The detailed emission sources and their respective shares for Riihimäki WWTP can be seen in Appendix 4.

4.2.3 Paroinen WWTP

Paroinen WWTP's average CF over the three-year period was ~67 kg CO_2eq PE $^{-1}$year^{-1}. The monthly CF, direct and indirect emissions can be seen in Figure 31.

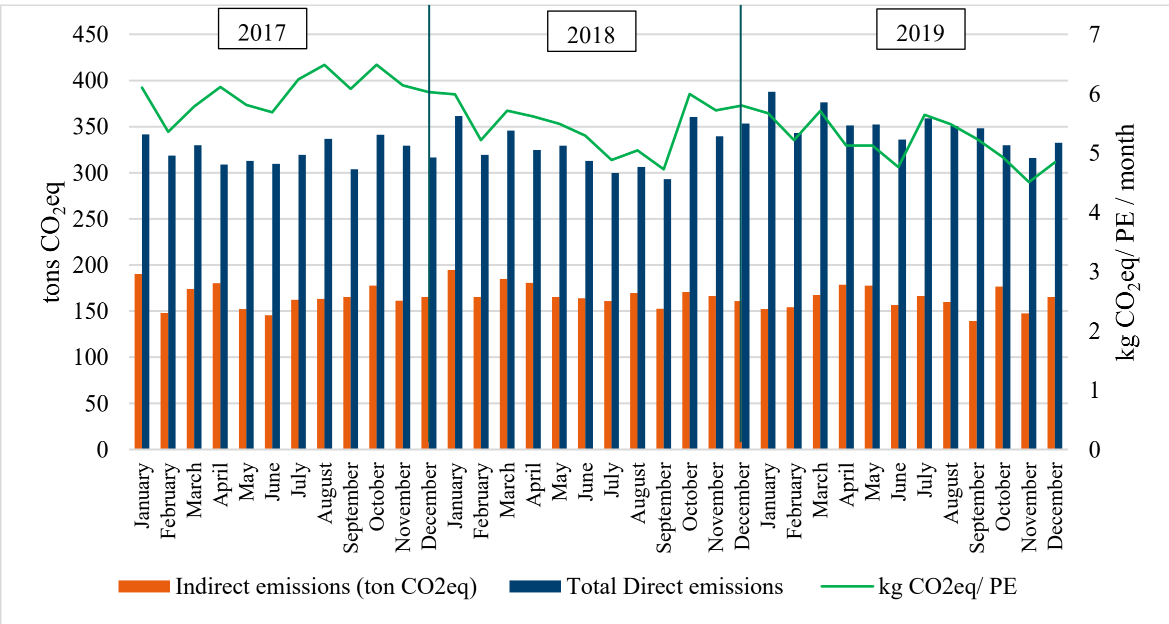


Figure 31 - Paroinen WWTP monthly carbon footprint with direct and indirect emissions

The shares of the total CO_2eq emissions can be seen in Figure 32. Approximately 66% of total emissions were attributed to direct emissions. Energy consumption was the second highest source for emissions with a share of ~24%. Chemical consumption emissions were the second highest with ~9% of total emissions. Transport emissions constituted minor shares with ~1%.

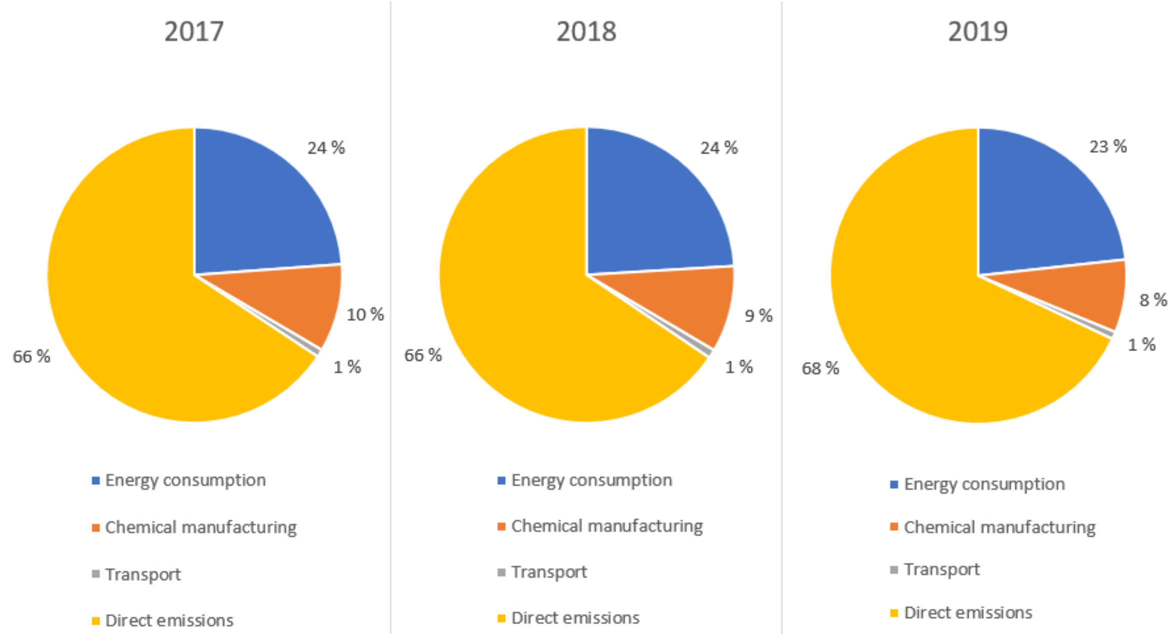


Figure 32 - Shares of Paroinen's total emissions by category

Paroinen WWTP's yearly average indirect emissions were stable over the study period. Energy consumption dominated the indirect emissions and constituted the highest share of total emissions among the sites. Contributing categories for indirect emissions can be seen in Figure 33.

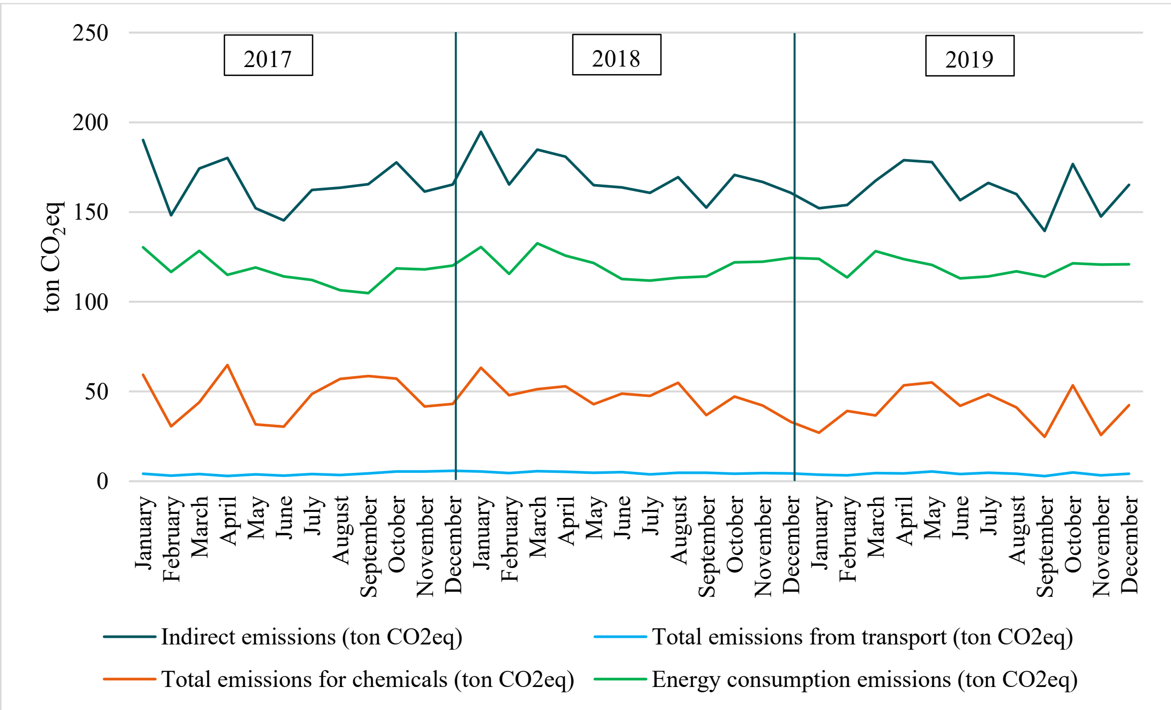


Figure 33 – Paroinen WWTP's total indirect emissions with contributing categories

Paroinen WWTP's direct emissions trends were heavily influenced by the N_2O emissions from wastewater treatment, as shown in Figure 34. There were no clear seasonal trends with direct emissions. Composting was the second highest while emissions from anaerobic digestion contributed the least.

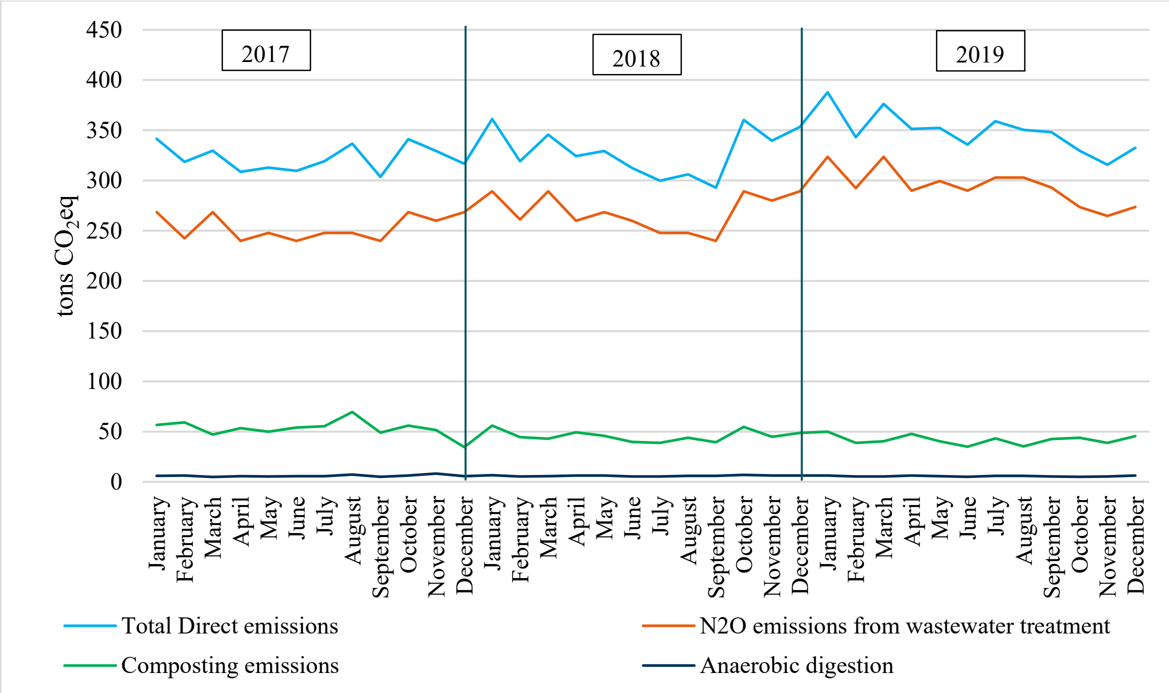


Figure 34 – Paroinen WWTP's direct emissions and its contributing categories

Paroinen WWTP sold excess heat produced on-site to the grid and hence the emission offsets from these sales were calculated. The offsets were then presented in terms of kg CO_2eq saved PE^{-1} $year^{-1}$ with a negative value. Figure 35 shows offset peaks occurring mostly during the summer/autumn, except the peak in December 2017 which was the only peak that occurred during winter.

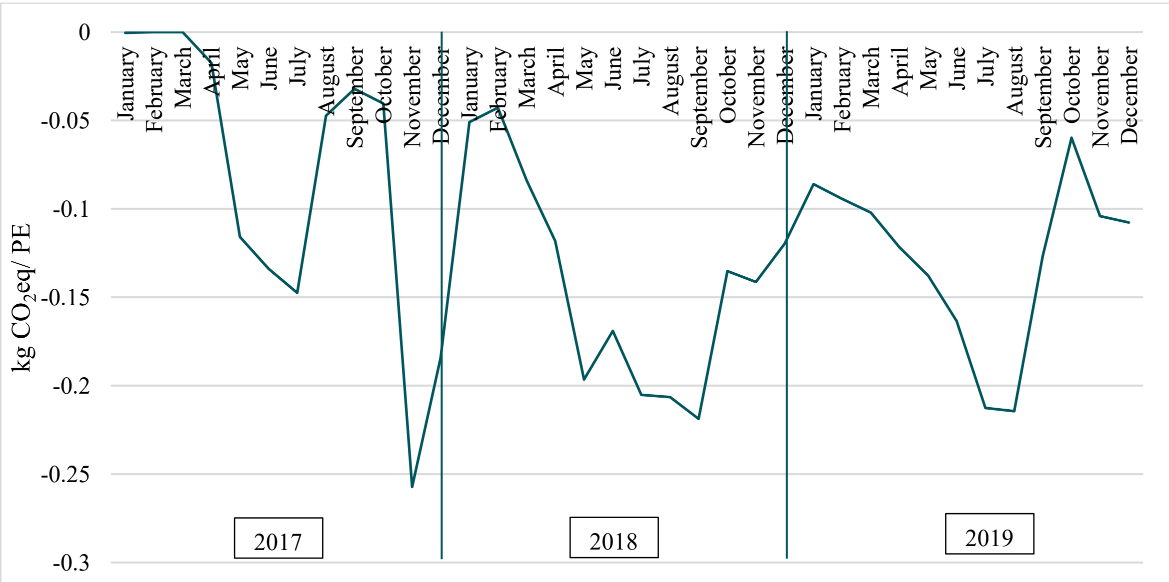


Figure 35 - Emission offsets of Paroinen WWTP per PE during the study period

In order to understand the trends and peaks of the offsets, the amount of district heat sold by the plant was investigated and shown in Figure 36. Paroinen WWTP sold most of its excess heat during the summer and sold more heat than was purchased during the years

2018 and 2019. This accounts for the occurrence of the offset peaks in the summer. The winter peak was attributed to the offsets from large amounts of sludge sent to ENVOR Oy for anaerobic digestion between October and December.

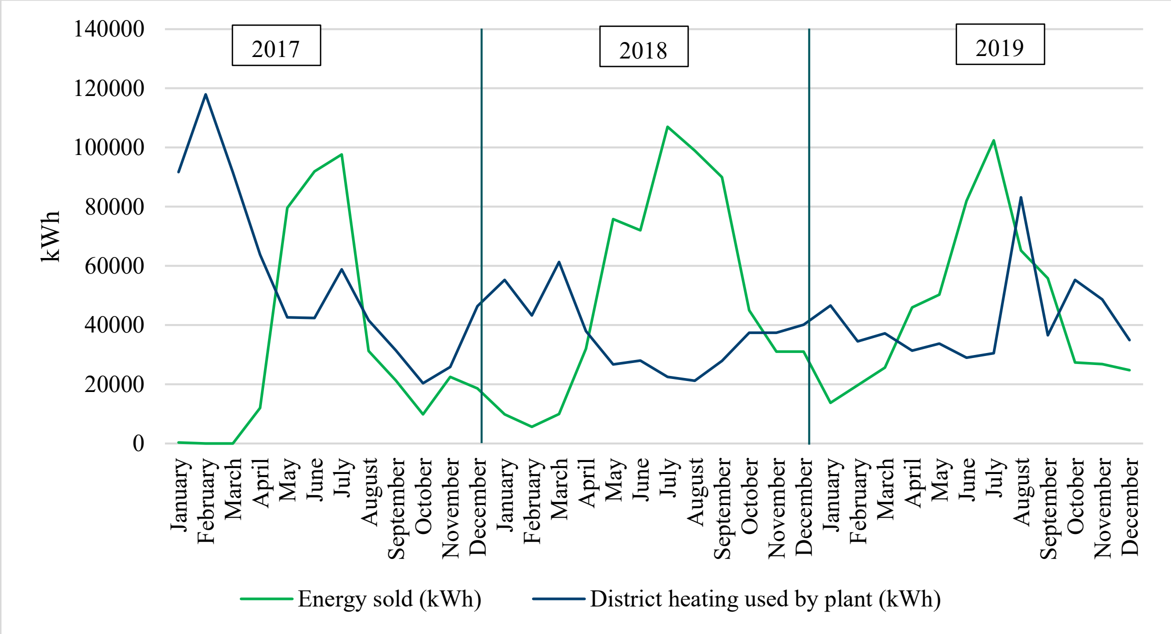


Figure 36 - District heat consumed and sold by the Paroinen WWTP

4.2.4 Mäkikylä WWTP

Mäkikylä WWTP had an average CF value of 58 kg CO_2eq PE $^{-1}$year^{-1}. It also had the highest emission offsets among the WWTPs for the studied period. The monthly CF, direct and indirect emissions can be seen in Figure 37.

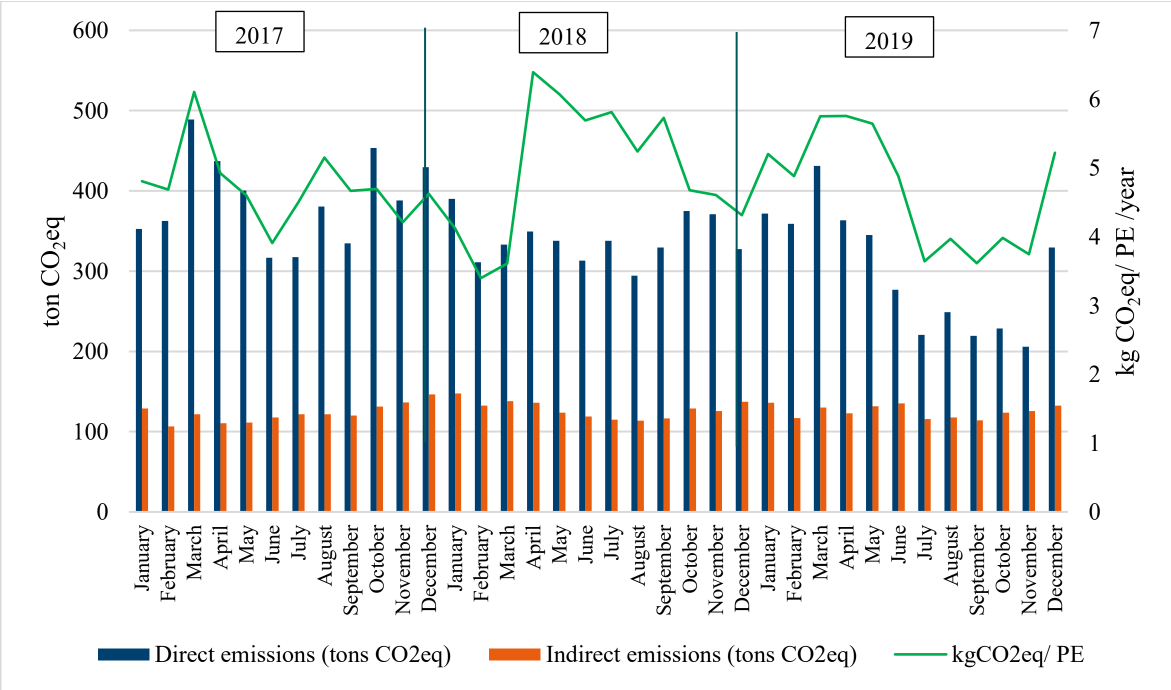


Figure 37 - Mäkikylä WWTP's monthly CF along with direct and indirect emissions

The share of total emissions by category can be seen in Figure 38. As with the other WWTPs, direct emissions had the largest share with ~73%, followed by energy consumption with ~18% of emissions. Chemical consumption constituted ~8% while transport emissions had minor shares of ~1% on average.

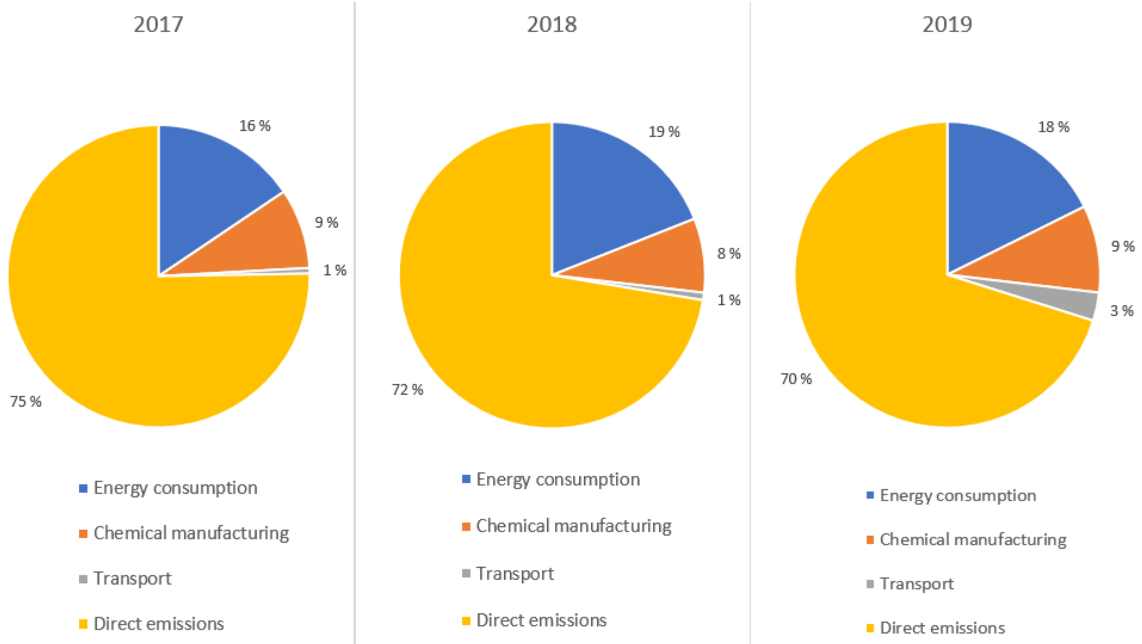


Figure 38 - Share of total emissions of Mäkikylä WWTP by category for the three-year period.

Energy consumption emissions had majority share of the indirect emissions. Chemical consumption emissions remained relatively constant over the study period. Transport emissions remained constant and rose sharply from May to December 2019. Total indirect emissions and their contributors over the study period can be seen in Figure 39.

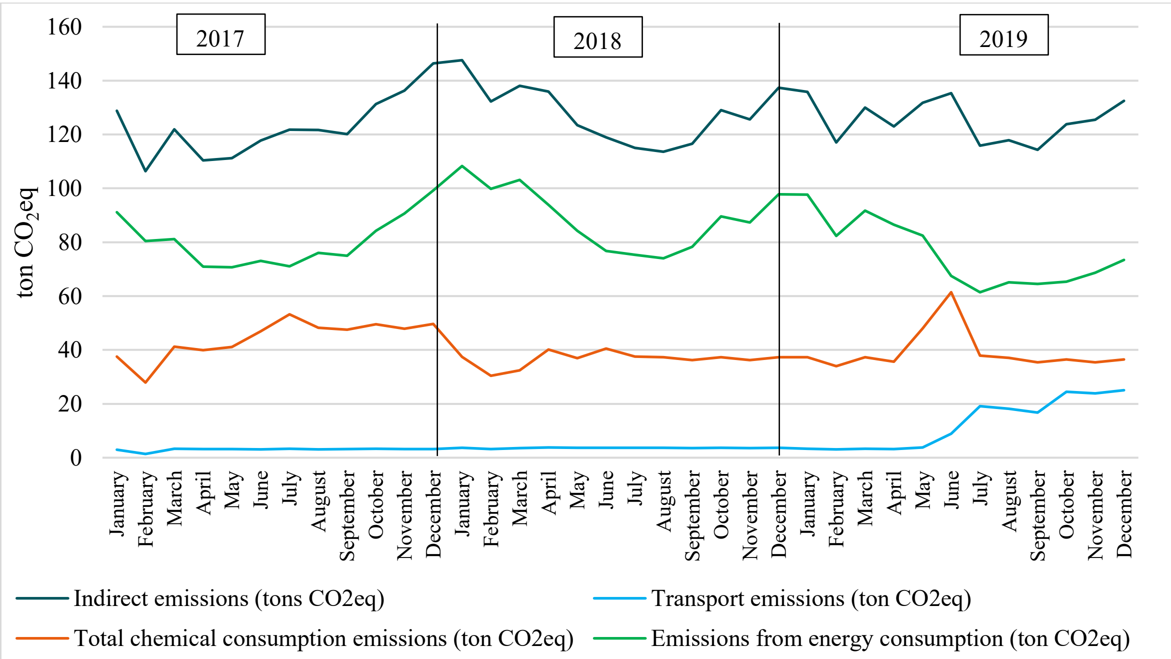


Figure 39 – Mäkikylä WWTP's indirect emissions and their contributing categories

N_2O emissions from wastewater treatment dominated the direct emissions as shown in Figure 40. Composting and anaerobic digestion emissions were stable but sharply declined after May 2019, when the biogas plant was sold.

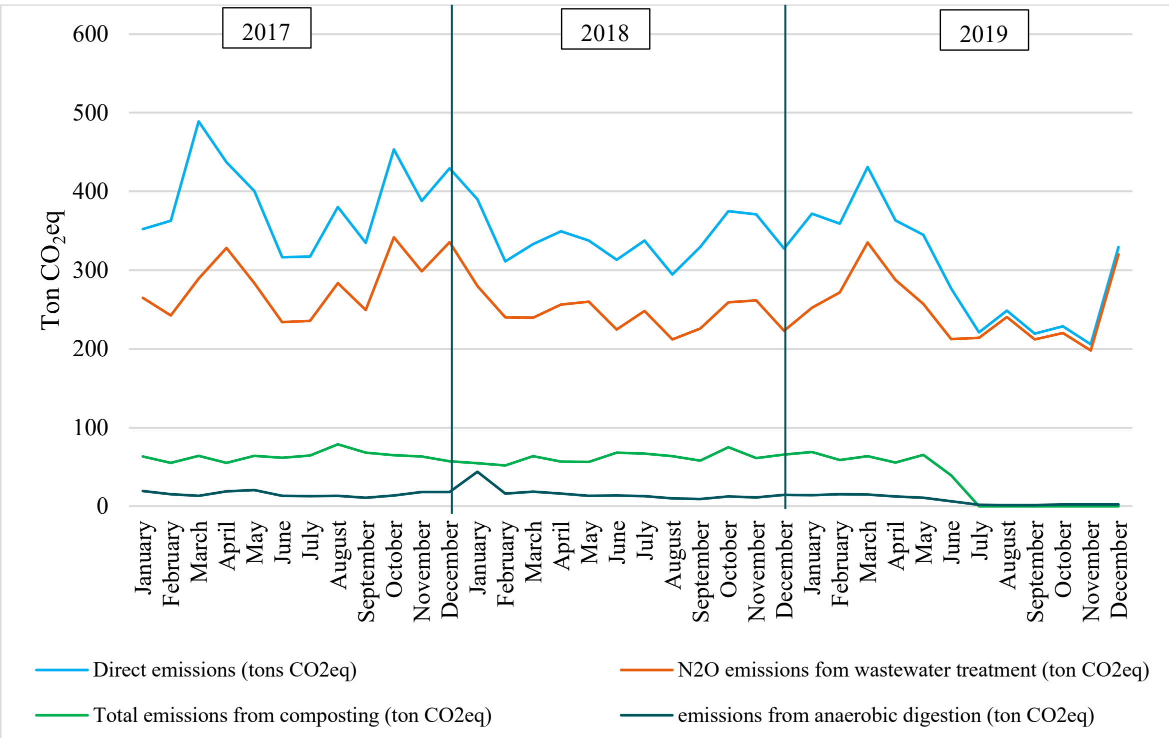


Figure 40 – Mäkikylä WWTP's direct emissions and contributing categories

Kouvola Vesi Oy, which owns the Mäkikylä WWTP, operated the biogas plant until June 2019, when it was sold to Gasum Oy. During the ownership by Kouvola Vesi Oy, biogas was sold to Gasum Oy. The emission offsets from the sale of biogas represented in kg CO_2eq saved PE^{-1} $month^{-1}$ can be seen in Figure 41. The offsets dipped after June 2019 after the biogas plant was sold and offsets were based on the different fate of the biogas from outsourced digestion. There were no clear seasonal trends for the emission offsets.

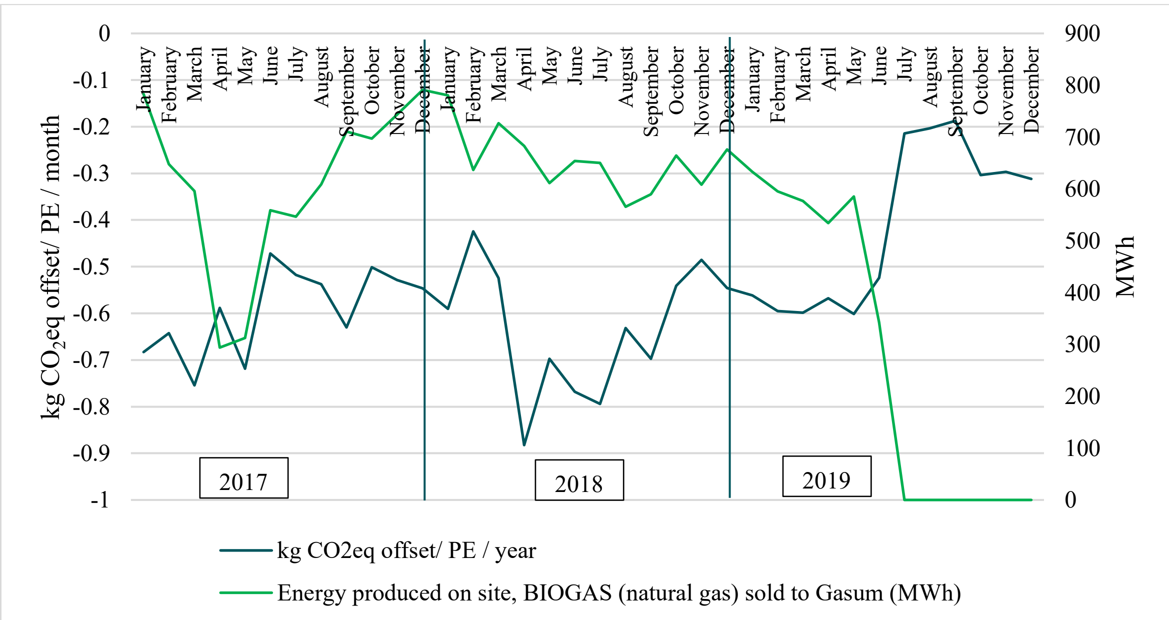


Figure 41 – Mäkikylä WWTP's monthly emission offsets and amount of sold biogas

4.3 Comparison of energy efficiency among sites

4.3.1 Electricity consumption and efficiency

This sub-chapter will compare the electricity consumption efficiencies of the WWTPs in terms of PE treated and 1 m^3 wastewater treated.

During the studied period, solar panels were operated at Mäkikylä and Taskila WWTPs which supplemented the electricity purchased by the plants. For the Taskila WWTP, the solar panels contributed ~1.3% of the yearly consumption. For Mäkikylä WWTP, the contribution of solar panels was negligible (~0.01% of the yearly consumption) since it began serving the plant at the last two months of the study period. Paroinen relied fully on the grid for electricity and had no electricity generated on-site over the study period. Riihimäki generated 2.8% of its electricity consumption in 2017 from biogas combusted in CHP units but this was discontinued due to a faulty CHP unit. Due to the small share of the electricity generated and consumed on-site, it can be concluded that the sites were mostly dependent on the grid for electricity.

For the biogas plant operated in conjunction with the Mäkikylä WWTP, the amount of electricity produced and used on-site was estimated based on the difference between the electricity purchased from the grid and electricity used by the biogas plant. It was estimated that ~16% of the electricity consumed by the biogas plant was produced on-site from natural gas and biogas. The amount of electricity used by the biogas plant was included in the consumption of the Mäkikylä WWTP for the CF calculation.

Figure 42 shows the amount of purchased electricity by each studied WWTP. Among the WWTP, Riihimäki purchased the least electricity while Taskila purchased the most on average.

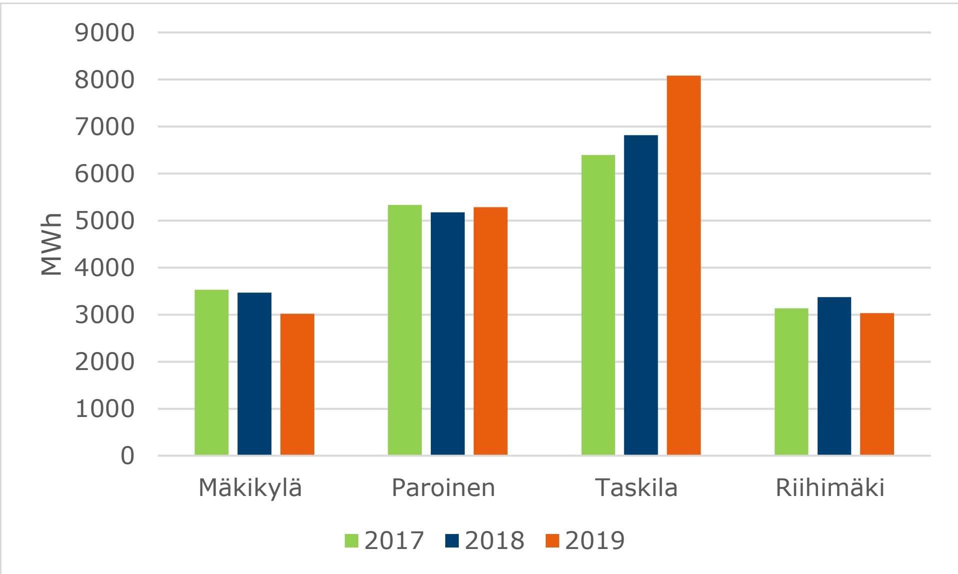


Figure 42 - Electricity purchased by each WWTP over the studied period

Since the main functional unit in this study was the PE, the electricity consumed per PE treated per year was calculated for more comparability. The results in Figure 43 show Mäkikylä being the lowest electricity consumer on average, per PE treated. Paroinen WWTP was the highest electricity consumer per PE treated.

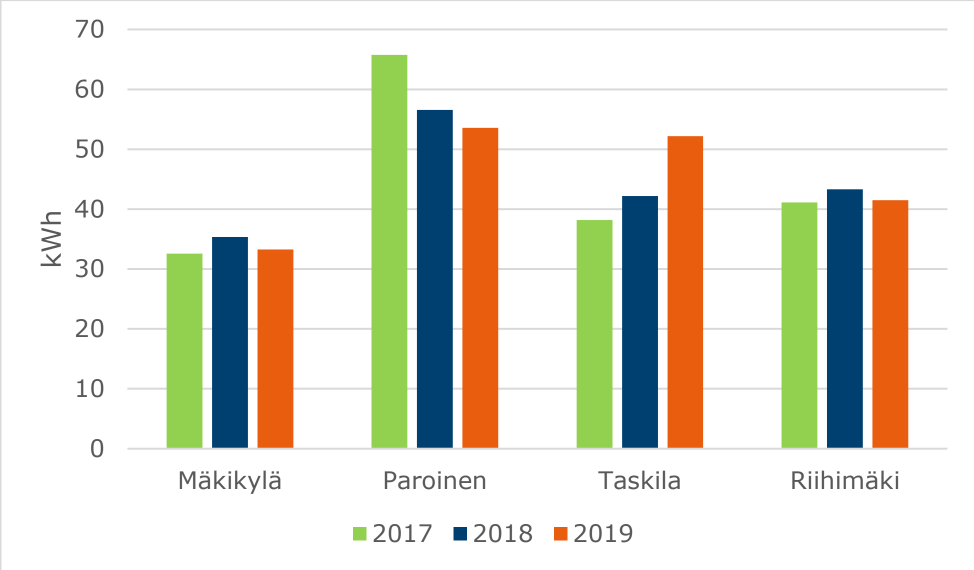


Figure 43 - Electricity consumed per PE treated/year

Mäkikylä WWTP consumed the least electricity while Riihimäki WWTP consumed the highest per m^3 treated wastewater, as shown in Figure 44.

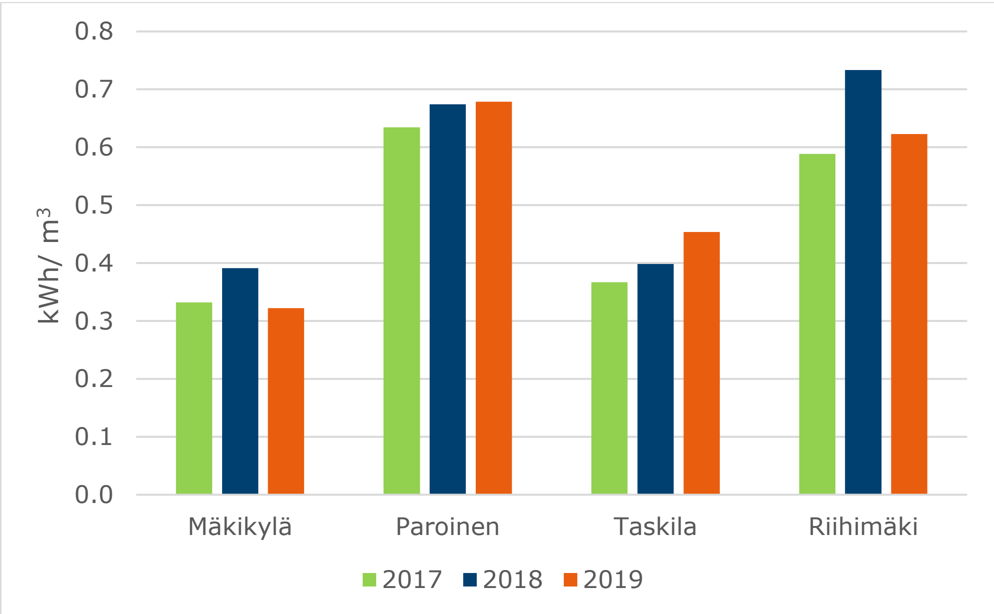


Figure 44 - Electricity consumption per m^3 wastewater treated for WWTPs studied

4.3.2 Heating

This chapter will compare the heat energy consumed by each WWTP

All sites except Taskila WWTP supplemented their purchased heat with heat generated from biogas combustion in gas boilers or heat recovered from effluent wastewater. Riihimäki and Mäkikylä WWTP's purchased heat energy was based on natural gas while Paroinen and Taskila used district heating. Riihimäki purchased natural gas which was combusted on-site to produce heat and hence was considered as direct emissions. District heating emissions shares in this study ranged from 1% to 5% of total emissions. Due to insufficient data, the percentage shares of heating attributed to biogas combustion on-site could not be estimated for the Paroinen and Riihimäki WWTPs.

The biogas plant operated in conjunction with the Mäkikylä WWTP utilized the heat from biogas for its own operations. According to data received by the WWTP, 63% of the biogas produced was sold to Gasum Oy while 31% was used to produce heat energy used by the biogas plant. These were average values over the whole studied period. The remaining amount of biogas was used with natural gas purchased for electricity generation on-site.

The total amount of heat energy purchased by the sites can be seen in Figure 45.

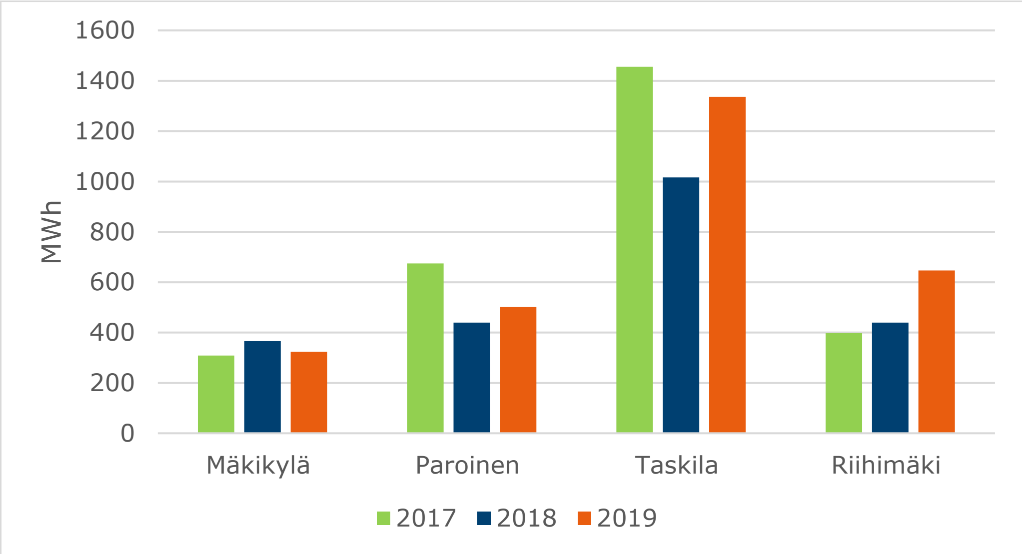


Figure 45 - Heat purchased by the studied WWTPs over the studied period

When normalized to PEs served, Taskila WWTP consumed the most heat per PE per year followed by Riihimäki, Paroinen and Mäkikylä WWTPs, as shown in Figure 46.

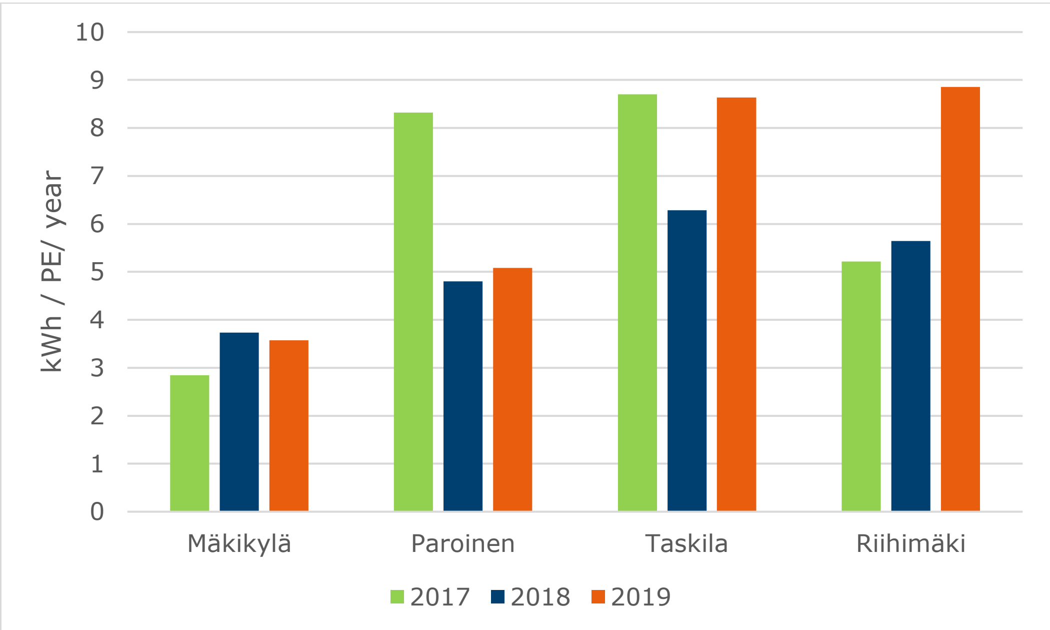


Figure 46 - Purchased heat per PE per year for the studied WWTPs

4.4 Comparison of transport emissions

Among the WWTPs, Taskila had the longest distance for chemical supply and the lowest for sludge treatment. For Riihimäki, Paroinen and Mäkikylä WWTPs, there were little difference in distances travelled for chemical supply. Mäkikylä WWTP had the longest average distance for treating sludge and screenings. The average distance covered to supply chemicals and transport sludge and screenings can be seen in Table 12.

Table 12 Average distances and mass transported for chemical supply, sludge and screenings treatment among the WWTPs

	Average distance for chemical supply (km)	Average distance for sludge and screenings treatment (km)	Average yearly mass transported (chemicals, sludge & screenings) (tons)
Paroinen	170	50	3932
Riihimäki	160	50	622
Taskila	400	14	36417
Mäkikylä	190	115	4317

In order to compare the transport emissions among the WWTPs, the total monthly transport emissions were normalized in terms of 1 kg substance transported. The average distances travelled were comparable between Paroinen, Riihimäki and Mäkikylä WWTP. Taskila however, had more than twice as much transport distance for chemicals than the other plants but the least for sludge treatment since the composting site was in Oulu. The results in Figure 47 show that Taskila had the lowest transport emissions per kg substance transported while Riihimäki had the highest on average.

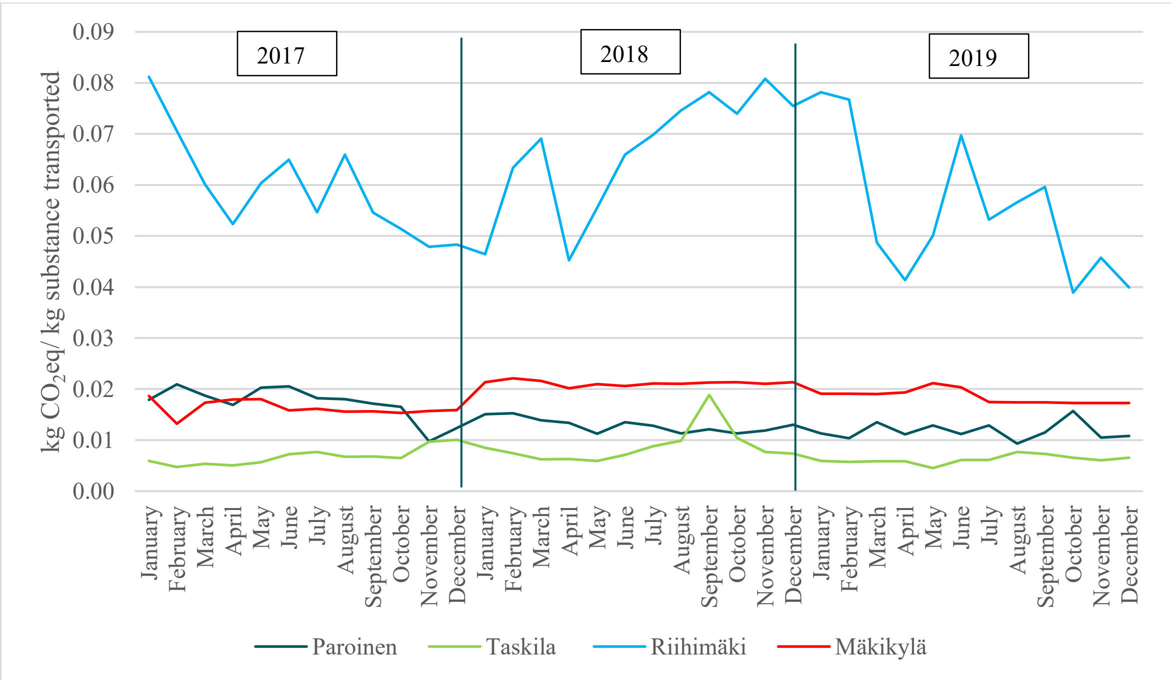


Figure 47 - Normalized transport emissions per kg substance transported, for all WWTPs.

4.5 Comparison of chemical consumption emissions

The total chemical emissions for each WWTP can be seen in Table 13. Taskila WWTP had the highest chemical consumption emissions, followed by Paroinen, Mäkikylä and Riihimäki WWTP.

Table 13 - Total chemical emissions for each WWTP

Year	Paroinen (ton CO2eq)	Taskila (ton CO2eq)	Riihimäki (ton CO2eq)	Mäkikylä (ton CO2eq)
2017	532,94	1504,31	38,88	473,76
2018	531,43	2057,79	39,49	421,27
2019	461,33	2016,11	33,89	443,04

The average shares of total chemical emissions for each chemical used by the WWTPs over the three-year period can be seen in Table 14. For the Taskila WWTP, polyaluminium chloride had majority share of chemical consumption emissions. For the Paroinen WWTP, the consumption of calcium carbonate dominated the chemical consumption emissions with ~56%. For Riihimäki WWTP, the emissions from chemical consumption were solely attributed to the polymer consumption. For the Mäkikylä WWTP, ferric sulphate constituted majority of the chemical consumption emissions.

Table 14 - Average shares of total chemical emissions for each chemical used by the WWTPs over the study period.

Chemical	Paroinen	Taskila	Riihimäki	Mäkikylä
PAC	29 %	32 %	N/A	15 %
Ferrous sulphate	0 %	N/A	0 %	N/A
Methanol	5 %	3 %	N/A	N/A
Polymers	10 %	10 %	100 %	7 %
Lime	N/A	1 %	N/A	9 %
Calcium carbonate	56 %	N/A	N/A	20 %
Ferric sulphate	N/A	13 %	N/A	44 %
Ferric chloride	N/A	N/A	N/A	4 %
Calcium hydroxide	N/A	11 %	N/A	N/A
Formic acid	N/A	4 %	N/A	N/A
hydrogen peroxide	N/A	6 %	N/A	N/A
Sulphuric acid	N/A	0 %	N/A	N/A
Sodium hydroxide	N/A	10 %	N/A	N/A
Peracetic acid	N/A	10 %	N/A	N/A
Citric acid	N/A	0 %	N/A	N/A
Sodium hypochlorite	N/A	0 %	N/A	N/A

N/A = not applicable to WWTP

4.6 Comparison of results for other functional units

In this section, the carbon footprint of the WWTPs in terms of their treatment efficiency will be shown. The results compared are the emissions per cubic meter of wastewater treated and emissions per kg total nitrogen (TP) and kg total phosphorous removed (TP).

4.6.1 Emissions per volume treated

The average emissions per cubic meter wastewater treated for each studied year can be seen in Figure 48. The emissions ranged from 0.6 kg – 0.8 kg CO_2eq / m^3 treated wastewater. Paroinen and Taskila WWTP had the highest emissions per cubic meter wastewater treated while Mäkikylä WWTP had the least. Taskila and Paroinen WWTPs had similar CFs in terms of volume treated, with their average emissions differing by only 0.01 kg CO_2eq / m^3.

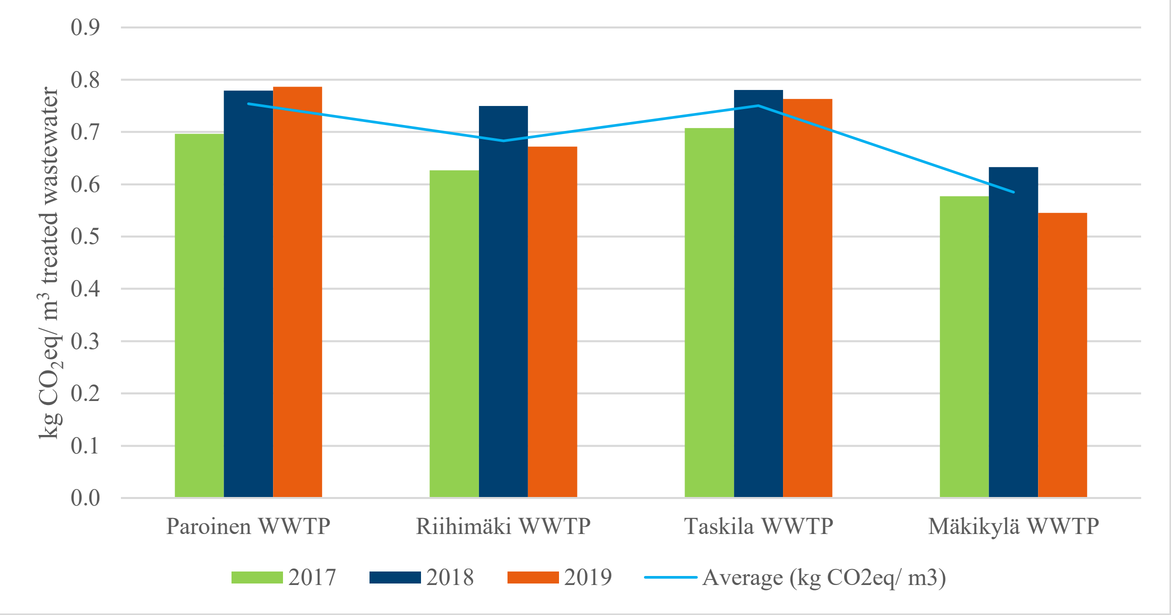


Figure 48 - Average CO_2 equivalent emissions per cubic meter wastewater treated for each WWTP

4.6.2 Total nitrogen and phosphorous removal

The carbon footprint represented in terms of nitrogen removed were compared among the studied WWTPs. The results for each WWTP can be seen in Figure 49. Riihimäki WWTP had the lowest emissions per kg TN removed while Taskila WWTP had the highest.

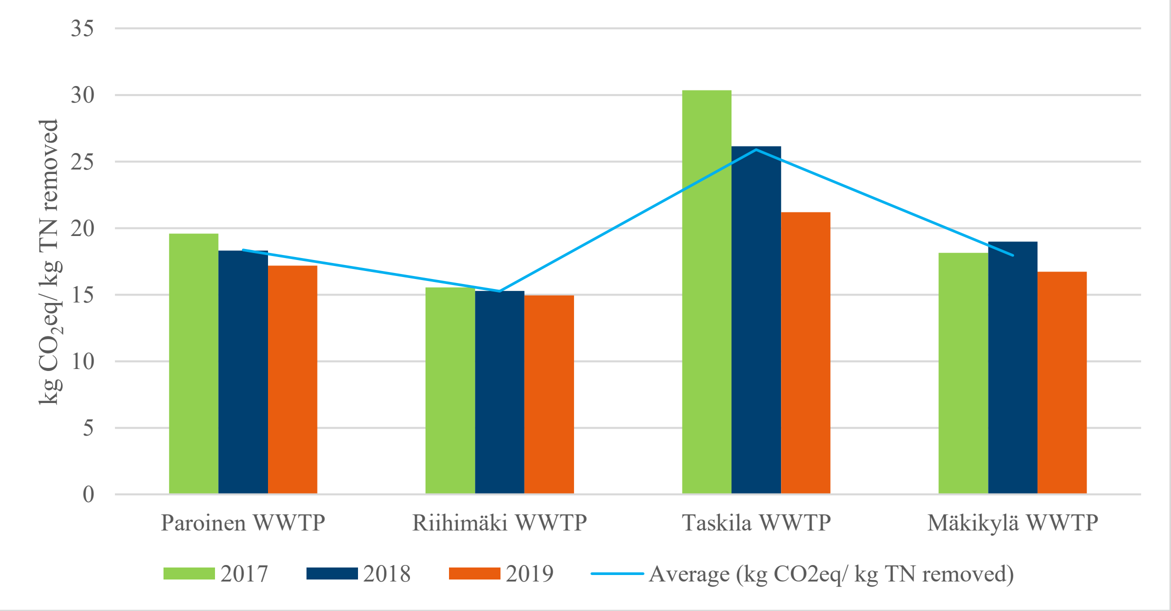


Figure 49 - average CO_2 equivalent emissions per kg TN removed over the studied period

CO_2 emissions per kg phosphorous removed ranged from 85 – 103 kg CO_2eq, as can be seen in Figure 50. Mäkikylä WWTP had the highest average, while Taskila WWTP had the lowest.

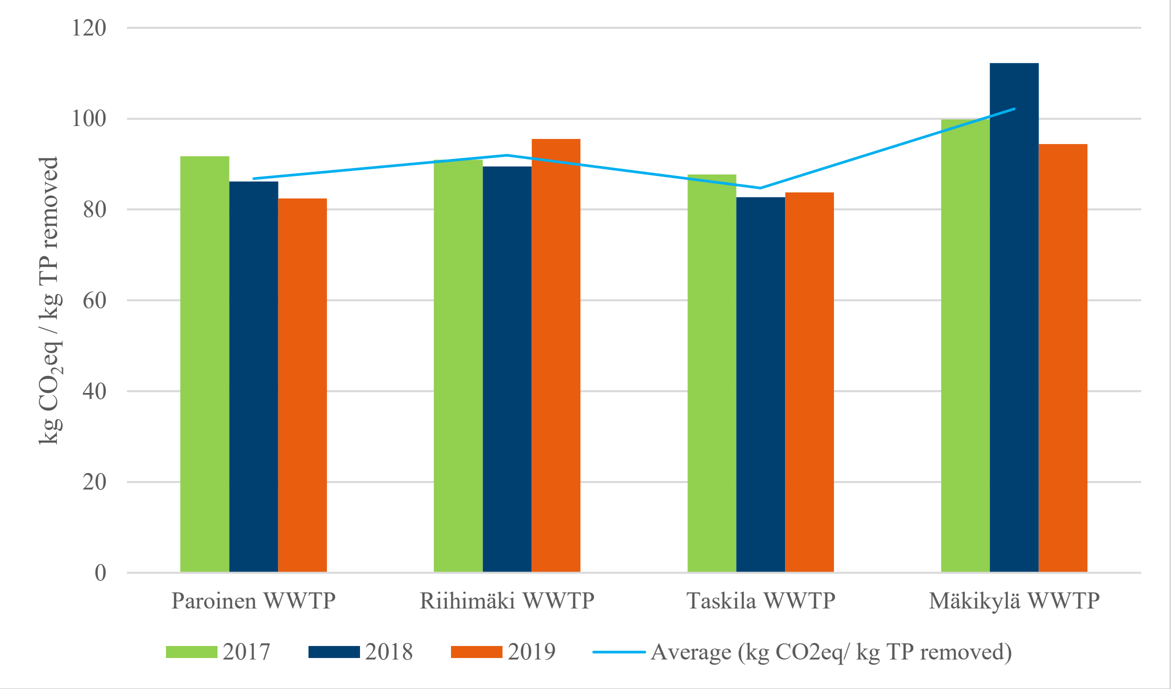


Figure 50 - kg CO_2eq / kg TP removed over the studied period for each WWTP

4.7 Sensitivity test for N_2O emission factors

Due to N_2O emissions from wastewater treatment contributing >50% of the total carbon footprint of the WWTPs, the impact of changing the N_2O EF on the total emissions was investigated. The EFs discussed in Chapter 3.7.5 (0.8% and 2.1% $TN_{influent}$) were used for the test. The percentage decrease and increase of the carbon footprint relative to the baseline EF (1.6% $TN_{influent}$) were then calculated. The results can be seen in Figure 51. Results showed minor differences between the sites. Taskila exhibited the most sensitivity to the EFs while Paroinen exhibited the least changes. Riihimäki had the second highest sensitivity and Mäkikylä being the third.

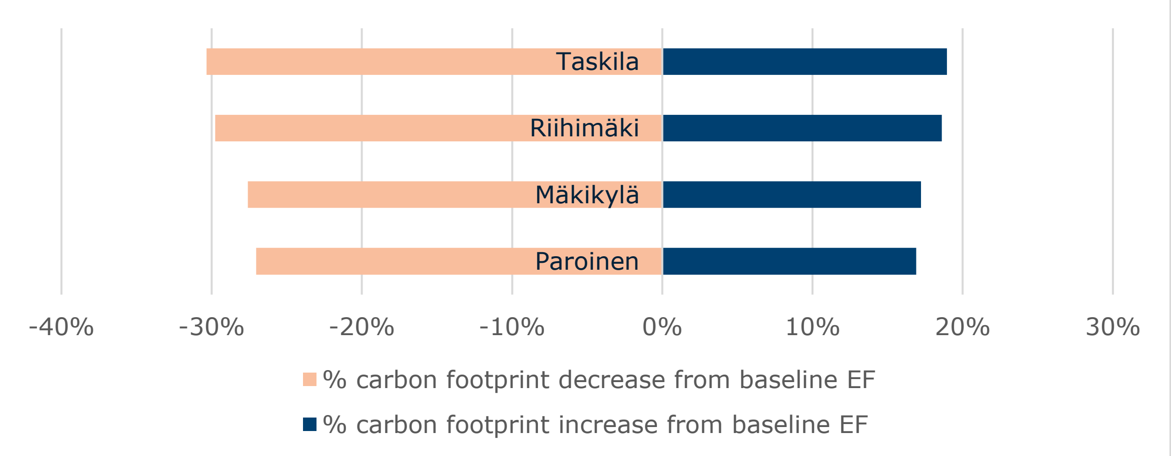


Figure 51 - Decrease and increase of total emissions compared to baseline N_2O EF

5 Discussion

This chapter will first discuss the overall results of the WWTPs before discussing each WWTP in the subsequent chapters.

The CF results of the WWTPs in this study fell within the range of studies done by Gustavsson & Tumlin (2013) and Maktabifard *et al.* (2019) (refer to Table 4). In relation to the volume of wastewater treated, all values also fell within the range given in the study by Maktabifard *et al.* (2019).

There was no strong dependence between the monthly direct or indirect emissions and the monthly carbon footprint of all sites in this study, based on the correlation results shown in Table 11. The reason for this could lie in the use of the PE as a functional unit. The three-month average PE values differed within and between the studied years for all WWTPs (refer to Figure 19). Thus, if there was a lower BOD_7 loading rate than the previous month, while total CO_2eq emissions remained the same, the resulting carbon footprint would be higher for that month due to lower PEs served. This is further supported by all sites showing moderate to strong negative correlations between monthly CFs and monthly PEs served (refer to Table 11). The moderate to strong negative correlation coefficients indicate high inverse proportionality between the monthly CF and monthly PEs served. Thus, the monthly CF showed a stronger dependence on the PEs served (and BOD_7 loading rates) than the monthly direct/indirect emissions.

All sites in this study attributed ~72% of the total GHG emissions to direct emissions. 59% was attributed to N_2O emissions from wastewater treatment, ~9% to CH_4 emissions (from sludge treatment, biogas production and combustion) and ~4% to N_2O emissions from composting. Due to the EF for N_2O emissions from wastewater treatment being dependent on the total nitrogen in the influent, majority of the direct emissions were dependent on the influent total nitrogen load. Direct emissions from screenings treatment were negligible for all WWTPs in this study, with an average of less than 1% of total GHG emissions. The emissions from screenings were dependent on the fossil carbon present. Since screenings comprise of random materials, an exact fossil carbon percentage was difficult to estimate. Therefore, an assumption of 20% fossil carbon had to be made, as was done in the study by Gustavsson & Tumlin (2013).

The average electricity usage per PE in this study was 44 kWh $PE^{-1}year^{-1}$. The study by Gustavsson & Tumlin (2013) resulted in an average of 54 kWh PE^{-1} $year^{-1}$. The energy intensive processes on-site were challenging to identify since the WWTPs only monitored few processes or just the total consumption of the WWTP. Based on the estimations given by the WWTPs, the most intensive processes were the activated sludge aeration and the pre-treatment phase.

Based on results in Chapter 4.4, WWTPs transporting higher masses benefitted from lower CO_2 emissions per kg substance transported. Thus, through “economies of scale”, WWTPs on a larger scale benefit from lower specific transport emissions with increasing masses of substance transported.

Among the chemicals used, coagulants contributed the most emissions for Taskila and Mäkikylä WWTPs, who used polyaluminium chloride and ferric sulphate. Since Riihimäki WWTP used ferrous sulphate (which has a zero EF value in this study), polymers accounted for all its chemical consumption emissions. The use of calcium carbonate accounted for more than half of Paroinen’s chemical consumption emissions due to the large amounts consumed by the plant (~1000 tons per year). Calcium carbonate was the second highest contributor for chemical consumption emissions after ferric sulphate, for the Mäkikylä WWTP.

5.1 Taskila WWTP

Taskila WWTP's carbon footprint during the studied period ranged from 75 - 86 kg CO_2eq PE^{-1} $year^{-1}$, which was the highest among the sites in this study.

Taskila WWTP had the lowest share for CH_4 emissions from sludge treatment among the WWTPs, with an average of ~6% of total CO_2eq emissions. Aside the fact that Taskila WWTP did not have surplus emissions from anaerobic digestion, a possible reason for the lower shares for CH_4 could be due to the fate of the waste sludge produced. According to data received from the site, not all the sludge was composted or further treated after the KemiCond process. For the years 2017 – 2018, ~60% of the waste sludge was spread on farmland following the KemiCond process. Since the disposal of sludge is excluded from the scope of this study, the emissions from the spread of sludge on farmland was excluded in this study. The lower CH_4 emissions, due to the boundary cut-off, explains the lower share of CH_4 emissions.

For indirect emissions, clear trends were seen for Taskila WWTP, whose peaks occurred in winter and spring months. Although district heating was consumed at a higher rate in the winter months, the peaks for the indirect emissions were also attributed to an increase in PAC and sodium hydroxide consumption which occurred at the same period.

Among the WWTPs, Taskila had the lowest share for energy consumption emissions. It also consumed the second least electricity per unit volume wastewater treated (refer to Figure 44). The site had a steady increase in consumption from June 2018, possibly attributed to the new MBR process installed which consumed additional electricity. Based on the energy consumption data received from the plant, the introduction of the MBR process led to an increase of ~17% of electricity consumed for the activated sludge process.

Taskila had the highest share for chemical manufacturing emissions among the WWTPs in this study. This is due to the plant consuming the most chemicals among the WWTPs, especially the additional chemicals used in the KemiCond process. PAC contributed most of the emissions with an average of ~32% of total chemical consumption emissions. This was due to PAC having the highest EF among the coagulants in this study.

Although the Taskila WWTP had the highest transport distances in this study, transport emissions still contributed a minor share of 2% of the total emissions, which was the highest average in this study. This is in line with the review in Chapter 2.4.4, whereby sites situated in northern Finland are expected to have longer transport distances due to more sparsely populated areas.

Due to the site's relatively lower nitrogen removal efficiency than the other WWTPs in this study (refer to Table 5), the emissions per total nitrogen removed were much higher (refer to Figure 51). However, it had the lowest emissions per total phosphorous removed among the WWTPs.

5.1.1 Recommendations for Taskila WWTP

Approximately 4% of Taskila WWTP's carbon footprint was associated with the KemiCond process (chemical consumption and transport emissions). The main benefits of the KemiCond process are to improve dewatering of sludge (reducing the amount of sludge), as well as hygienization of sludge and reduction of bad odor. The site should assess if the reduction of sludge transport emissions (due to higher dewatering from KemiCond process) outweighs the emissions from the consumption and transport of KemiCond process chemicals. If so, the site can continue utilizing the KemiCond process. However, if the emissions from the KemiCond process outweighs the transport emissions of sludge that doesn't undergo KemiCond, then the site can consider alternatives. An alternative could be replacing the KemiCond process with on/off-site anaerobic digestion of the waste sludge produced.

Depending on the fate of the biogas produced, the WWTP can benefit from the emission offsets from the biogas produced either on/off-site. Due to the large scale of the WWTP and the higher magnitude of waste sludge produced, the biogas potential of the waste sludge would be high, resulting in higher emission offsets. Furthermore, approximately 60% of the waste sludge was spread on farmland following the KemiCond process during 2017-2018 which could have been utilized for anaerobic digestion.

The use of polyaluminium chloride (PAC) alone constituted ~5% of the total carbon footprint of the site. The use of ferric sulphate accounted for just <1% of the total carbon footprint even though the site used ~500 tons more ferric sulphate than PAC. The site can therefore consider changing from PAC to other chemicals with lower emission factors. Based on the chemical EFs in Appendix 1, ferric sulphate and ferrous sulphate had the lowest EFs in this study and could be possible alternatives.

5.2 Riihimäki WWTP

Riihimäki WWTP had the lowest carbon footprint among the sites with an average of 45 kg CO_2eq PE^{-1} $year^{-1}$. This was close to the average value of 46 kg CO_2eq PE^{-1} $year^{-1}$ estimated by Gustavsson & Tumlin (2013) in their study. It was also close to the study done by Mölsä (2019) on the Viikinmäki WWTP in Finland, which resulted in 51 kg CO_2eq PE^{-1} $year^{-1}$ (41 kg CO_2eq for wastewater and 9.7 kg CO_2eq for sludge treatment). The reason for the site having the lowest footprint could be due to the dairy wastewater treated. The dairy wastewater increased the BOD_7 loading, resulting in an increase of PEs served. The ratio of TN to PEs was greatly reduced (refer to Figure 21). The lower TN to PE ratio resulted in lower N_2O emissions from wastewater treatment per PE served. Due to the high GWP of N_2O, the site benefited from a lower carbon footprint due to higher PEs served, relative to the TN loading to the site.

Another contributing factor for Riihimäki having the lowest carbon footprint is due to the electricity consumption EFs. Riihimäki had the lowest EF for electricity consumption among the WWTP for each of the studied years (see Appendix 2), which suggests lower fossil fuel in the energy mix. Additionally, it purchased the least amount of electricity among the WWTPs, as shown in Figure 42. This was expected considering it was the smallest WWTP in term of scale. Due to shares of chemicals and transportation being very low for Riihimäki, the energy consumption dominated the shares of indirect emissions. The low EF for electricity consumption, coupled with the high share of energy consumption, led to low indirect emissions for Riihimäki. Furthermore, the use of ferrous sulphate had zero emissions in this study and did not contribute to indirect emissions.

Riihimäki WWTP had the highest share for direct emissions among the sites in this study with an average of ~74% of total GHG emissions. N_2O emissions from wastewater constituted ~63% of the total emissions over the studied period. Since Riihimäki combusted natural gas on-site for heat energy, these were considered as direct emissions.

Natural gas combustion for heat accounted for ~4% of total GHG emissions. CH_4 from sludge treatment accounted for ~8% of total GHG emissions on-site. (See Appendix 4 for detailed results).

Riihimäki WWTP's indirect and direct emissions experienced the least variation among the WWTPs over the study period. For direct emissions, this was due to the TN loading to the plant having the least standard deviation, suggesting more stable loading rates. Since N_2O emissions were based on TN loading rates, it implied more stable N_2O emissions and least variations compared to the other WWTPs. Additionally, the monthly PEs served over the study period had the second least standard deviation. This suggests relatively more stable BOD_7 loading over the study period. The lower variation in indirect and direct emissions, coupled with the lower variation in PEs served, resulted in minimal changes in CF values over the study period, compared to the other WWTPs. This is shown in Table 10 where the maximum and minimum CFs differed by only ~0.4 kg CO_2eq / PE/ year. It should be noted that the plant could only provide yearly consumption values for heat energy consumed, but not monthly values. This yearly total was then divided equally among the months and peaks were therefore not possible to identify during winter. It is expected that the monthly CF would have fluctuated more, provided the winter consumption data was more accurate.

Riihimäki WWTP used ferrous sulphate as a coagulant during the study period. Since the chemical supplier (Kemira) assigned a zero-emission factor for ferrous sulphate (due to it being a by-product), it was assigned a zero-emission factor in this study as well (refer to Chapter 3.6.2). This significantly reduced the share for chemical consumption and total indirect emissions. However, based on scenarios where the EF for PAC was used in lieu of the ferrous sulphate EF, the site's final CF increased by ~2 kg CO_2eq/ PE/year and still had the lowest footprint among the sites. Thus, the consumption of ferrous sulphate did not have a significant effect on Riihimäki WWTP's CF, regardless whether or not an EF value was assigned to it. If an EF was assigned to ferrous sulphate, more variability in the indirect emissions would be noticeable, due to the varying amounts of monthly ferrous sulphate consumed by the plant.

In terms of nitrogen removal, Riihimäki WWTP also had the lowest CF with an average of 15.5 kg CO_2eq/ kg TN removed. This is attributed to the site having the highest nitrogen removal efficiency among the sites in this study with an average of 73% over the study period. A possible reason could be the incoming dairy wastewater from Valio Oy. The dairy wastewater contains a lot of organics which could promote the denitrification process. Also, this is evident in the site not consuming external carbon sources in its denitrification process.

5.2.1 Recommendation for Riihimäki WWTP

Riihimäki WWTP purchased natural gas which was combusted on-site to produce heat. The heating of the site accounted for ~4% of the total carbon footprint. This was the highest share for heat energy used on-site among the WWTPs in this study. This was due to the EF for heat production from natural gas being higher than the EFs for district heating among other WWTPs in this study (refer to Appendix 2). It is recommended that the site considers switching from natural gas to district heating, provided the district heat EF is less than that of the heat produced from natural gas.

5.3 Mäkikylä WWTP

Mäkikylä WWTP's monthly CF did not exhibit any seasonal trends although total direct and indirect emissions were lower in the summer, on average. This could be linked to the PEs served by the plant. The PEs served by Mäkikylä had the second highest variability over the studied period, based on the standard deviation test results in Table 9. The increased variability of PEs served resulted in an increase in CF variability. This is also reflected in Table 9, where Mäkikylä also has the second highest standard deviation for monthly CFs over the study period. With the increase in monthly CF variability, seasonal trends would be less visible.

Energy consumption emissions had the highest shares for indirect emissions. Mäkikylä purchased the second least electricity and consumed the least electricity per PE served, and per m^3 wastewater treated. Due to the WWTP owning a CHP unit, additional electricity was produced on-site from biogas combustion and this supplemented the purchased electricity. The peak formation of indirect emissions during the winter was as a result of increased heating consumed at the WWTP. Since the EF for electricity consumption was the highest for the Mäkikylä WWTP, it is suggested that the CF of the plant could be further reduced with a lower EF for electricity production.

Mäkikylä WWTP had the longest distance for sludge treatment after they stopped operating the biogas plant and had to transport the excess sludge off-site. The transport of sludge off-site increased the total share of transport emissions from 1% to 3% between 2018 and 2019, even though sludge was only transported for half of the year in 2019. This suggests the transport of sludge could have higher emissions if the sludge is transported the same distance all year-round.

Majority of direct emissions were dictated by N_2O from wastewater treatment, and directly related to the influent total nitrogen load of the WWTPs. Due to nitrogen loading being lower in the summer on average, the resulting direct emissions also displayed similar trends with lower emissions in the summer.

The biogas plant operated in conjunction with the Mäkikylä WWTP, co-digested biowaste from various municipal sources in addition to the waste sludge from the WWTP. The feedstock for the anaerobic digestion process comprised of ~70% waste sludge and ~30% biowaste. Thus, the WWTP benefitted from an increase in biogas production due to the additional feed of biowaste co-digested.

Based on Figure 41, the biogas plant sold more biogas during the winter & spring months than in the summer. However, the emission offsets in terms of PE did not exhibit peaks during the winter and spring. The emission offset peak that occurred between March and April of 2018 was mainly attributed to a drop of ~55000 PEs served. This resulted in a higher emission offset per PE served. This is also evident in the monthly CF, where there is a sudden spike between March and April 2018, although direct and indirect emissions did not rise as much. Since the PE served was based on BOD_7 loading rates, a possible explanation could be an analytical error was made during the BOD_7 analysis of the influent between March and April 2018.

5.3.1 Recommendations for Mäkikylä WWTP

Since Mäkikylä WWTP's EF for electricity production was the highest among the sites, it is suggested that the plant could change electricity suppliers to ones having lower EFs for electricity production. This will further reduce the shares for energy consumption for the plant.

The Mäkikylä WWTP began transporting sludge for outsourced anaerobic digestion from June 2019. Due to the outsourced facility located 200 km away, combined with the large amounts of sludge transported, the transportation emissions for sludge between June and December 2019 was ~2% of the overall carbon footprint for 2019. It is evident that the transportation emissions spiked after the biogas plant was sold to Gasum. It is therefore recommended that the Mäkikylä WWTP considers a closer facility to outsource the sludge treatment from in order to reduce the transportation emissions.

5.4 Paroinen WWTP

Paroinen WWTP's monthly CF did not exhibit any seasonal trends over the study period. The dynamic of the monthly CF was mostly influenced by the PEs served. This is further supported by Table 11, where Paroinen WWTP had the highest negative correlation coefficient between monthly CF and monthly PEs served. The site also had the lowest share for N_2O emissions from wastewater treatment, among the WWTPs in this study. This is also supported by Figure 51, where Paroinen WWTP had the least sensitivity to the N_2O emission factors. Additionally, the site had the lowest positive correlation coefficient between the monthly direct emissions and the monthly CF, which further suggests the least influence from direct emissions on the CF compared to the other WWTPs.

Energy consumption emissions contributed the most shares for indirect emissions. Paroinen had the highest shares for electricity consumption among the WWTPs. This is also evident in Figure 43, where Paroinen had the highest electricity consumption per PE wastewater treated. Conversely, district heating consumption had the lowest share of total emissions among the WWTPs. During the years 2018 and 2019, Paroinen sold more heat than was used in the plant (refer to Figure 36). This suggests that the plant had produced excess heat from biogas production and relied less on the grid for district heat. The peak offsets from the sale of district heat to the grid occurred during the summer, due to less heat required by the plant than the winter months.

The use of calcium carbonate contributed ~56% of total chemical consumption emissions due to the large amounts consumed (over 1100 tons per year). PAC followed with ~27% of chemical manufacturing emissions. A possible reason for the large amounts of calcium carbonate consumed could be due to the low alkalinity of the incoming influent wastewater.

Sludge that by-passed digestion in 2017 was composted on-site and only sent off-site from October to December 2017. From January 2017 to September 2017, there were no offsets from outsourced digestion since there was no sludge sent off-site. Since the emissions from composting were higher than anaerobic digestion emissions (refer to Figure 34), Paroinen would have benefited from the carbon offsets if the sludge was sent-off site for anaerobic digestion. However, depending on the boundaries considered, they would have also benefitted from selling the compost as fertilizer. Since the disposal aspect was not included in the boundary of this study, those offsets were not estimated.

5.4.1 Recommendations for Paroinen WWTP

Based on Figure 43, it is evident that Paroinen consumes the most electricity per PE treated. This could be due to the additional reject water aeration which might be the cause of the increased electricity consumed. The site can consider aerating the reject water in the main biological reactors. This is based on the assumption that the energy efficiency in the side stream reject water treatment might be lower than the biological reactors. Since there was no energy metering for the processes on-site, Paroinen WWTP can monitor the electricity consumption of the reject water aeration and can confirm/reject this assumption. Also, the site can consider supplementing electricity through the utilization of biogas in a CHP unit, use of solar panels or other forms of renewable energy on-site. Although this will constitute a small share of the total consumption, this would slightly reduce the electricity consumption from the grid and the associated emissions. Since Paroinen WWTP had the lowest EF for district heating among the sites, it can rely more on the grid for district heat and utilize more biogas for electricity production on-site.

The consumption of high amounts of calcium carbonate indicates low levels of alkalinity in the wastewater. Since denitrification produces alkalinity during the biological process, the site can consider enhancing the denitrification process (without methanol) to reduce the dependence on pH adjustment chemicals. The site can also consider replacing calcium carbonate with another chemical whose emission factor is lower.

5.5 Impact of N_2O EF on carbon footprint results

From Figure 51, the percentage changes in CFs among the WWTPs did not differ significantly when the N_2O EFs were changed. Taskila WWTP had the highest uncertainty and Paroinen WWTP had the least. Paroinen WWTP had the least share for N_2O emissions from wastewater treatment and had the least uncertainty.

The reason for Paroinen WWTP having the lowest sensitivity to the N_2O EF was due to the site having the largest shares for indirect emissions (~37% of total emissions). This reduced the overall share of direct emissions and resulted in less sensitivity. As discussed in Chapter 2.4.1, N_2O measurement studies on WWTPs having side stream process had higher EFs on average. Since the site treats the supernatant from sludge dewatering in a separate side stream process, there is a possibility that the sensitivity would be higher, based on literature findings.

Based on Figure 21, the nitrogen load normalized to PE indicates that Taskila WWTP has a higher concentration of nitrogen load per PE. Since the EF for N_2O was based on the nitrogen concentration of the influent, having a higher concentration would lead to more N_2O emissions per PE. This could explain the reason for Taskila WWTP having higher uncertainty than expected. By this logic, Riihimäki WWTP should have had the least sensitivity due to the TN loading per PE being the least. However, it had the second highest sensitivity. This is probably due to the site having the highest share for N_2O emissions from wastewater treatment (and direct emissions in general) caused by very low indirect emission shares. Hence, the site's CF was heavily dependent/sensitive to N_2O and direct emissions. This is further supported by Table 11, where Riihimäki WWTP had the highest positive correlation coefficient between monthly CFs and direct emissions.

The unit of the N_2O EF used could have significant impacts on the final carbon footprint result. Representing the N_2O EF in terms of influent nitrogen load does not account for the nitrification and denitrification processes of the WWTP. Since literature studies suggest nitrification as the main process responsible for N_2O emissions, representing the EF in terms of total nitrogen loading to the plant will not account for the nitrification process.

The EF expressed in terms of nitrogen denitrified would lead to higher emissions for sites having higher total nitrogen removal rates and lower emissions for sites with lower denitrification rates. If a site has high nitrification rate but low denitrification rates, the resulting N_2O emissions would be low even though nitrification is suggested to be the main pathway process.

If indeed the nitrification process is the major pathway for N_2O formation, an EF accounting for nitrification would be more suitable. N_2O measurement studies that expressed the EF to account for nitrification exist. An example is the study done by Bellandi *et al* (2018) which expressed the EF as N_2O emitted per unit NH_4^+ removed, to account for the nitrification process. However, at the time of this study most literature studies still express the EF in terms of influent nitrogen. Since more data was available for this EF, it was decided to base the N_2O EF in this study on nitrogen load of the influent.

Due to the complexity of N_2O formation and its highly process-dependent nature, the only accurate way for wastewater treatment plants to estimate their N_2O emissions is to conduct an online measurement campaign or through advanced emission modelling. Ideally, the monitoring campaign should last at least a year in order to capture any seasonal dynamic emissions at the plant.

6 Conclusion

The carbon footprint assessment was performed for four wastewater treatment plants in Finland. The goal of this study was to quantify the total emissions of the Riihimäki, Paroinen, Taskila and Mäkikylä WWTPs, identify the major GHG contributors, and provide recommendation for the WWTPs to reduce their CF. A brief guideline was also given to assist WWTPs in general, to assess their carbon footprint.

Carbon footprint results ranged from 45 – 82 kg CO_2eq PE^{-1} $year^{-1}$ with an average value of 63 kg CO_2eq PE^{-1} $year^{-1}$. Fugitive emissions constituted majority of the carbon footprint with an average of 72% of all CO_2eq emissions. Energy consumption was the second highest source of emissions with an average of 19% of all CO_2eq emissions. Chemical consumption and transport emissions had shares of 8% and 1% of total CO_2eq emissions respectively.

Results of the study indicated a heavy dependence of the carbon footprint on N_2O emissions from wastewater treatment which accounted for over 50% of total emissions for all WWTPs in this study. There were no seasonal trends for the CF although indirect emissions appeared to be higher during the winter periods, while direct emissions were lower on average during the summer periods. The lower direct emissions during summer was due to the dominance of N_2O emissions from wastewater treatment, which was based on the incoming total nitrogen loading to the WWTPs. Since the total nitrogen loading was lower during the summer, the resulting direct emissions were lower as well.

For chemical consumption, it was discovered that polyaluminium chloride (PAC) had the highest EF while ferrous sulphate had zero emissions in this study. Thus, switching to a coagulant with a lower EF such as ferrous sulphate could reduce the total emissions. It should be noted that the supplier of ferrous sulphate did not have any LCA results for ferrous sulphate production. Since the supplier converted their ferrous sulphate waste to a final product with minimal energy consumptions, it is expected that the EF for production would be slightly more than zero. The use of calcium carbonate was found to constitute major shares of Paroinen and Mäkikylä WWTPs. This suggests that pH adjustment chemicals can potentially be the most emission-intensive chemicals, depending on the level of alkalinity of the wastewater treated.

It was recommended for plants to consider changing energy suppliers to more eco-friendly ones with lower production EFs or supplement their consumption with energy generated on-site (for example through biogas). Additionally, the sites should consider utilizing more waste sludge for outsourced or on-site anaerobic digestion. This will either generate more energy that can be used on-site or result in higher offsets if the biogas is replacing fossil fuels elsewhere. Thirdly, the use of district heating rather than natural gas combustion on-site was suggested, due to the average EF for district heating in Finland being lower than the EF for natural gas combustion.

For the boundaries selected in this study, it was discovered that the treatment and transport of screenings constituted less than 1% of all emissions on average, for all WWTPs. Also, the CH_4 emissions from sludge storage in this study were negligible for all WWTP, constituting less than 1% of all emissions on site. Thus, for simplification, these emissions could be excluded from the boundaries in other future studies. Although transport emissions were very low for all WWTPs, these are dependent on transportation distances and quantities transported and could vary based on the location and scale of the WWTP. Transportation emissions should therefore be included in the carbon footprint boundary for WWTPs.

Limitations in this study were attributed to insufficient data regarding fugitive emissions on-site. Fugitive emissions of N_2O and CH_4 dominated the total emissions and these emissions were calculated based on emission factors. The EFs for N_2O (from wastewater treatment) were further based on the nitrogen loading to the WWTPs, which assumes a complete nitrification process. Complete nitrification might not occur in all WWTPs. Due to the site-specific nature of N_2O emissions from wastewater treatment, the use of a single EF from literature would not be as accurate in representing the actual process conditions at the site. The second limitation was regarding the process-specific energy consumptions on-site. It was discovered that most of the WWTPs only monitored the whole consumption of the WWTP, or just a specific process (for example aeration) while the consumption of other processes such as pumping, or sludge dewatering were unknown. These were estimated by the plant operators and this made it difficult in accurately assessing the most energy intensive operations on-site.

Since the Finnish energy sector's emissions are declining gradually over time, it is anticipated that the energy consumption emissions will constitute minor shares of the carbon footprint in Finnish WWTPs in the future. However, it is uncertain how the introduction of innovative treatment processes for emerging micropollutants or resource recovery could affect the energy intensity of the wastewater treatment processes in the future.

With the anticipated reduction in energy consumption emissions, the shares of total GHG emissions in WWTPs attributed to direct emissions are predicted to increase in the future. The WWTPs would therefore be able to assess majority of their carbon footprint by simply quantifying these fugitive emissions. Due to the high plant-specific nature of direct emissions (especially N_2O emissions), advanced modelling or on-site measurements of these emissions would be needed for more accurate CF assessment. Since fugitive emissions are linked to process conditions, the dynamics of these emissions might change depending on the season and process conditions. Thus, it is important to capture the seasonal dynamics of the fugitive emissions by implementing measurement campaigns of a minimum period of one year. The data obtained from these measurement campaigns, coupled with treatment process data, can then be used to calibrate any models used for quantifying these emissions.

A further research proposal could include a life cycle cost analysis of WWTPs which could subsequently be combined with LCA results, such as carbon footprint analysis. The operational cost and carbon footprint of different treatment process/chemical alternatives could be assessed. For example, an alternative chemical might be cheaper and have a lower EF but could also have a lower treatment efficiency. Thus, multi-objective optimization can be done in the WWTP. This will ensure a more sustainable management of the WWTPs operations.

Each WWTPs in this study had unique treatment processes. The Paroinen WWTP included a reject water treatment unit, the Riihimäki WWTP had a dairy wastewater pre-aeration unit, while the Taskila WWTP included an MBR and KemiCond process. For sludge treatment, the Mäkikylä WWTP co-digested waste sludge with biowaste. The MBR and KemiCond process used in the Taskila WWTP are not mainstream wastewater and sludge treatment processes in Finland at the time of this study. For the Mäkikylä WWTP, anaerobic co-digestion of biowaste and sewage sludge resulted in high biogas production and high emission offsets. A typical Finnish WWTP of similar scale utilizing only sewage sludge as digester feed may not achieve similar offsets. The Riihimäki WWTP did not consume external carbon due to it having the highest BOD_7 to TN ratio levels and possibly higher process temperature due to the dairy wastewater. This ratio was much higher than the other WWTPs and probably not as common in a typical Finnish WWTP. Thus, in terms of representativeness of Finnish WWTPs, more nation-wide CF assessment data for WWTPs would need to be compiled and benchmarked. However, some of the processes mentioned for the case study plants, such as the MBR process, could become mainstream for WWTPs in the future.

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List of Appendices

Appendix 1: General EFs used among the WWTPs in this study

		value	unit	Source
Chemical Manufacturing	Polyaluminium chloride (PAC)	455	g CO_2eq/ kg	Kettunen (2020)
	Lime	300	g CO_2eq/ kg	(EEA, 2009)
	Polymer (Superfloc A-130)	3290	g CO_2eq/ kg	Kettunen (2020)
	Polymer (Superfloc C-492 HMW)	4200	g CO_2eq/ kg	Kettunen (2020)
	Polymer (Superfloc C-494 HMW)	4150	g CO_2eq/ kg	Kettunen (2020)
	Polymer (Superfloc C-496 HMW)	4150	g CO_2eq/ kg	Kettunen (2020)
	Ferric sulphate	150	g CO_2eq/ kg	Kettunen (2020)
	Ferric chloride	395	g CO_2eq/ kg	INCOPA (2014)
	Calcium carbonate	269	g CO_2eq/ kg	(Sustainable Solutions Corporation, 2016)
	Sodium carbonate	1840	g CO_2eq/ kg	(Prashantsinh *et al.* 2015)
	Methanol	462	g CO_2eq/ kg	Kajaste *et al.* 2018
	Formic acid	2490	g CO_2eq/ kg	Kettunen (2020)
	Hydrogen peroxide	491	g CO_2eq/ kg	Kettunen (2020)
	Sulphuric acid	4	g CO_2eq/ kg	EPA (1995)
	Sodium hydroxide	244	g CO_2eq/ kg	Kettunen (2020)
	Ferrous sulphate	0	g CO_2eq/ kg	Kettunen (2020)
Transport emissions	40 ton capacity trucks	55	g CO_2eq/ ton.km	lipasto, VTT
	40 ton capacity trucks (empty load)	1234	g CO_2eq/km	lipasto, VTT
	6 ton capacity trucks	110	g CO_2eq/ ton.km	lipasto, VTT
	6 ton capacity trucks (empty load)	292	g CO_2eq/km	lipasto, VTT
	Freight ships	8.1	g CO_2eq/ ton.km	lipasto, VTT
	Freight trains	21	g CO_2eq/ ton.km	(ECA, 2016)
Direct emissions	Composting of sludge	10	g CH_4 kg DS^{-1}	IPCC, (2006)
	Screenings treatment	0.473	kg CO_2eq/ kg TS	Gustavsson & Tumlin (2013)
	Sludge storage	8.9	g CH4-C.ton DM^{-1} day^{-1}	Willén *et al* (2016)
	Leakages from anaerobic digestion	0.4%	produced biogas	Tauber *et al* (2019)
	Methane slip from combustion units	1 %	of CH_4 combusted	Schaum *et al*. (2015)
	biogas potential	308.46	Nm^3 biogas. t-VS^{-1}	Kiselev *et al* 2019

	denisty of methane at standard temperature and pressure	0.72	$kg.Nm^{-3}$	Suomen kaasuyhdistys, 2016
	% of methane in biogas	60 %	of produced biogas	Kiselev *et al* 2019
Direct emissions	Composting of sludge	0.6	$g\ N_2O.\ kg\ DS^{-1}$	IPCC (2006)
	N2O emissions from wastewater treatment	1.6%	$TN_{influent}$ emitted as N_2O -N	IPCC (2019)
EFs used for emission savings calculation	Well-to-tank production of CNG	28.8	g CO_2e/kWh	(Hagos and Ahlgren, 2018)
	Emissions from natural gas vehicles (passenger vehicles)	60.4	g CO_2e /Km	(Hagos and Ahlgren, 2018)
	Energy consumption per km in natural gas vehicles	0.49	kWh/km	(Research Institute of Finland Ltd, 2017)
	Emissions of natural gas vehicles per kWh	123.27	g CO_2e/ kWh	Calculated based on the 2 EFs above
	Average emissions of electricity production in Finland	264.04[a] 289.67[b] 276.86[c]	g CO_2e/ kWh	(Finnish Energy, 2020)
	Average emissions of district heating in Finland	147.1[a] 148.14[b] 130[c]	g CO_2e/ kWh	(Finnish Energy, 2020)

a= 2017, b= 2018, c=2019

Appendix 2: EFs for electricity production given by WWTP energy suppliers

Year	Mäkikylä ($g\ CO_2eq/kWh$)	Paroinen ($g\ CO_2eq/kWh$)	Taskila ($g\ CO_2eq/kWh$)	Riihimäki ($g\ CO_2eq/kWh$)
2017	264	248,5	249	198
2018	289,7	269,2	276	226
2019	276.9	258,9	262,5	198

Appendix 3 EFs for district heat production for WWTPs

Year	Mäkikylä (g CO_2eq/kWh)	Paroinen (g CO_2eq/kWh)	Taskila (g CO_2eq/kWh)	Riihimäki (g CO_2eq/kWh)
2017	248[b]	116,5	227	248[b]
2018	248[b]	123,2	219	248[b]
2019	248[b]	125	218	248[b]

b = emission factor for natural gas used in heating (Gode et. al, 2012)

Appendix 4: Source of emissions in Riihimäki WWTP and their respective shares

Source	2017 Share	2018 Share	2019 Share
Nitrous oxide emissions from wastewater treatment	61,5 %	57,3 %	60,2 %
Purchased electricity	18,6 %	22,1 %	18,4 %
CH4 from composting	7,1 %	7,3 %	7,0 %
N2O from composting	4,0 %	4,2 %	4,0 %
natural gas used for heating	3,0 %	3,2 %	4,9 %
emissions from screenings treatment	1,1 %	1,2 %	1,1 %
Polymers used	1,2 %	1,1 %	1,0 %
Transport of Ferrous sulphate	0,2 %	0,2 %	0,2 %
Transport of polymers	0,0 %	0,0 %	0,0 %
Transport of sludge	0,9 %	0,9 %	0,7 %
Transport of screenings	0,0 %	0,0 %	0,0 %
CH4 slip in combusiton units	0,9 %	1,0 %	0,9 %
CH4 leakages from digestion	0,4 %	0,4 %	0,4 %
CH4 from digestion storage	0,1 %	0,1 %	0,1 %
Ferrous sulphate used	0,0 %	0,0 %	0,0 %
emissions from co-composting organics	1,0 %	1,0 %	1,0 %

Appendix 5: Source of emissions in Taskila WWTP and their respective shares

Source	2017 share	2018 share	2019 share
N2O from wastewater treatment	58,9 %	57,7 %	59,4 %
Emissions from electricity consumption	12,9 %	14,1 %	11,5 %
CH4 from composting	6,4 %	5,2 %	6,1 %
Emissions from use of Sodium hydroxide	1,3 %	1,4 %	1,4 %
N2O from composting	3,6 %	2,9 %	3,5 %
Emissions from use of PAC	3,6 %	6,7 %	3,7 %
Emissions from use of citric acid	0,0 %	0,0 %	0,1 %
Emissions from use of sodium hypochlorite	0,0 %	0,0 %	0,2 %
Emissions from screenings treatment	0,7 %	0,4 %	0,4 %
Emissions from district heating	2,7 %	1,7 %	2,1 %
Emissions from use of ferric sulphate	2,2 %	2,0 %	1,0 %
Transport of raskas sooda	0,0 %	0,0 %	0,1 %
Transport of lime	0,0 %	0,0 %	0,0 %
Transport of PAC	0,4 %	0,8 %	0,4 %
Transport of Superfloc A-130 (Polymer)	0,0 %	0,0 %	0,0 %
Transport of Superfloc C-494HMW (Polymer)	0,1 %	0,1 %	0,1 %
Transport of hydrogen peroxide	0,0 %	0,0 %	0,0 %
Transport of ferric sulphate	0,8 %	0,7 %	0,4 %
Transport of methanol	0,0 %	0,0 %	0,1 %
Transport of DEX-A375 (Formic acid)	0,0 %	0,0 %	0,0 %
Transport of DEX-550 (hydrogen peroxide)	0,0 %	0,0 %	0,0 %
Transport of sulphuric acid	0,0 %	0,0 %	0,0 %
Transport of caustic soda (for KemiCond process)	0,2 %	0,2 %	0,2 %
Transport of screenings	0,0 %	0,0 %	0,0 %
Transport of citric acid	0,0 %	0,0 %	0,0 %
Transport of sodium hypochlorite	0,0 %	0,0 %	0,0 %
Emissions from use of polymers	1,5 %	1,1 %	1,7 %
Emissions from the use of Formic acid	0,7 %	0,5 %	0,5 %
Emissions from co-composting organics	0,6 %	0,5 %	0,5 %
Transport of sludge	0,4 %	0,4 %	0,4 %
Emissions from use of Methanol	0,4 %	0,4 %	0,5 %
Emissions from use of Lime	0,3 %	0,0 %	0,0 %
Emissions from use of Sulphuric acid	0,0 %	0,0 %	0,0 %
Emissions from the use of Hydrogen peroxide	0,7 %	0,6 %	1,0 %
Emissions from peracetic acid use	1,4 %	1,3 %	1,3 %
CH4 from sludge storage	0,0 %	0,0 %	0,0 %
Emissions from use of sodium carbonate	0,0 %	1,4 %	3,4 %

Appendix 6: Source of emissions in Mäkikylä WWTP and their respective shares

Source	2017 share	2018 share	2019 share
Nitrous oxide emissions from wastewater treatment	55,2 %	52,3 %	59,2 %
Purchased electricity for WWTP	11,5 %	13,3 %	14,3 %
Methane emissions from composting	7,3 %	7,8 %	4,0 %
N2O emissions from composting	4,1 %	4,4 %	2,3 %
Emissions from Lime consumption	0,1 %	2,0 %	0,0 %
Emissions from $CaCO_3$ consumption	3,0 %	0,0 %	1,9 %
Purchased electricity for Biogas plant	2,9 %	4,1 %	1,8 %
Natural gas for heating (WWTP)	1,3 %	1,7 %	1,7 %
Emissions from natural gas supplementing biogas for electricity production	2,9 %	1,6 %	0,9 %
Emissions from natural gas supplementing biogas for heating (biogas plant)	1,6 %	0,9 %	0,5 %
Emissions from ferric chloride consumption	3,2 %	3,7 %	4,6 %
Emissions from PAC consumption	0,9 %	1,3 %	1,5 %
Emissions from polymer consumption	0,5 %	0,5 %	0,6 %
Transport of chemicals	0,6 %	0,8 %	0,8 %
Transport of sludge	0,0 %	0,0 %	2,2 %
Transport of screenings	0,0 %	0,0 %	0,0 %
Methane slip in combustion units	1,5 %	1,6 %	0,6 %
Methane leakage from digestion process	1,6 %	1,8 %	1,0 %
CH4 from digestate storage	0,0 %	0,0 %	0,0 %
Emissions from screenings treatment	0,8 %	1,2 %	1,4 %
Emissions from co-composting organics	1,0 %	1,1 %	0,6 %

Appendix 7: Source of emissions in Paroinen WWTP and their respective shares

Source	2017 share	2018 share	2019 share
N_2O from wastewater treatment	51,9 %	53,9 %	57,6 %
emissions from purchased electricity	22,6 %	23,3 %	22,3 %
CH_4 from composting	6,9 %	5,9 %	5,2 %
Emissions from $CaCO_3$ consumption	5,0 %	5,4 %	4,0 %
N_2O from composting	3,9 %	3,3 %	3,0 %
emissions from purchased district heat	1,3 %	0,9 %	1,0 %
emissions from ferrous sulphate use	0,0 %	0,0 %	0,0 %
emissions from methanol use	0,6 %	0,5 %	0,2 %
emissions from polymer use	1,0 %	0,7 %	0,8 %
emissions from PAC use	2,5 %	2,3 %	2,6 %
emissions from ferrous sulphate transport	0,3 %	0,3 %	0,3 %
emissions from methanol transport	0,2 %	0,1 %	0,1 %
emissions from lime transport	0,3 %	0,3 %	0,2 %
emissions from PAC transport	0,1 %	0,0 %	0,1 %
emissions from polymer transport	0,0 %	0,0 %	0,0 %
emissions from sludge transport	0,1 %	0,2 %	0,2 %
Transport of screenings	0,0 %	0,0 %	0,0 %
CH_4 slip from combustion units	0,8 %	0,7 %	0,6 %
CH_4 leakage from anaerobic digestion	0,3 %	0,3 %	0,2 %
CH_4 emissions from sludge storage	0,0 %	0,0 %	0,0 %
CH_4 from outsourced sludge treatment (digestion)	0,1 %	0,3 %	0,3 %
Emissions from treatment of screenings	0,8 %	0,6 %	0,4 %
emissions from co-composting organics used	1,3 %	1,1 %	0,9 %