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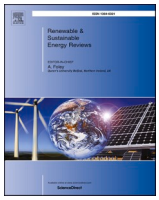

Net-zero carbon condition in wastewater treatment plants: A systematic review of mitigation strategies and challenges

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ABSTRACT

The wastewater sector accounts for up to 7 and 10% of anthropogenic CH_4 and N_2O emissions, respectively. Nowadays wastewater treatment plants are going through a paradigm shift to approach a net-zero carbon condition. Numerous ongoing measures have taken place to identify the sources of greenhouse gases and minimize the carbon footprint. This paper systematically reviews all known practices leading towards net-zero carbon wastewater treatment. The greenhouse gas emissions from the wastewater sector are identified and carbon footprint quantification tools, such as reliable models and emission factors are compared. The direct process emissions can contribute to over 60% of the carbon footprint in wastewater treatment plants, while around 30% of the carbon footprint is due to energy-related indirect emissions. Therefore, greenhouse gas mitigation via process optimization and energy usage in wastewater treatment plants are comprehensively described. The implantation of novel nitrogen removal processes can reduce both greenhouse gas emissions and energy consumption. Other techniques such as source separation systems can potentially allow mitigation of N_2O emissions by 60% while avoiding energy-intensive nitrogen fertilizer production. Nutrient recovery methods are another approach which offer negative value for the net carbon footprint. Recovering N_2O for energy production is a promising method which can lead to both direct and indirect carbon footprint reductions. Ultimately, to achieve full decarbonization any remaining emissions need to be offset, including carbon footprint of chemicals usage and transportation.

1. Introduction

The biological wastewater treatment process has historically been focused on efficient removing organic pollutants and nutrients to reduce the impact of wastewater on the aquatic environment. However, the wastewater sector is now undergoing a paradigm shift towards an integrated operation which is focused on resource recovery and net-zero carbon condition [1]. It is estimated that wastewater treatment plants (WWTPs) are responsible for nearly 5% of the global non-CO_2 greenhouse gas (GHG) emissions and this is projected to increase by 22% by 2030 [2,3].

GHG emissions from WWTPs are classified as either direct or indirect. The direct, non-biogenic GHG emissions, termed also scope 1 emissions, take place during wastewater and sludge treatment processes [4]. The direct CO_2 emissions are considered carbon neutral due to their biogenic nature, i.e., produced by the biodegradation of organic compounds in wastewater. This approach is in contrast with other sectors, such as transportation and energy, where the fossil-based CO_2 is the major contributor to GHG emissions. On the other hand, non-CO_2 direct emissions, including methane (CH_4) and nitrous oxide (N_2O), are of a significant concern due to a high global warming potential (GWP) of those gases. It is widely acknowledged that the wastewater treatment sector significantly contributes to CH_4 and N_2O emissions, estimated at 7–10% for each gas [5,6]. However, these percentages can vary depending on site-specific factors, such as treatment technologies, plant size, and regional differences in wastewater management practices. Based on the latest Intergovernmental Panel on Climate Change (IPCC) report [7], the GWP in a 100 years horizon is 27.2, 29.8 and 273 for non-fossil origin CH_4, fossil-origin CH_4 and N_2O, respectively.

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Nomenclature and abbreviations

Item	Description
AD	Anaerobic Digestion
ADM	Anaerobic Digestion Model
ANAMMOX	Anaerobic Ammonia Oxidation
AnAOB	Anaerobic Ammonia Oxidizing Bacteria
AOB	Ammonia Oxidizing Bacteria
ASM	Activated Sludge Model
ASMN	Activated Sludge Model for Nitrogen
BEAM	Biosolids Emissions Assessment Model
BNR	Biological Nutrient Removal
BOD	Biochemical Oxygen Demand
BW	Black Water
CANDO	Coupled Aerobic-Anoxic Nitrous Decomposition Operation
CE	Circular Economy
CF	Carbon Footprint
CH_4	Methane
CHP	Combined Heat and Power
CO_{2e}	Carbon Dioxide Equivalent
COD	Chemical Oxygen Demand
CWA	Clean Water Act
DAMO	Denitrifying Anaerobic Methane Oxidation
DAMO/A	Denitrifying Anaerobic Methane Oxidation-Anammox
DEFRA	Department for Environment, Food and Rural Affairs
DNRA	Dissimilatory Nitrate Reduction to Ammonium
DO	Dissolved Oxygen
DW	Dry Waste
ECAM	Energy Performance and Carbon Emissions Assessment and Monitoring
EF	Emission Factor
EPA	Environmental Protection Agency
EU	European Union
FOG	Fat-Oil-Grease
FTIR	Fourier Transformed Infrared Spectroscopy
FU	Functional Unit
FW	Food Waste
GC	Gas Chromatography
GEA	German Environmental Agency
GHG	Greenhouse Gas
GW	Grey Water
GWP	Global Warming Potential
HRT	Hydraulic Retention Time
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
LCA	Life Cycle Assessment
ML	Machine Learning
MLR	Mixed Liquor Recirculation
N	Nitrogen
N_2	Dinitrogen
N_2O	Nitrous Oxide
N_2OR	Nitrous Oxide Reductase
NH_2OH	Hydroxylamine
NH_4^+	Ammonium
NO	Nitric Oxide
NO_2^-	Nitrite
OFMSW	Organic Fraction of Municipal Solid Waste
P	Phosphorus
PANDA	Partial Nitrification, Denitratation and Anaerobic Ammonia Oxidation
PD	Partial Denitrification
PD/A	Partial Denitrification-Anammox
PE	Population Equivalent
PN/A	Partial Nitritation-Anammox
SBR	Sequencing Batch Reactor
SCENA	Shortcut Enhanced Nutrient Abatement
SCND	Shortcut Nitrification-Denitrification
SHARON	Single Reactor System for High Activity Ammonium Removal over Nitrite
SNAD	Simultaneous Partial Nitritation-Anammox and Denitrification
SRT	Solid Retention Time
TCD	Thermal Conductivity Detector
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
TSS	Total Suspended Solids
UK	United Kingdom
US	United States
VS	Volatile Solid
WEF	Water Environmental Federation
WFD	Water Framework Directive
WRRF	Water Resource Recovery Facility
WWTP	Wastewater Treatment Plant

The indirect GHG emissions, referred to as scope 2 emissions, are due to energy and electricity consumption by WWTPs. The remaining indirect emissions, referred to as scope 3 emissions, are associated with transportation and production of chemicals outside of the plant [2]. The total sum of GHG emissions from all the scopes in known as the carbon footprint (CF) and reported based on the carbon dioxide equivalent (CO_{2e}) unit [8].

In comparison with the transportation and energy sectors, the wastewater sector has fewer studies on net-zero carbon pathways [1]. However, many countries are beginning to move towards low-carbon operation and undertaking actions to decarbonize municipal water management [9]. Net-zero targets are included in near-term policies and plans, and the discussion on the net-zero condition has been evolving recently in line with the targets of the Paris Agreement [10].

Metcalf et al. [11] outlined criteria for a net-zero transition in wastewater treatment, including the overall energy balance, process-related GHG emissions, chemical consumption and sludge disposal. Therefore, the net-zero pathways for wastewater sector is more diverse and complex compared to other sectors. A multi-criteria approach that considers both wastewater reclamation and other sustainability goals need to be defined specifically for the wastewater sector [1]. The net-zero carbon condition refers to a prospective WWTP which aims to offset certain CF that is sum of scope 1,2 and 3 emissions in wastewater treatment and thus attain net-zero GHG emission. The ne-zero carbon WWTP employs novel strategies to mitigate N2O and CH4 emissions via process modification. Furthermore, this plant needs to mitigate CF via energy optimization as well as chemicals and transportation optimization. The number of scientific papers on the topic has increased rapidly in the last decade (Fig. 1), confirming growing attention from the scientific community.

This review aims to provide comprehensive insights into effectively reducing CF and achieving a net-zero carbon condition in municipal WWTPs. The paper covers crucial aspects, including relevant regulations, GHG sources, and quantification methods and tools for CF assessment. Process optimization, energy usage, and reduction and offsets of indirect emissions are discussed to minimize CF. Practices for net-zero carbon wastewater treatment and a multi-criteria approach to decarbonize WWTPs are thoroughly reviewed with an on environmental protection, resource recovery, and reducing impact. GHG emissions in WWTPs, particularly N_2O and CH_4, and their sources are explored.

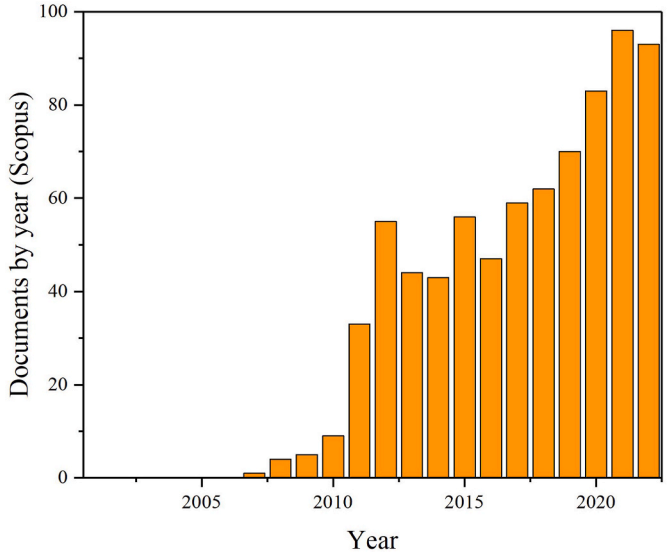

Fig. 1. Number of publications covering CF of WWTPs documented in the Scopus database since 2007 (analysis based on the keywords: CF and wastewater).

Methods for estimating the emissions and effective mitigation strategies are also discussed. Energy optimization in WWTPs is addressed, emphasizing reduced consumption and increased production through co-digestion and biogas utilization. The review explores the optimization of chemical dosages and transportation to minimize emissions. Additionally, the study investigates the potential of recovering N_2O from wastewater as an energy source. The findings of study carry significant implications, supporting climate change mitigation, environmental protection, and sustainable wastewater treatment with a reduced CF. The review underscores the need for comprehensive strategies to achieve net-zero carbon conditions in municipal WWTPs.

2. Evolving wastewater regulations and recommendations towards a net-zero carbon condition

Fig. 2 presents a summary of historical regulations and recommendations in the wastewater sector and outlines the recent trends that would affect the sustainability of wastewater treatment. For a long time, the regulations focused on protecting the aquatic environment, while only recently the efficient use/recovery of resources and reducing the impact on the entire environment have been postulated.

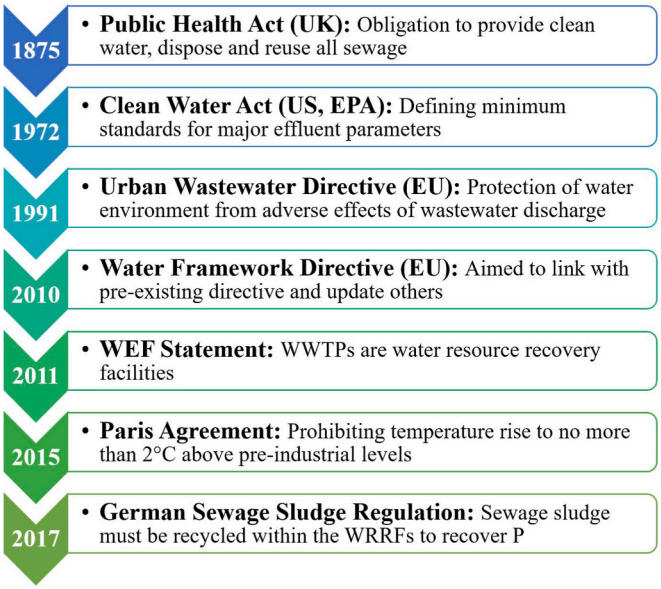

Fig. 2. Summary of significant wastewater management regulations/recommendations.

2.1. Protection of the aquatic environment

Based on the Public Health Act established in United Kingdom (UK) in 1875, the authorities were obliged to provide clean water, dispose and reuse all sewage. A century later (1972), the Environmental Protection Agency of United States (US EPA) published its definition of the minimum standards for secondary treatment, including major parameters, such as total suspended solids (TSS) and biochemical oxygen demand (BOD). A few of US EPA regulations brought changes in planning and designing WWTPs, including the Clean Water Act (CWA) (1972) that defined the minimum standards for discharging wastewater. Further bio-solids regulations were set in 1993 for pathogen and heavy metal content as well as safe handling and use of bio-solids [11].

Meanwhile, across the European Union (EU), the first directive concerning municipal wastewater treatment (91/271/EEC) was adopted in 1991 to protect the aquatic environment from unfavorable effects of municipal wastewater discharge. In 1998, another directive (98/15/EC) clarified the requirements in relation to the discharges from municipal WWTPs to areas which are potentially subject to eutrophication [12].

In the beginning of 21st century, the EU Water Framework Directive (WFD) was adopted, establishing a framework for water policy (2000/60/EC). The WFD established water quality standards and discharge controls to mitigate the impact of human activities on surface water quality [13]. In 2015, the United Nations set a global target recommendation through its sustainable development goals to reduce untreated wastewater by half by the year 2030 [14].

2.2. Efficient reuse and recovery of resources

In 2011, Water Environmental Federation (WEF) stated "*WWTPs are not waste disposal facilities, but rather water resource recovery facilities that produce clean water, recover nutrients (such as phosphorus (P) and nitrogen (N)), and have the potential to reduce the nation's dependence upon fossil fuel through the production and use of renewable energy*" [15]. *In 2012, WEF officially started using the novel term of water resource recovery facility (WRRF), instead of WWTP* [16].

In 2017, according to the new German Environmental Agency regulation (GEA) [17], sewage sludge must be recycled to recover P in plants with population equivalent (PE) over 50,000. The regulation is aimed at closing the P cycle and replacing over 50% of the imported P.

2.3. Reducing impact on the entire environment

The landmark Paris Agreement [18] aimed at limiting the global warming by prohibiting the temperature rise <2 °C above the pre-industrial levels, while IPCC [7] encouraged to follow even more ambitious goal leveled at 1.5 °C. In order to achieve this milestone, countries are aiming to achieve net-zero carbon condition by 2050. In 2019, the European Green Deal was introduced, which re-evaluated the EU's commitment to the net-zero concept with the introduction of the 'Fit for 55' package. The package outlines the path towards carbon-neutral transition, including short-term targets for 2030, such as a minimum reduction of 55% in GHG emissions compared to the 1990 level [19].

According to the International Energy Agency (IEA), developing and deploying clean energy technologies is crucial for achieving net-zero carbon conditions [20]. In practice, achieving net-zero globally will require a two-pronged approach, i.e. removing carbon from the atmosphere and restricting human-produced emissions. Based on the IPCC [7] definition, the net-zero carbon entails balancing anthropogenic GHG

emissions with their intentional removal from the atmosphere. Although the proposed net-zero pathways can be found in various sectors, no framework for the wastewater sector has been complied yet.

Municipal WWTPs can be a significant element of circular economy (CE) due to incorporation of resource recovery and energy production, while minimizing the environmental impact on the aquatic environment. Salminen et al. [21] presented a concept of a water-smart CE which would decrease energy and water loss and recover valuable resources and reuse wastewater. The authors suggested taxation of water abstraction and tax relief on the recycled materials and instruments targeting pollutants in wastewater.

To proactively respond to climate change and reduce GHG emissions, China committed in 2020 to peak carbon emissions by 2030 and achieve carbon neutrality by 2060, known as the "dual carbon" goal [22]. According to the European Environment Agency, the EU's GHG emissions fell by 23.2% since 1990. The EU's global contribution declined from 15% to 8% between 1990 and 2018. Despite this, the EU aims to cut carbon emissions by 55% by 2030 (compared to 1990 level) and achieve carbon neutrality by 2050 as per their new policy announcement [23]. Under the Paris Agreement, Australia also set its target to reduce GHG emissions in 2030 by 26–28% from 2005 level and achieve net-zero emission by 2050 [24]. Germany and the UK led 50% of the EU's GHG emission reduction from 1990 to 2019. Romania, France, Italy, Poland, and the Czech Republic contributed 33% of the EU's total reduction during that period. Over 29 years, the UK and Germany were responsible for nearly half of the overall net reduction in the EU-KP. In 2019, Germany, Spain, and Poland played a significant role in the EU-KP's net GHG reduction, accounting for over half of the total absolute reduction [25]. In the same year, the Netherlands' GHG emissions dropped by 3.2% compared to 2018, with total emissions approximately 18% lower than the 1990 levels [26].

3. Identification of sources of GHG emissions in wwtps

The largest contribution to the total CF in WWTPs comes from scope 1 CF, which is due to the direct fugitive emissions from wastewater treatment and anaerobic digestion (AD) processes. A few studies which are presented in Table 1 [4,27,28] reported that the direct emissions contributed to over 60% of the total CF.

The indirect energy-related emissions, which are caused by the electricity purchased from the grid, contributed on average to the total CF with a lower share of approximately 20%. Those emissions play a minor role especially in the countries, such as Finland and Austria, that have a high share of renewable energy sources [27,28]. In contrast, Hu et al. [29] analyzed the CF of WWTPs in China and found a lower share of 29% for the direct emissions, while 45% of the total CF was related to sludge handling. This high share resulted from a large number of studied WWTPs performing sludge incineration and landfilling. In the case of plants without on-site electricity production (via e.g., biogas production through AD and energy co-generation), a dominant percentage share can also be attributed to the indirect emissions [30]. The indirect emissions related to chemicals consumed at the plant and transportation showed a marginal effect on the CF of WWTPs [4].

Kadam et al. [31] presented a discussion on achieving carbon neutrality in municipal WWTPs based on feasible technical aspects. Their proposed plant incorporates various advanced primary treatment techniques, leading to efficiently organic recovery and reducing the oxygen demand for oxidation of organic compounds. Consequently, biogas production during biological processes significantly increases. The optimized approach also involves converting concentrated primary sludge to biomethane through anaerobic digestion. Additionally, H_2 gas derived from N upgrades plays a crucial role in enhancing biomethane quality by reducing CO_2 content in the biogas. These findings demonstrate the potential of municipal WWTPs to achieve high process efficiency and energy utilization. To achieve a net-zero carbon condition, a hybrid system integrating both GHG emission reduction solutions and energy recovery is suggested [32]. Specific solutions for the three scopes of emissions are discussed in Sections 5 to 7.

Wu et al. [33] performed a comprehensive CF analysis of different wastewater treatment configurations. The authors reported that scope 1 emissions accounted for 23–83% of the total CF in different configuration scenarios. Scope 1 emission could be the primary contributor to CF for configurations with anaerobic processes. This is due to the reduction in scope 2 and scope 3 emissions resulting from energy recovery and reduced sludge production. Recent studies by Yao et al. [34] reported N_2O emissions alone to exceed 50% of the total CF based on site-specific monitoring of BNR WWTPs employing such configurations as A/O, anaerobic-anoxic-oxic (A_2O), and SBR. The contribution of scope 2 emissions is also highly variable and can range from 14 to 68% of the total CF of WWTP (Table 1). Scope 2 emissions could be minimized by reducing the energy consumption, but most wastewater treatment configurations with relatively low GHG emissions involve energy recovery, which offsets the scope 2 emissions. Scope 3 emissions play a minor role in the overall GHG emissions in WWTPs, contributing only 1–13% to the overall CF.

3.1. WWTP emission hotspots

Fig. 3 shows the potential sources of different GHGs that are emitted directly and indirectly. The preliminary stage of wastewater treatment is known as a potential source of CH_4 emissions. Anaerobic conditions often prevail in sewer systems, and intense CH_4 fluxes have been

Table 1
CF distribution in WWTPs - overview.

Reference -*number of studied plants*	Contribution to the total CF (%)										
	Scope 1					Scope 2	Scope 3				
	Bioreactor	AD	Recipient	Sludge handling	Total		Grit and screening handing	Chemicals	Sludge handing (3rd parties)	Transport	Total
Parravicini et al. [27] – *2 WWTPs*	42.9	26.3	9.1	–	78.4	15.5	–	4.2	–	1.7	5.8
Hu et al. [29] – *344 WWTPs*	29		–	–	29	26	–	–	45	–	–
Maktabifard et al. [30] – *6 WWTPs*	58	3	1	10	72	26	1	1	–	–	2
Awaitey [28] – *4 WWTPs*	52	1.5	2	11	66.5	18	0.5	8	–	7	15.5
Min	29	3	0	0	29	15.5	0	0	0	0	0
Max	58	26.3	9.1	11	78.4	26	1	8	45	7	15.5
Average	45.5	10.3	4	10.5	61.5	21.4	0.8	4.4	15	2.9	17.1
Deviation	10.9	11.3	3.6	0.5	19.2	4.7	0.3	2.8	21.2	2.9	16.8

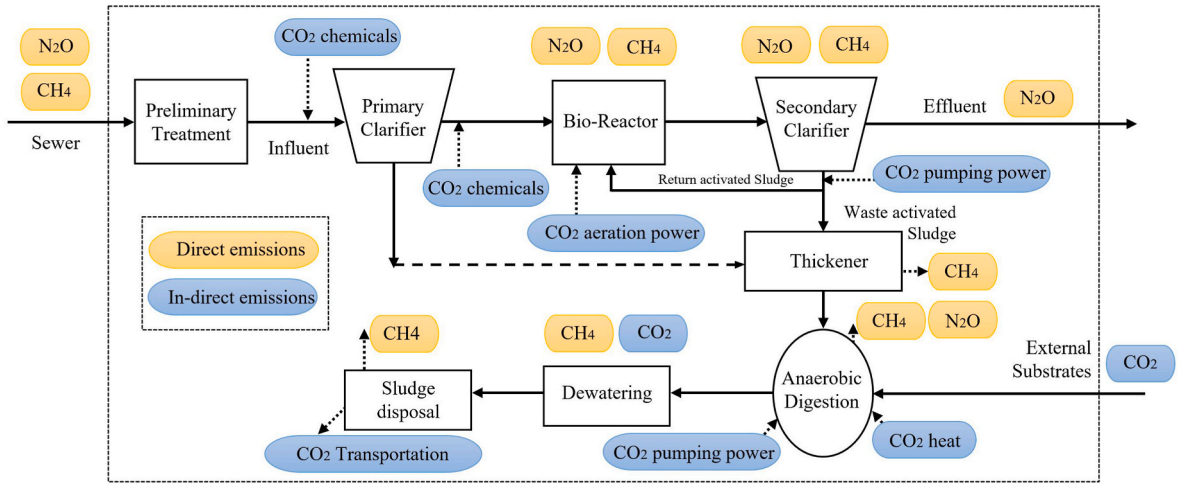

Fig. 3. Sources of the direct and indirect GHG emissions in WWTPs.

reported at the headworks of the WWTP, especially in aerated grit chambers. Other potential CH_4-forming spots are primary clarifiers, and then anaerobic compartments in the biological reactors. CH_4 can subsequently be stripped in the aerobic compartments of the reactors [35]. Due to the leakage of gases from internal facilities in the plant, biogas production installations may become significant CH_4 sources. Aerobically stabilized sludge, when stored for several months, can also contribute to CH_4 emissions due to the fact that anaerobic conditions start to prevail in the long-term perspective [36]. The effluent BOD load can contribute to further direct CH_4 emissions from the natural processes in recipients [27].

The GHG emissions from the preliminary treatment and sludge treatment stages do not attract as much attention as the emissions from the biological stage employing biological nutrient removal (BNR). Bioreactors have been recognized as the main source of GHG emissions in WWTPs. Campos et al. [37] reported that, the dominant N_2O production occurs in the bioreactors (90%) while for CH_4 emissions the sludge line with AD was responsible for over 70% and the remaining portion originates from bioreactors. N_2O produced under favorable conditions (either anoxic or aerobic) is emitted via either saturation-induced liquid-gas transfer or stripped to the air via aeration [38]. The effluent N load also becomes the source of direct N_2O emissions in the recipients.

3.2. N_2O emissions

WWTPs are considered significant anthropogenic sources of atmospheric N_2O, contributing 3–10% of the total emissions [5]. The latest IPCC report [7] shows a progressive increase in the atmospheric N_2O concentration, which has been levelled at the rate of 0.95 ppb/year for the last decade (2010–2019). The report emphasizes that wastewater treatment is the fourth largest sector responsible for N_2O emissions. The N_2O emissions from this sector increased from 0.2 Tg N/year to 0.35 Tg

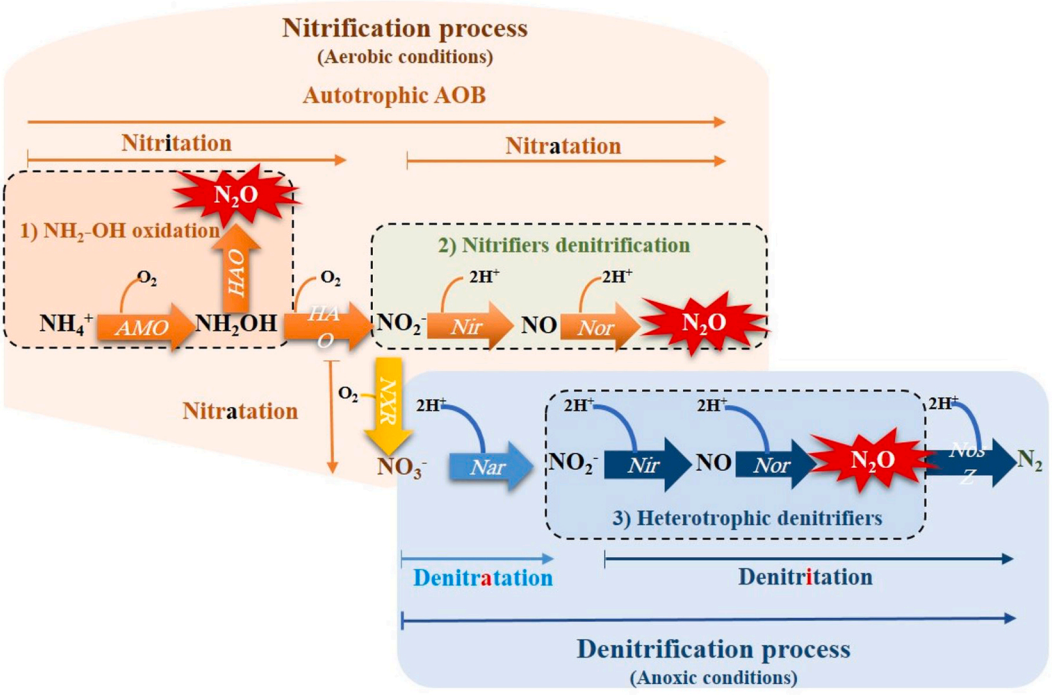

Fig. 4. Biological pathways of N_2O production and sink in the bioreactors (responsible genes: *AMO*: ammonia monooxygenase; *HAO*: hydroxylamine oxidoreductase; *NXR*: nitrite oxidoreductase; *Nar*: nitrite oxidoreductase; *Nir*: nitrite reductase; *Nor*: nitrite reductase; *NosZ*: Nitrous oxide reductase).

N/year between the 1980s and 2010s.

The N_2O emissions occur dynamically and normally remain beyond the control of the plant operators. Even small changes, such as dissolved oxygen (DO) variations, can significantly affect the liquid N_2O production by microorganisms [3].

3.2.1. Identification of the dominant pathways

N_2O production pathways involve three microbiological reactions, which require either aerobic or anoxic conditions (Fig. 4). In two pathways mediated by ammonia oxidizing bacteria (AOB), N_2O can be an intermediate product of hydroxylamine (NH_2OH) oxidation and the final product of autotrophic denitrification. Moreover, N_2O is an intermediate product of denitrification by heterotrophs. If that process is not disturbed, N_2O is further reduced to dinitrogen (N_2) in the final step of denitrification and this pathway can thus be a sink of N_2O [39]. Based on a literature review of N_2O emissions from WWTPs, Vasilaki et al. [39] formulated four main operational factors responsible for liquid N_2O production in the conventional nitrification-denitrification systems, including (i) low DO conditions, nitrite (NO_2^-) accumulation and changes in the ammonium (NH_4^+) concentration in aerobic compartments, (ii) low chemical oxygen demand (COD) to N ratio and NO_2^- accumulation in anoxic bioreactors, (iii) alternation of the anoxic to aerobic environments in intermittent compartments, (iv) sudden fluctuations (shocks) in the processes conditions.

Diverse contributing factors and dynamic reactions can occur simultaneously in the bioreactors, which are beyond operational control in full-scale WWTPs. Furthermore, heterotrophic denitrification can consume liquid N_2O produced via the different pathways. Regardless of the various production and consumption pathways of liquid N_2O, the stripping process should be considered as the process leading to N_2O emissions.

3.2.2. Additional sources of N_2O emissions in WWTPs

Although the direct N_2O process emissions released from bioreactors has the highest contribution to the total CF of WWTPs, a minor fraction of N_2O is also emitted from other units of the plant. Hwang et al. [40] performed N_2O measurements in different units of a WWTP, including the primary clarifier and secondary clarifier, as well as the sludge thickener and anaerobic digester. Small quantities of N_2O (0.012 g N_2O/kg TN) were produced in the digester, accounting for less than 1% of the total N_2O emissions. Regarding the primary and secondary clarifier, N_2O emission factors (EFs) of 0.22–0.26 g N_2O/kg TN were reported based on the full-scale measurements of Hwang et al. (2016). For comparison, Solís et al. [41] applied an EF of 0.01 kg N_2O–N/kg TN to predict the direct N_2O emissions from sludge storage, assuming uncovered storage throughout the year. Another study by Caniani et al. [42] found interesting results in the disinfection unit, observing high N_2O EF of 0.008 kg CO_{2e}/kg COD, due to the interaction between disinfecting agent and NH_2OH.

3.3. CH_4 emissions

The latest IPCC [7] report shows a progressive increase in the atmospheric concentration of CH_4, which has been levelled at the high rate of 9.3 ppb/year in the last decade. On a global scale, the wastewater sector accounts for approximately 5–7% of anthropogenic CH_4 emissions, ranking fifth after livestock (32%), oil and gas (25%), landfills (13%), and coal mining (11%) [43]. In the United States, CH_4 emissions from wastewater treatment increased from 10% to 14% between 1990 and 2019 [44]. A recent study by Song et al. [45] utilized the updated data from municipal WWTPs in US and estimated 10.9 ± 7.0 MMT CO_{2e}/year of annual CH_4 emissions which is about the double amount of emissions estimated by IPCC [36] (4.3–6.1 MMT CO_{2e}/year).

CH_4 is mainly produced during the anaerobic degradation of organic matter in bioreactors and digesters. The effectively utilized CH_4 can assist the plant to move towards energy neutrality and indirectly offset CO_2 emissions. On the other hand, CH_4 would increase the plant CF when emitted through incomplete combustion or leakage [46].

Studies at a regional level are usually based on CH_4 EFs [47]. Case studies of CH_4 emissions at the plant level are less frequent in the literature [48]. Zhao et al. [49] found that the CH_4 emissions from municipal WWTPs in China were over three times higher compared to the US plants. Although an empirical approach is highly uncertain, it is still a suitable way to estimate CH_4 emissions from WWTPs. Zhang et al. [6] concluded that it would be crucial to perform extensive measurements under various treatment processes, scales, and locations to obtain more accurate CH_4 emission predictions.

A full-scale study of Ribera-Guardia et al. [50] reported the long-term CH_4 emissions from aerobic compartments of a plug-flow bioreactor. The peak CH_4 emissions were found in the first aerobic zone, while decreasing towards the end of the bioreactor. The authors assumed that CH_4 was produced under anaerobic conditions in the initial stages of the plant and then stripped in the aerobic compartment of bioreactors. The highest liquid CH_4 concentrations were detected in the plant influent (0.55 mg CH_4/l) and reject water from the anaerobic digesters (0.52 mg CH_4/l).

Hwang et al. [30] determined the CH_4 EFs in different units of a municipal WWTP in South Korea. The sludge thickening process produced the largest CH_4 emissions (2.09 g CH_4/kg BOD) considerably more than aeration basin (0.72 g CH_4/kg BOD), primary clarifier (0.26 g CH_4/kg BOD) and secondary clarifier (0.67 g CH_4/kg BOD). CH_4 was primarily released from the digesters with a high CH_4 EF of 227 g CH_4/kg BOD. Wei et al. [51] analyzed four scenarios of sludge treatment and disposal. The highest contribution to CH_4 emissions was incineration (45.1%), followed by sanitary landfills (23%), land utilization (17.7%), and building materials (14.2%).

In a plant-wide model, implemented by Solís et al. [52], CH_4 emissions were calculated by a modified Anaerobic Digestion Model No. 1 (ADM1) [53]. Fugitive emissions were included as 2.7% of the produced biogas which was un-combusted in combined heat and power (CHP) units or slipped from anaerobic digester. The remaining amount of biogas was assumed to be completely combusted and CH_4 was fully converted to CO_2 while generating heat and electricity. It was assumed that CH_4 from the digester effluent was fully stripped to the atmosphere. Furthermore, the direct emissions due to sludge storage were estimated at 8.7 kg CH_4 per ton of volatile solids (VS). Overall, for the studied WWTP with the flow rate of 21,000 m^3/d, the CF was 19,000 kg CO_{2e}/d and CH_4 emissions contributed 5.8% to the total CF.

4. Estimation methods of GHG emissions

4.1. GHG emissions inventories

European inventory: The annual EU GHG inventory 1990–2019 report [54] contains the CH_4 and N_2O emissions from the wastewater sector. Both N_2O and CH_4 emissions account for 0.6% of the total EU GHG emissions in 2019 with 35.5 Mt CO_{2e} (domestic wastewater) and 9.3 Mt CO_{2e} (industrial wastewater). Between 1990 and 2019, the total emissions from wastewater treatment in EU decreased by 43.7%. Due to the application of new wastewater treatment processes, CH_4 and N_2O emissions from domestic wastewater decreased by approximately 50% and 17%, respectively [54].

US inventory: According to the US GHG Emissions and Sinks 1990–2020 Inventory Report [55], the wastewater treatment sector significantly contributed to the total N_2O country-wide emissions. It was the second largest anthropogenic source of N_2O emissions in 2020, levelled at 23.5 Mt CO_{2e}.

The wastewater sector contributed to 5.5% of N_2O fluxes, which resulted in 0.4% of the total GHG emission in the US. Comparing to 1990, a 41.8% increase was reported due to the country's growing population and a peaking protein consumption. The wastewater sector was ranked seventh of CH_4 emission sources with its 18.3 Mt CO_{2e}

fluxes. During the period of 1990–2002, CH_4 emissions remained stable, but a decreasing trend was observed later due to a reduction in the use of on-site septic systems [55].

Australian inventory: In the quarterly updated national inventory GHG (Australia's Greenhouse Accounts [56]), the levels of ozone layer depleting gases are given for different sectors, including wastewater handling. In 2010, wastewater treatment contributed to the total Australia's emissions in 0.51%, reflecting 0.8 Mt CO_{2e}. After a decade, the emissions increased to 0.9 Mt CO_{2e} constituting 0.72% of the total GHG emissions in the continent.

Chinese inventory: The most recent Chinese national GHG report from 2014 revealed that N_2O emissions from WWTPs were 1.1 Mt of N_2O, accounting for 5.6% of the national N_2O emissions. CH_4 emissions from wastewater treatment were 2.7 Mt of CH_4, accounting for 4.9% of China's total CH_4 emissions [6].

4.2. Direct measurements

There are various methods on *in-situ* measurements of GHG emissions that can be applied in WWTPs. In hermetic facilities, the direct GHG emissions can be analyzed continuously in the ventilation air by combining the concentration results with the airflow parameters [35]. In the case of open-to-air plants, collection of flux gas requires a dedicated sampling technique. In order to aggregate samples in open-surface tanks, different gas hoods and flux chambers are installed in multiple measurement points to address spatial and temporal variability along the wastewater train. In both cases, liquid N_2O concentrations should be simultaneously examined [57] and they need to be complemented by the analysis of conventional qualitative and quantitative operating parameters. The analytical techniques for measuring GHG concentration in collected flux-gas samples include Fourier Transformed Infrared Spectroscopy (FTIR) or Gas Chromatography with a Thermal Conductivity Detector (GC TCD).

Duan et al. [58] provided a critical review on using isotope technology for direct N_2O measurements, including both natural abundance and labelled isotope approaches. However, the authors emphasized that the accuracy and reliability of these techniques requires further improvement.

In order to reliably validate N_2O emission models, data from full-scale measurement campaigns are essential. However, the number of plants which measure N_2O emissions is currently very limited [59]. The long-term (yearly) N_2O monitoring data are available in the literature for Viikinmäki WWTP in Helsinki [60] and Kralingseveer WWTP [35].

4.3. CF calculation tools

General requirements that are dedicated to CF calculation can be found in International Organization for Standardization (ISO) 14,064 standard [61]. The IPCC (2006) with 2019 refinement for the national GHG inventories [36] is a frequently applied guidance note for the estimation of GHG emissions from WWTPs.

Life cycle assessment (LCA) is a broad technique to systematically evaluate multiple environmental impacts associated with municipal water cycle infrastructure, including wastewater collection and treatment [62]. The CF is part of LCA that quantifies a negative impact on the climate change. The LCA can be an accurate tool for calculating the indirect GHG emissions including high level of details in inventory data [63]. The direct GHG releases to air from the wastewater and sludge lines of WWTPs are incorporated via additive calculation procedures, often supported by secondary data estimations sourced from, either empirical or mechanistic models as well as in-situ measurement campaigns [63].

Several tools are available specifically for the CF assessment of WWTPs, usually as open-source files or applications. These are designed to deliver CF results based on the user's data input comprising operational parameters of the analyzed WWTP or its part. Table 2 shows an overview of CF calculation tools specifically dedicated to WWTPs. The Carbon Footprint Calculation Tool (CFCT) [64] has been utilized frequently in the literature [4,30] which is an adjustable spreadsheet developed originally for Swedish case studies [65] and has been updated since then.

Table 2
Overview of the CF assessment tools for WWTPs.

	Tool name (if available)	Country of origin	Year	Methodology	Open source
1	Quantifying the GHG emissions of WWTPs (as part of a thesis project)	Netherlands	2009	ISO	YES - source code for MatLab
2	Biosolids Emissions Assessment Model (BEAM)	Canada	2010–2011	IPCC, WRI	YES - Excel
3	Worldbank Organization - Sustainable Urban Energy and Emissions Planning Toolkits - Energy Balance and GHG Inventory Spreadsheet	USA	2011–2012	IPCC	YES - Excel
4	Waste Sector GHG Protocol Calculation Tool	France	2013	GHG Protocol	YES - Excel
5	Energy Performance and Carbon Emissions Assessment and Monitoring (ECAM)	Germany	2015	IPCC	YES - online
6	GESTABoues for wastewater treatment sludge management	France	2016	Own - based on literature	YES - after free registration
7	C-FOOT-CTRL	UK/Greece	2019	Own - based on literature	NO
8	Queensland Water GHG Calculator	Australia	2010–2019	National Greenhouse and Energy Reporting Standard	YES - excel
9	Calculation of the CF from Swedish WWTPs (SVU 12–120)	Sweden	2012–2021	Own - based on literature	YES - excel

When planning the CF calculation for a WWTP, two main approaches for setting boundary conditions of the CF analysis can be applied. The first method covers a full life cycle of a WWTP within the construction, operation and demolishment stages [66]. Another method would be an in-depth analysis of the operation stage with detailed considerations of process emissions [4].

Overlooking the construction and demolition phases of WWTPs and supporting buildings is a common simplification in LCA. The impacts of these phases are usually considered insignificant compared to the emissions from the operational stage [67]. The inclusion of new aspects in WWTPs, such as energy and resource recovery, would broaden the

scope of carbon accounting. Therefore, Li et al. [46] proposed a system expansion approach, while assuming that the products recovered from wastewater, such as struvite, vivianite, biodiesel, bioplastics, biochar and protein, should substitute similar products on the market. This would result in offsetting carbon emissions from the system.

4.4. Empirical EFs

Simple empirical models are the most common approach to estimate N_2O emissions from wastewater and N_2O EFs are advantageous for providing a better insight into the WWTPs CF. It is important to note that the specific functional units (FUs) used to report N_2O EF might vary in the literature. The influent total Kjeldahl nitrogen (TKN) load was used most frequently, but the total removed N load or influent NH_4 load have occasionally been reported in the literature as well (Table 3).

The latest IPCC guidelines [7] recommended 1.6% kg N_2O/kg influent N-load as the EF based on the results of various monitoring campaigns. The reported N_2O EF for other processes, such as nitrite "shunt" or deammonification, are typically higher than full nitrification/denitrification. For example, Gustavsson and La Cour Jansen [68] estimated 6% kg N_2O/kg $NH_{4\ removed}$, EF for the nitritation process, while the EF for partial nitrification anammox process was 3% kg N_2O/kg $N_{\ removed}$ [69].

Continuous, long-term monitoring campaigns can substantially improve the N_2O EF accuracy [70]. A high inconsistency in the annual EFs (0.1–8% of the N load) was found after monitoring fourteen full-scale WWTPs in Switzerland, but the EFs were strongly correlated with the effluent NO_2^- concentrations. The authors proposed a national EF calculated from the weighted (weights based on the fraction of N loading in the country-scale) EFs of carbon removal (EF: 0.1–8% depending on the expected variability in plant performance), nitrification only (EF: 1.8%), and full N removal (EF: 0.9%). This approach allowed to estimate country-specific N_2O emissions from WWTPs. In Switzerland, the average EFs and total annual N_2O emissions ranged between 0.9 and 3.6% and 410–1690 ton N_2O (corresponding to 0.3–1.4% of the total GHGs), respectively [70].

The fixed EF approach neglects the impact of operational conditions and wastewater characteristics on N_2O emissions. Valkova et al. [3] suggested that the N_2O EF by IPCC overestimates N_2O emissions for the plants with the high efficiency of total nitrogen (TN) removal. The authors proposed that the annual direct N_2O emissions can be predicted based on the annual average TN removal efficiency. In ten Austrian WWTPs with high N removal efficiency (83–92%), the N_2O EF of 0.12% $\pm$ 0.1% kg N_2O/kg TKN was found. The N_2O EFs do not capture emission dynamics, potentially leading to overestimation or underestimation of emissions [71]. To improve the accuracy of N_2O emissions and CF assessment in process optimization and mitigation strategies for individual WWTPs, local measurements and mathematical models are preferred over fixed EFs.Table 3 shows significant variations in N_2O EFs depending on the estimation methodology, plant location, and study conditions. The scale of estimation, from global inventories through individual facilities, also impacts the results. Nayeb et al. [72] found high uncertainty in the CF estimation using national-scale inventories. Maktabifard et al. [30] demonstrated over 50% uncertainty in total CF estimation when considering a wide range of N_2O EFs from literature. High uncertainties were especially observed in the facilities dominated by the direct emissions.

Table 3
Measured and estimated N_2O EFs from WWTPs in different studies.

Reference	N_2O EF benchmark			Type of study
	kg N_2O/kg TKN	kg N_2O/kg $N_{removed}$	kg N_2O/kg $NH_{4\ removed}$	
Solís et al. [41]	0.55%			Practical modelling
Li et al. [73]				Practical modelling
Maktabifard et al. [59]	0.94%			Practical modelling
Zaborowska et al. [74]	1.6%			Practical modelling
Domingo-Félez and Smets [75]			1.2–4.6%	Theoretical modelling
Massara et al. [76]	1–11%			Theoretical modelling
Pocquet et al. [77]			1%	Theoretical modelling
Ni et al. [78]	0.69–3.5%			Practical modelling
Foley et al. [79]		1.57%		LCA
Gruber et al. [80]	1–2.4%			Long term monitoring campaign
Sun et al. [81]	0.2–1.6%			Sampling measurements
Kosonen et al. [60]	1.9%			Long term monitoring campaign
Wang et al. [82]	0.095–3.44%			Online measurements
Hwang et al. [40]	1.606%			Sampling measurements
Duan et al. [83]	0.58%			Long term monitoring campaign
IPCC [36]	1.6%			Inventory report
EEA [84]	0.035%			Inventory report
NGER [85]	0.7–0.8%	1%		Inventory report
WERF [86]		0.01–2.5%		Inventory report
IPCC [87]		0.32–0.5%		Inventory report

4.5. Mathematical models

Mechanistic models are considered powerful tools for determining liquid N_2O production. These models can help identify the dominant N_2O production pathways and mitigation strategies. In comparison with empirical models, the use of mechanistic models may significantly reduce the uncertainty of the total CF results, as the CF results are highly sensitive to N_2O emissions [59].

Single-pathway (AOB denitrification/NH_2OH oxidation) and two-pathway models have been suggested for N_2O production by AOBs, while heterotrophic denitrification could be modelled as three or four-step process [88]. These models are added as extensions to existing activated sludge models (ASMs). For example, Kim et al. [89] combined the ASM for Nitrogen (ASMN) [90] and ADM1 [53] in the ASMN for GHGs (ASMN_G). Blomberg et al. [91] extended the ASM3 [92] with a dynamic N2O prediction and implemented the model in a full-scale WWTP in Finland. Zaborowska et al. [93] extended the ASM2d (ASM2d-N_2O) and included both N and P removal processes to model a full-scale WWTP in Poland. Research attention has primarily focused on modelling N2O emissions, but efforts are being made to develop models considering CO_2 and CH_4 emissions from wastewater (ASMs) and sludge treatment (ADMs). Mannina et al. [94] proposed a simple mass balance-based model with N_2O from nitrification and denitrification. A modified Benchmark Simulation Model No. 2 (BSM2) incorporated GHG emissions were considered (N_2O and CO_2 from AS, CO_2 and CH_4 from AD) along with the indirect emissions from the electricity and chemicals [95,96]. These models investigated the influence of different operational scenarios on the effluent quality, operational cost and GHG emissions. The DO control in the aerobic zone was a commonly studied strategy for GHG mitigation strategy. Other model-based studies considered the influent COD/N ratio [94], TSS control in the primary clarifier and CEPT [95,97], MLR ratio [96] and sidestream deammonification [74]. The advantage of the modelling approach was the possibility to take into account the processes interactions and identification of

trade-offs between conflicting objectives [98]. In the approach proposed by Arnell et al. [97], the plant-wide model was combined with LCA to account for the global environmental impact due to external resource use.

The models of N_2O production/consumption should be integrated with a stripping model to predict N_2O emission. Baresel et al. [99] initially applied the saturation-induced liquid-gas transfer and the stripping N_2O emission model in the anoxic and aerobic compartments, respectively. These models were adopted in several subsequent studies [59,91,93].

The inclusion of all the known N_2O production pathways may result in over parameterized complex models. These models require more calibration efforts due to a higher number of coefficients. Moreover, calibration of the models by using short-term measurement data would reduce the accuracy and increase the uncertainty of the model [59].

Although mechanistic models have explicit advantages in determining N_2O mitigation strategies for WWTPs, their practical use in GHG inventories is still limited. Another emerging mathematical model is Machine Learning (ML), which can transform WWTP data into knowledge. By training ML models with outputs from ASMs, the prediction capabilities can be improved. These trained ML models are highly effective for online monitoring and providing decision support and are widely used in industry. Limitations of ML approaches may arise from data scarcity for learning and testing. To address this, Mehrani et al. [100] proposed a hybrid model combining a mechanistic model with ML to simulate N_2O production in a sequencing batch reactor. Szelag et al. [101] examined various ML models for N_2O emission prediction, using global sensitivity analysis to select a ML model as an alternative to the mechanistic model.

5. Mitigation of GHG emissions via process modification

From the CF perspective of WWTPs, the direct emissions from the treatment processes usually play the most significant role. This chapter presents N_2O mitigation strategies in order to reach a net-zero condition and explores the implementation of innovative removal processes, source separation, and nutrient recovery for decarbonizing WWTPs.

5.1. Approaches to N_2O mitigation strategy

N_2O mitigation strategies are mainly focused on [83]: (i) Optimizing aeration mode and DO set-point, (ii) Preventing DO gradients via mixing optimization, (iii) Avoiding NH_4^+ peaks, (iv) Preventing NO_2^- accumulation, (v) Ensuring complete denitrification by supplying sufficient carbon source or/and increasing hydrolysis in the primary clarifiers.

Duan et al. [83] evaluated N_2O production by adopting a multi-pathway mechanistic model. The employed multi-criteria strategy in an Australian plant, which was focused on reducing DO levels, resulted in 35% mitigation of N_2O emissions with the EF reduced from 0.89 to 0.58% of the influent TN load. This could mainly be associated with the declines in N_2O generated via the NH_2OH oxidation pathway. Based on lower DO levels simultaneous nitrification and denitrification was encouraged and thus nitrite accumulation was minimized, leading to less N_2O production via AOB denitrification pathway. Another study by Mampaey et al. [102] reported a decline of over 50% in N_2O emissions when one-stage granular SHARON (partial nitrification) reactor cycles were reduced by 1 h, which also falls in the mitigation category of optimizing aeration.

One of the most effective ways to recognize N_2O mitigation strategies is testing different operational modes [39]. Rodriguez-Caballero et al. [103] suggested that the optimum control strategy in sequencing batch reactors (SBRs) would be shortening aerobic-anoxic cycles (20 min of aeration with a short anoxic phase).

5.2. Novel processes for N removal

Nitrogen removal systems, which are based on anaerobic ammonia oxidation (anammox), can be categorized into partial nitritation-anammox (PN/A), simultaneous PN/A and denitrification (SNAD), partial denitrification-anammox (PD/A), and denitrifying anaerobic methane oxidation (DAMO)-anammox (DAMO/A) systems (Fig. 5). PN/A and SNAD can effectively treat ammonia-rich wastewater, while reducing energy consumption and sludge production compared to the traditional nitrification-denitrification processes. In contrast, PD/A and DAMO/A are used to treat NO_3^--rich wastewater, with a focus on reducing energy consumption (no oxygen demand and reducing C/N ratio to < 3) and achieving sustainable removal efficiencies [104]. Therefore, anammox-based systems are becoming a promising technology in WWTPs for energy neutrality [105].

There are three main pathways of N_2O production in anammox-based systems, including NH_2OH oxidation ($(NH_4^+ \rightarrow NH_2OH \rightarrow N_2O)$) and autotrophic denitrification both mediated by AOB ($NO_2^- \rightarrow NO \rightarrow N_2O$ or $NH_2OH \rightarrow N_2O$ or $NH_2OH + NO \rightarrow N_2O$), as well as heterotrophic denitrification ($NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O$) mediated by diverse groups of denitrifiers. The N_2O consumption ($N_2O \rightarrow N_2$) is a unique N_2O degradation (nosZ-dominated) catalyzed by denitrifiers [106]. Other microorganisms present in anammox-based systems, such as anaerobic ammonia oxidizing bacteria (AnAOB), dissimilatory nitrate reduction to ammonia (DNRA) heterotrophs and DAMO autotrophs do not contribute to N_2O emission in anammox systems [107–109].

PN/A. The reported overall N_2O emissions in PN/A systems are highly variable (dependent on the scale) and range from 0.08% to 6.6% of N-load [5,110]. The mitigation of N_2O emission in PN/A systems can be achieved by adjusting various operation parameters, such as NO_2^-/NH_4^+ and C/N ratios, aeration strategies, DO set points, temperatures, and pH [111,112].

N_2O emission is dominated by NH_2OH oxidation pathway at low DO levels (<0.5 mg O_2/L), low inorganic carbon, high pH or low NO_2^- concentration, while AOB denitrification pathway dominates at higher inorganic carbon, lower pH or higher NO_2^- concentration [5]. Properly selecting intermittent aeration strategies can lead to frequent shifts in the predominating pathways of N_2O production and help mitigate N_2O emissions [113]. Beier et al. [114] aimed to predict the production of N_2O in a side-stream system using a novel modelling approach. During the periods when the system was aerated, the average EF was achieved 0.05 mg $N_2O/mgNH_4^+$. During the anoxic phases, the production factor was determined to be 33%. The overall EF of oxidized NH_4^+ was found to be 27%, with denitrification being the primary source of N_2O production.

A high NO_2^-/NH_4^+ ratio can result in an increased N_2O production [115], but a low NO_2^- and high NH_4^+ loading is expected to minimize N_2O production rates [116].

SNAD. N_2O is produced through the oxidation of NH_2OH by AOB and reduction of NO_2^- to NO, then to N_2O under aerobic conditions [117]. N_2O production is also linked to heterotrophic denitrification under anoxic conditions [118]. The N_2O production in SNAD systems was found to be slightly lower (by 4%) compared to PN/A systems due to lower NO_2^- accumulation [82]. This reduction may be due to the consumption of NO_2^- and/or NO during anammox metabolism [119]. Moreover, complete heterotrophic denitrification by the NosZ enzyme was identified as a promising strategy for mitigating N_2O emissions [120]. For comparison, N_2O emission was mitigated by 80% and 70% in a SNAD biofilter compared to traditional nitrification and nitrification-denitrification biofilters [117].

PD/A. In PD/A systems, diverse microbial communities compete for NO_2^-. The presence of AnAOB and heterotrophic denitrifying/Dissimilatory Nitrate Reduction to Ammonium (DNRA) in this system is crucial for N_2O mitigation. N_2O is not produced during anammox metabolism and should not be produced by heterotrophs catalyzing partial denitrification (PD) ($NO_3^- \rightarrow NO_2^-$) (Fig. 5) [121]. Low N_2O emissions were

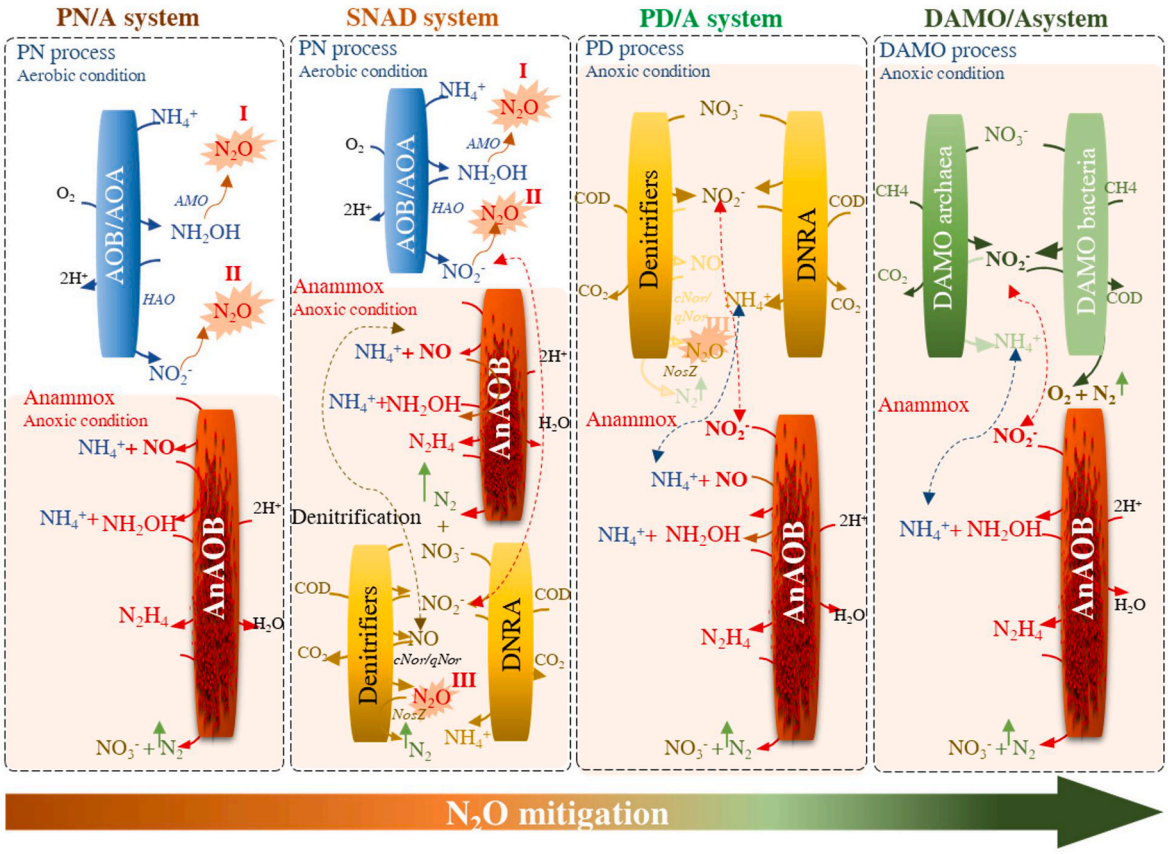

Fig. 5. Anammox based N removal systems.

observed in PD/A systems either with or without COD addition. Mitigation of N_2O was successful (EF = 0.002%) with low levels of COD addition and remained low during long-term PD/A performance [122]. However, adding COD resulted in approximately 50% reduction in N_2O emissions compared to the period without COD addition, indicating further reduction through the activity of NosZ enzyme in denitrifiers [122]. The N_2O conversion to N_2 gas was comparable to N_2O production from NO_2^- by heterotrophs under COD-limited conditions [123]. Thus, PD/A and DAMO/A have gained a significant attention for applications in WWTP as an alternative process to nitrification-denitrification and pursuing efficient N removal in a sustainable way. The most important challenge for a stable PD/A performance is a balance in NO_2^- by the activities of coexisting denitrifiers and AnAOB [123]. Moreover, low process temperatures and low influent C/N ratios could limit the startup of PD/A systems [121]. In contrast, heterotrophs can produce N_2O when PD is not successfully performed to only produce NO_2^- during unbalanced COD addition [124]. For example, 30% of total N_2O was observed to be produced in the AnAOB-dominated anoxic zone, potentially by heterotrophic denitrification or other unknown pathways [125]. Proper control of denitrifiers through adjusting COD addition (proper C/N ratio and COD type) appears to be crucial for N_2O mitigation in PD/A systems [126]. Comprehensive research should be taken for a deep understanding PD/A microbial metabolism and microbial growth rates, which are needed to improve the efficiencies of N and COD removal rates in WWTPs in the future.

DAMO/A. DAMO microorganisms can be classified as DAMO archaea and DAMO bacteria. DAMO-archaea can reduce NO_3^- to NO_2^- (substrate for AnAOB) by CH_4 oxidation under anoxic conditions, leading to N_2O and CH_4 mitigation. In contrast, DAMO-bacteria perform full denitrification ($NO_3^- \rightarrow N_2$) by utilizing CH_4 as an electron donor [127]. Combining DAMO microorganisms and AnAOB have many advantages: i) CH_4 could be decreased to 15% in the effluent wastewater [128]; ii) minimal amounts of N_2O were emitted [129]; iii) excellent nitrogen removal efficiency (99%) was achieved and cost savings were 49% [130]. The DAMO/A systems exhibit negligible N_2O emissions due to the absence of N_2O-generating enzymes in DAMO archaea and AnAOB, while DAMO bacteria effectively remove excess NO_2^- [104]. Thus, DAMO/A has significantly attracted attention for applying it in WWTP as an alternative process to nitrification-denitrification and realizing optimal N removal efficiency and environmental sustainability. However, according to Samedo et al. [131], Thauera linaloolentis strain 47 Lol^T contains NosZ, which converts N_2O to N_2. However, Mania et al. [132] found that when NO_3^- levels are high, the regulation of NosZ activity and N_2O reduction may not be possible. Consequently, elevated NO_3^- levels can significantly contribute to environmental impact and result in higher net N_2O emissions [131]. More research is still needed to understand DAMO/A microbial metabolism.

5.3. *Source separation systems*

In source separation systems, black water (BW), grey water (GW) and food waste (FW) are separated from municipal wastewater and solid waste as shown in Fig. 6. Separating the different kinds of wastewater at source can potentially increase the treatment efficiency, nutrient recovery and biogas yield. Kjerstadius et al. [133] compared the CF of conventional systems and a source separation system in Swedish WWTPs. The LCA showed that the source separation system could decrease the overall CF by 34 kg CO_{2e}/PE·year, primarily due to an increasing the biogas production.

Other studies, e.g. Remy [134], were in-line with the findings of Kjerstadius et al. [133] regarding the reduction of the net CF after source separation. In contrast, Thibodeau [135] reported a CF of 65 kg CO_{2e}/PE·year for a source separation scenario, which was higher by 12 kg CO_{2e}/PE·year in comparison with the conventional system. The wet

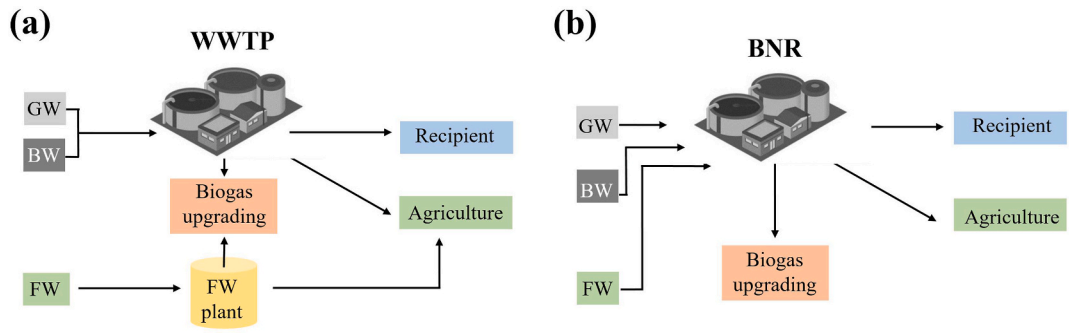

Fig. 6. (A) Conventional systems vs (B) source separation system of wastewater treatment (adopted from Kjerstadius et al. [133]).

fraction of BW was entirely returned to agriculture, leading to an increase in the CF resulting from transportation. However, the increased CF could be balanced out by selling bio-products as fertilizer.

Another benefit of source separation could be an increased recycle of nutrients to farmlands. The range of excess return of nutrients (N and P) was 1.7–4.3 kg N/PE·year and 0.06–0.54 kg P/PE·year, respectively [133–135]. The increased recovery of nutrients and utilizing them on farmlands could partially offset CF [4] and further decrease the indirect CF of WWTPs. Besson et al. [136] performed an LCA study and compared the environmental impacts of a central WWTP with source separation systems. Their results showed that for maximizing nutrient recovery and reducing GHG emissions, urine and BW separation are more favorable scenarios than a centralized WWTP. The separation allowed for mitigating N_2O emissions by 60% and avoiding energy-intensive N fertilizer production (Habor-Bosch process), which emits a large amount of GHGs (8.6 kg CO_{2e}/kg N) [136].

In another study, Tian et al. [137] utilized urine wastewater derived from a source separation system to pretreat WAS for AD. The authors observed a significant improvement in cumulative methane production, showing a 23% increase after pretreatment with urine wastewater at a volumetric proportion of 1:8. The presence of urea and hydrolyzed free ammonia in urine wastewater had a synergistic effect on sludge disintegration, leading to a reduction in sludge volume and increased availability of substrates for biological treatment processes. These findings highlight the potential of urine wastewater for pretreatment to enhance the efficiency of AD processes and improve biogas production from WAS. Badeti et al. [138] conducted a theoretical investigation of the impact of urine diversion on the biological treatment processes at a decentralized WWTP. Their simulations showed that 33% of aeration energy could be saved with 90% urine diversion. Furthermore, direct N_2O and CO_2 emissions in the treatment processes could be reduced by 98% and 25%, respectively, while indirect GHG emissions could be reduced by 20%. Source separating sanitation was considered in a new city district of Hiedanranta, Finland, Lehtoranta [139] considered source separating sanitation and applied LCA to compare it with a conventional sanitation system. The study revealed that with a separating system, 3 to 10 times more N could be recovered compared to the conventional system. Based on their environmental performance, source separating systems with improved nutrient recovery showed reduced climate and eutrophication impacts.

5.4. Systems with nutrient recovery

5.4.1. P recovery from wastewater

The P load in urban wastewater could theoretically replace up to 50% of the mineral P fertilizer used in agriculture. Moreover, P mining from the phosphate rock is associated with environmental concerns, such as air pollution and water contamination [140]. Amann et al. [141] used LCA to evaluate 18 P recovery technologies in terms of the energy demand and GWP. The analysis showed that a wide-range of GHG emissions can be estimated through the implementation of different P recovery strategies. The recovery from the liquid phase reduces the net GHG emissions. For the recovery from sludge, high GHG emissions were attributed to the processes which were already practiced in the full-scale. The recovery from sewage sludge ash showed the highest potential for an efficient P recycling. This method could annually decrease the cumulative energy demand and GWP by 0.096% and 0.1% per PE, respectively [141].

Another LCA study by Duan et al. [142] analyzed and compared the environmental impacts of different P removal and recovery methodologies. For chemical P removal and recovery pathways, the net specific CF was 44.5 kg CO_{2e}/kg $P_{removed}$ and −3.76 kg CO_{2e}/kg $P_{recovered}$, respectively. The negative value indicated the potential savings of CF due to P recovery. In another review, Zhao et al. [143] explored potential pathways for P recovery in WWTPs with a special focus on the A-2B-centered process, which involves an anaerobic fixed bed reactor to capture COD for improving energy efficiency. Simultaneously, this process allows for P recovery through further treatment with P crystallization. The A-2B-centered process exhibited the lowest specific energy consumption and GHG emissions when considering the amount of P recovered, indicating. Its promising approach in terms of both energy efficiency and environmental impact. The management of P resources in WWTPs encompasses multifaceted concerns related to environmental protection, energy efficiency, and GHG emissions. By exploring and implementing efficient recovery paths like the A-2B-centered process, WWTPs can take significant steps towards sustainable P management.

5.4.2. N recovery from wastewater

Recovery of N as a nutrient would decrease both direct environmental impacts and energy consumption. Nitrification-denitrification could be energy intensive and without any additional benefits aside from meeting the effluent standards. N recovery allows for the simultaneous treatment of wastewater, while consuming less energy and recovering resources. Beckinghausen et al. [144] performed a comprehensive review of approximately 50 N recovery techniques. The authors focused on techniques that could produce fertilizers. The highlighted technique was permeable membrane, which consumed smaller amount of energy (1–1.2 kWh/kg N). Moreover, it showed the potential to recover NH_4^+ from wastewater and the market value of the recovered product (ammonium sulphate). Other techniques, such as vacuum membrane distillation, can recover large amounts of energy in the form of heat (27 kWh/kg N), but the process is highly energy demanding (60 kWh/kg N). These techniques require further investigation to evaluate the costs of energy, as the amount of recovered heat highly effects the overall energy balance [145]. Innovative technologies, such as membranes, sorbents, electrodialysis, bioelectrochemical process, and even microalgae, are used to enhance nutrient recovery from wastewater, moving beyonnd conventional crystallization methods. Moreover, at the full-scale scale, some physicochemical methods, such as air stripping and struvite formation, are employed to recover P and N from side-streams [146].

6. Mitigation of GHG emissions via energy usage optimization

Wastewater treatment has a high potential to decarbonize and move towards net-zero carbon condition, since water reclamation from wastewater is considered to be the most energy-intensive water infrastructure [1].

6.1. CF due to energy consumption

Energy efficiency has received growing attention as the number of WWTPs is growing globally and the effluent quality criteria become more strict [147]. The water industry is estimated to account for 2.1% in the US and 0.8% in the EU of the total electricity consumption [148]. Around 23 MMT CO_{2e} was generated during electricity consumption, which accounts for 37.7% of the entire CO_{2e} release in the water industry [149]. Aeration, pumping, and heating are the main energy consumers in WWTPs [149]. In a typical WWTP, aeration can account for up to 60% of all electricity usage, followed by sludge treatment and return activated sludge (RAS) recirculation from secondary clarifiers [150].

Decarbonization of the energy sector can lead to a reduction in the indirect emissions in WWTPs. Coal-fired electricity production is being replaced by natural gas and more countries are substituting fossil fuels with renewable resources, such as wind, solar, hydropower, and nuclear power [149]. Hydropower and nuclear power can be seen as the cleanest energy source with a maximum carbon intensity of 40 g CO_{2e}/kWh and 110 g CO_{2e}/kWh respectively, while coal is the most carbon-intensive energy source (Table 4) with the maximum carbon intensity of 1689 g CO_{2e}/kWh [151]. Currently, about 88% of the grid energy is satisfied by fossil fuels. Over the next 20 years, it is planned to increase the usage of renewable energy by 25%, which would decrease the CF related to the energy mix by up to 42%.

Delre et al. [132] evaluated the effect of electricity usage on the CF of seven different WWTPs located in Denmark and Sweden. In the Danish WWTPs, where the energy mix mostly consists of fossil fuels, energy consumption accounted for up to 29% of the CF. In the Swedish WWTPs, where nuclear and hydropower account for most of the energy mix, the CF-related energy consumption was as low as 3%. In the US, the fuel mix varies substantially by the state, resulting in energy carbon intensity ranging from 0.12 kg CO_{2e}/kWh in Idaho to 1.15 kg CO_{2e}/kWh in Washington, DC [82]. As shown in Fig. 7, the Midwest and Southwest regions contribute heavily to CF. This is due to the heavy reliance on fossil fuels in these regions. In the Midwest, 66.5% of the energy mix is satisfied by coal, while only 6% of the energy mix is from renewable energy sources. In contrast, in the Southwest regions, natural gas and coal make up 45% and 22% of the energy mix. On the other hand, approximately 50% of the energy mix in some regions is fueled by renewable energy, and thus the specific GHG emissions were found as low as 0.38 kg $CO_{2e}/m^3_{wastewater}$ [148].

Although the energy mix has a noticeable effect on the indirect CF of WWTPs, other factors also play an important role. For example, the Owls Head WWTP (New York, US) has a substantially lower CF related to energy compared to the Gloversville-Johnstown WWTP (New York, US), even though the energy mix is similar in those plants. This is because Owls Head WWTP uses chlorine as a disinfectant, while Gloversville-Johnstown WWTP uses ultraviolet (UV) for effluent disinfection, which requires electricity consumption [82].

Table 4
EFs for different sources of electric energy [151].

Energy Source	min (g CO_{2e}/kWh)	max (g CO_{2e}/kWh)
Coal	675	1689
Oil	510	1170
Natural gas	290	930
Biogas	50	700
Hydropower	3	40
Nuclear power	4	110

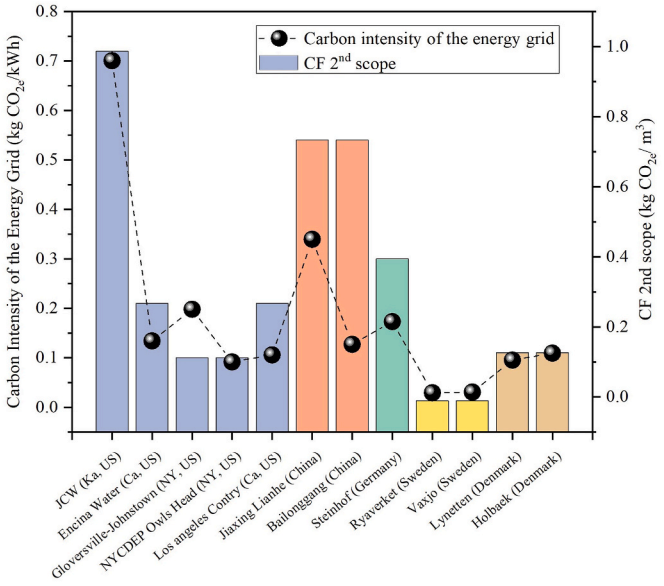

Fig. 7. Comparison between carbon intensity of the energy in different countries and scope 2 emissions of selected WWTPs [82,152].

6.2. Reducing energy consumption

Minimizing energy consumption can play a pivotal role in the CF reduction. There are two major approaches to decrease the power use in WWTPs: (i) reduction through operational changes and (ii) application of innovative, energy-efficient wastewater treatment processes [153]. Table 5 shows the energy mix and CF reduction under different scenarios in several case studies.

The DO, solids retention time (SRT), and mixed liquor recirculation (MLR) are crucial operational parameters in the bioreactor, which highly determine the operation of the aeration system and electricity consumption. Energy consumption could be decreased by 7–9% through the adjustment of SRT, DO, and MLR [154]. Mamais et al. [150] also obtained a 4500 MWh/year energy reduction by optimizing the SRT and DO concentration in a WWTP. Borzooei et al. [159] concluded that theoretically a reduction of up to 5000 MWh/year in energy consumption would be possible through operational changes in the largest Italian WWTP (2.1 million PE).

The advanced equipment also brings benefits to the CF. Li et al. [157] installed a TN online monitoring equipment at both influent and effluent, with a carbon source dosing system. As a consequence, the TN treatment efficiency increased by 1%, while the energy consumption and carbon emission decreased by 5.6% and 12.7%, respectively. Li et al. [157] updated the traditional blower to magnetic suspension centrifugal blowers, resulting in a 15–24% decrease in the aeration energy and a 4.6–7.7% decrease in the CF. Additionally, Ke and Lu [160] noted that the energy efficiency of the magnetic suspension blower could be improved by 20%.

ML is one of the novel techniques for reducing the energy demand of WWTPs. Cao and Yang [161] developed a model (OS-LEM) to control DO supply of aerobic bioreactors, which can reduce the energy consumption by 40% in comparison with the conventional on/off DO control. Similarly, Ramli et al. [158] used ML to build a prediction model to reduce the energy consumption of WWTPs by forecasting it one month ahead of time, which estimated a 2.2% reduction in energy consumption.

Table 5
Reported energy reductions in WWTPs under different scenarios.

Reference	Scenario	GHG emission related to energy ($kgCO_{2e}/m^3$)	Reduction in Energy Demand		Total GHG emission decrease (%)	Remarks
			(MWh/year)	%		
Mamais et al. [150]	Reduction in SRT and DO	–	4500	–	–	Psyttalia (Greece) WWTP in Greece was analyzed.
Daskiran et al. [154]	Reduction in SRT and MLR	0.1	3510	–	0%	Emission rates matched the active scenario in the WWTP
	Reduction in SRT and DO	0.06	3530	–	−15%	The emission rates were about 0.17 kgCO2/m3 more than the active scenario in the WWTP
	Reduction in MLR and DO	0.05	4661	–	−9%	The emission rates were about 0.1 kgCO2/m3 more than the active scenario in the WWTP
Borzooei et al. [155]	Reduction in SRT	–	5000	–	–	In Castiglione Torinese WWTP (Italy) different scenarios where SRT was optimized were tested
Longo et al. [147]	Minimizing energy consumption of pumps, motors, and other electro-mechanic devices	–	–	10–20%	–	WWTP in Missouri Southeast, USA
Torregrossa et al. [156]	optimizing pumping, blowers, and AD	–	–	50–80%	–	Various WWTPs in Europe
Li et al. [157]	Upgrading to a magnetic suspension blower	–	–	15–24%	4.6–7.7%	14 WWTPs in North-Central China were examined
	Accurate aeration system modification	–	–	6.3%	10.37%	The implementations were studied on two WWTP in China
	Intelligent precise aeration system for sewage treatment	–	–	5.3%	10.93%	The implementations were studied on a WWTP in China
Ramli et al. [158]	Optimizing pumping, blowers, and AD	–	–	3%	–	WWTP in Peninsulay (Malaysia)

6.3. Increasing energy production inside WWTPs

6.3.1. Co-digestion

Co-digestion is proposed as a straightforward approach for improving macronutrient balance, correcting moisture, and/or diluting inhibitory or toxic compounds [162], which possessed a positive effect on energy production and CF reduction. Most energy neutral plants located in developed countries adopt biogas recovery through co-digestion [163]. Koch et al. [164] reported more than double energy recovery when adding a ratio of 10% FW as a co-substrate in a WWTP. The biogas output increased by 17% by co-digesting 7% of fat with the mixed sewage sludge [30]. Sarpong and Gude [165] reported that the utilization of fat-oil-grease (FOG) for co-digestion with low-strength wastewater can increase energy production by 0.08 kWh/m^3, while the total required energy of the studied plant was reported as 0.32 kWh/m^3. Wickham et al. [166] demonstrated a biogas production increase of up to 191% using carbonated soft drinks as a co-substrate. An increase of 81% in biogas production was achieved by adding 3% of glycerol as a co-substrate [167]. Similarly, biogas production was increased by up to 209% by using grease trap water as a co-substrate [168]. Many plants, employing co-digestion, have reached 100% energy neutrality and even become net energy positive, including Zurich Wedholzli WWTP-Switzerland (42 GWh/year), Point Loma WWTP-US (193 GWh/year), Grevesmuhlen WWTP-Germany (193 GWh/year), and Sheboygan Regional WWTP-US (32 GWh/year) [169]. Table 6 provides the potential increase in the biogas production rate for various co-substrates.

6.3.2. Biogas utilization and upgrading

The produced biogas can be utilized internally for valorization in combustion engines to supply electricity and heat generation. Most WWTPs that produce biogas through AD in developing countries do not have thermal energy recovery or electricity production, thus the biogas is flared and wasted [163]. A decade ago, the USEPA estimated that if 544 US WWTPs with AD utilize CHP, the reduction in energy consumption would be equivalent to the emissions from 430,000 cars [173]. The Psyttalia WWTP deployed CHP and revealed the lowest annual energy consumption among 10 studied WWTPs in Greece, at 15 kWh/PE

Table 6
Biogas production rate increase after using different co-substrates.

Co-substrate	Condition	Energy production without co-substrate (kWh/d)	Energy Production with co-substrate (kWh/d)	Biogas Production without co-substrate (L/gVS/day)	Biogas Production with co-substrate (L/gVS/day)	Reference
Microalgae	–	774	1189	–	–	[170]
Cheese whey	–	774	1531	–	–	
The organic fraction of municipal solid waste (OFMSW)	Volumetric ratio 75:25; HRT 20 d	7688	8418	–	–	[171]
Dry waste (DW)	Volumetric ratio 80:20; Hydraulic Retention Time (HRT) 16 d	677	10,639	–	–	
OFMSW + DW	Volumetric ratio 68:23:9; HRT 18 d	6037	8972	–	–	
Grease (G)	TS: G; 50:50	–	–	0.48	0.78	[172]
Septage (SP)	TS:SP; 90:10	–	–	0.48	0.46	
Whey (W)	TS: W; 70:30	–	–	0.48	0.75	

[150].

The produced biogas can be upgraded to biomethane, which has the same CH_4 quality as natural gas and could be utilized as a fuel for vehicles or pumped into the grid [174]. Biogas upgrading, involving CO_2 removal, employs gas sorption and separation techniques. Physical and chemical scrubbings effective but energy-intensive, while pressure swing adsorption is less energy-intensive, but more complex. Membrane separation technologies are environmentally friendly with a relatively low energy consumption, whereas cryogenic separation demands high energy for cooling [175]. Following the technological advances in biogas upgrading, a transition away from CHP and towards upgrading biogas to biomethane has been trending [176]. Biogas is already being converted to biomethane in 15 EU countries and 10 of those countries are injecting biomethane into the natural gas grid. The countries with the highest number of biomethane plants include Germany (185 plants), UK (80 plants), and Sweden (61 plants), while the other countries are lagging behind [174].

To address GHG release from biogas upgrading, emerging technologies focus on CO_2 utilization through methanation, combining CO_2 with H_2 to produce CH_4 via chemical and biological processes. Implementing this technology requires investment and operational costs, especially for H_2 production through electrolysis. The biomethanation process efficiency is a limitation, but benefits include carbon neutrality and continuous fuel delivery for grid balance [175].

Nguyen et al. [169] compared the CHP and biomethane for biogas utilization from WWTPs. It was concluded that the CHP was effective with a negative CF, while the biogas upgrading to biomethane resulted in a higher CF due to a high energy input during the upgrading process. Ravina and Genon [177] also concluded that CHP of biogas with the thermal energy utilization produces the lowest CO_{2e} ($-0.277tCO_2/t$ biogas). The biomethane upgrade (-0.13 tCO_2/t biogas) to the Italy's national grid enabled to store biomethane at a lower cost for further utilization. Thus, upgrading biogas to biomethane could be a more sustainable scenario than on-site biogas consumption in CHP units, especially if the gas energy content is not fully cogenerated and the produced thermal energy is not capitalized. Regarding the CH_4 loss during upgrading, a sustainable system should reduce CH_4 slip below 4%, with a target value of 0.05% [177].

7. Mitigation of GHG emissions via chemicals and transportation optimization

To achieve full decarbonization and the net-zero carbon condition in WWTPs, both upstream and downstream, indirect emissions are ought to be reduced. Ambiguous GHG emissions from WWTPs are primarily related to chemical agent usage and transportation of by-products (grit, screenings, and sludge) [28].

7.1. Reducing chemical consumption

Chemicals are used at different stages of wastewater treatment and sludge management. To minimize their contribution to the CF, not only consumption cuts can be implemented, but also some sustainability criteria introduced to the purchase procedure may result in reducing the indirect GHG emissions. Factors linked to the usage of chemical agents in the context of upstream indirect emissions are leveled at 1964 kg CO_{2e}/kg for inorganic agents and 1909 kg CO_{2e}/kg for organic agents (Ecoinvent database 3.6).

The inorganic agents are commonly used to support enhanced biological P removal. Metal salts, such as aluminum (III) sulphate, iron (III) chloride, and lime in the form of $Ca(OH)_2$ or CaO_2, are added to trigger precipitation. The use of lime compounds results in a pH increase, which requires the addition of acid chemicals to adjust the pH level of the WWTP's effluent. Moreover, both metal salts and calcium agents result in an increase in sludge volume, leading to higher GHG emissions associated with sludge disposal. It is preferable to use recycled inorganic chemical agents instead of new resources in order to avoid emissions related to the extraction and manufacturing processes of raw materials. Organic agents and polymers are regularly applied for flocculation, coagulation, and dewatering processes. Recycling could effectively reduce the indirect GHG fluxes. Instead of synthetic, polymers of bio-based origin can be applied as a more environmentally friendly choice [178,179].

The application of chemical agents influences the entire wastewater treatment process and its stages [180]. The enhancement of primary sedimentation via chemically forced flocculation contributes to a higher chemical consumption in the overall GHG balance. Ultimately, it becomes an advantage due to the biogas and energy production bust. In contrast, the chemical agents used to support dewatering processes may become a target in lowering CO_{2e} indirect emissions via overall chemical agent consumption cuts at WWTPs.

7.2. Reducing transport-related CF

Conveying activities and services, such as upstream transportation of chemical agents and downstream transport of wastewater treatment by-products (grit, screenings, and sludge), contribute to the indirect emissions of WWTPs. It is often challenging to implement a transport-related emission reduction plan based on switching to more environmentally-friendly means of transportation, such as rail transport, or shortening the transportation distance. Therefore, more advanced reduction scenarios are sought.

According to the Department for Environment, Food and Rural affairs (DEFRA) [181] EF database, the amount of GHG emitted to the atmosphere depends on (1) engine standard, which translates into the type of fuel combusted, (2) size of the vehicle – rigid, and (3) the laden. Emission reduction can be achieved via a detailed transportation plan, including optimization of laden for each material transported, adaptation of the vehicle rigid type (elimination of oversizing), and optimization of fuel type engines.

Transport-related concerns resulted in the promotion of various biofuels: especially bioethanol from sugar and starch crops and biodiesel from oilseed crops [182]. Triggered by the Renewable Energy Directive I and II [183,184], the process of replacing fossil-origin gasoline and diesel with biofuels presents at least 65% GHG savings in comparison with the conventional fuels.

8. Recovering N_2o from wastewater as an energy source

Recovering N_2O as an energy source would be a win-win strategy. The process would be a step-forward towards net-zero operation by offering both the possibility of energy generation and mitigation of N_2O emissions. There are limited studies available in the literature that investigate N_2O recovery, which has also been being used as a powerful oxidant for energy generation. Wu et al. [185] suggested that applying Coupled Aerobic-Anoxic Nitrous Decomposition Operation (CANDO) might be one of the bioprocesses with a high potential to achieve energy neutral wastewater treatment paradigm. The process involves three steps: i) partial nitrification of NH_4^+ to NO_2^-; ii) partial anoxic reduction of NO_2^- to N_2O; and 3) N_2O conversion to N_2 with energy recovery by either catalytic decomposition or co-combustion of N_2O with CH_4. Yu et al. [186] documented a novel process that recovers N_2O by inhibiting the activity of the nitrous-oxide reductase (N_2OR) in denitrifying bacteria. N_2O recovery was demonstrated in batch experiments in which NO was the only electron-acceptor. N_2O recovery level in their study reached approximately 70% of the total gas production.

The most investigated N_2O recovery process is CANDO, which has been tested with various configurations and real wastewater [187]. However, achieving stable long-term operation requires further experiments. Promising alternatives include autotrophic photoelectrotrophic denitrification, sulfur-driven denitrification, and hydrogenotrophic denitrification [188]. These autotrophic processes are currently

immature technologies and need intensive investigations. N_2O recovery from high-strength wastewater is more favorable than low-strength (mainstream) wastewater due to a higher energy and economic potential. Recently, studies such as Deng et al. [189] and Huo et al. [190] have developed reliable models to simulate N_2O recovery from Fe(II) EDTA-NO. The simulation results of Huo et al. [190] suggested that with a S/N mass ratio of around 1, high-purity N_2O could be more rapidly and efficiently recovered from Fe(II)EDTA-NO (N_2O recovery efficiency reached up to 85%). Deng et al. [189] highlighted that sufficiently increasing the headspace volume of the reactor was considered an ideal strategy to obtain ideal N_2O production of 86.6%. Duan et al. [188] critically reviewed the progress of research on N_2O recovery and counted (i) unstable nitritation, (ii) the low energy potential and (iii) environmental risks associated with intentional N_2O emissions in mainstream treatment, as technical and economic challenges.

9. Challenges and limitations while implementing net-zero carbon practices

Both direct (fugitive, process-related) and indirect (due to energy and chemicals consumption, transportation) GHG emissions have some potential for reduction. However, implementing mitigation strategies for the net-zero carbon concept in WWTPs comes with recognized limitations and trade-offs. Effluent quality, operational cost and GHG emissions are potentially conflicting objectives [95,97,191]. For example, energy savings in the AS bioreactor, achieved by decreasing aeration intensity, may result in increased N_2O emissions [192]. Co-digestion of external substrates reduces energy-related emissions and increases process-related GHG emissions [30]. Those studies proposed either separate evaluation criteria or integrated plant performance indicators. LCA complements decision-making to emphasize environmental impact [97]. In a full-scale WWTP modelling study [74], three sustainability criteria were considered: effluent TN concentration, overall energy balance and GHG footprint. Investigating the DO set-point in the aerobic zone and MLR rate showed improvements in GHG emissions and effluent quality. Technological upgrades, such as CEPT and deammonification in reject water treatment, enhanced energy neutrality and reduced GHG footprint, but did not improve effluent quality simultaneously.

10. Conclusions

Several steps may be considered when moving WWTPs towards net-zero carbon conditions. The direct emissions from treatment processes can contribute to more than 60% of the CF in WWTPs, while energy consumption with over 30% contribution to the CF has also a high potential for decarbonization of WWTPs. The remaining emissions are of lesser importance and can be offset by optimizing chemical usage and transportation. The first major step towards net-zero carbon condition is identification of emission hotspots and quantification of those emissions via either measurements, reliable models or EFs. The second step is GHG mitigation via process modification and energy usage optimization. The implementation of novel N removal processes such as PD/A and DAMO/A, could reduce GHG emissions and energy consumption while ensuring reliable N removal efficiencies. Other techniques such as source separation systems could potentially allow mitigation of N_2O emissions by 60% while avoiding energy-intensive N fertilizer production. Nutrient recovery methods are another approach which offered negative value for the net CF. Permeable membrane N recovery offered 1 kWh/kg N of energy savings and P recovery led to -3.76 kg CO_{2e}/kg $P_{recovered}$ CF savings. Upgrading biogas to biomethane could be a more sustainable scenario than on-site biogas consumption in CHP units, especially if the thermal energy is not capitalized. Recovering and utilizing N_2O for energy production is a promising method which leads to both direct and indirect CF reductions.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

Data availability

No data was used for the research described in the article.

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