Validation of an evidence-based methodology to support regional assessment and decarbonisation of wastewater treatment service in Italy

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Abstract

In this paper, a new regional methodological approach for determining direct and indirect emissions from wastewater treatment plants (WWTPs) is proposed. Additionally, an entire territorial wastewater treatment service located in the northern Italy and serving 411484 PE was assessed. The most accurate emission factor identification is presented using appropriate on-site measurements, monitoring different aerated operational units, and sampling several streams in 12 relevant WWTPs of different treatment capacities, ranging from 3000 to 73000 PE. Dissolved greenhouse gas (GHG) concentrations from 0.2 to 24 mgN_2O/L, 0.1 to 1 mgCH_4/L, and 1.8 to 52 mgCO_2/L in effluent flows were detected. Specific carbon footprints resulted in the emissions of 0.04–0.20 tonCO_{2eq}/PE/y, varying as per the size of the plant. The most impactful categories were identified for indirect emissions associated with dissolved GHGs discharged in the surface water body and due to energy consumption, which accounted for 13–70% and 10–40%, respectively. The overall territorial carbon footprint of the wastewater treatment service was also quantified to provide evidence-based decision support system (DSS) and prepare systemic mitigation strategies.

Keywords: carbon footprint, greenhouse gas, wastewater, emission factor, decarbonisation

Nomenclature

AD: Activity Data

ASMN: Activated Sludge Models for Nitrogen

BSM2-e: Benchmark Simulation Model No 2 Emission

BSM2G: Benchmark Simulation Model No 2 Greenhouse Gas

C: Carbon

CF: Carbon Footprint

CFCT: Carbon Footprint Calculation Tool

CF-TOOL CTRL: Carbon Footprint Control

CHEApet: Carbon Heat Energy Assessment Plant Evaluation Tool

CO_{2eq}: Equivalent CO_2

COD: Chemical Oxygen Demand

COD_{eff}: Effluent Chemical Oxygen Demand

COD_{in}: Influent Chemical Oxygen Demand

COD_{rem}: Removed Chemical Oxygen Demand

DEEM: Diffusive Emissions Estimation Model

DSS: Decision Support System

ECAM: Energy Performance and Carbon Emissions Assessment and Monitoring

GHG: Greenhouse gas

GWP: Global Warming Potential

IA: Intermittent Aeration

K: Potassium

MLE: Modified Ludzack−Ettinger

PE: Population Equivalent

SCENA: Short Cut Enhanced Nutrients Abatement

SCF: Specific Carbon Footprint

EF: Emission Factor

TN: Total Nitrogen

TN_{eff}: Effluent Total Nitrogen

TN_{rem}: Removed Total Nitrogen

TP: Total Phosphorus

TS: Total Solids

TSS: Total Suspended Solids

WESTWeb: Water Energy Sustainability Tool

WWEECarb: Water and Waste Environmental Engineering Carbon Footprint

WWTP: WasteWater Treatment Plant

1. Introduction

Several regions are working towards low carbon and circular economy (Low-carbon economy - Regional Policy, 2014) and actions to decarbonise urban water management can have a relevant impact (Climatesmartwater.org, 2018) especially when territorial pathways are developed.

In recent years, environmental legislations such as the Urban Wastewater Treatment Directive (UWWTD) (DIRECTIVE 91/271/EC), currently under revision, introduced more stringent quality standards for the effluents of the wastewater treatment plants (WWTPs) that led to increase in energy consumption and greenhouse gas (GHG) emissions (Gu et al., 2016). In many European countries, urban water cycle accounts for 1–3% of the total electric energy consumption (Longo et al., 2016) and 3–10% of the global warming potential (GWP) by contributing towards GHG emissions into the atmosphere, both as direct and indirect footprints (Samuelsson et al., 2018). As per the U.S. Environmental Protection Agency (EPA), the emissions of CO_2, CH_4, and N_2O constitute the largest part of CF of WWTPs, and global GHG emissions of the wastewater sector are predicted to increase by up to 27% by 2030 (Caniani et al., 2019; Huang et al., 2020).

In this context, several national and international initiatives and activities have been started to support the transition to low-carbon regions and urban water utility (WacClim, 2020; Crippa et al., 2019). In Italy, the National Regulatory Authority for Energy, Networks, and Environment (ARERA) introduced in 2017 the 'Carbon Footprint of the wastewater treatment service' as one of the key performance indicators to analyse and assess the technical quality standard of the activities carried out by water utilities. On one hand this regulatory driver can make a substantial shift towards the low-carbon water utilities, on the other hand the Authority could only guide the operators by indicating the adoption of the general ISO 14064-1:2019. In fact, while for energy audits, a standardised European methodology was developed in the H2020 project ENERWATER to assess the energy footprint of WWTPs (Longo et al., 2019), for CF, same standard approach is missing. Today several tools and software have been developed in recent years to quantify CF although they do not follow any specific normalised or standard methodologies (Mannina et al., 2016). Moreover, one of the most critical aspects is the appropriate identification of representative and validated emission factors (EFs). In fact, some referenced databases were developed to collect and update the EFs, mainly at the international or national level, such as emission factor database (EFDB) by Intergovernmental Panel on Climate Change (IPCC). Nevertheless, the wastewater sector is typically characterised by wide local variations in influent characteristics and different process parameters or operative conditions (Vasilaki et al., 2019). Therefore, site-specific EFs need to be considered (Parravicini et al., 2016) through long-term sampling campaigns, especially for evaluating direct emissions from the main processes and indirect dissolved GHGs present in the final effluents.

This paper goes beyong the current state of the art because a new normalised methodological approach according to the guidelines of ISO 14064 (WWEECarb) is proposed for the determination both of direct and indirect emissions in WWTPs and of the overall CF of regional wastewater service carried out by a water utility. Most of the considered emissions factors, were validated by site-specific measurements campaigns and emissions categories included both fossil and biogenic origin of the main GHGs. This validated approach can contribute to the standardization of the methodology for carbon footprint assessment in wastewater treatment service and to identify mitigation actions and priorities for regional decarbonisation.

This methodology was applied to territorial wastewater service (52 municipalities and 1376 square kilometres) located in the northern Italy (411484 PE) managed by a single public-owned water utility. The most accurate EF identification is presented by using the real operational data and appropriate measurements of emitted and dissolved GHGs from different operational units of 12 WWTPs.

2. Materials and methods

2.1 General Guideline ISO 14064-1

ISO 14064-1 standard provides '*specifications with guidance at the organization level for quantification and reporting of greenhouse gas emissions and removals*'. Several aspects and steps should be followed to apply the ISO standard at organisation level: 1) reporting boundaries should be defined; 2) direct and indirect emissions must be aggregated into inventory categories, including direct emissions and removals, and indirect emissions from imported energy, transportation, products used by the organisation, associated with the use of products from the organisation, and other sources; and 3) different GHGs (i.e. CO_2, CH_4, N_2O, NF_3, and SF_6) associated with the type of organisation have to be separately quantified, and 4) anthropogenic biogenic, non-anthropogenic biogenic, or fossil origin of CO_2 should be distinguished. Moreover, CF determination should be annually referred, and the result should be reported in tonnes of CO_{2eq} using appropriate GWPs (IPCC, 2019). Finally, based on the general principles of consistency and accuracy, ISO 14064-1 requires the definition of quantitative and/or qualitative uncertainty associated with the method used for the quantification of the emissions.

2.2 Description of the territorial wastewater service

The proposed approach was applied to determine the CF of wastewater services in the region managed by the Italian water utility Alto Trevigiano Servizi, consisting of 52 municipalities in the province of Treviso (Italy), with a total served population of 411484 PE. The regional wastewater treatment service is operated by 5 WWTPs with a design treatment capacity of more than 40000 PE, by 3 WWTPs with the capacities range of 15000–40000 PE, 27 WWTPs with less than 15000 PE capacity, and 28 septic tanks. The representative WWTPs to be monitored over the long-term, were selected by considering both the treatment capacity of the WWTPs and the representative characteristics. Therefore, site-specific EF measurements were carried out in 12 WWTPs (8 WWTPs with capacities higher than 15000 PE and 4 smaller WWTPs), covering approximately 90% of the total served population. The characteristics of these WWTPs in terms of process configuration, influent loads and removals efficiencies are summarized in Table 1. The selected WWTPs included both conventional activated sludge in the Modified Ludzack−Ettinger (MLE) configuration (n = 10) and intermittent aeration (IA) processes (n = 2). The innovative technology of Short Cut Enhanced Nutrients Abatement (SCENA) (n = 1) (https://www.smart-plant.eu/) for the nitrite treatment of nutrient-rich anaerobic rejected liquor from the sludge line was studied. All the aerated units of different water and sludge lines (aerated degritting unit, biological reactor, aerobic stabilisation, via-nitrite supernatant treatment, and biofilter) were monitored. The operation data were collected for all the plants over one whole year. The CF results of the monitored WWTPs were reported as the entire data (tonCO_{2eq}/y) and specific coefficients (SCF, $tonCO_{2eq}/PE/y$). SCFs were used to quantify the GHG impacts of the unselected WWTPs.

Septic tank emissions were also calculated according to the methodology mentioned in IPCC (2019), considering proposed EFs for both direct and indirect contributions of the dissolved GHGs in the effluent.

Table 1. Selected WWTPs for on-site measurements

	Design capacity	Measured units	Biological treatment	COD influent	TN influent	TP influent	COD removal	TN removal	TP removal
	PE	-	-	tonCOD/y	tonTN/y	tonTP/y	%	%	%
WWTP1	73000	Biological reactor	MLE	1919	198	30	93%	84%	90%
WWTP2	40000	Biological reactor	MLE	1394	69	21.6	92%	84%	96%
WWTP3	40000	Biological reactor, Via-nitrite supernatant treatment, Biofilter	MLE, via-nitrite supernatant treatment	2037	165	24	90%	70%	49%
WWTP4	70000	Biological reactor, Biofilter	IA	1876	137	21.7	88%	71%	70%
WWTP5	32000	Biological reactor	MLE	1253	103	21.1	93%	88%	79%
WWTP6	45000	Biological reactor, Aerobic stabilisation	MLE	432	49	5.8	87%	85%	82%
WWTP7	9500	Biological reactor, Biofilter	MLE	273	34	3.9	93%	85%	72%
WWTP8	18000	Biological reactor, Aerated degritting unit	MLE	318	44	4.9	86%	88%	58%
WWTP9	22000	Biological reactor, Aerated degritting unit	MLE	444	40	7.8	90%	80%	60%
WWTP10	3000	Biological reactor,	MLE	58	8.6	1.1	87%	88%	43%
WWTP11	4500	Biological reactor,	MLE	93	11.7	1.1	93%	87%	60%
WWTP12	10000	Biological reactor, Aerated degritting unit	IA	237	18.4	2.5	89%	88%	73%

WWTP=wastewater treatment plant; PE=population equivalent; MLE= Modified Ludzack−Ettinger; IA=intermittent aeration; COD=chemical oxygen demand; TN=total nitrogen; TP=total Phosphorus.

2.3 Direct emissions and dissolved gases measurements campaigns

Each selected WWTP was monitored for one month during the on-site campaign. Aerated points were considered as direct emission sources (aerated de-sanding units, aerobic biological processes, aerobic stabilisations of sludge, and biofilters). Measurements were performed for at least one week in each sampling unit. Monitoring was continuously carried out for the biggest WWTPs (WWTP1, 2, 3, 4, 5, and 6) and discontinuously for the smaller ones (WWTP7, 8, 9, 10, 11, and 12). Experimental equipment for the analysis of direct GHG mainly consisted of two devices.

For continuous measurements, the gas analyser (MIR 9000-CLD type, ENVEA, IT) with a membrane air dryer Mgf Sky 30/7M was used under controlled thermal conditions with the online acquisition of emitted GHG concentrations (one data every 5 min). The measurements of CH_4, CO_2, and N_2O were performed using the standard infrared absorption (UNICHIM method and ISTISAN 91/41 Report). The system was also calibrated each week using gas cylinders at the standard concentrations (CH_4 at 40.6 ppm, CO_2 at 2.33%, and N_2O at 160 ppm). The gaseous samples were conveyed to the analyser through a heated gas line at 120 °C. A cooling device (HIREF solution, https://hiref.it/) was used to reduce the temperature of the gases to 4 °C, thus, minimising the water vapour content. Every 3 h, a compressor was activated for ambient air sampling as a zero reference. In addition, a floating chamber was used to convey the samples to the analyser. For the design and construction of the chamber, guideline indications reported for similar measurements were followed (Spinelli et al., 2018; Caniani et al., 2019; Yver Kwok et al., 2015). The floating system was made from high-density polyethylene with a total volume of 310 L and a bottom area of 1 m^2.

Some specific sampling campaigns were carried out in the discontinuous mode for WWTPs with less than 25000 PE capacity. Specifically, peristaltic pump and gas bags (5 L) were used to acquire the gaseous samples (three replicates for each point). Subsequently, GHG concentrations were determined using photoacoustic spectroscopy (Brüel & Kjaer Multi-gas Monitor Type 1302).

Moreover, composite liquid samples from the influent and effluent of each WWTP were collected twice a week. The dissolved CO_2, CH_4, and N_2O were measured to calculate the possible indirect emissions of the GHG mass loads discharged in the water bodies. In this case, it was assumed that the dissolved gases are totally stripped after the discharge in the water body. Since the fate of dissolved GHGs is not uniquely predictable, the hypothesis allows to be conservative for the footprint calculation. Stripping the pre-treatment at 20°C (ultrasonic sonication SONOREX model), followed by photoacoustic spectroscopy determination (Brüel & Kjaer Multi-gas Monitor Type 1302) were carried out. Finally, the main conventional physical and chemical characterisations (chemical oxygen demand (COD), total suspended solids (TSS), total nitrogen (TN), and total phosphorus (TP)) of the wastewaters were analysed (APHA, 2015).

3. Results and Discussion

3.1 Audit of predictive tool and models

Technical and scientific literature shows that several tools and software solutions have been developed for different purposes. Simple stationary (i.e., Carbon Footprint Calculation Tool (CFCT)) or dynamic calculations (Mannina et al., 2016) have been proposed in recent decades (Table 2). Simulation models (Models 6, 7, 8, and 9) were implemented to calculate the emissions generated mainly from biological processes, such as activated sludge model (ASM) and benchmark simulation model (BSM), while specific tools were implemented for determining the CF of a single WWTP (Tools 1, 2, and 3) or for the entire water and wastewater service (Tools 4, 5, and 10). Direct emissions in most of these existing tools are mainly considered from the biological processes of the water line and only in some few cases from biogas production. Other aerated stages, such as degritting units, aerobic stabilisations of sludge, biofilters, and anaerobic supernatant treatments, are not usually considered, notwithstanding their wide impact on the global emissions of the plants (Demir et al., 2019). Moreover, most of the analysed models and tools focus mainly on direct N_2O emissions, while

CH_4 is generally considered only from the sludge line. Nevertheless, depending on the wastewater treatment configurations and influent characteristics, methane emissions could also represent a relevant contribution to the mainstream water line, even higher than N_2O emissions (Zhan et al., 2017). Moreover, CO_2 concentrations have typically accounted only for the fossil origin, while biogenic part, derived from microbial respiration during the biological processes has usually not been quantified. Finally, only a few studies have considered the dissolved fractions in the liquid of the gases, even when their contributions seemed relevant. In fact, the last report of the European Commission (JRC, 2020) also underlined the importance of considering the GHG impacts of an integrated system, including sewer network, wastewater treatment, sludge disposal, and final discharge into water bodies.

Additionally, even when emissions sources are located outside the physical boundaries of the WWTPs, they are strictly associated with the water utility management choices. Thus, indirect impacts due to energy consumption, chemical dosing, transport, and waste disposal have to be considered in wastewater CF assessment (Brown et al., 2010).

Currently, in the existing models, EFs are usually set by using internal libraries, without the possibility of editing default values and considering case-specific factors, which are crucial to achieve real evidence-based results in heterogeneous sectors such as the wastewater treatment service.

A few applications included CO_{2eq} mitigations, such as carbon sequestration from the soils and substitution of mineral fertiliser when sludge is applied in agriculture fields.

In contrast, normalised approach proposed in this study (WWEECarb) was applied to the entire wastewater service considering: 1) all main GHGs, both biogenic and of fossil origin; 2) direct emissions generated from different operational units of the WWTP; 3) indirect emissions due to energy and chemical consumptions and transportation; 4) dissolved gases present in the effluent; 5) emissions and removals related to sludge disposal and reuse; and 6) editable EFs, derived from both onsite measurement campaigns and technical literature libraries. Finally, the standard deviations of EFs were considered to evaluate the uncertainty and accuracy of the CF results.

Table 2. Comparison between existing carbon footprint tool and models and this specific case study methodology

MODEL/TOOL	Application	GHGs	Emissions Categories						Mitigation	EFs
			Direct GHGs	*Dissolved GHGs*	*Sludge Disposal*	*Energy*	*Chemicals*	*Transports*	*Carbon sequestration and minimisation*	
1 -CFCT	WWTP	CH_4, N_2O, fossil CO_2	✓*	✓	✓	✓	✓	✓	✓	Editable
2 -CF-TOOL CTRL	WWTP	N_2O, CO_2	✓**		✓	✓	✓			Default
3-CHEApet	WWTP	CH_4, N_2O, CO_2	✓**	✓ only N_2O	✓	✓	✓	✓		Default
4-WESTWeb	Water Service	CH_4, N_2O, CO_2, NO_x, PM, SO_x, VOC, CO	✓**		✓	✓	✓			Default
5-ECAM	Water service	CH_4, N_2O, fossil CO_2	✓**	Not specified	✓	✓			✓	Editable
6-DEEM	Biological Unit	N_2O, CO_2	✓**							Default
7-ASMN	Biological Unit	N_2O	✓**							Default
8-BSM2G	WWTP	CH_4, N_2O, CO_2	✓**		✓	✓	✓			Default
9-BSM2-e	WWTP	CH_4, N_2O, CO_2	✓**		✓	✓	✓			Default
10-WWEECarb	**WW Service**	**CH_4, N_2O, fossil and biogenic CO_2**	**✓§**	**✓**	**✓**	**✓**	**✓**	**✓**	**✓**	**Editable**

CFCT= Carbon Footprint Calculation Tool; CF-TOOL CTRL= Carbon Footprint Control; CHEApet= Carbon Heat Energy Assessment Plant Evaluation Tool; WESTWeb= Water-Energy Sustainability Tool; ECAM= Energy Performance and Carbon Emissions Assessment and Monitoring; DEEM= Diffusive Emissions Estimation Model; ASMN= Activated Sludge Models for Nitrogen; BSM2G= Benchmark Simulation Model No 2 Greenhouse Gas; BSM2-e= Benchmark Simulation Model No 2 Emission, WWEECarb= Water and Waste Environmental Engineering Carbon Footprint; WWTP= wastewater treatment plant; WW= wastewater; GHGs=greenhouse gases; EFs=emission factors

* Unique Direct EF from the whole water line
**Direct EFs from biological reactor and sludge line
§Direct EFs from aerated de-sanding units, aerobic biological process, aerobic stabilization of sludge, fugitive emissions from sludge line and biofilters

1 Gustavsson & Tumlin, 2013, 2 Baeza et al., 2017, 3 https://www.waterrf.org/research/projects/demonstration-carbon-heat-energy-assessment-and-plant-evaluation-tool-cheapet, 4 https://west.berkeley.edu/model.php, 5 http://wacclim.org/ecam/sources.php, 6 Guo et al., 2012; Mannina et al., 2016), 7 Guo et al., 2012; Mannina et al., 2016, 8 Flores-Alsina et al., 2012; Mannina et al., 2016, 9 Mannina et al., 2016; Sweetapple et al., 2013, 10 This study

3.2 Proposed methodology for carbon footprint estimation in wastewater service

The methodology was developed based on ISO 14064-1:2019 standard and adapted to a wastewater service with systemic and territorial approaches. In this context, the operational control principle defined by UNI ISO 14064-1:2019 was applied as a reporting boundary criterion for CF quantification. The reporting boundaries were set considering: 1) the physical operative limits of the WWTPs to define the direct emissions, 2) impacts of energy and chemical supplies, 3) waste and reagent transportations, and 4) emissions caused by the final sludge disposal or recovery/valorisation. Specifically, direct emissions included the GHGs from: i) biogas combustion, ii) different aerated units (aerated degritting unit, biological reactor, aerobic sludge stabilisation, and biofilter) of the WWTP, and iii) fugitive gases of the sludge line. In contrast, emissions from the dissolved gases on the water body, energy and chemical consumptions, transportation, and sludge disposal were considered as indirect. The indirect emissions from sludge reuse and related mitigations, such as carbon sequestration and synthetic fertiliser substitution, were estimated. CF was calculated based on the contribution of three main GHGs: methane, carbon dioxide, and nitrous oxide (Nguyen et al., 2019). Each relevant GHG contribution, fossil or biogenic CO_2 was distinguished based on the origin of the emission (Table 3).

Table 3. Emissions categories, including the distinction of CO_2 origin and the separate quantification for each GHG

ISO 14064-1:2019 Category	Proposed declined categories	CO_2 origin	Considered GHG
Direct emissions	Direct emission from combustion	Biogenic	tonCO_{2eq}/y, reported separately for N_2O, CH_4 and CO_2
	Direct emissions from WWTP processes in mainstream	Biogenic	
	Direct fugitive emissions	Biogenic	
Indirect emissions from imported energy	Indirect emissions from energy consumption	Non biogenic	tonCO_{2eq}/y
Indirect emissions from transportation	Indirect emissions from waste transport	Non biogenic	tonCO_{2eq}/y, reported separately for N_2O, CH_4 and CO_2
	Indirect emissions from chemical transport	Non biogenic	

Indirect emissions from products used by the organization	Indirect emissions from chemical consumption	Non biogenic	$tonCO_{2eq}/y$
Indirect emissions from other sources	Indirect emissions on the water body	Biogenic	$tonCO_{2eq}/y$, reported separately for N_2O, CH_4 and CO_2
	Indirect emissions from sludge composting	Biogenic	

Emissions were determined based on Equation 1.

$$Emission\ contribution\ \left(\frac{tonCO_{2eq}}{y}\right) = Activity\ data\ \left(\frac{quantity}{y}\right) * EF\ \left(\frac{tonGHGs}{quantity}\right) * GWPs\ \left(\frac{tonCO_{2eq}}{tonGHG}\right) \quad \text{(Eq. 1)}$$

where GWPs are referred in IPCC Fifth Assessment Report, AR5 (IPCC, 2019).

In general, activity data (AD) represents the quantity, generated or used, of energy, mass, or volume, representing the key parameter for each emission category. The types of AD considered in this study are listed in Table 4. The EFs used were collected from the literature, guidelines, and databases or measured with specific on-site campaigns (Table 4 and Figure 1).

Table 4. Emissions categories, activity data and type of EFs

Number	Declined category	Activity data	Emission factor (EF)	Ref.
(1)	Direct emission from combustion	Biogas produced (Nm^3/y), methane content ($\%CH_4$)	g CO_2/kg burned CH_4 g CH_4/kg burned CH_4 g N_2O/kg burned CH_4	International databases (IPCC, 2019b)
(2)	Direct emissions from WWTP processes in mainstream	Influent and effluent COD and TN loads (ton/y)	kg N_2O/kgTN_{rem} kg CH_4/kg COD_{in} kg CO_2/kg COD_{rem}	Measured in this study Literature, international databases (Aboobakar et al., 2013; Ahn et al., 2010; Caniani et al., 2019; Foley, de Haas, Hartley, et al., 2010; Joss et al., 2009; Masuda et al., 2018; Ribera-Guardia et al., 2019; Wang et al., 2016);
(3)	Direct fugitive emissions	Biogas produced (Nm^3/y) and methane content ($\%CH_4$); sludge produced (ton/y) and characterisation (%TS, %N, %P, %K, %C)	kgN_2O/tonTS kgCH_4/tonTS kgCO_2/tonTS	Literature (Kirkeby et al., 2005; Majumder et al., 2014; Scheutz & Fredenslund, 2019; Willén et al., 2016)
(4)	Indirect emissions for energy consumption	Electricity consumption (MWh/y), other fuel combustion also renewable (Nm^3/y or l/y)	$tonCO_{2eq}$/GWh	National and International databases (IPCC, 2019a; ISPRA, 2018)

(5)	Indirect emissions for waste transport	km travelled	gN_2O/km gCH_4/km gCO_2/km	National databases http://www.sinanet.isprambiente.it/it/sia-ispra/fetransp
(6)	Indirect emissions for chemical transport	km travelled	gN_2O/km gCH_4/km gCO_2/km	National databases http://www.sinanet.isprambiente.it/it/sia-ispra/fetransp
(7)	Indirect emissions for chemical consumption	Quantity used (kg/y)	$kgCO_{2eq}$/kg reagent	Literature, international databases (Gustavsson & Tumlin, 2013)
(8)	Indirect emissions on the water body	Effluent COD and TN loads (ton/y)	g CO_2/kgCOD_{eff} g CH_4/kgCOD_{eff} g N_2O/kgTN_{eff}	Measured in this study
(9)	Indirect emissions and mitigation for sludge disposal	Dried sludge	kg N_2O /tonTS kg CH_4/tonTS kg CO_2/tonTS	Literature, international databases (Boldrin et al., 2009; Chai et al., 2015; Chen & Kuo, 2016; Han et al., 2018; IPCC, 2019c; Kirkeby et al., 2005; Piippo et al., 2018; Yuan et al., 2018; https://www.climfoot-project.eu/)
(10)	Indirect emissions from sludge reuse	Dried sludge	kg N_2O /tonTS kg CH_4/tonTS kg CO_2/tonTS	(Boldrin et al., 2009; Bruun et al., 2006; IPCC, 2019c; Kirkeby et al., 2005)
(11)	Mitigations: carbon sequestration and synthetic fertiliser substitution	Dried sludge	kg N_2O /tonTS kg CH_4/tonTS kg CO_2/tonTS	(Foley, de Haas, Yuan, et al., 2010; Kirkeby et al., 2005)

TS=total solids, COD_{in}= influent chemical oxygen demand; COD_{eff}= effluent chemical oxygen demand; COD_{rem}= removed chemical oxygen demand; TN_{rem}= removed total nitrogen; TN_{eff}= effluent total nitrogen; K=potassium; C=carbon.

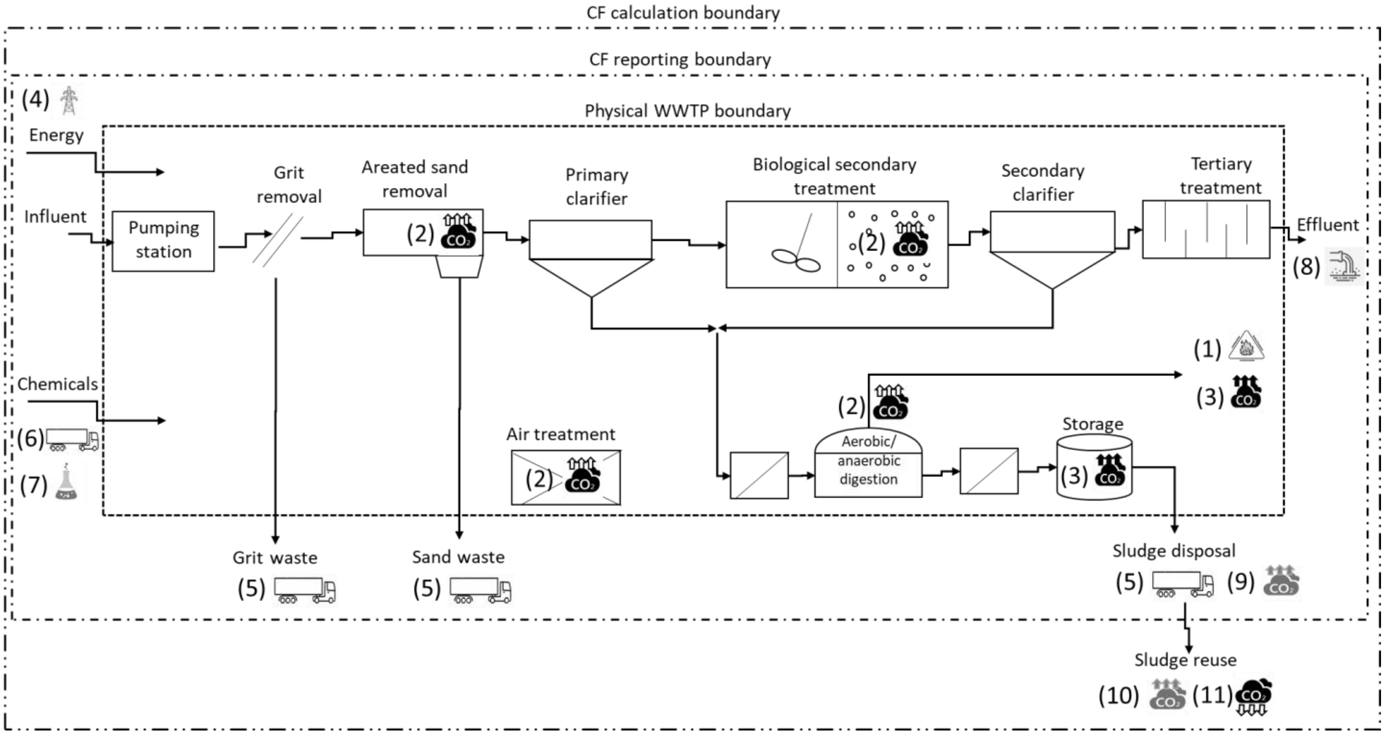

Figure 1. General WWTP scheme and emissions categories in respect to operative, carbon footprint (CF) reporting and calculation boundaries

The logical flow scheme of the proposed approach according to ISO 14064-1 is reported in Figure 2.

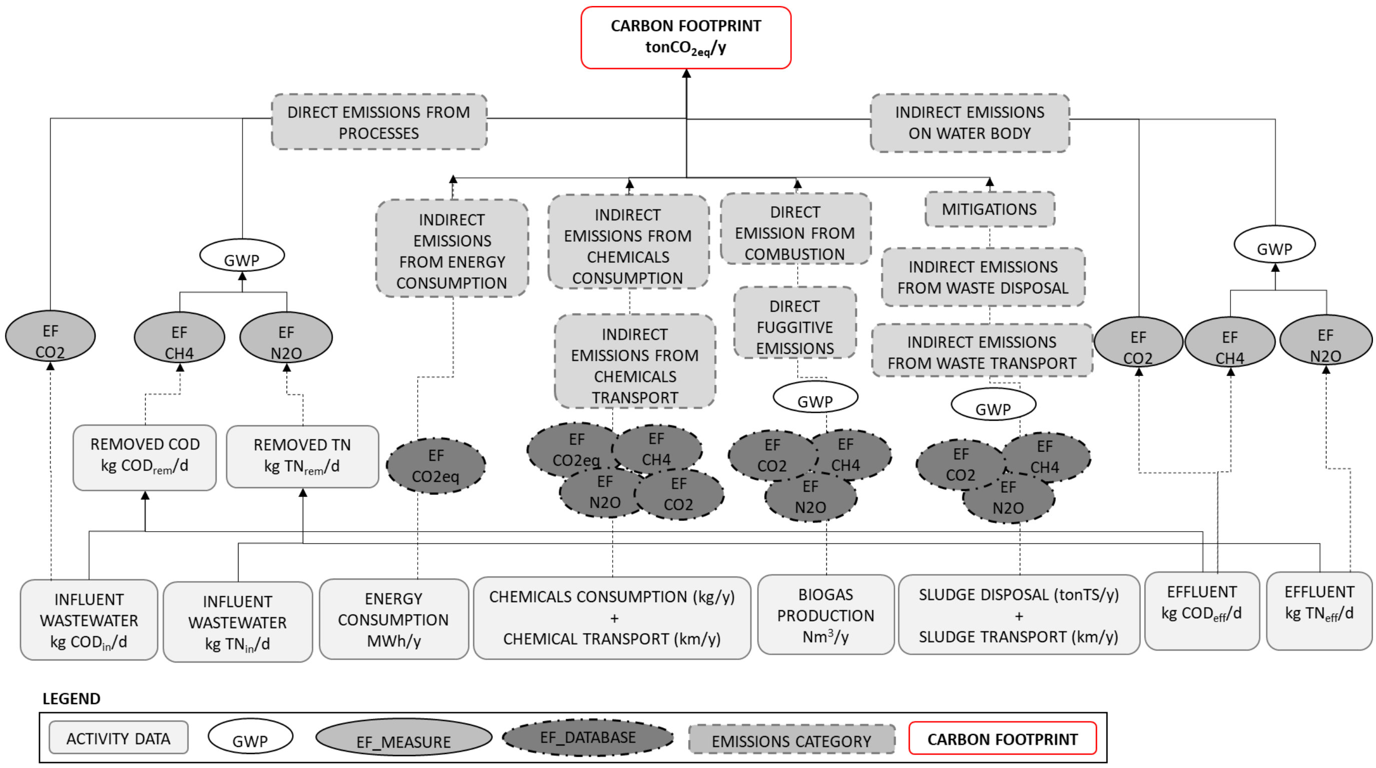

Figure 2. Logical flow scheme of the proposed methodology

The overall calculation of CF, considering the on-site measured emissions, was carried out by following several phases: 1) elaboration of 1-year plant operation data of 12 WWTPs, such as temperature, wastewater flowrate, airflow rate, influent and effluent COD and TN mass loads; 2) calculation of the mass loads of emitted CO_2, CH_4, and N_2O; 3) normalisation at 20 °C; and 4) conversion into CO_2 equivalent.

3.3 Measurement campaigns: direct emitted GHG concentrations

Results from the measurement campaigns showed concentration variability in GHGs emitted from the biological processes of different WWTPs, as reported in previously literature studies (Tumendelger et al., 2019). The average values of N_2O concentrations were less than 4 mg/m^3 of air for the largest WWTPs (WWTP1–6), while they ranged from 4 to 15 mg/m^3 for the smallest plants (WWTP7–12) (Figure 2). A similar behaviour was observed for CO_2 quantification. Values lower than 5 g CO_2/m^3 were detected for the largest WWTPs and in the range of 7–16 g CO_2/m^3 in other cases (Figure 2). Especially for N_2O emissions, the difference in the contributions of the largest WWTPs could be justified by the usually higher efficiencies of the aeration systems and more elevated removals, as reported by Valkova et al. (2020). In contrast, for directly emitted CH_4 concentrations, a strict correlation was not identified with the size of the WWTPs (Figure 3).

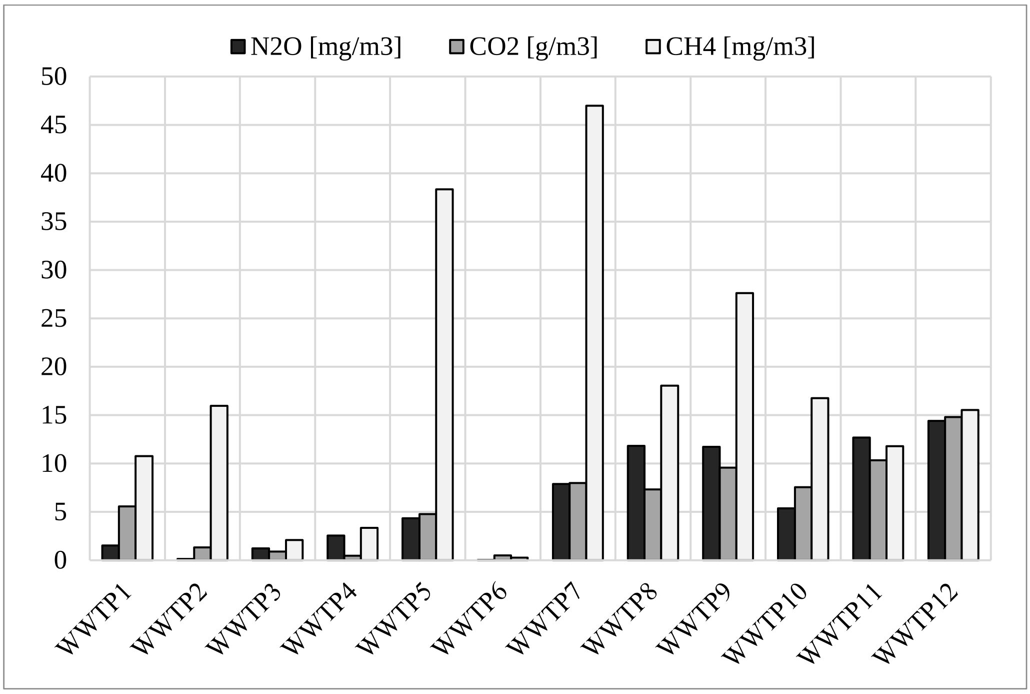

Figure 3. Direct emitted GHGs concentrations from biological processes related to the capacity of the plants, from WWTP1 (highest capacity) to WWTP12 (smallest capacity).

Furthermore, an important relationship was observed during the rain events that occurred during the sampling campaigns. It was evident (example shown for WWTP1 in Figure 3) that wet periods affected the direct GHG emissions with higher concentrations, mainly for N_2O and CH_4, both emitting from the aerated degritting units and biological processes. It can be noticed that the emitted CH_4 and N_2O detected during the rain events were more than 3–6 times higher than the those detected during the dry period (Figure 4a, c, b, and d). These variations were probably dependent on sewage system characteristics and a corresponding increase in the dissolved gases in the influent flow during the rain events. Urban sewer systems can cause several contaminant degradations, which can generate dissolved gaseous sub-products, as previously reported by Jin et al. (2019).

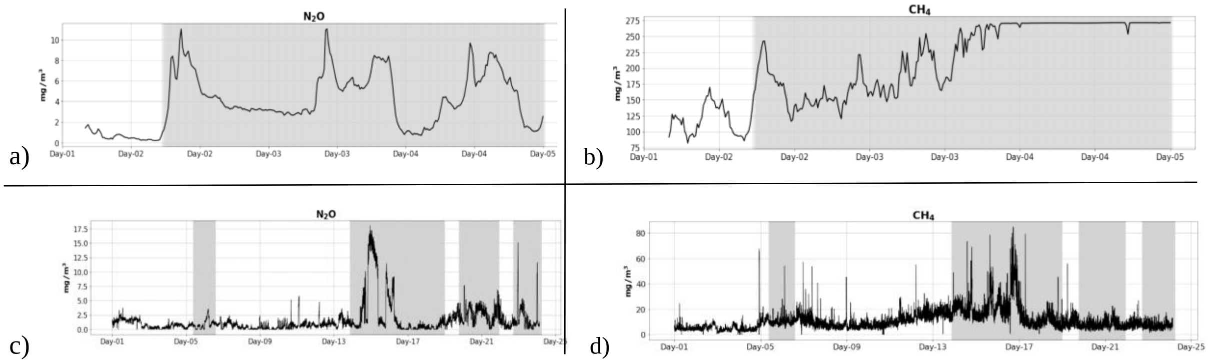

Figure 4. Direct GHGs concentrations during the measurement campaigns, including wet period: a) N_2O concentrations and b) CH_4 concentrations from aerated degritting unit, c) N_2O concentrations and d) emitted CH_4 concentrations from biological treatment.
LEGEND: xx Wet period ☐ Dried period

Moreover, from the measurement campaigns, the directly emitted GHG concentrations were also found to be spatially varying at different points of the same biological reactor, as discussed by Pan et al. (2016). For WWTP5, the average emitted concentrations at the beginning and end of the aerobic reaction volume decreased from 6 ± 17 mgN_2O/m^3, 50 ± 41 $mgCH_4/m^3$, and 5 ± 2 gCO_2/m^3 to 3 ± 5 mgN_2O/m^3, 22 ± 28 mg CH_4/m^3, and 5 ± 2 g CO_2/m^3, respectively. It was observed that for N_2O and CH_4, higher concentrations were observed in the initial section of the reaction volume, where the influent macro-contaminant mass loads (COD and TN) were also probably higher. On the other hand, CO_2 concentrations, mainly generated from biomass respiration, remained almost stable throughout the entire biological unit (Zhan et al., 2017). For the calculation of the emitted mass loads, average concentrations of GHGs for each unit were considered.

3.4 Measured direct emissions factors

GHG loads were calculated from the measured concentrations and normalised to identify different EFs reported in terms of average values and standard deviations in Figures 5, 6, and 7 for N_2O, CH_4, and CO_2, respectively. Biological treatment was typically characterised by the highest emissions, with the values ranging from $6.7 * 10^{-8}$ to 0.02 kg N_2O/kgN_{rem}, $2.2 * 10^{-8}$ to 0.003 kg $CH_4/kgCOD_{in}$, and $1.8 * 10^{-5}$ to 3.1 kg $CO_2/kgCOD_{rem}$. It was also observed that the EF values were inversely proportional to the WWTP size, resulting in higher specific values for the smallest WWTPs (WWTP7–12). In the short cut biological process treating nutrient-rich anaerobic rejected liquor via nitrite resulted in significantly high EFs in terms of N_2O (0.27 kg N_2O /kgTN_{rem}), while CH_4 and CO_2 impacts were consistent with other biological treatments. Aerated degritting units showed the values of less than $2.6 * 10^{-4}$ kg N_2O /kgN_{rem}, in the range of $1.3 * 10^{-4}$–$1.5 * 10^{-3}$ kg CH_4/kgCOD_{in} and $4.1 * 10^{-3}$–0.1 kgCO_2/kgCOD_{rem}. Aerated sludge stabilisation, measured with a continuous monitoring campaign for WWTP 6, yielded the EFs for CO_2 ($1.3 * 10^{-3}$ kg CO_2/kgTS) higher than those of N_2O and CH_4 ($8.9 * 10^{-8}$ kg N_2O/kgTS and $3.8 * 10^{-7}$ kg CH_4/kgTS, respectively). Finally, for biofilters, the normalised values per cubic meter of the treated air changed from $1.2 * 10^{-6}$ to $6 * 10^{-6}$ kg N_2O /m^3, $2.6 * 10^{-5}$ to $7.5 * 10^{-5}$ kg CH_4/m^3, and $4 * 10^{-4}$ to $8.3 * 10^{-4}$ kg CO_2/m^3, highlighting the significant impact of methane on the final global emission from these air treatment units.

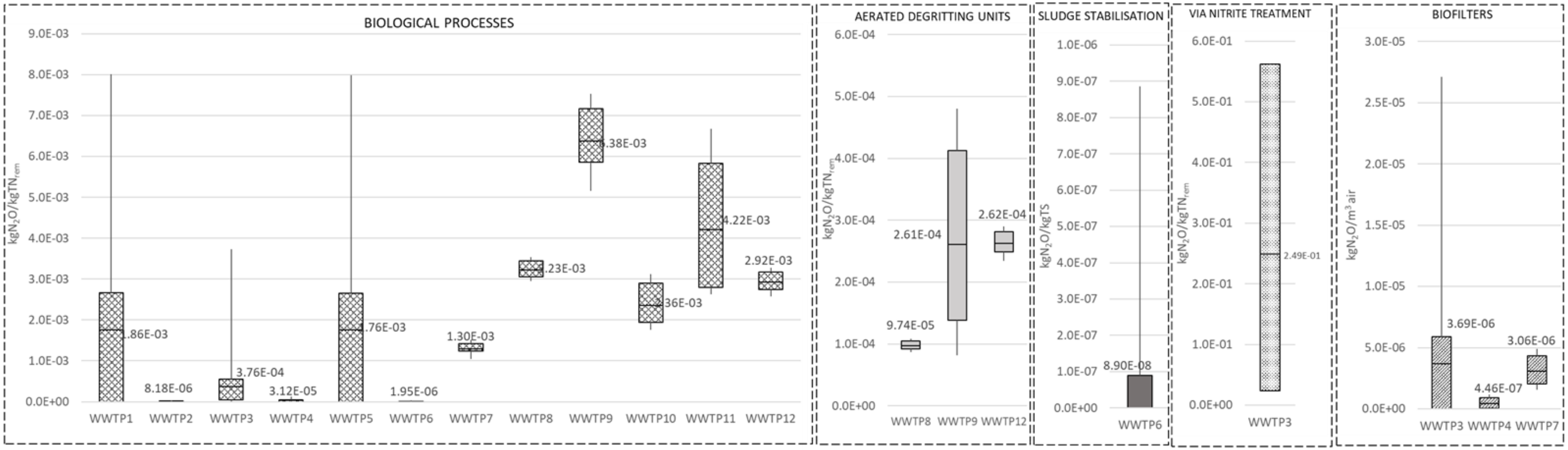

Figure 5. Emission factors for direct N_2O emissions from WWTPs grouped for operational unit

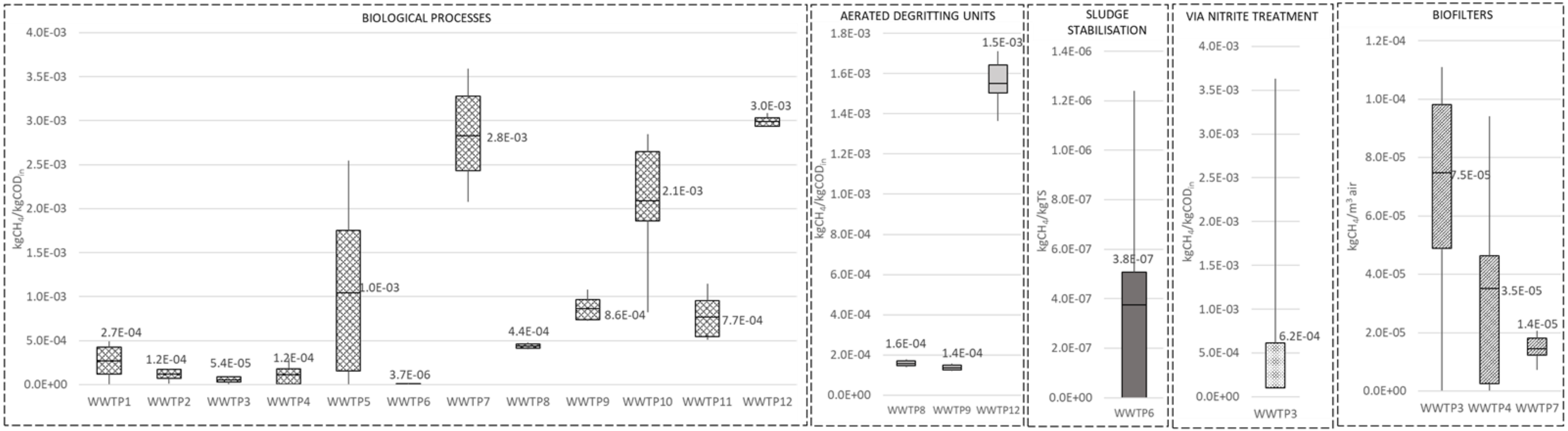

Figure 6. Emission factors for direct CH_4 emissions from WWTPs grouped for operational unit

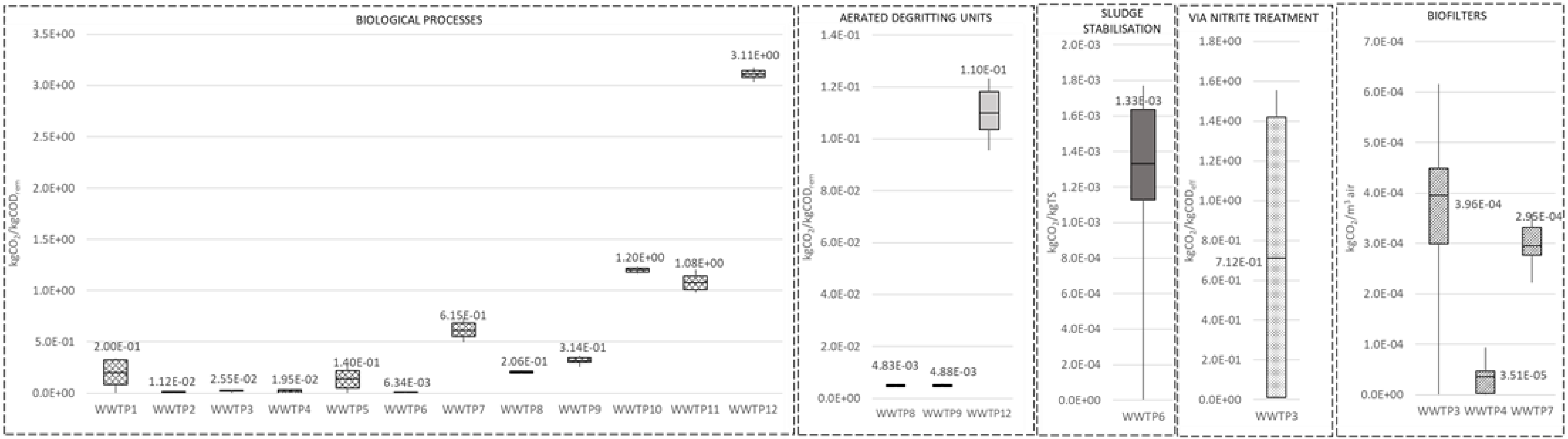

Figure 7. Emission factors for direct CO_2 emissions from WWTPs grouped for operational unit

3.5 Measured indirect emission factors

The dissolved GHG concentrations in the effluents of the WWTPs varied in the range of 0.2–24 mg/L for N_2O, 0.1–1 mg/L for CH_4, and 1.8–52 mg/L for CO_2. Specifically, for the largest WWTPs (from 1 to 6), the average values were 4.6 ± 9.4 mg/L for N_2O, 0.4 ± 0.3 mg/L for CH_4, and 30 ± 19 mg/L for CO_2. Meanwhile, the values were generally found to be lower and respectively equal to 0.8 ± 0.5, 0.3 ± 0.2, and 16.4 ± 2.5 mg/L for the smaller WWTPs (from 7 to 12). Scientific literature on the measured values of dissolved GHGs in the effluents of WWTPs is scarce. In general, these values have been found to range from 0.009 to 24 mg/L for N_2O, 0.009 to 4.5 mg/L for CH_4, and 245 to 1352 mg/L for CO_2 (Caniani et al., 2019; Masuda et al., 2015, 2018; Vieira et al., 2019). This variability indicates the need for site-specific campaigns for properly evaluating the indirect emissions due to dissolved GHG contributions.

Moreover, during the sampling campaigns, dissolved GHGs in the influent streams of the WWTPs from the sewage systems were monitored. Specifically, in the largest WWTPs (from 1 to 6), the dissolved GHG influent concentrations were found to be 12.5 ± 26, 1.1 ± 1.2, and 76 ± 86 mg/L for N_2O, CH_4, and CO_2, respectively, while the values were found to be 0.5 ± 0.4 mg N_2O /L, 0.3 ± 0.2 mg CH_4/L, and 50 ± 16 mg CO_2/L for the smaller WWTPs (from 7 to 12). These aspects indicated that a relevant contribution of dissolved GHG comes directly from the sewage networks, especially for plants with higher capacities. Furthermore, for smaller WWTPs, the dissolved GHG concentrations in the effluent were higher than those of the influent stream. This confirmed that for smaller WWTPs, which are usually subjected to more limiting operative conditions (such as low carbon:nitrogen (C:N) ratio, unstable or optimised process parameters, and inefficient aeration supply) (Kumar et al., 2021), additional contributions of GHGs were generated during the biological processes, which remained dissolved in the liquid stream.

The EFs of dissolved GHG emission category varied in the ranges of 33–782 g N_2O /kgN_{eff}, 4–132 g CH_4/$kgCOD_{eff}$, and 553–358 g CO_2/$kgCOD_{eff}$ for N_2O, CH_4, and CO_2, respectively (Table 5). The standard deviations of the dissolved GHG EFs obtained for the six continuously monitored WWTPs were significantly relevant and heterogeneous, ranging from 34% to 136% with respect to the average values. Moreover, the EF values were found to be inversely proportional to the WWTP size, resulting in the higher specific factors for the smallest WWTPs (Table 5).

Table 5. Emission factors (EFs) for indirect emissions on water body due to dissolve GHGs in WWTPs effluent and related standard deviations

		WWTP1	WWTP2	WWTP3	WWTP4	WWTP5	WWTP6	WWTP7	WWTP8	WWTP9	WWTP10	WWTP11	WWTP12
DISSOLVED GHG_EF	**PE_ COD**	44499	31819	46511	53713	30645	20166	6241	7258	10139	1335	2125	5410
gN_2O/ $kgTN_{eff}$	**Mean**	**46**	**53**	**33**	**108**	**782**	**305**	**98**	**129**	**538**	**346**	**140**	**122**
	Dev std	*23*	*23*	*21*	*147*	*330*	*171*	-	-	-	-	-	-
gCH_4/ $kgCOD_{eff}$	**Mean**	**7**	**4**	**39**	**10**	**133**	**41**	**8**	**11**	**10**	**9**	**11**	**16**
	Dev std	*5*	*5*	*24*	*8*	*63*	*2*	-	-	-	-	-	-
gCO_2 /$kgCOD_{eff}$	**Mean**	**790**	**1328**	**5358**	**1330**	**2546**	**3603**	**853**	**378**	**1014**	**735**	**553**	**3434**
	Dev std	*551*	*503*	*3082*	*1667*	*865*	*3508*	-	-	-	-	-	-

3.6 Specific GHG contributions on measured direct and dissolved emissions

The main contributions of different GHGs to the direct and dissolved emission categories were calculated as a percentage of the total emissions (Figure 8). The reported percentages of different GHG contributions were expressed as the percentage of equivalent CO_2. The values, as the gaseous flows were directly transferred to the atmosphere, showed that the main impacts were attributed to CO_2 for smaller plants (< 15000 PE), while N_2O was the main responsible in largest WWTPs (> 15000 PE). On the other hand, CH_4 emissions accounted for about 11–12 % for all the sizes. For indirect emissions into the water body, the main contributor was N_2O, followed by CO_2 and CH_4.

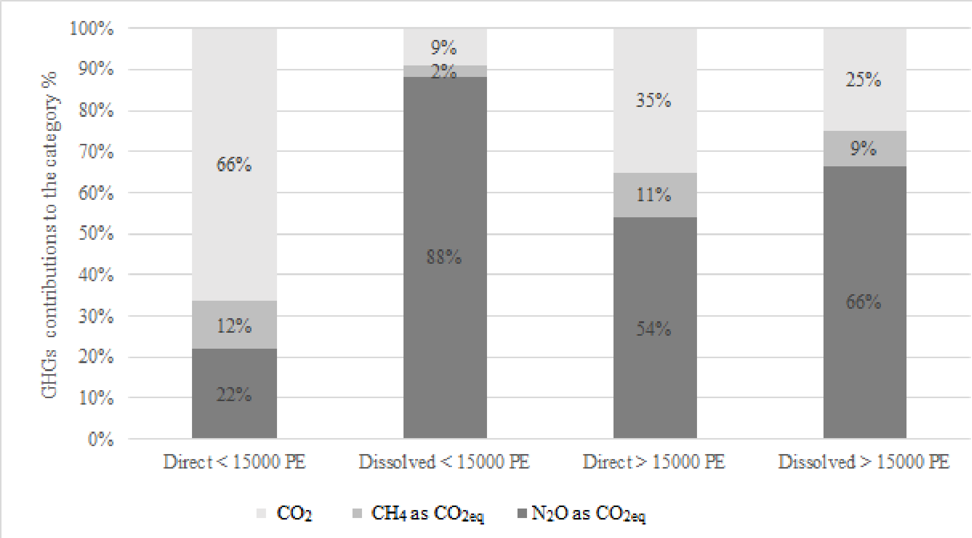

Figure 8. GHGs contributions to the category's direct emissions from processes and indirect emission to the water body in relation with the WWTP size (<15000 PE and >15000 PE).

3.7 Carbon footprint results

After the long-term sampling periods, the entire CF of each selected WWTP was calculated by adding the different contributions of the categories based on the normalised proposed approach (Table 6). The final values ranged from a minimum 210 tonCO_{2eq}/y (WWTP10) to a maximum 4047 tonCO_{2eq}/y (WWTP 3). In general, the most impactful categories (Figure 9) were the indirect emissions associated with dissolved GHGs present in the water body, which influenced 13–70% of the CF of each WWTP. Indirect emissions due to energy consumption accounted for 10–40% and, as expected, this category is directly related to the carbon and nitrogen removed loads in the different WWTPs (Table 6). Moreover, direct emissions from treatment processes contribution approximately 19% on an average. The disposal of sewage sludge and use of chemicals affected 6–34% and 1–9%, respectively. The impacts and their variabilities, especially for direct and dissolved GHG contributions, further underlined the already discussed importance of the measurement campaigns to obtain more appropriate and proper data for the specific conditions of each plant.

Table 6. Carbon footprint of the selected monitored WWTPs

	Direct emission from combustion	Direct emissions from WWTP processes in mainstream	Direct fugitive emissions	Indirect emissions from energy consumption	Indirect emissions from waste transport	Indirect emissions from chemical transport	Indirect emissions from chemical consumption	Indirect emissions on the water body	Indirect emissions from sludge composting	Indirect emissions from sludge reuse*	Mitigations from carbon sequestration and synthetic fertiliser substitution*	Total WWTP CF
	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$	$tonCO_{2eq}/y$
WWTP1	0	862	3.2	881	30	2.2	61	511	777	220*	-314*	3128
WWTP2	0	17	2.2	570	20	1.2	20	316	490	129*	-190*	1436
WWTP3	398	310	252	770	14	3.2	110	1933	257	63*	-101*	4047
WWTP4	530	87	335	370	28	4.1	189	1493	493	132*	-204*	3528
WWTP5	0	244	1.9	627	20	1.4	49	3136	437	124*	-186*	4517
WWTP6	0	7	2.7	679	30	3.1	210	872	598	139*	-219*	2402
WWTP7	0	725	2.6	215	4	1.3	22	149	59	18*	-29*	1179
WWTP8	0	79	1.9	189	8	1.5	16	215	116	31*	-44*	627
WWTP9	0	185	2.2	187	6	0.9	42	1222	106	29*	-42*	1752
WWTP10	0	66	0.0	39	2	0.2	4	100	0	0*	0*	210
WWTP11	0	103	0.0	69	3	0.3	4	61	0	0*	0*	240
WWTP12	0	717	1.9	99	5	0.5	19	174	71	20*	-28*	1087
											TOTAL	**24154**

***Emissions Categories excluded from reporting boundaries**

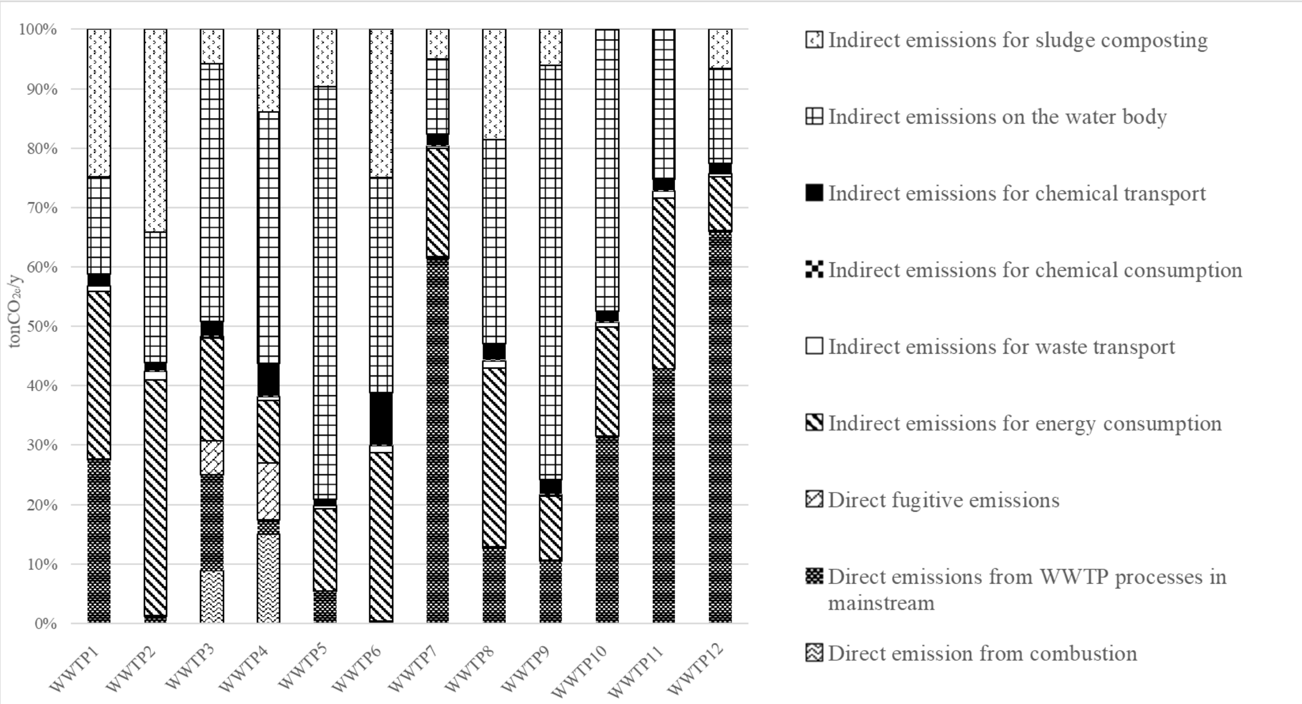

Figure 9. Categories contributions (%) to the total carbon footprint for each selected WWTP

The SCFs of the WWTPs were finally calculated and were found to be between 0.04 and 0.20 $tonCO_{2eq}$/PE/y (Figure 10). The obtained SCFs were indirectly dependent on the size of the plants and were more widely distributed for the WWTPs with less than 15000 PE capacity. Similarly, the average SCFs were found to be 0.091 ± 0.038 $tonCO_{2eq}$/PE/y for the WWTPs with more than 15000 PE capacity and 0.153 ± 0.045 $tonCO_{2eq}$/PE/y for smaller facilities. These SCF results were found to be more variable and almost 1.7 times higher than those of the larger size (>15000 PE). Notwithstanding that the calculation procedure was not perfectly comparable, these results were consistent with those mentioned in other published research which reported the values in the range of 0.07–0.108 $tonCO_{2eq}$/PE/y (Gustavsson & Tumlin, 2013), 0.023–0.1 $tonCO_{2eq}$/PE/y (Maktabifard et al., 2020) and 0.062–0.161 $tonCO_{2eq}$/PE/y (Mamais et al., 2015).

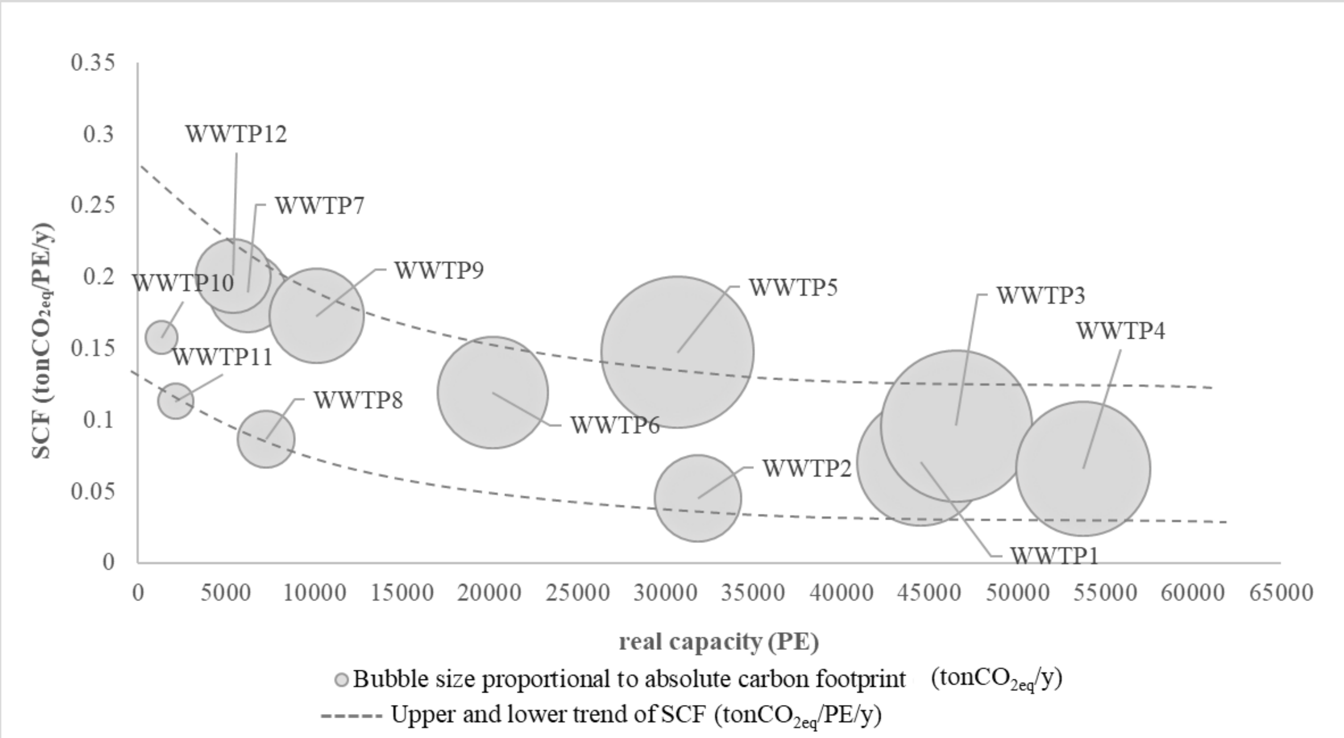

Figure 10. Specific Carbon Footprints (SCF) of the selected WWTPs

The CF was finally extended at the regional level, including for all plants managed by the water utility, as well as the septic tanks, resulting in 2396 and 1588 tonCO_{2eq}/y emitted by the non-monitored WWTPs and septic tanks, respectively, yielding a total territorial CF of the wastewater service of 28137 ± 8497 tonCO_{2eq}/y. Biogenic CO2 had a 25% impact on total CF. The highest contribution to territorial CF was from WWTPs with more than 15000 PE capacity, accounting for 68%. Nevertheless, the presence of several minor plants should be considered as their contributions were not found to be negligible, with the impacts of 16% from the plants with capacities ranging from 15000 to 5000 PE and 10% for the smaller WWTPs. Finally, septic tank emissions accounted for 6% to the total CF.

4. Conclusion

Regional CF of wastewater treatment service was determined by properly applying for the first time the methodology ISO14064-1 to identify specific emission categories. The proposed new evidence-based methodology enables the calculation of the Carbon Footprint accounting all the emissions categories in clear operational and reporting boundaries within cities and regions. The calculations were validated measuring the most impacting categories of direct and dissolved GHGs emissions with specific sampling campaigns. The uncertainties related to the EFs let to long-term on-site measurements of relevant GHGs. In fact, calculations carried out using only the available EFs mentioned in the guidelines may significantly differ from the actual site conditions. The measurement campaigns mentioned in this study enabled us to define the site-specific EFs for the main direct sources from the treatment processes (degritting units, aerobic biological treatments, biofilters, and aerobic sludge stabilisations) in 12 different sized WWTPs. Furthermore, dissolved GHGs discharged in the final water bodies were analysed and were found to significantly impact the entire CF quantification. The results, in terms of the EFs (average values and standard deviations), showed a high variability in the emissions, based on the WWTP size and specific operative conditions, especially for biological processes. The average values were found to be $3.4 * 10^{-3}$ $kgN_2O/kgTN_{rem}$, $1.7 * 10^{-3}$ $kgCH_4/kgCOD_{in}$, and 1.1 $kgCO_2/kgCOD_{rem}$ for the smallest (< 15000 PE) WWTPs. On the other hand, the biggest (> 15000 PE) WWTP EFs resulted in $6.6 * 10^{-4}$ $kgN_2O/kgTN_{rem}$, $2.7 * 10^{-4}$ $kgCH_4/kgCOD_{in}$, and 0.07 $kgCO_2/kgCOD_{rem}$. The analytical campaigns of this study also highlighted that biogenic CO_2 emitted from the biological processes significantly impacted the global CF of the 12 WWTPs, accounting for 35–66% of the total directly emitted CO_{2eq}. The overall territorial CF of the wastewater service was 28137 ± 8497 $tonCO_{2eq}/y$, including of the non-monitored WWTPs and septic tanks. The most impacting categories for most plants were: i) indirect emissions associated with the dissolved GHGs present in the water body, ii) indirect emissions due to energy consumption, followed by iii) direct emissions from treatment processes, iv) disposal of sewage sludge and v) use of chemicals.

Finally, this approach can support territorial water utilities not only to assess their carbon footprint with normalized approach, but also to develop regional mitigation scenarios and decisions towards low-carbon water utilities. In fact, since the Specific Carbon Footprint coefficients change based on the plants size, mitigation actions could consider the population distribution in the territory and both centralized and decentralized systems. Moreover, mitigations for wastewater service decarbonisation according to the shown most impacting emission categories could be prioritised as following: 1) acquire renewable energy sources to reduce the indirect emissions from fossil primary energy production; 2) optimize efficiency and kinetics of biological removal of organic and nutrients loads and aeration efficiency in order to reduce dissolved GHGs in the final effluent, 3) reduce direct emissions mainly avoiding uncontrolled transitory phases or limiting operative conditions in the biological reactors; 4) promote less impacting sludge disposal destination especially avoiding landfill and 5) use chemical reagents characterized by lower Emission Factors for their primary production. The proposed methodological approach coupled with accurately planned site-specific long term measurement campaigns could further boost and address the decarbonization of the wastewater service in territories.

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References

Aboobakar, A., Cartmell, E., Stephenson, T., Jones, M., Vale, P., & Dotro, G. (2013). Nitrous oxide emissions and dissolved oxygen profiling in a full-scale nitrifying activated sludge treatment plant. *Water Research*, *47*(2), 524–534. https://doi.org/10.1016/j.watres.2012.10.004

Ahn, J. H., Kim, S., Park, H., Rahm, B., Pagilla, K., & Chandran, K. (2010). N2O emissions from activated sludge processes, 2008-2009: Results of a national monitoring survey in the united states. *Environmental Science and Technology*, *44*(12), 4505–4511. https://doi.org/10.1021/es903845y

ARERA (2017). *562/2017/R/IDR*, URL: https://www.arera.it/it/docs/17/562-17.htm (accessed 10 June 2021).

Boldrin, A., Andersen, J. K., Møller, J., Christensen, T. H., & Favoino, E. (2009). Composting and compost utilization: Accounting of greenhouse gases and global warming contributions. *Waste Management and Research*, *27*(8), 800–812. https://doi.org/10.1177/0734242X09345275

Brown, S., Beecher, N., & Carpenter, A. (2010). Calculator tool for determining greenhouse gas emissions for biosolids processing and end use. *Environmental Science and Technology*, *44*(24), 9509–9515. https://doi.org/10.1021/es101210k

Bruun, S., Hansen, T. L., Christensen, T. H., Magid, J., & Jensen, L. S. (2006). Application of processed organic municipal solid waste on agricultural land - A scenario analysis. *Environmental Modeling and Assessment*, *11*(3), 251–265. https://doi.org/10.1007/s10666-005-9028-0

Caniani, D., Caivano, M., Pascale, R., Bianco, G., Mancini, I. M., Masi, S., Mazzone, G., Firouzian, M., & Rosso, D. (2019). CO2 and N2O from water resource recovery facilities: Evaluation of emissions from biological treatment, settling, disinfection, and receiving water body. *Science of the Total Environment*, *648*, 1130–1140. https://doi.org/10.1016/j.scitotenv.2018.08.150

Chai, C., Zhang, D., Yu, Y., Feng, Y., & Wong, M. S. (2015). Carbon footprint analyses of mainstream wastewater treatment technologies under different sludge treatment scenarios in China. *Water (Switzerland)*, *7*(3), 918–938. https://doi.org/10.3390/w7030918

CHEApet, URL: https://www.waterrf.org/research/projects/demonstration-carbon-heat-energy-assessment-and-plant-evaluation-tool-cheapet, (accessed 28 June 2021)

Chen, Y. C., & Kuo, J. (2016). Potential of greenhouse gas emissions from sewage sludge management: A case study of Taiwan. Journal of Cleaner Production, 129, 196–201. https://doi.org/10.1016/j.jclepro.2016.04.084

Climatesmartwater.org, 2018 Roadmap-to-a-Low-Carbon-Urban-Water-Utility-2018.pdf

CLIMFOOT, URL: https://www.climfoot-project.eu/, (accessed 21 June 2021)

Crippa, M., Gabriel, O., Diego, G., Marilena, M., Edwin, S., Eleonora, L. V., Efisio, S., Fabio, M.-F., & Jos, O. (2019). Fossil CO2 and GHG emissions of all world countries. In *Journal of Geophysical Research: Atmospheres* (Vol. 105, Issue D2). https://doi.org/10.2760/56420

Demir, Ö., & Yapıcıoğlu, P. (2019). Investigation of GHG emission sources and reducing GHG emissions in a municipal wastewater treatment plant. *Greenhouse Gases: Science and Technology*, *9*(5), 948–964. https://doi.org/10.1002/ghg.1912

EUROPEAN PARLIAMENT AND COUNCIL. (2000). *DIRECTIVE 2000/60/EC of 23 October 2000 establishing a framework for Community action in the field of water policy*.

Foley, J., de Haas, D., Hartley, K., & Lant, P. (2010). Comprehensive life cycle inventories of alternative wastewater treatment systems. *Water Research*, *44*(5), 1654–1666. https://doi.org/10.1016/j.watres.2009.11.031

Foley, J., de Haas, D., Yuan, Z., & Lant, P. (2010). Nitrous oxide generation in full-scale biological nutrient removal wastewater treatment plants. *Water Research*, *44*(3), 831–844. https://doi.org/10.1016/j.watres.2009.10.033

Gu, Y., Dong, Y. N., Wang, H., Keller, A., Xu, J., Chiramba, T., & Li, F. (2016). Quantification of the water, energy and carbon footprints of wastewater treatment plants in China considering a water-energy nexus perspective. *Ecological Indicators*, *60*, 402–409. https://doi.org/10.1016/j.ecolind.2015.07.012

Gustavsson, D. J. I., & Tumlin, S. (2013). Carbon footprints of Scandinavian wastewater treatment plants. *Water Science and Technology*, *68*(4), 887–893. https://doi.org/10.2166/wst.2013.318

Han, Z., Sun, D., Wang, H., Li, R., Bao, Z., & Qi, F. (2018). Effects of ambient temperature and aeration frequency on emissions of ammonia and greenhouse gases from a sewage sludge aerobic composting plant. *Bioresource Technology*, *270*(September), 457–466. https://doi.org/10.1016/j.biortech.2018.09.048

Huang, F., Shen, W., Zhang, X., & Seferlis, P. (2020). Impacts of dissolved oxygen control on different greenhouse gas emission sources in wastewater treatment process. *Journal of Cleaner Production*, *274*, 123233. https://doi.org/10.1016/j.jclepro.2020.123233

IPCC (2019). URL: https://www.ipcc.ch/report/ar5/syr/ (accessed 10 June 2021)

IPCC (2019a). *Chapter 5 Incineration and Open Burning*.

IPCC (2019b). *Guidelines for National Greenhouse Gas Inventories - Wastewater treatment and discharge*.

IPCC (2019c). N2O Emissions From Managed Soils, and CO2 Emissions From Lime and Urea Application. *2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories*, 1–48.

ISO 14064-1 (2019). *Greenhouse gases - Part 1: Specification for the quantification, monitoring and reporting of organization emissions and removals*.

ISPRA (2018). *Fattori di emissione atmosferica di gas a effetto serra e altri gas nel settore elettrico*.

Jin, P., Gu, Y., Shi, X., & Yang, W. (2019). Non-negligible greenhouse gases from urban sewer system. *Biotechnology for Biofuels*, *12*(1), 1–11. https://doi.org/10.1186/s13068-019-1441-8

Joss, A., Salzgeber, D., Eugster, J., König, R., Rottermann, K., Burger, S., Fabijan, P., Leumann, S., Mohn, J., & Siegrist, H. R. (2009). Full-scale nitrogen removal from digester liquid with partial nitritation and anammox in one SBR. *Environmental Science and Technology*, *43*(14), 5301–5306. https://doi.org/10.1021/es900107w

Kirkeby, J., Gabriel, S., & Christensen, T. H. (2005). *Miljøvurdering af genanvendelse og slutdisponering af spildevandsslam-en livscyklus screening af fire scenarier*. 53.

Kumar, A., Thanki, A., Padhiyar, H., Singh, N. K., Pandey, S., Yadav, M., & Yu, Z. G. (2021). Greenhouse gases emission control in WWTS via potential operational strategies: A critical review. *Chemosphere*, *273*, 129694. https://doi.org/10.1016/j.chemosphere.2021.129694

Longo, S., Mauricio-Iglesias, M., Soares, A., Campo, P., Fatone, F., Eusebi, A. L., Akkersdijk, E., Stefani, L., & Hospido, A. (2019). ENERWATER – A standard method for assessing and improving the energy efficiency of wastewater treatment plants. *Applied Energy*, *242*(March), 897–910. https://doi.org/10.1016/j.apenergy.2019.03.130

Longo, S., D'Antoni, B. M., Bongards, M., Chaparro, A., Cronrath, A., Fatone, F., Lema, J. M., Mauricio-Iglesias, M., Soares, A., & Hospido, A. (2016). Monitoring and diagnosis of energy consumption in wastewater treatment plants. A state of the art and proposals for improvement. *Applied Energy*, *179*, 1251–1268. https://doi.org/10.1016/j.apenergy.2016.07.043

Low-carbon economy - Regional Policy, 2014Low-carbon economy - Regional Policy - European Commission (europa.eu) Circular cities and regions initiative | European Commission (europa.eu)

Mannina, G., Ekama, G., Caniani, D., Cosenza, A., Esposito, G., Gori, R., Garrido-Baserba, M., Rosso, D., Olsson, G. (2016). Greenhouse gases from wastewater treatment - A review of modelling tools. *Science of the Total Environment*, 551-552, 254-270. http://dx.doi.org/10.1016/j.scitotenv.2016.01.163

Majumder, R., Livesley, S. J., Gregory, D., & Arndt, S. K. (2014). Biosolid stockpiles are a significant point source for greenhouse gas emissions. *Journal of Environmental Management*, *143*, 34–43. https://doi.org/10.1016/j.jenvman.2014.04.016

Maktabifard, M., Zaborowska, E., & Makinia, J. (2020). Energy neutrality versus carbon footprint minimization in municipal wastewater treatment plants. *Bioresource Technology*, *300*(December 2019), 122647. https://doi.org/10.1016/j.biortech.2019.122647

Mamais, D., Noutsopoulos, C., Dimopoulou, A., Stasinakis, A., & Lekkas, T. D. (2015). Wastewater treatment process impact on energy savings and greenhouse gas emissions. *Water Science and Technology*, *71*(2), 303–308. https://doi.org/10.2166/wst.2014.521

Masuda, S., Sano, I., Hojo, T., Li, Y. Y., & Nishimura, O. (2018). The comparison of greenhouse gas emissions in sewage treatment plants with different treatment processes. *Chemosphere*, *193*, 581–590. https://doi.org/10.1016/j.chemosphere.2017.11.018

Masuda, S., Suzuki, S., Sano, I., Li, Y. Y., & Nishimura, O. (2015). The seasonal variation of emission of greenhouse gases from a full-scale sewage treatment plant. *Chemosphere*, *140*, 167–173. https://doi.org/10.1016/j.chemosphere.2014.09.042

Nguyen, T. K. L., Ngo, H. H., Guo, W., Chang, S. W., Nguyen, D. D., Nghiem, L. D., Liu, Y., Ni, B., & Hai, F. I. (2019). Insight into greenhouse gases emissions from the two popular treatment technologies in municipal wastewater treatment processes. *Science of the Total Environment*, *671*, 1302–1313. https://doi.org/10.1016/j.scitotenv.2019.03.386

Pan, Y., Van Den Akker, B., Ye, L., Ni, B. J., Watts, S., Reid, K., & Yuan, Z. (2016). Unravelling the spatial variation of nitrous oxide emissions from a step-feed plug-flow full scale wastewater treatment plant. *Scientific Reports*, *6*(October 2015), 1–10. https://doi.org/10.1038/srep20792

Parravicini, V., Svardal, K., & Krampe, J. (2016). Greenhouse Gas Emissions from Wastewater Treatment Plants. *Energy Procedia*, *97*, 246–253. https://doi.org/10.1016/j.egypro.2016.10.067

Piippo, S., Lauronen, M., & Postila, H. (2018). Greenhouse gas emissions from different sewage sludge treatment methods in north. *Journal of Cleaner Production*, *177*, 483–492. https://doi.org/10.1016/j.jclepro.2017.12.232

Ribera-Guardia, A., Bosch, L., Corominas, L., & Pijuan, M. (2019). Nitrous oxide and methane emissions from a plug-flow full-scale bioreactor and assessment of its carbon footprint. *Journal of Cleaner Production*, *212*, 162–172. https://doi.org/10.1016/j.jclepro.2018.11.286

Samuelsson, J., Delre, A., Tumlin, S., Hadi, S., Offerle, B., & Scheutz, C. (2018). Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. *Water Research*, *131*, 299–309. https://doi.org/10.1016/j.watres.2017.12.018

Scheutz, C., & Fredenslund, A. M. (2019). Total methane emission rates and losses from 23 biogas plants. *Waste Management*, *97*, 38–46. https://doi.org/10.1016/j.wasman.2019.07.029

SINANET, URL: http://www.sinanet.isprambiente.it/it/sia-ispra/fetransp, (accessed 21 June 2021)

Spinelli, M., Eusebi, A. L., Vasilaki, V., Katsou, E., Frison, N., Cingolani, D., & Fatone, F. (2018). Critical analyses of nitrous oxide emissions in a full scale activated sludge system treating low carbon-to-nitrogen ratio wastewater. *Journal of Cleaner Production*, *190*, 517–524. https://doi.org/10.1016/j.jclepro.2018.04.178

Tumendelger, A., Alshboul, Z., & Lorke, A. (2019). Methane and nitrous oxide emission from different treatment units of municipal wastewater treatment plants in Southwest Germany. *PLoS ONE*, *14*(1), 1–17. https://doi.org/10.1371/journal.pone.0209763

Valkova, T., Parravicini, V., Saracevic, E., Tauber, J., Svardal, K., & Krampe, J. (2020). A method to estimate the direct nitrous oxide emissions of municipal wastewater treatment plants based on the degree of nitrogen removal. *Journal of Environmental Management*, *279*(April 2020). https://doi.org/10.1016/j.jenvman.2020.111563

Vasilaki, V., Massara, T. M., Stanchev, P., Fatone, F., & Katsou, E. (2019). A decade of nitrous oxide (N2O) monitoring in full-scale wastewater treatment processes: A critical review. *Water Research*, *161*, 392–412. https://doi.org/10.1016/j.watres.2019.04.022

Vieira, A., Marques, R., Raposo, R., Martins, M., Alves, R., Povoa, P., Irizar, I., Beltrán, S., Craamer, P., Urchegui, G., & Oehmen, A. (2019). The impact of the art-ICA control technology on the performance, energy consumption and greenhouse gas emissions of full-scale wastewater treatment plants. *Journal of Cleaner Production*, *213*, 680–687. https://doi.org/10.1016/j.jclepro.2018.12.229

WACCLIM, URL: http://wacclim.org/ecam/sources.php, (accessed 28 June 2021)

Wang, Y., Fang, H., Zhou, D., Han, H., & Chen, J. (2016). Characterization of nitrous oxide and nitric oxide emissions from a full-scale biological aerated filter for secondary nitrification. *Chemical Engineering Journal*, *299*, 304–313. https://doi.org/10.1016/j.cej.2016.04.050

WESTWEb, URL: https://west.berkeley.edu/model.php, (accessed 28 June 2021)

Willén, A., Rodhe, L., Pell, M., & Jönsson, H. (2016). Nitrous oxide and methane emissions during storage of dewatered digested sewage sludge. *Journal of Environmental Management*, *184*, 560–568. https://doi.org/10.1016/j.jenvman.2016.10.025

Yuan, J., Li, Y., Chen, S., Li, D., Tang, H., Chadwick, D., Li, S., Li, W., & Li, G. (2018). Effects of phosphogypsum, superphosphate, and dicyandiamide on gaseous emission and compost quality during sewage sludge composting. *Bioresource Technology*, *270*(September), 368–376. https://doi.org/10.1016/j.biortech.2018.09.023

Yver Kwok, C. E., Müller, D., Caldow, C., Lebègue, B., Mønster, J. G., Rella, C. W., Scheutz, C., Schmidt, M., Ramonet, M., Warneke, T., Broquet, G., & Ciais, P. (2015). Methane emission estimates using chamber and tracer release experiments for a municipal waste water treatment plant. *Atmospheric Measurement Techniques*, *8*(7), 2853–2867. https://doi.org/10.5194/amt-8-2853-2015

Zhan, X., Hu, Z., & Wu, G. (2017). Greenhouse Gas Emission and Mitigation in Municipal Wastewater Treatment Plants. In IWA (Ed.), *Water Intelligence Online* (Vol. 16). IWA Publishing. https://doi.org/10.2166/9781780406312