

## Chapter 6

# Full-scale emission results (N<sub>2</sub>O and CH<sub>4</sub>)

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### SUMMARY

This chapter reviews the studies from N<sub>2</sub>O and CH<sub>4</sub> monitoring campaigns in full-scale wastewater treatment plants (WWTPs) and sewer networks. The focus is on greenhouse gas (GHG) emissions from WWTPs as more literature is available. The analysis classifies quantified N<sub>2</sub>O and CH<sub>4</sub> emission factors (EFs), triggering operational conditions and formation pathways for different configurations. Control strategies to minimize N<sub>2</sub>O emissions are proposed for different process groups. The main reasons for EF discrepancies are discussed. Overall, N<sub>2</sub>O emission factors for processes treating low-strength wastewater streams range between 0.003 and 5.6% of the N-load (average equal to 0.9% of the N-load). Emissions higher than mainstream process average emissions have been reported in sequencing batch reactors (average equal to 3.6% of the influent N-load) and step-fed plug flow reactors. In full-scale sidestream processes, less than 15 monitoring campaigns have reported EFs (average equal to 2.5% of the N-load). Differences in the EFs among the process groups are partially attributed to disparities in the control strategies (i.e. aeration control), configuration, and operational and environmental conditions that favour the preferred enzymatic pathways. Overall, triggering operational conditions for elevated N<sub>2</sub>O emissions in full-scale wastewater treatment processes include (i) increased NH<sub>4</sub><sup>+</sup> concentrations leading to a high ammonia oxidation rate (AOR) and increased production of intermediates (e.g. NH<sub>2</sub>OH, NO<sup>-</sup>, etc.), (ii) improper aeration control (i.e. inadequate aeration and non-aeration duration, over-aeration, under-aeration), (iii) NO<sub>2</sub><sup>-</sup> accumulation triggering the nitrifier denitrification pathway, and (iv) sudden shifts in incomplete heterotrophic denitrification (i.e. due to excess dissolved oxygen (DO), chemical oxygen demand (COD) limitation etc.). The N<sub>2</sub>O monitoring strategies can also influence the reliability of the quantified EFs. Due to temporal variation of N<sub>2</sub>O emissions, short-term studies are not sufficient to quantify annual EFs. The analysis showed that the average EF for processes treating low-strength streams monitored for less than a week is 0.66% of the influent N-load. On the other hand, processes monitored over 6 months have an

average EF equal to 1.74%. Compared with  $N_2O$ ,  $CH_4$  quantification from full-scale WWTPs is less investigated, while it also contributes significantly to the overall plant carbon footprint. The results of full-scale  $CH_4$  quantification studies are summarized in this chapter. Emissions of  $CH_4$  in WWTPs mainly originate from the influent, anaerobic wastewater treatment and anaerobic sludge handling processes. The amount of  $CH_4$  emissions varies greatly with different configurations of WWTPs. For WWTPs without anaerobic sludge handling processes, the  $CH_4$  emissions can mainly be traced back to the  $CH_4$  dissolved in the influent. When anaerobic treatment is applied in WWTPs for wastewater COD removal, its  $CH_4$  emissions might substantially increase the overall plant carbon footprint. GHG monitoring campaigns carried out in WWTPs should include the monitoring of fugitive  $CH_4$  emissions. Finally,  $CH_4$  and  $N_2O$  emissions reported from sewer networks are also summarized in this chapter.

The last part of the chapter summarizes some mitigation strategies applied at full-scale to control fugitive CHG emissions from WWTPs and sewers.

**Keywords:** Full-scale greenhouse gas emissions, methane, nitrous oxide, sewer networks, wastewater treatment plants

## TERMINOLOGY

Term	Definition
Activated sludge	Flocs of sludge particles containing microbes, which are formed in the presence of oxygen in aeration tanks.
Activated sludge process	The wastewater treatment process which applies activated sludge to speed up the decomposition of contaminants in wastewater. Oxygen is provided in the aeration tank in favour of the metabolization of activated sludge, to convert contaminants into harmless products. After the aeration tank, the mixed activated sludge goes to a clarifier to separate the sludge and treated water. The treated water will undergo further treatment.
Aeration	The introduction of air into the aeration tank for the oxidation of organic, nitrogenous and phosphorous compounds by microbes, and also for keeping the activated sludge suspended and well mixed.
Aerobic	Conditions with free oxygen in the wastewater.
Ammonia monoxygenase	An enzyme catalysing $NH_4^+$ oxidation to $NH_2OH$ .
Anaerobic	Conditions without atmospheric or dissolved molecular oxygen in the wastewater.
Anoxic	Conditions of oxygen deficiency and presence of oxidized nitrogen species.
Biomass	A clump of organic material consisting of living organisms, which lives on the substrates in wastewater, or the dead organism debris.
Chemical oxygen demand	An indication of the amount of organic materials in wastewater. It refers to the amount of oxygen equivalent consumed in the chemical oxidation of organic matter by a strong oxidant such as potassium dichromate.
Dissolved oxygen	Molecular oxygen dissolved in wastewater.
Greenhouse gas	Gas that absorbs and emits radiant energy within the thermal infrared range and contributes to the global warming effect.
Heterotrophic denitrification	A series of reduction reactions from nitrate to nitrogen gas by heterotrophic denitrifiers under anoxic conditions, with organic carbon as the electron donor for the reactions.
Nutrient	Substances such as nitrogenous compounds and phosphorous or organic matter that can be assimilated by microbes to promote the metabolism and growth of microbes in the reactor.
Organic matter	The organic waste of plant or animal origin from homes or industry, mainly volatile fraction of solids.

Oxidation	Oxidation is the addition of oxygen, removal of hydrogen, or the removal of electrons from an element or compound. In wastewater treatment, organic matter is oxidized to more stable substances.
pH	An indication of the acidity or alkalinity of solutions.
Reactor	Containers of different size or design which can hold the activated sludge to conduct wastewater treatment processes.
Wastewater	The used water and solids from a community that flow into a treatment plant. Storm water, surface water, groundwater infiltration and a fraction of industrial wastewater also may be included.
EF	Emission factor

## 6.1 INTRODUCTION

Nitrous oxide (N<sub>2</sub>O), is a potent greenhouse gas (GHG), 298 times stronger than CO<sub>2</sub> in terms of global warming potential (IPCC, 2013). N<sub>2</sub>O can be generated in large amounts and stripped in the atmosphere during biological nutrient removal (BNR) at wastewater treatment plants (WWTPs). In the past few years, concern regarding the quantification and investigation of N<sub>2</sub>O, from full-scale wastewater treatment processes has increased. There are three main biological pathways for N<sub>2</sub>O production in BNR systems. N<sub>2</sub>O can be formed during the autotrophic oxidation of ammonia to nitrite/nitrate through the activity of ammonia oxidizing bacteria (AOB) under aerobic conditions (nitrification/nitritation). The N<sub>2</sub>O production by AOB can be due to the autotrophic denitrification of nitrite (nitrifier denitrification pathway) and due to incomplete oxidation of hydroxylamine (NH<sub>2</sub>OH) (NH<sub>2</sub>OH oxidation pathway). N<sub>2</sub>O is also an intermediate during the reduction of nitrate/nitrite to nitrogen gas through the activity of heterotrophic denitrifying bacteria under anoxic conditions (heterotrophic denitrification pathway). There is a wide variety of different BNR processes applied at wastewater facilities to treat the incoming wastewater (i.e. with different numbers of compartments/zones for nitrification and denitrification, recirculation flows, flow-patterns and feeding strategies). Studies have shown that the direct N<sub>2</sub>O emissions of BNR processes in WWTPs can contribute up to ~78% of the operational carbon footprint (Daelman *et al.*, 2013). There are recent studies reporting even higher percentages; for example, N<sub>2</sub>O contributes up to 86% of the carbon footprint in the study of Kosonen *et al.* (2016), compared to direct methane emissions (CH<sub>4</sub>).

Significant N<sub>2</sub>O emissions have been reported from the biological treatment of high-strength wastewater streams. The anaerobic supernatant is a by-product from the treatment of the primary and secondary sludge via anaerobic digestion when the digestate is dewatered. This stream is small in volume (1–2% compared to the mainstream line), but very concentrated in nutrients and is conventionally recycled back to the primary treatment increasing the loads (and thus, the energy requirements and costs) of the mainstream biological treatment (i.e. contains 10–20% of the WWTP nitrogen load). For this purpose, BNR technologies (such as partial-nitritation-anaerobic ammonium oxidation (PN-anammox), nitritation-denitrification, etc.) have been developed to treat high-strength streams in a cost and energy efficient way (Lackner *et al.*, 2014; Zhou *et al.*, 2018). In the sidestream biological processes, favourable conditions for N<sub>2</sub>O generation can prevail (i.e., NO<sub>2</sub><sup>-</sup> accumulation, elevated NH<sub>4</sub><sup>+</sup> concentrations, etc.). Studies have shown that biological processes treating high-strength streams can contribute over 90% of the total direct N<sub>2</sub>O emissions compared to the mainstream BNR processes (Schaubroeck *et al.*, 2015).

The recent mitigation roadmap to carbon neutrality in urban water published by the Water and Wastewater Companies for Climate Mitigation (WaCCliM) project and the International Water Association (IWA) (Ballard *et al.*, 2018), states that direct N<sub>2</sub>O emissions in water utilities, should be considered for carbon footprint assessment, reporting and mitigation. However, in practice, the quantification of direct N<sub>2</sub>O emissions at WWTPs via monitoring campaigns is not a regulatory requirement. Therefore, wastewater utilities usually estimate N<sub>2</sub>O emissions via theoretical methods, that is based on the population equivalent of the WWTP (IPCC, 2006); the latter can significantly

underestimate the actual emissions (Cadwallader & VanBriese, 2017). The 2019 IPCC Refinement of the 2006 IPCC Guidelines has significantly increased the suggested default EF; they propose a value equal to 1.6% of the influent N-load.

Full-scale monitoring campaigns have been implemented in full-scale BNR processes to provide insights into the dynamics and triggering mechanisms for N<sub>2</sub>O generation. However, results were variable and there is still not a consensus to explain the exact causes. The application of different WWTP configurations and different biological treatments is a main reason that explains the variation in results. The sampling strategy and duration also play an important role. Most of the studies were performed over a short-term (days–weeks) showing only diurnal emission patterns. The sampling strategy (grabbing samples or online monitoring) is also a factor that can lead to an over or underestimation of the N<sub>2</sub>O emissions. Additionally, N<sub>2</sub>O fluxes were characterized by significant spatial and temporal variability due to the different interacting biological processes that consume or produce N<sub>2</sub>O and the variation in operational conditions (Daelman *et al.*, 2015; Gruber *et al.*, 2020). Mechanistic process-based models have been developed over recent years aiming to integrate N<sub>2</sub>O emissions generation of different processes in the design, operation and optimization of biological processes (Domingo-Félez *et al.*, 2017; Mannina *et al.*, 2016; Massara *et al.*, 2017). However, their online integration for the reliable quantitative estimation of N<sub>2</sub>O emissions and offline integration for long-term quantitative purposes remain challenging (Haimi *et al.*, 2013; Mampaey *et al.*, 2019).

WWTPs also emit CH<sub>4</sub> (Daelman *et al.*, 2013; Ribera-Guardia *et al.*, 2019). Emissions of CH<sub>4</sub> in WWTPs mainly originate from the influent, anaerobic wastewater treatment and anaerobic sludge handling processes and can present large variations from plant to plant. For WWTPs without anaerobic sludge handling processes, the majority of the CH<sub>4</sub> emitted originates from the dissolved CH<sub>4</sub> in the influent formed in sewer networks. For WWTPs with anaerobic sludge handling processes, anaerobic sludge treatment and handling facilities contribute the most to the CH<sub>4</sub> emissions in plants. CH<sub>4</sub> emissions can substantially contribute to the carbon footprint of a WWTP, especially in those facilities with low N<sub>2</sub>O emissions. Despite of its importance in the overall emitted GHG, there are only a few studies in the literature reporting CH<sub>4</sub> emissions from full-scale systems.

Finally, sewer systems also present fugitive greenhouse gas emission, with CH<sub>4</sub> being the main greenhouse gas produced although N<sub>2</sub>O has also been reported. The reporting of emissions from sewers is much more scarce as compared to WWTPs but its important contribution to the overall CH<sub>4</sub> emissions of wastewater systems cannot be neglected.

## 6.2 N<sub>2</sub>O EMISSIONS FROM FULL-SCALE WWTP MONITORING RESULTS

This chapter reports emission factors (EFs) for the main BNR processes for wastewater treatment and proposes mitigation measures (Table 6.1). Monitoring campaigns to quantify and mitigate N<sub>2</sub>O emissions have been performed over recent years in different WWTP configurations. Our observations to date confirm that due to differences in monitoring strategies (i.e. length of monitoring period) and design and operational conditions, universally acceptable configuration-based or performance based EF estimation modes are not yet available. The challenge of evaluating and mitigating N<sub>2</sub>O emissions from BNR processes is further complicated by practical and technological hurdles that are related with the little field data regarding N<sub>2</sub>O emissions for several BNR processes and other operational constraints.

Mainstream process groups include biological nutrient removal systems targeting N-removal (N-BNR) (aerobic/anoxic compartments), biological nutrient removal systems targeting both N and P removal (NP-BNR) (anaerobic/anoxic/aerobic compartments) and conventional activated sludge (CAS) systems (only aerobic reactors). Oxidation ditch (OD) reactor types and sequencing batch reactor (SBR) types have been considered as distinct process groups. Sidestream processes including partial-nitrification reactors, 1-step and 2-step PN-anammox and nitrification-denitrification configurations are also categorized as a distinct process group. Other processes with fewer than two case studies

**Table 6.1** Methods and main findings of studies resulting in mitigation measures (The abbreviations are explained below\*).

Process	EF range (% N-load)	Main findings	Mitigation measures	Source
Partial Nitrification – Anammox (1 reactor)	0.17–3.9	<ul style="list-style-type: none"> <li>• Smoother aeration transitions during normal reactor operation connected with lower N<sub>2</sub>O emissions; comparison with experiments</li> <li>• Prolonged anoxic periods leading to increased N<sub>2</sub>O emissions</li> <li>• Over-aeration significantly impacting on N<sub>2</sub>O emissions</li> <li>• Nitrification-denitrification SBR: the accumulation of N<sub>2</sub>O at the end of the SBR anoxic phase is stripped in the subsequent aerobic phase and can have a significant impact on the amount of N<sub>2</sub>O emitted</li> <li>• DO and conductivity have been linked with emissions in nitrification-denitrification SBR systems</li> </ul>	<ul style="list-style-type: none"> <li>• Optimize the aeration regime by introducing aeration control and ensuring smooth shift patterns in the aeration</li> <li>• Preferably operate under shorter cycles and short aeration intervals to avoid accumulation of NO<sub>2</sub><sup>-</sup></li> <li>• Step feeding and use of conductivity as a surrogate to estimate the effluent NH<sub>4</sub>-N concentration of the reactor and optimize the anaerobic supernatant feeding load (avoid either FA accumulation or high AOR that trigger N<sub>2</sub>O)</li> <li>• Frequent alternation of aerobic/anoxic phases to avoid nitrite accumulation</li> </ul>	<p>Castro-Barros <i>et al.</i> (2015); Kampschreur <i>et al.</i> (2009a, b); Weissenbacher <i>et al.</i> (2010); Joss <i>et al.</i> (2009); Christensson <i>et al.</i> (2013)</p>
Partial Nitrification/– Anammox (2 reactors) and nitrification-denitrification systems	2.3–7.6	<ul style="list-style-type: none"> <li>• Nitrification: N<sub>2</sub>O formation higher during non-aerated periods</li> <li>• Splitting the anoxic period: average anoxic N<sub>2</sub>O formation rate decreased</li> <li>• Shorter cycles can reduce the N<sub>2</sub>O EF at the expense of higher NO<sub>3</sub><sup>-</sup> concentrations</li> <li>• Anammox reactor: NO<sub>2</sub><sup>-</sup> accumulation potentially increasing N<sub>2</sub>O emissions</li> </ul>	<ul style="list-style-type: none"> <li>• Apply continuous aeration in nitrification reactor</li> <li>• Operate under lower DO setpoint and control the aeration rate. It is preferred that DO &gt; 1.5 mg/L. Lower DO levels have been linked with elevated N<sub>2</sub>O generation in nitrification-denitrification SBR systems.</li> <li>• Avoid anoxic phases in nitrification reactors</li> <li>• Operate a one-reactor nitrification-anammox process; potentially emitting less N<sub>2</sub>O due to limited NO<sub>2</sub><sup>-</sup> accumulation</li> </ul>	<p>Mampaey <i>et al.</i> (2016); Kampschreur <i>et al.</i> (2008); Vasilaki Gustavsson and la Cour Jansen (2011); Ahn <i>et al.</i> (2010)</p>

(Continued)

**Table 6.1** Methods and main findings of studies resulting in mitigation measures (The abbreviations are explained below\*) (Continued).

Process	EF range (% N-load)	Main findings	Mitigation measures	Source
N-BNR	0.018–4	<ul style="list-style-type: none"> <li>N<sub>2</sub>O emissions have been correlated with increased abundances of AOB &amp; lower counts of N<sub>2</sub>O-reducers; AOB abundance favoured by higher NO<sub>3</sub><sup>-</sup> &amp; NO<sub>2</sub><sup>-</sup> concentrations</li> <li>DO exhibiting a significant influence on the N<sub>2</sub>O production</li> <li>N<sub>2</sub>O production mainly in the aerated zones, minor N<sub>2</sub>O consumption &amp; minor stripping effect in the anoxic zones</li> <li>N<sub>2</sub>O emitted directly from the aeration basin: low COD:N ratio limiting denitrification &amp; leading to 5-times higher N<sub>2</sub>O emissions</li> <li>N<sub>2</sub>O dynamics not significantly influenced by DO variations (within the range of 1.5–2 mg/L)</li> <li>Daily N<sub>2</sub>O peaks occurring under higher aeration flow rates (more intense N<sub>2</sub>O stripping) and under elevated bulk NO<sub>2</sub><sup>-</sup> concentrations in the bioreactor and under poor plant aeration performance and insufficient DO</li> <li>Low EF: diluted influent (groundwater infiltration) as the most probable reason</li> </ul>	<ul style="list-style-type: none"> <li>Avoid NO<sub>2</sub><sup>-</sup> accumulation, low temperatures &amp; excess DO in the anoxic bioreactors to enable complete heterotrophic denitrification &amp; hinder nitrifier denitrification</li> <li>Apply proper control of DO in aerated compartments</li> <li>Apply the BP-ANN model as a convenient &amp; effective method for the description of N<sub>2</sub>O emissions in an A/O</li> <li>Studies in full-scale MLE reactors have not suggested process/study specific mitigation measures</li> </ul>	<p>Castellano-Hinojosa <i>et al.</i> (2018); Sun <i>et al.</i> (2017); Kosonen <i>et al.</i> (2016); Aboobakar <i>et al.</i> (2013); Samuelsson <i>et al.</i> (2018); Masuda <i>et al.</i> (2018); Rodriguez-Caballero <i>et al.</i> (2014); Pan <i>et al.</i> (2016); Ahn <i>et al.</i> (2010); Bellandi <i>et al.</i> (2018); Spinelli <i>et al.</i> (2018); Townsend-Small <i>et al.</i> (2011); Foley <i>et al.</i> (2010); Foley <i>et al.</i> (2015)</p>

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**Table 6.1** Methods and main findings of studies resulting in mitigation measures (The abbreviations are explained below\*) (Continued).

Process	EF range (% N-load)	Main findings	Mitigation measures	Source
NP-BNR	0.068–3.4	<ul style="list-style-type: none"> <li>N<sub>2</sub>O emitted mainly from the oxic zone, with the emission levels increasing greatly from the beginning of the oxic zone towards the anoxic zone</li> <li>NO<sub>2</sub><sup>-</sup> accumulation directly triggering N<sub>2</sub>O production</li> <li>Both diurnal &amp; seasonal N<sub>2</sub>O emission levels fluctuating strongly; N<sub>2</sub>O generated &amp; emitted more in summer than in winter</li> <li>Other factors influencing the N<sub>2</sub>O emission: low DO/temperature</li> <li>Microbial population &amp; aeration strategy as key factors of N<sub>2</sub>O generation &amp; emission</li> <li>Risk of elevated emissions in processes with plug-flow pattern with step feeding</li> <li>Significant spatial variability of N<sub>2</sub>O generation within the reactor</li> </ul>	<ul style="list-style-type: none"> <li>Increase DO availability for both AOB &amp; NOB and improve the growth conditions of AOB</li> <li>Apply a step-stage aeration mode with varying aeration intensities (location-specific emission patterns for a plug-flow process)</li> <li>Ensure better mixing via a higher horizontal plug-flow rate combined with an appropriate vertical airflow flux; the large cross-section widths reduced using partition walls to elevate flow velocities under a constant A<sup>2</sup>/O tank working volume</li> <li>Avoid incomplete/intermittent nitrification &amp; over-aeration during the aerobic processes to achieve lower N<sub>2</sub>O emissions</li> <li>Apply uniform spatial DO profiles to promote SND that probably leads to less N<sub>2</sub>O emissions</li> <li>Perform flow equalization to control the peaking factor of the influent N-loading to the AS</li> <li>Ensure a sufficiently long SRT to prevent NO<sub>2</sub><sup>-</sup> accumulation during nitrification</li> <li>Avoid the COD limitation of the denitrification process by minimizing the pre-sedimentation of organic carbon in the influent &amp; dosing additional organic carbon</li> <li>Avoidance of over-aeration (DO 1–2 mg/L) and control of mixed liquor recirculation rates to exceed 500% has been shown to reduce N<sub>2</sub>O emissions</li> </ul>	<p>Wang <i>et al.</i> (2016b); Li <i>et al.</i> (2016); Ren <i>et al.</i> (2013); Foley <i>et al.</i> (2010); Foley <i>et al.</i> (2015); Yan <i>et al.</i> (2014); Zaborowska <i>et al.</i> (2019)</p>

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**Table 6.1** Methods and main findings of studies resulting in mitigation measures (The abbreviations are explained below\*) (Continued).

Process	EF range (% N-load)	Main findings	Mitigation measures	Source
SBR	0.58–5.6	<ul style="list-style-type: none"> <li>NH<sub>4</sub><sup>+</sup> accumulation leading to a high AOR during the aerobic SBR phases and, finally, to the increased production of intermediates (e.g. NH<sub>2</sub>OH)</li> <li>Low DO during nitrification majorly influencing N<sub>2</sub>O production</li> <li>Cycles with long aerated phases showing the largest N<sub>2</sub>O emissions, with a consequent increase in the carbon footprint</li> <li>Transient NH<sub>4</sub><sup>+</sup> &amp; NO<sub>2</sub><sup>-</sup> concentrations &amp; transition from anoxic to aerobic possibly involved in the increased N<sub>2</sub>O production</li> <li>Spatial variability of N<sub>2</sub>O generation within the reactor has been reported due to elevated NH<sub>4</sub><sup>+</sup> concentration in the feeding point</li> </ul>	<ul style="list-style-type: none"> <li>Apply intermittent aeration and reduce the aerated periods to decrease the NO<sub>2</sub><sup>-</sup> accumulation</li> <li>Increase the aeration rate during the feeding period &amp; decrease it to a proper level for nitrification in the aerobic stage</li> <li>Alternatively, change the operational SBR mode (from feeding under synchronous aeration to feeding with anoxic stirring) to ensure enough COD provision/better utilization of influent COD for denitrification</li> <li>Allow the system to consume N<sub>2</sub>O through denitrification. Extend denitrification length or if required supply external carbon source during denitrification</li> <li>Continuous aeration at DO equal to ~0.5 mg/L favouring simultaneous nitrification-denitrification can also reduce the N<sub>2</sub>O</li> </ul>	<p>Ni <i>et al.</i> (2013); Rodriguez-Caballero <i>et al.</i> (2015); Sun <i>et al.</i> (2013); Duan <i>et al.</i> (2020); Gruber <i>et al.</i> (2020); Foley <i>et al.</i> (2010); Foley <i>et al.</i> (2015)</p>
OD	0.03–2.8	<ul style="list-style-type: none"> <li>Both nitrifying &amp; denitrifying zones are potential hotspots of N<sub>2</sub>O production</li> <li>Relatively low emissions due to strong dilution effect (relatively long HRT), AOB ≈ NOB (minor NO<sub>2</sub><sup>-</sup> accumulation, less likely N<sub>2</sub>O production via nitrifier denitrification), more uniform DO profile in the OD process (SND promoted)</li> <li>Aerated zones: N<sub>2</sub>O fluxes correlated with location-specific pH, AS mixed liquor temperature, DO, NH<sub>4</sub><sup>+</sup> &amp; NO<sub>2</sub><sup>-</sup> concentrations &amp; interactive combinations</li> <li>Anoxic zones: N<sub>2</sub>O fluxes correlated with location-specific sCOD, pH, AS mixed-liquor temperature, DO, NO<sub>2</sub><sup>-</sup> &amp; NO<sub>3</sub><sup>-</sup> concentrations &amp; interactive combinations</li> </ul>	<ul style="list-style-type: none"> <li>Essential to control DO at proper level during nitrification/denitrification &amp; enhance the utilization rate of influent organic carbon for denitrification</li> <li>Multivariate analysis can be applied (i.e. clustering, classification) to investigate the combined effect of operational variables on N<sub>2</sub>O emissions</li> </ul>	<p>Sun <i>et al.</i> (2015); Ren <i>et al.</i> (2015); Daelman <i>et al.</i> (2015); Vasilaki <i>et al.</i> (2018); Ahn <i>et al.</i> (2010); Yan <i>et al.</i> (2014); Masuda <i>et al.</i> (2018); Chen <i>et al.</i> (2019); Foley <i>et al.</i> (2010); Foley <i>et al.</i> (2015); Ekström <i>et al.</i> (2017)</p>

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Table 6.1 Methods and main findings of studies resulting in mitigation measures (The abbreviations are explained below\*) (Continued).

Process	EF range (% N-load)	Main findings	Mitigation measures	Source
CAS	0.36–1.8	<ul style="list-style-type: none"> <li>Nitrification is the main driving force behind N<sub>2</sub>O emission peaks</li> <li>Compared to other parameters (e.g. sludge concentration/retention time), air flow-rate variations possibly influencing the N<sub>2</sub>O emissions; high N<sub>2</sub>O emissions under conditions of over-aeration or incomplete nitrification along with NO<sub>2</sub><sup>-</sup> accumulation</li> <li>The treatment of the anaerobic supernatant in mainstream CAS systems can trigger significant N<sub>2</sub>O emissions</li> </ul>	<ul style="list-style-type: none"> <li>Reduce the aeration rate to decrease the N<sub>2</sub>O emission rate and energy consumption required for aeration</li> <li>Add an anoxic zone &amp; recirculation to a non-BNR system for nitrification; otherwise, high N<sub>2</sub>O emissions expected in case of increased DO</li> <li>Control the DO; dynamic changes in DO concentrations reported as being responsible for N<sub>2</sub>O emission peaks</li> <li>Avoid the concurrence of decreased DO &amp; NO<sub>2</sub><sup>-</sup> accumulation</li> </ul>	Chen <i>et al.</i> (2016); Ribeiro <i>et al.</i> (2017); Ahn <i>et al.</i> (2010); Gruber <i>et al.</i> (2020); Brotto <i>et al.</i> (2015)
Other/generic	0.004–2.8	<ul style="list-style-type: none"> <li>Investigate possible links between WWTP operating conditions &amp; N<sub>2</sub>O emission fluxes</li> <li>Aerobic zones: N<sub>2</sub>O fluxes correlated with location-specific pH, AS mixed liquor temperature, DO, NH<sub>4</sub><sup>+</sup> &amp; NO<sub>2</sub><sup>-</sup> concentrations &amp; interactive combinations</li> <li>Anoxic zones: N<sub>2</sub>O fluxes correlated with location-specific sCOD, pH, AS mixed-liquor temperature, DO, NO<sub>2</sub><sup>-</sup> &amp; NO<sub>3</sub><sup>-</sup> concentrations &amp; interactive combinations</li> <li>In cases of low external C-source availability, internally stored compounds (e.g. polyhydroxyalkanoates (PHAs)) can be alternatively utilized. The latter is likely to increase the N<sub>2</sub>O production during denitrification</li> </ul>	<ul style="list-style-type: none"> <li>BNR processes: Avoid high NH<sub>4</sub><sup>+</sup> &amp; NO<sub>2</sub><sup>-</sup> &amp; DO concentrations &amp; transients</li> <li>Aerobic processes: avoid incomplete/intermittent nitrification &amp; over-aeration</li> <li>Rely on more uniform spatial DO profiles to promote SND</li> <li>Minimize peak N-flow (flow equalization)</li> <li>pH maintained 6 ≤ pH ≤ 7</li> <li>Provision of sufficient C-source to increase the possibility of N<sub>2</sub>O consumption through denitrification.</li> <li>DO must be controlled at approximately 2 mg/L while aeration minimized to avoid stripping</li> <li>Perform advanced N-removal (e.g. nitrification-denitrification or partial nitrification-anammox) only after optimizing the process parameters. BP-ANN and data-driven models suitable for the description of N<sub>2</sub>O emissions in other WWTPs with different configurations (e.g. A<sup>2</sup>/O, SBR &amp; nitrification-anammox), if influent/environmental parameters &amp; N<sub>2</sub>O emission data can be investigated through full-, pilot- or lab-scale experiments</li> </ul>	Wang <i>et al.</i> (2016a); Ahn <i>et al.</i> (2010); Samuelsson <i>et al.</i> (2018); Kosonen <i>et al.</i> (2016); Mello <i>et al.</i> (2013); Filali <i>et al.</i> (2013); Foley <i>et al.</i> (2010); Foley <i>et al.</i> (2015); Baresel <i>et al.</i> (2016); Gruber <i>et al.</i> (2020)

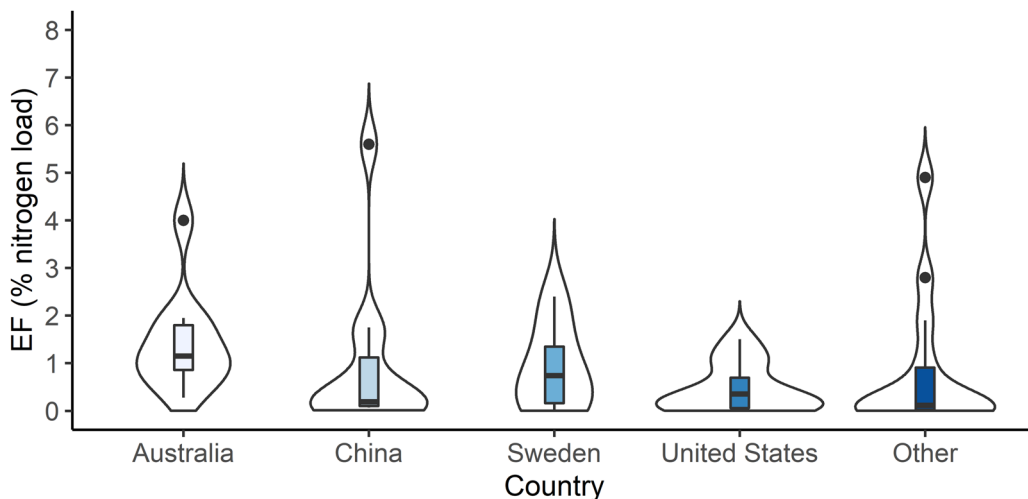
\*SBR: sequencing batch reactor; DO: dissolved oxygen; FA: free ammonia; AOR: ammonia oxidation rate; AOB: ammonia oxidizing bacteria; NOB: nitrite oxidizing bacteria; COD: chemical oxygen demand; BP-ANN: back propagation-artificial neural network; A/O: anoxic/oxic reactor; MLE: modified Ludzack-Ettinger; N-BRN: nitrifying – biological nitrogen removal; NP-BNR: biological nitrogen and phosphorus removal; A<sup>2</sup>/O: anaerobic/anoxic/oxic reactor; SND: simultaneous nitrification denitrification; AS: activated sludge; SRT: sludge retention time; HRT: hydraulic retention time; OD: oxidation ditch; sCOD: soluble chemical oxygen demand; CAS: conventional activated sludge.

which do not belong to the aforementioned process groups are categorized separately. These include intermittently aerated or simultaneous nitrification-denitrification reactors (i.e., Filali *et al.*, 2013; Gruber *et al.*, 2020; Mello *et al.*, 2013), systems with external carbon dosage (Ahn *et al.*, 2010) and biofilm reactors for C (i.e. Townsend-Small *et al.*, 2011) or N removal (i.e., Bollon *et al.*, 2016).

In total ~67% of the analysed mainstream reactors, have reported the quantified EFs in terms of the %N-load. Approximately 12% of the studies have reported the EFs in terms of N-removed.

There is a significant variation in the  $N_2O$  emissions of full-scale wastewater treatment processes. The  $N_2O$  emissions range reported in literature is between 0.003% of the influent N-load for a mainstream BNR system treating municipal low-strength wastewater, diluted by groundwater and marine intrusions and 7.6% of the  $NH_4-N$  load for a sidestream short cut enhanced nutrient abatement (SCENA) SBR treating anaerobic digestion supernatant. Generally, BNR processes treating high strength streams have been associated with high risk of elevated  $N_2O$  emissions. This is mainly due to the high ammonia oxidation rate (AOR) and  $NO_2^-$  accumulation typically observed in such systems (Desloover *et al.*, 2011; Gustavsson & la Cour Jansen, 2011; Kampschreur *et al.*, 2008). Discrepancies in the EFs observed in the different process groups can, to some extent, be attributed to variations in operational characteristics and control parameters. In addition to reactor configuration, emission rates depend on the operational/environmental conditions and preferred enzymatic pathways (Wan *et al.*, 2019).

Figure 6.1 shows boxplots of the observed EFs (with respect to the influent N-load) of mainstream processes in different countries. The width of the violin plot outlines surrounding the boxplots represents the data kernel density distribution of the EFs. Overall, ~60% of the monitoring campaigns in processes treating low-strength streams have been performed in China (18%), the United States (18%), Australia (14%) and Sweden (10%). Overall, the highest EFs have been reported in Australia. The median EF in Australia is 1.35% of the N-load (average equal to 1.6%). The lowest EFs have been

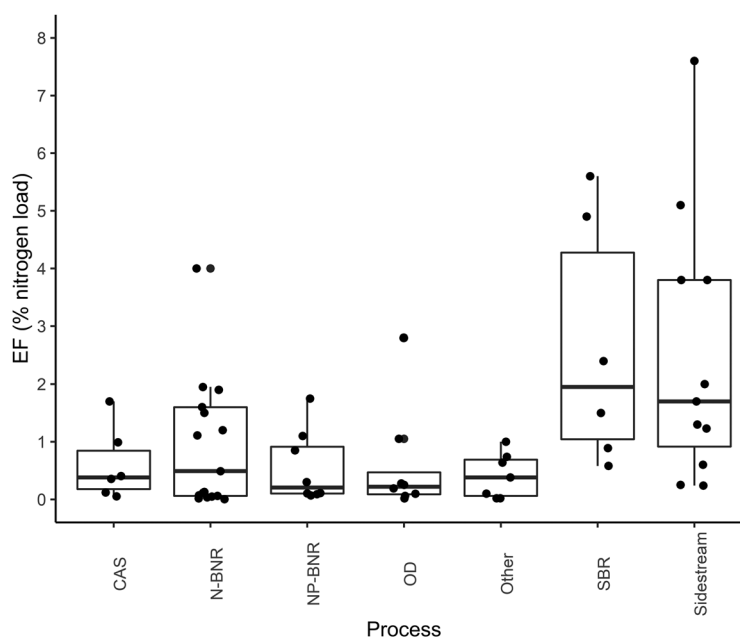


**Figure 6.1** Boxplots of the reported EFs (% N load) with respect to the WWTP in different countries using violin plot outlines. The rectangles represent the interquartile range. The median is denoted by the black horizontal line dividing the box into two parts. The dots represent the values exceeding 1.5 times the interquartile range. The upper and lower whiskers stand for values higher or lower than the interquartile range, respectively (within 1.5 times the interquartile range above and below the 75th and 25th percentile, respectively). The violin plot outlines show the kernel probability density of the EF in mainstream and sidestream processes; the width of the violin plot outlines represents the proportion of the data located there.

reported in China; the median EF is equal to 0.2% of the N-load (average 0.8% of the N-load). In the United States the median EF is 0.3%, while in Sweden the median EF is 0.74% of the N-load (averages equal to 0.4% and 0.9% of the N-load, respectively.)

The majority of the processes monitored in Australia are step-feed reactors. Higher than average N<sub>2</sub>O emissions have been reported for step-feed reactors. Moreover, the majority of the WWTPs studied in China do not have anaerobic digestion on-site. The anaerobic supernatant is a by-product from the treatment of the primary and secondary sludge via anaerobic digestion when the digestate is dewatered. This stream is small in volume (1–2% compared to the mainstream line), but very concentrated in nutrients and is conventionally recycled back to the primary treatment increasing the loads (and thus, the energy requirements, costs and potentially the N<sub>2</sub>O emissions) of the mainstream biological treatment. The majority of the studied processes in Sweden and Australia belong to WWTPs with anaerobic digestion on-site. It is possible that WWTPs that recycle anaerobic supernatant that contains 10–20% of the WWTP nitrogen load, have a higher risk of increased N<sub>2</sub>O emissions. The sampling protocols and duration of monitoring campaigns also vary significantly between the different countries. For instance, long-term monitoring of N<sub>2</sub>O emissions (>6 months) has been performed mainly in China via grab-samples collected bi-monthly. [Vasilaki \*et al.\* \(2019\)](#) showed that low-frequency (i.e. bimonthly) grab-sampling might underestimate emissions due to limitations in sampling duration (i.e. it does not occur during night-time) or due to short-term process perturbations triggering elevated emissions not coinciding with the sampling days.

[Figure 6.2](#) shows the EF range for the different groups of mainstream processes and sidestream processes. As a general remark, the majority of the EFs in processes treating low-strength wastewater



**Figure 6.2** Boxplots visualizing the EF range for the different groups of mainstream processes and sidestream processes (adapted from [Vasilaki \*et al.\* 2019](#)). The rectangles represent the interquartile range. The median is denoted by the black horizontal line dividing the box into two parts. The dots represent values exceeding 1.5 times the interquartile range. The upper and lower whiskers represent values higher or lower the interquartile range, respectively (within 1.5 times the interquartile range above and below the 75th and 25th percentile, respectively).

range from 0.1% to 2% of the influent N-load. Higher than average emissions have been reported in SBRs and step-fed plug-flow reactors. The potential for N<sub>2</sub>O emissions from reactors treating high-strength wastewater streams is considered higher compared to the mainstream BNR processes. This is mainly because the nitrification/partial-nitrification occurring during sidestream treatment is linked with higher ammonia oxidation rate (AOR) and NO<sub>2</sub><sup>-</sup> accumulation (Desloover *et al.*, 2011; Gustavsson & la Cour Jansen, 2011; Kampschreur *et al.*, 2008).

The benchmarking of the EF for groups of processes remains challenging, mainly due to differences in the strategies applied during monitoring, the operational and environmental conditions and the duration of monitoring campaigns. Additionally, limited information exists on the N<sub>2</sub>O emissions for several other processes (e.g. biofilm-based processes or partial-nitrification-anammox systems, etc.) (Sabba *et al.*, 2018; Vasilaki *et al.*, 2019).

Process characteristics, EFs, N<sub>2</sub>O triggering mechanisms, operational conditions and mitigation measures for processes treating low-strength and high-strength wastewater streams are analysed in Sections 6.2.1 and 6.2.2, respectively.

High sensitivity of the quantified EF between different monitoring strategies and monitoring campaign durations has been reported (Vasilaki *et al.*, 2019). When considering the duration of the monitoring campaign, studies lasting over a year result in a median EF equal to 1.7% of the N-load. On the other hand, most of the monitoring campaigns lasting less than one month have reported EFs less than 0.3% of the N-load. Therefore, short-term monitoring periods may fail to capture underlying seasonal variations in the N<sub>2</sub>O formation (or be affected by short-term process perturbations) and, consequently, result in unreliable EFs. Similarly, the studies monitoring N<sub>2</sub>O emissions in mainstream wastewater processes continuously (i.e. online via gas analysers), have quantified higher N<sub>2</sub>O EFs than studies monitoring N<sub>2</sub>O emissions discontinuously (i.e. offline via grab samples). The average EFs of mainstream wastewater processes monitored continuously and discontinuously are 1.2% and 0.44% of the N-load, respectively. Low-frequency sampling campaigns have a high risk of not sufficiently capturing short-term changes in pollutant concentrations, operational conditions and system disturbances impacting N<sub>2</sub>O generation.

The reliability of the monitoring campaigns also depends on the amount and location of the sampling points (Gruber *et al.*, 2020). Significant spatial variations of the N<sub>2</sub>O emissions have been reported in complete mixing reactors (Duan *et al.*, 2020). The variability was attributed to gradients in the nutrients within the reactor and elevated NH<sub>4</sub><sup>+</sup> concentrations close to the feeding area causing increased AOR and triggering N<sub>2</sub>O emissions. The use of one gas chamber for N<sub>2</sub>O emissions collection in complete mixing reactors might result in unreliable quantification of N<sub>2</sub>O EFs. The latter can have significant implications, since one gas chamber is conventionally used for sampling in complete mixing reactors, whereas several sampling points are suggested for reactors operating in plug-flow mode (Duan *et al.*, 2020). On the other hand, Gruber *et al.* (2020) observed negligible spatial variability of N<sub>2</sub>O emissions in a complete mixing reactor monitored with three gas chambers in different locations within the reactor. Therefore, additional studies are required to determine the optimum N<sub>2</sub>O sampling points and understand under which conditions nutrient gradients are observed.

Differences in the N<sub>2</sub>O emissions have been also reported in parallel reactors. Chen *et al.* (2019), studied parallel OD reactors and observed deviations in the N<sub>2</sub>O emissions behaviour under similar NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and dissolved oxygen (DO). They suggested that the reliable quantification of WWTP N<sub>2</sub>O EFs requires monitoring of all plant reactors. The opposite has been reported by Daelman *et al.* (2015) who observed similar N<sub>2</sub>O emission patterns in two parallel OD reactors.

Generally, the quantification of reliable annual EFs requires sampling campaigns lasting at least 1 year. Additionally, a decision tree for the selection of the monitoring strategy has been developed by Gruber *et al.* (2020). They define specific criteria for the selection of sampling points and location. Influent compositions, feeding locations and homogeneity, and the key performance indicators (i.e. removal efficiencies) should be considered to decide whether similar N<sub>2</sub>O emissions are expected in parallel reactors. Similarly, plug-flow type reactors featuring spatial variability of concentrations and aeration intensity require multiple sampling points.

The variability of EF reported in full-scale wastewater treatment processes can be attributed to complex relationships between emitted N<sub>2</sub>O and operational conditions and different configurations (i.e., SBR, continuous systems), loads (i.e., NH<sub>4</sub><sup>+</sup> concentrations), feeding strategies and operational control (i.e., DO set-points).

The conditions leading to elevated N<sub>2</sub>O emissions or N<sub>2</sub>O generation are usually associated with N-forms build-up in the reactor (i.e., NH<sub>4</sub><sup>+</sup>, NH<sub>2</sub>OH, NO<sup>-</sup>, NO<sub>2</sub><sup>-</sup>). Depending on the BNR process and the acclimatized biomass in the reactor, the accumulation of N intermediates does not necessarily have to be very high to trigger N<sub>2</sub>O pathways. The accumulation mainly depends on the influent dynamics or on improper process operation and/or design.

During nitrification, NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup> concentration can significantly affect the N<sub>2</sub>O emissions (Law *et al.*, 2012; Wunderlin *et al.*, 2012). High NO<sub>2</sub><sup>-</sup> accumulation, that is the toxic product of aerobic NH<sub>3</sub> oxidation in AOB, has also been linked with elevated N<sub>2</sub>O emissions, especially under low DO concentrations (Desloover *et al.*, 2011; Kampschreur *et al.*, 2008; Law *et al.*, 2012; Massara *et al.*, 2017; Peng *et al.*, 2015; Tallec *et al.*, 2006). Different N<sub>2</sub>O production dynamics can be potentially triggered under the same NO<sub>2</sub><sup>-</sup> concentration depending on the type of AOB. It has been also reported that AOB can adapt to different environments with different NO<sub>2</sub><sup>-</sup> concentrations. Overall, N<sub>2</sub>O generation has been associated with higher NO<sub>2</sub><sup>-</sup> concentrations in wastewater treatment processes (Foley *et al.*, 2010). DO is also considered an important parameter affecting N<sub>2</sub>O emissions (Kampschreur *et al.*, 2009b), with sub-optimum DO concentrations generally increasing N<sub>2</sub>O emissions. AOB can use nitrite instead of oxygen as an electron acceptor (Kampschreur *et al.*, 2009a, b), in oxygen limiting conditions, generating N<sub>2</sub>O emissions. At present, establishing a generic optimum DO concentration threshold to minimize N<sub>2</sub>O emissions for nitrifying systems is not possible since other compounds (i.e. N compounds discussed above) have a simultaneous effect on N<sub>2</sub>O generation. An optimal DO level for minimal N<sub>2</sub>O emissions can be established for each system taking into consideration the concentration of other compounds that affect these emissions. Overall, in aerated reactors/zones, higher emissions are expected under high NH<sub>4</sub><sup>+</sup> concentrations, high AOR, sub-optimum DO (under or over-aeration) or NO<sub>2</sub><sup>-</sup> build-up (Desloover *et al.*, 2011; Kampschreur *et al.*, 2008). Sub-optimum pH and short solids retention times (SRTs) have been reported to influence N<sub>2</sub>O production in AOB.

Additionally, feeding mainstream reactors with high-strength streams (i.e., anaerobic supernatant) can create peak nutrient loadings increasing the risk of elevated N<sub>2</sub>O emissions. In the studied processes, WWTPs that have anaerobic digestion on-site have median EFs equal to 1.5% of the N-loading (average equal to 1.47%). On the other hand, processes that are not fed with anaerobic supernatant (i.e., WWTPs applying sludge dewatering and drying) have a median EF equal to 0.11% of the N-load (average 0.47% of the N-load).

Sub-optimum DO, chemical oxygen demand (COD), pH and SRT can also result in nitrite and N<sub>2</sub>O accumulation during denitrification (Schulthess *et al.*, 1994; Yang *et al.*, 2012). Low values of COD/N can result in incomplete denitrification and, therefore, N<sub>2</sub>O accumulation via the heterotrophic denitrification pathway (Wunderlin *et al.*, 2012).

Seasonal environmental variations, can influence the bacterial community structure in WWTPs (Flowers *et al.*, 2013) and the N<sub>2</sub>O emissions. Temperature can significantly affect the AOB specific growth rate during nitrification (Van Hulle *et al.*, 2010). The higher temperature also decreases the N<sub>2</sub>O solubility, thus intensifying the N<sub>2</sub>O stripping to the atmosphere (Reino *et al.*, 2017). Adouani *et al.* (2015) reported an increased sensitivity of the N<sub>2</sub>O reductase activities at lower temperatures compared to other denitrification enzymes and, therefore, to incomplete denitrification. Other seasonal variations (e.g., influent loading, wet and dry season) can affect the enzymatic reactions and the emissions. Vasilaki *et al.* (2018) observed peaks of N<sub>2</sub>O emissions coinciding with precipitation events, at low temperatures. Further investigation is required to understand the impact of seasonal effects on the N<sub>2</sub>O emissions (Gruber *et al.*, 2020; Vasilaki *et al.*, 2019).

Disturbances in the process can affect short-term (i.e., 1 day) or even longer period (i.e., >1 week) N<sub>2</sub>O generation (Vasilaki *et al.*, 2018). Gruber *et al.* (2021) observed in an SBR reactor, that N<sub>2</sub>O

emission peaks, nitrification failure, poor activated sludge settleability and high turbidity of treated effluent, were all linked to a less diverse microbial community and changes in community mixture. Specifically, a decrease in abundance of filamentous and nitrite oxidizing bacteria was reported.

## 6.2.1 Processes treating low strength streams

### 6.2.1.1 N-BNR and NP-BNR processes

This section discusses findings regarding  $N_2O$  generation in BNR processes. The Modified Ludzack-Ettinger (MLE) process is the most studied N-removal configuration. In total, 41% of the N-BNR systems are MLE processes. The MLE process consists of anoxic and aerobic tanks and a secondary settler. The influent wastewater is first fed to the anoxic tank for denitrification and next to the aerobic zone for nitrification. The process uses an internal recycle flow from the aerobic tank to the head of the anoxic tank providing nitrate for denitrification. After anoxic and aerobic processes, the wastewater is fed to the secondary settler. A part of the sludge, the return activated-sludge, returns to the head of the anoxic zone to increase the mixed liquor volatile suspended solids (MLVSS) concentration in the reactor. In total, the N-BNR configurations consist of a broad category of processes with anoxic and oxic compartments. Step-feed plug-flow reactors with alternating anoxic/oxic zones and reactors with small anoxic compartments (for predenitrification) and aerobic compartments with and without recirculation of nitrates belong to this category.

Similarly, the anaerobic/anoxic/aerobic ( $A^2/O$ ) process is the most studied N and P-removal configuration. In total 64% of the NP-BNR systems are  $A^2/O$  processes. The  $A^2/O$  process is a modification of the MLE process. The process consists of an anaerobic zone followed by the same configuration of MLE. The return activated sludge goes to the head of the anaerobic tank. The anoxic tank is used to decrease the amount of nitrate, in the anaerobic tank, that returns from the activated sludge. Overall, the NP-BNR process group includes configurations with anaerobic, anoxic and aerobic compartments, such as reversed- $A^2/O$  configurations ( $A^2/O$  systems where the anaerobic and anoxic compartments are reversed) or  $A^2/O$  systems with a predenitrification zone.

The median EF of N-BNR processes is 0.5% of the influent N-Load, while the median EF of NP-BNR processes is 0.2% of the influent N-Load. In N-BNR configurations, the  $N_2O$  emissions range between 0.003% and 4% of the influent N-load (Foley *et al.*, 2010; Spinelli *et al.*, 2018). In NP-BNR configurations, the  $N_2O$  emissions range between 0.07% and 1.75% of the influent N-load (Wang *et al.*, 2016b; Yan *et al.*, 2014). MLE and  $A^2/O$  are the most studied configurations; around 54% of the monitoring campaigns have been performed in these two systems.

Overall, in N-BNR and NP-BNR systems,  $N_2O$  emission peaks have been reported during the transition from non-aerated to aerated zones/compartments (i.e. Rodriguez-Caballero *et al.* 2014; Sun *et al.* 2017). This can be partially due to incomplete denitrification and accumulation of dissolved  $N_2O$  under anoxic conditions. Elevated emissions have been also linked with excess DO in anoxic compartments, inhibiting complete denitrification (Castellano-Hinojosa *et al.*, 2018). Therefore, process control in the anoxic compartments should target the minimization of  $NO_2^-$  accumulation and excess DO and the avoidance of COD limitation. This will facilitate complete heterotrophic denitrification and  $N_2O$  consumption.

In aerobic compartments, peak  $N_2O$  fluxes have coincided with peak nutrient loads and low DO concentrations (Wang *et al.*, 2011, 2016b); the integration of flow equalization can control the influent N-loading peaks to the systems. Moreover, close to the inlet of aerobic compartments with a plug-flow pattern, AOB abundances and high  $NO_2^-$  concentrations can result in an increase in the  $N_2O$  emissions. Risk of elevated emissions has also been reported in processes with plug-flow pattern and step feeding. Pan *et al.* (2016) showed an EF equal to 0.7% of the influent N-load in the first step of a plug-flow reactor and 3.5% in the second step. The increased  $N_2O$  emissions in the second step were attributed to the recirculated stream being directed only at the first step causing dilution; the MLVSS concentration in the second step was 40% lower than that in the first step (70% less biomass compared to the first step). The higher specific AOR in this stage triggered the  $N_2O$  generation. It is

important to note that in reactors with plug-flow pattern, the effect of the N-load, DO concentration and temperature on N<sub>2</sub>O emissions varies along the reactor (Aboobakar *et al.*, 2013). Thus, the dominant N<sub>2</sub>O triggering conditions can also vary.

Low EFs have been reported in reactors treating diluted low-strength wastewater (i.e. due to groundwater infiltration) (Bellandi *et al.*, 2018; Spinelli *et al.*, 2018). Low EFs have also been reported in the majority of the A<sup>2</sup>/O and reversed A<sup>2</sup>/O processes, with the median N<sub>2</sub>O EF ~0.11% of the influent N-load. However, it must be noted that the seasonal variability of the N<sub>2</sub>O emissions in A<sup>2</sup>/O reactors has not been studied adequately. The majority of the monitoring campaigns lasted less than 3 months. Wang *et al.* (2016b) showed that the EF of an A<sup>2</sup>/O process has strong temporal patterns and varied between 0.1 and 3.4% of the influent N-load between different months within 1 year. The effect of environmental conditions on N<sub>2</sub>O generation is discussed in Section 6.2.

Both the nitrifier denitrification pathway and the NH<sub>2</sub>OH oxidation pathway have been suggested as major contributors to the N<sub>2</sub>O emissions in aerated compartments/zones. The nitrifier denitrification pathway is considered the main triggering mechanism in aerobic compartments (i) when NO<sub>2</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and O<sub>2</sub>-limiting conditions co-exist (Wang *et al.*, 2016b), (ii) when NO<sub>2</sub><sup>-</sup> is correlated with N<sub>2</sub>O emissions, (iii) when increasing N<sub>2</sub>O emissions are observed under DO limitation where sufficient O<sub>2</sub> is provided to the AOB for the oxygenation of NH<sub>3</sub> to NH<sub>2</sub>OH but not for aerobic respiration; NO<sub>2</sub><sup>-</sup> is potentially used as alternative electron acceptor to complete nitrification (Aboobakar *et al.*, 2013; Castellano-Hinojosa *et al.*, 2018; Sun *et al.*, 2017; Wang *et al.*, 2011), and (iv) under shock loads of toxic compounds, where the AOB likely activate their denitrification pathway (Rodriguez-Caballero *et al.*, 2014). In anoxic compartments, the nitrifier denitrification pathway has been suggested as the main contributor to N<sub>2</sub>O generation, when excess DO is observed (Castellano-Hinojosa *et al.*, 2018). The NH<sub>2</sub>OH oxidation pathway is significantly promoted at higher DO concentrations (Blomberg *et al.*, 2018; Zaborowska *et al.*, 2019) and when N<sub>2</sub>O emissions increase together with the AOR increase (Ni *et al.*, 2015; Pan *et al.*, 2016). Finally, heterotrophic denitrification is mainly triggered under carbon-limiting conditions (low COD/N ratio) and excess DO in anoxic compartments (Andalib *et al.*, 2017; Wunderlin *et al.*, 2012).

### 6.2.1.2 Sequencing batch reactors (SBR)

The SBR process uses a fill-and-draw complete mixing reactor operating in batch reaction steps. The biological removal and clarification occur in the same tank.

Mainstream SBRs have reported higher N<sub>2</sub>O emissions compared to the other mainstream process groups. EFs range between 0.89% of the influent N-load for an SBR that receives the anoxic selector effluent and operating under feeding (intermittent aeration), aerobic (intermittent aeration), settling and decanting sequences (Duan *et al.*, 2020) and 5.6% of the influent TN-load for an SBR operating under aerated feeding, aerobic and anoxic settling and decanting sequences (1 h each) (Sun *et al.*, 2013). The average EF from SBR reactors is 3.6% of the influent N-load (median: 3.65% of the influent N-load).

Overall, elevated emissions are attributed to (i) NH<sub>4</sub><sup>+</sup> accumulation leading to high AOR during the aerobic SBR phases and to increased production of intermediates (e.g., NH<sub>2</sub>OH, NO<sup>-</sup>, etc.), (ii) long aerated cycles, (iii) transitions from anoxic to aerobic phases possibly triggering increased N<sub>2</sub>O production, (iv) rapid changes in the NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub><sup>-</sup> concentrations within the cycle, (v) accumulation of dissolved N<sub>2</sub>O during anoxic settling and decanting that is stripped in the subsequent aerobic phase and (vi) accumulation of NO<sub>2</sub><sup>-</sup>.

Intermittent aeration and short aerated periods have been suggested to reduce the NO<sub>2</sub><sup>-</sup> accumulation in SBR systems and subsequently N<sub>2</sub>O emissions. Duan *et al.* (2020), however, showed that elevated DO concentrations (up to 8 mg/L) during intermittent aeration can also be responsible for elevated emissions in the SBR systems and should be avoided. The authors used a multi-pathway N<sub>2</sub>O model (Peng *et al.*, 2016) to design a mitigation strategy that was implemented in the studied system. They showed that continuous aeration at DO equal to ~0.5 mg/L that favours simultaneous

nitrification-denitrification (SND) can be an effective operational strategy for SBR reactors. The SND operation mode resulted in 35% reduction of the  $N_2O$  emissions compared to intermittent over-aeration. The reduction was due to the reduction of DO concentration during feeding and aerated phases that can enhance denitrification during aerated periods and minimize  $NO_2^-$  accumulation.

Additionally, in SBR reactors the supply of an external carbon source during denitrification can secure sufficient COD provision and better utilization of influent COD for denitrification (promoting complete heterotrophic denitrification). This allows the system to consume  $N_2O$  during denitrification and avoid stripping of residual liquid  $N_2O$  in the subsequent aerated phases, thus, reducing  $N_2O$  emissions. A cycle configuration with a sequence of aerobic phases (adjusted on site) followed by short non-aerated periods has been proposed as an effective control mechanism to reduce  $N_2O$  generation (Rodriguez-Caballero *et al.*, 2015).

In SBR reactors, elevated  $N_2O$  emissions are attributed to the  $NH_2OH$  pathway when elevated DO is observed during feeding and when high  $NH_4^+$  concentrations are observed without simultaneous  $NO_2^-$  increase in the aerated phases. The nitrifier denitrification pathway is the main  $N_2O$  triggering mechanism when low DO concentrations in aerobic phases are linked with the  $N_2O$  generation and when certain  $NO_2^-$  accumulation under aerobic conditions is observed in the reactor. In cases where  $N_2O$  generation continues when the aeration finishes, both the nitrifier denitrification and heterotrophic denitrification can contribute to the  $N_2O$  formation in the reactor. Finally, the correlation between  $N_2O$  emission and influent COD/N, indicates that the incomplete heterotrophic denitrification is mainly responsible for the  $N_2O$  generation.

#### 6.2.1.3 Oxidation ditch (OD)

An OD is a modified activated sludge biological treatment process; the removal of biodegradable organics is achieved by applying long SRTs. ODs are considered to approach complete mixing systems, but they can also operate in plug-flow mode.

The  $N_2O$  emissions of OD reactor types range from 0.03% of the N-load for an OD reactor favouring simultaneous nitrification denitrification (Ahn *et al.*, 2010) to 2.8% of the N-load for a system consisting of an anaerobic/anoxic/oxic plug-flow reactor followed by two parallel Carrousel reactors (Daelman *et al.*, 2015). The median EF is equal to 0.2% of the influent N-load (average equal to 0.3% of the N-load).

Overall, relatively low emissions have been reported in OD systems; this is attributed to the strong dilution effect (relatively long hydraulic retention time), to the abundance of AOB and nitrite oxidizing bacteria (NOB), and to the more uniform DO profile in the OD process especially when SND is promoted (Li *et al.*, 2016). Abundance of NOB and denitrifiers has been reported in OD systems as contributing to the consumption of  $NO_2^-$  during nitrification. The latter reduced  $NO_2^-$  accumulation and facilitated complete heterotrophic denitrification (Sun *et al.*, 2015). It is important to note, though, that the majority of the OD reactors have been monitored with gas hoods. The use of floating hoods to monitor GHG emissions in OD systems when aerated with surface aerators has been criticized due to the turbulence commonly observed at the surface affecting the capturing of the emissions in the hood (Ye *et al.*, 2014).

Elevated emissions have been linked to  $NH_4^+$  concentration peaks. In a simulation study, Ni *et al.* (2013) observed that more than 90% of the  $N_2O$  emissions were attributed to aerated zones with  $DO > 2$  mg/L and  $NH_4-N$  concentration peaks (up to  $\sim 9$  mg/L). Inadequate anoxic zones, inhibiting complete denitrification have been also reported in OD systems. OD systems with surface aerators are prone to developing zones with reduced DO, inhibiting complete nitrification, that results in nitrite accumulation and increased  $N_2O$  emissions.

A similar  $N_2O$  emissions pattern has been reported in two OD reactors operating under different control and design (Chen *et al.*, 2019; Daelman *et al.*, 2015). Both systems were monitored over a long term; an increasing trend in  $N_2O$  emissions coincided with increase in water temperature whereas, low emissions were observed under lower water temperature. Further studies are required



to understand the exact triggering mechanisms at decreasing temperatures and investigate if this N<sub>2</sub>O pattern is process-specific.

All N<sub>2</sub>O generation pathways have been reported in OD reactors. Incomplete heterotrophic denitrification has been attributed to the competition of the denitrification steps and the preference of the heterotrophic denitrifiers to reduce NO<sub>3</sub><sup>-</sup> instead of N<sub>2</sub>O under electron donor limitation (Pan *et al.*, 2013). Additionally, heterotrophic denitrification and nitrifier denitrification are the main N<sub>2</sub>O triggering mechanisms at insufficient anoxic conditions. Under these conditions NO<sub>2</sub><sup>-</sup> accumulation is expected. The NH<sub>2</sub>OH oxidation pathway will be triggered in periods with influent NH<sub>4</sub><sup>+</sup> concentration peaks, high ammonia oxidation rate and elevated DO concentrations. Vasilaki *et al.* (2018), showed that the relationships between N<sub>2</sub>O emissions and other variables monitored in an OD (i.e. NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, DO) are dynamic and affected by seasonal variations. The preferred N<sub>2</sub>O pathways were found to be dependent on time and operational conditions.

#### 6.2.1.4 Conventional activated sludge systems

CAS systems consist of aerobic reactors (1-step feed or multiple-step feed) without anoxic compartments. They are characterized by a median EF equal to ~0.4% of the influent N-load (average equal to 0.71%). The NH<sub>4</sub><sup>+</sup> removal is between 38% and 53%. The EF in CAS systems ranges from 0.05% of the N-load (translated to 9% of the NH<sub>4</sub>-N removed) (Chen *et al.*, 2019) to 1.7% of the N-load (Gruber *et al.*, 2020).

Peak loads and recirculation of the anaerobic supernatant can be responsible for the N<sub>2</sub>O fluxes observed in CAS systems, whereas high aeration rates have been reported, enhancing N<sub>2</sub>O stripping (Chen *et al.*, 2016). Additionally, the spatial variation of nutrients in step-fed CAS systems can result in incomplete denitrification and affect the AOR during nitrification (due to uneven substrate-biomass distribution in all feeding points), hence, increasing the total N<sub>2</sub>O emissions (Pan *et al.*, 2016). The treatment of the anaerobic supernatant in mainstream CAS systems has been reported to trigger significant N<sub>2</sub>O emissions. Gruber *et al.* (2020), monitored the N<sub>2</sub>O emissions in two parallel CAS systems and found that elevated emissions were observed solely in the reactor treating the anaerobic supernatant. N<sub>2</sub>O emissions can be reduced by up to 80% when influent N-loads are reduced by 30%.

Tumendelger *et al.* (2014) reported that the NH<sub>2</sub>OH oxidation pathway was responsible for up to 90% of the N<sub>2</sub>O formation under high DO (~2.5 mg/L at the middle and close to the outlet of the aerobic tank) in a CAS system (site preference (SP) isotopic analysis). Both AOB pathways contributed almost equally to N<sub>2</sub>O emissions generation at DO levels of ~1.5 mg/L, whereas nitrifier denitrification dominated at DOs lower than 1.5 mg/L. Overall, in activated sludge systems the reduction of aeration rates can decrease the N<sub>2</sub>O fluxes stripped and the control of DO has been proposed as a key measure to mitigate N<sub>2</sub>O emissions. Additionally, the addition of an anoxic zone to avoid the concurrence of decreased DO and NO<sub>2</sub><sup>-</sup> accumulation can have a positive impact on the N<sub>2</sub>O generation.

#### 6.2.2 Processes treating high strength (high nitrogen loading) streams

Sidestream processes, such as the partial-nitrification-anammox and nitrification-denitrification are emerging for the low-cost treatment of high-strength municipal wastewater streams (Lackner *et al.*, 2014; Zhou *et al.*, 2018). In the nitrification-denitrification process, ammonium is firstly oxidized to nitrite (nitrification) and then it is reduced to nitrogen gas (denitrification) under anoxic conditions. In the partial-nitrification-anammox process, ammonium is partially oxidized to nitrite and then ammonia and nitrite are converted to nitrogen gas and nitrate under oxygen-free conditions by anaerobic ammonium oxidizers (anammox).

N<sub>2</sub>O monitoring studies have been performed in less than 15 sidestream processes. There is a need to improve the understanding of N<sub>2</sub>O generation in sidestream processes. For instance, the N<sub>2</sub>O emissions were equal to 7.6% of the NH<sub>4</sub>-N load in a SCENA process and contributed up to 97% of the operational carbon footprint of the process (Vasilaki *et al.*, 2020). Additionally, the seasonal variation (~1 year) of N<sub>2</sub>O emissions in sidestream reactors has not been assessed.

The average EF from full-scale nitrification and partial-nitrification reactors is equal to 4.3% of the influent N-load. One-stage granular anammox reactors have an average EF of 1.1% of the influent N-load. [Zhuang \*et al.\* \(2020\)](#) showed that in a high-rate anammox granular sludge reactor, N<sub>2</sub>O emissions were mainly generated in anammox flocs (~10% total biomass) compared to anammox granules. They reported that the N<sub>2</sub>O reduction in flocs was inhibited due to the accumulation of NO. Anammox bacteria concentrations were higher in granules and scavenged NO that was inhibiting the N<sub>2</sub>O reduction. In comparison, emissions in lab and pilot-scale single-stage granular anammox reactors ranged from 0.1 to 12.19% of influent N-load ([Wan \*et al.\*, 2019](#)). Therefore, additional studies are required to establish reliable ranges of EFs in sidestream processes and gain insights into the mitigation of N<sub>2</sub>O emissions. Low emissions have been also reported in moving bed biofilm reactor (MBBR) anammox technologies. [Christensson \*et al.\* \(2013\)](#) reported that ~0.75% of the N-reduced were emitted as N<sub>2</sub>O at a full-scale deammonification MBBR. Process disturbances and a DO concentration lower than 1 mg/L can increase the N<sub>2</sub>O emissions. The authors concluded that stable operation at DO equal to 1 mg/L can result in average daily N<sub>2</sub>O of 0.06% of N-reduced. In pilot-scale MBBR-anammox and integrated fixed-film activated sludge (IFAS) – anammox systems [Liu \*et al.\* \(2014\)](#) reported N<sub>2</sub>O EFs equal to 0.52% and 1.7% of the total Kjeldahl nitrogen (TKN) load, respectively.

In the sidestream reactors, the rate of aeration and the DO concentration can significantly impact both the N<sub>2</sub>O emissions generation and the N<sub>2</sub>O mass fluxes stripped in the atmosphere ([Harris \*et al.\*, 2015](#); [Rathnayake \*et al.\*, 2015](#)). The influence of the aeration regime on the N<sub>2</sub>O generation varies; this can be partially due to the different configurations. For example, [Mampaey \*et al.\* \(2016\)](#) and [Stenström \*et al.\* \(2014\)](#) reported an increase of N<sub>2</sub>O emissions with lower DO concentrations in a PN-anammox system and a sidestream nitrification-denitrification SBR, respectively. [Vasilaki \*et al.\* \(2020\)](#) observed increased dissolved N<sub>2</sub>O concentration peaks at DO levels lower than 1 mg/L in a SCENA SBR system. The authors reported a Spearman's correlation coefficient between dissolved N<sub>2</sub>O concentration and DO equal to -0.7. On the other hand, [Kampschreur \*et al.\* \(2009a\)](#) could not identify a relationship between the N<sub>2</sub>O increase and the higher aeration flowrate during a prolonged aeration experiment in a single-stage nitrification-anammox reactor. As a general remark, it is suggested to have DO concentrations higher than 1 mg/L.

In one-stage PN-anammox reactors, elevated N<sub>2</sub>O emissions have been reported during shifts from low to high aeration and linked with high NH<sub>4</sub><sup>+</sup> concentrations and high AOR. Additionally, in nitrification-denitrification SBRs the aerobic dissolved N<sub>2</sub>O concentration has been correlated with the decrease of the average aerobic conductivity rate (Spearman's correlation coefficient equal to 0.7) and the changes of conductivity between sequential cycles. Higher emissions have been also linked with high ammonia removal efficiencies ([Vasilaki \*et al.\*, 2020](#)). This means that elevated emissions are due to AOR or higher than average NO<sub>2</sub><sup>-</sup> accumulation. N<sub>2</sub>O emissions have also increased due to the stripping of the accumulated N<sub>2</sub>O in the previous anoxic cycle (accumulated due to incomplete denitrification). In that case, step-feeding, control of initial NH<sub>4</sub><sup>+</sup> concentrations and aeration duration can mitigate the N<sub>2</sub>O peaks.

In anammox reactors, a non-negligible generation of N<sub>2</sub>O emissions has been reported. [Kampschreur \*et al.\* \(2008\)](#) observed an EF equal to 0.6% of the influent N-load for the anammox compartment of a full-scale two-stage PN-anammox system treating anaerobic supernatant. Given that N<sub>2</sub> is recognized as the end-product of the anammox process ([Jetten \*et al.\*, 2005](#)), the authors assumed that AOB from the nitrification compartment infiltrated the anammox reactor. [Yan \*et al.\* \(2019\)](#) observed, via laboratory experiments, that the increase of the COD/N ratio from 0 to 1 can decrease the N<sub>2</sub>O generation by 16.7% in a CANON process coupled with denitrification. Therefore, low carbon dosage can be a mitigation strategy for the CANON process or anammox reactors infiltrated with AOB from the nitrification compartment in two-stage PN-anammox processes.

It must be noted, though, that N<sub>2</sub>O generation depends not only on a single operational variable but also on the combined effect of several variables (temperature, NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, DO, aeration rate). This is supported by [Wan \*et al.\* \(2019\)](#) who found that higher temperatures resulted in increased N<sub>2</sub>O

emissions in the presence of COD and in decreased N<sub>2</sub>O emissions in the absence of COD in a one-stage PN-anammox reactor. The latter was attributed to increased anammox activity and reduction of NO<sub>2</sub><sup>-</sup> accumulation at higher temperature.

N<sub>2</sub>O emissions elevated during shifts from low to high aeration. Under these operational conditions the NH<sub>2</sub>OH pathway has been reported as a main generation mechanism (Castro-Barros *et al.*, 2015). At elevated NH<sub>4</sub><sup>+</sup> or DO in the reactor, N<sub>2</sub>O production by nitrifier denitrification is enhanced, while NH<sub>2</sub>OH oxidation is relatively unimportant (Harris *et al.*, 2015). Both NH<sub>2</sub>OH oxidation and nitrifier denitrification can be the main contributors to N<sub>2</sub>O accumulation across a range of conditions with varying concentrations of NH<sub>4</sub><sup>+</sup>, O<sub>2</sub>, and NO<sub>2</sub><sup>-</sup>. Harris *et al.* (2015) concluded that when N<sub>2</sub>O emissions are relatively low under optimal reactor operation the current understanding of N<sub>2</sub>O production and isotopic fractionation is incomplete and needs further investigation.

### 6.3 CH<sub>4</sub> EMISSIONS FROM FULL-SCALE WWTPS

Compared with N<sub>2</sub>O, CH<sub>4</sub> emissions from full-scale WWTPs is less investigated, while it contributes significantly to the overall plant carbon footprint. The results of full-scale CH<sub>4</sub> quantification studies are summarized in Table 6.2. Emissions of CH<sub>4</sub> in WWTPs mainly originate from the influent, anaerobic wastewater treatment and anaerobic sludge handling processes. CH<sub>4</sub> emissions thus vary greatly with different WWTP configurations. For WWTPs without anaerobic sludge handling processes, the majority of the CH<sub>4</sub> may be traced back to the dissolved CH<sub>4</sub> in the influent, which was likely formed in sewer networks. For WWTPs with anaerobic sludge handling processes, anaerobic sludge treatment and handling facilities may contribute the most to CH<sub>4</sub> emissions in plants. When anaerobic treatment is applied in WWTPs for wastewater COD removal, its CH<sub>4</sub> emissions might substantially increase the overall plant carbon footprint.

#### 6.3.1 WWTPs without anaerobic sludge handling

In WWTPs without anaerobic sludge treatment, the largest CH<sub>4</sub> emission source is often the aerobic tank and headworks (especially aerated grit chamber) via the stripping of CH<sub>4</sub> dissolved in the influent. The biological generation of CH<sub>4</sub> requires strict anaerobic conditions. Due to the short residence time, and periodical exposure to oxygen and nitrate or nitrite, it is often not believed that CH<sub>4</sub> can be produced from the headworks or from the aerobic/anoxic wastewater treatment processes (Ribera-Guardia *et al.*, 2019). Instead, it is more likely to be generated in pressurized sewer mains (see next section). By measuring liquid and gas CH<sub>4</sub> concentration, mass balance analyses have been performed in some studies (Daelman *et al.*, 2013; Noyola *et al.*, 2018; Yan *et al.*, 2014), suggesting dissolved CH<sub>4</sub> in the influent could be the main source of CH<sub>4</sub> emissions in WWTPs without anaerobic sludge treatments. In two studied WWTPs in China without sludge stabilization processes, Yan *et al.* (2014) observed 80–98% of total CH<sub>4</sub> was emitted from the wastewater treatment line, and the remaining from headworks. With mass balance analysis, it was concluded that the majority of the CH<sub>4</sub> emissions originated from the CH<sub>4</sub> dissolved in the influent. Similar observations were reported by Daelman *et al.* (2013). In two Dutch WWTPs without anaerobic sludge digestion, 86% and 77% of the total methane emissions stemmed from the influent. Nevertheless, in some cases, CH<sub>4</sub> may be generated during the wastewater treatment processes. A WWTP in Japan without anaerobic sludge digestion saw its CH<sub>4</sub> mainly (86.4%) emitted from the aerobic tank. Considering the relatively small amount of CH<sub>4</sub> in the influent, the CH<sub>4</sub> emitted is likely formed during the wastewater treatment processes under anaerobic conditions (Masuda *et al.*, 2015). Wang *et al.* (2011) also reported CH<sub>4</sub> formation during the wastewater treatment processes, emitting a significant amount of CH<sub>4</sub>.

#### 6.3.2 WWTPs with anaerobic sludge handling

Anaerobic sludge digestion is a commonly practised technology for sludge stabilization. During anaerobic sludge digestion, biodegradable organic matters are degraded in the absence of oxygen, to

Table 6.2 A summary of CH<sub>4</sub> quantification results in full-scale WWTPs.

WWTP	Wastewater treatment process	Influent organic strength (mgO <sub>2</sub> /L)	Sludge treatment process	Contribution from Headworks (%)	Contribution from secondary treatment (%)	Contribution from sludge management (%)	Emission factor (kg CH <sub>4</sub> /kg COD <sub>Influent</sub> )	Contribution of total CH <sub>4</sub> emissions to the overall carbon footprint	Source
Durham WWTP, USA	Aerobic treatment	BOD: 250	Aerobic sludge digestion	36	51	13	0.16% (BOD)	Not measured	Czepiel <i>et al.</i> (1993)
Jinan WWTP, China	Anaerobic/anoxic (AAO) process	COD: 200	Sludge dewatering, drying	9.3	80% (40% from anaerobic tank and 40% from aerobic tank)	9.5	0.08%	Not measured	Wang <i>et al.</i> (2011)
Papendrecht WWTP, Netherlands	Anaerobic tank followed by anoxic/oxic carousel reactors	Not available	Sludge dewatering	50	47	3	0.87%	17%	Daelman <i>et al.</i> (2013); Daelman <i>et al.</i> (2012)
Kortenoord WWTP, Netherland	Anaerobic tank followed by anoxic/oxic carousel reactors	Not available	Sludge dewatering	45	45	10	0.53%	13%	Daelman <i>et al.</i> (2013); Daelman <i>et al.</i> (2012)
Kralingseveer WWTP, Netherlands	Anoxic/aerobic plug flow followed by carousel reactors with surface aerators	COD:539	Anaerobic sludge digester; digestate stored for up to 5 days	<31	Not provided	72 (50% from dewatered sludge storage and buffer tank)	0.8–1.2% (seasonal variation)	5–36%	Daelman <i>et al.</i> (2013); Daelman <i>et al.</i> (2012)
Granollers WWTP, Spain	Anoxic/oxic plug-flow reactors	COD:750	Anaerobic sludge digestion	Not provided	Not provided	Not provided	0.016%	Not measured	Rodriguez-Caballero <i>et al.</i> (2014)

(Continued)

Table 6.2 A summary of CH<sub>4</sub> quantification results in full-scale WWTPs. (Continued).

WWTP	Wastewater treatment process	Influent organic strength (mgO <sub>2</sub> /L)	Sludge treatment process	Contribution from Headworks (%)	Contribution from secondary treatment (%)	Contribution from sludge management (%)	Emission factor (kg CH <sub>4</sub> /kg COD <sub>Influent</sub> )	Contribution of total CH <sub>4</sub> emissions to the overall carbon footprint	Source
Beijing WWTP1, China	Oxidation ditch process	COD: 306–689	Sludge thickening, drying and storage	1%	98% (mainly from influent stripping)	Likely negligible	0.17–0.39%	19%	Yan <i>et al.</i> (2014)
Beijing WWTP2, China	Reversed AAO process	COD: 353–687	Sludge thickening, drying	11%	89% (mainly from influent stripping)	Likely negligible	0.10–0.19%	15.8%	Yan <i>et al.</i> (2014)
Beijing WWTP2, China	AAO process	COD: 353–687	Sludge thickening, drying	19.8%	80% (mainly from influent stripping)	Likely negligible	0.06–0.11%	6.1%	Yan <i>et al.</i> (2014)
Sendai WWTP, Japan	Pseudo Anoxic-oxic process	COD: 110	Sludge dewatering, storage	8.2	86.4	<5.4	1.0%	8.3%	Masuda <i>et al.</i> (2015)
La Roca del Vallès WWTP, Spain	SBR for COD and N removal	COD: 600	No sludge stabilization	N/A	N/A	N/A	0.02%	Not measured	Rodriguez-Caballero <i>et al.</i> (2015)
Akiu WWTP, Japan	Oxidation ditch	BOD: 130	Sludge thickening, storage		97.7 (Grit chamber+OD)	<1	1.3% (BOD)	<4%	Masuda <i>et al.</i> (2018)
Hirose-gawa WWTP, Japan	Anoxic-oxic process	BOD: 210	Sludge thickening, storage	<75	23.5	Not clear	0.98% (BOD)	<5%	Masuda <i>et al.</i> (2018)
Kamiyagari WWTP, Japan	Pseudo anoxic-oxic process	BOD: 150	Sludge thickening, storage	68.1	22.6	<5.6	0.3% (BOD)	<5%	Masuda <i>et al.</i> (2018)
Girona WWTP, Spain	Modified Ludzack-Eittinger (MLE) configuration	COD: 410	Anaerobic digestion	N/A	N/A	N/A	0.28–0.49%	45–57%	Ribera-Guardia <i>et al.</i> (2019)

CH<sub>4</sub>-rich biogas, which can be captured for energy recovery. Undesirable leaks of the generated CH<sub>4</sub> could contribute significantly to the plant overall carbon footprint. In WWTPs with anaerobic sludge digestion, its related CH<sub>4</sub> emissions could contribute the majority of the total CH<sub>4</sub> emissions. Daelman *et al.* (2012) found 72 ± 23% of the total CH<sub>4</sub> emissions originated from the anaerobic sludge handling facilities: the gravitational thickener for the primary sludge, the centrifuge, the buffer tank for the effluent of the digester, the storage tank that contains the dewatered sludge and methane leakage from the gas engines. Recent studies focusing on methane losses from 23 biogas plants, including those from WWTP facilities, found an average CH<sub>4</sub> emission rate of 10.4 kgCH<sub>4</sub>/h with an average loss of 4.6% of the produced CH<sub>4</sub> (Scheutz and Fredenslund, 2019; Tauber *et al.*, 2019). Importantly, Pan *et al.* (2016) identified that the anaerobic sludge drying lagoon could also produce a large amount of CH<sub>4</sub>. During a long-term sludge drying process, the degradable organics are converted to CH<sub>4</sub> under anaerobic conditions. Without capturing the produced biogas, the CH<sub>4</sub> emissions from a long-term sludge drying lagoon would represent a quarter to two-thirds of the overall GHG emissions from the investigated WWTP.

### 6.3.3 WWTPs with anaerobic wastewater treatment technologies

While most WWTPs rely on anoxic/aerobic technologies for COD removal, anaerobic technologies (e.g., upflow anaerobic sludge blanket reactor and anaerobic lagoon) are also applied in WWTPs for COD removal. The anaerobic COD removal wastewater treatment processes often lead to substantial CH<sub>4</sub> emissions. During anaerobic wastewater treatment, biodegradable organics are converted to CH<sub>4</sub>. Methane is regarded poorly soluble in water with a relatively high Henry's Law constant. It was previously believed that dissolved methane was saturated at equilibrium with the gas phase methane concentration. However, studies have found dissolved methane is often supersaturated in bulk liquid, and can be several times higher than the predicted equilibrium concentration (Hartley and Lant, 2006). The ratio of the actual dissolved methane concentration to the calculated value from Henry's Law is used to describe the extent of methane supersaturation. For anaerobic treatment systems receiving municipal wastewater, the degree of methane supersaturation measured in many studies falls in the range of 1.34 to 6.9, with a median value of 1.64 (Crone *et al.*, 2016; Hartley and Lant, 2006). Inadequate liquid-to-gas mass transfer of methane due to the lack of mixing and low liquid velocities inherent to the reactor design, results in the observed supersaturation of methane (Crone *et al.*, 2016).

The relatively high dissolved CH<sub>4</sub> concentration in the anaerobic treatment effluent leads to substantial release of CH<sub>4</sub> in downstream processes. Existing quantification studies are mostly conducted in lab-scale and pilot-scale reactors. According to the data summarized by Crone *et al.* (2016), nearly half (49%) of the total CH<sub>4</sub> generated during the anaerobic wastewater treatment is lost in the effluent, which is subject to release in downstream processes. The aerobic activated sludge process is reportedly able to remove 80% of the dissolved CH<sub>4</sub> (Daelman *et al.*, 2012). With COD removal efficiency of anaerobic treatment technologies in the range of 55–80%, the dissolved CH<sub>4</sub> in the anaerobic treatment effluent could lead to CH<sub>4</sub> emissions of about 1.4–2% of the influent COD (kgCH<sub>4</sub>/kgCOD<sub>influent</sub>). In comparison, for WWTPs without anaerobic wastewater treatment, the total CH<sub>4</sub> emissions account for 0.02–1.2% of the influent COD (Table 6.2). The anaerobic wastewater treatment process could produce CH<sub>4</sub> emissions higher than an entire WWTP implementing anoxic/aerobic wastewater treatment processes. The CH<sub>4</sub> emissions resulting from the anaerobic wastewater treatment process is still one of the major obstacles for its wide application.

It is clear that CH<sub>4</sub> emissions represent a significant portion of the overall carbon footprint in WWTPs while rarely being the dominant one. The contribution of CH<sub>4</sub> emissions varied mostly from 4% to 19% of the overall carbon footprint (Table 6.2). In cases when N<sub>2</sub>O emissions are particularly low, the CH<sub>4</sub> emissions could be the dominant source (45–57%) of overall GHG emissions, as reported by Ribera-Guardia *et al.* (2019). Overall, CH<sub>4</sub> emissions from WWTPs should be monitored, especially in facilities where anaerobic treatment is implemented.

## 6.4 GHG EMISSIONS FROM SEWER NETWORKS

### 6.4.1 Reported CH<sub>4</sub> emissions from sewer networks

Anaerobic conditions in sewer pipes together with the high biodegradable COD concentration in the sewage favour the accumulation of methane as the end-product of the methanogenic archaea present in the sewer networks. There are not many studies focusing on the quantification of the overall CH<sub>4</sub> emissions from full-scale sewer systems, probably due to the complexity of the monitoring and the limited accessibility of some parts of the network. To date, overall methane emission data is only available for single pipe rising main and gravity sewers, calculated through the dissolved methane concentration data and following the methods explained in Chapter 4.

The overall methane emission potential of the monitored rising main sewers varies substantially, ranging from 0.04 to 0.32 kg CO<sub>2</sub>-equivalent/m<sup>3</sup> with an average value of 0.18 kg CO<sub>2</sub>-equivalent/m<sup>3</sup> of wastewater transported. Table 6.3 summarizes the studies reporting CH<sub>4</sub> emissions from sewer networks in the literature.

The majority of the methane formed in rising mains will be eventually stripped to the atmosphere via ventilation in gravity sewers or at WWTPs during the treatment of wastewater, mainly because methane oxidation in sewers is expected to be a slow process (Valentine & Reeburgh, 2000). Therefore, rising main data can be used to calculate potential overall emission rates from sewer systems.

In some other studies, the quantification of overall CH<sub>4</sub> emissions has been carried out by direct measurement of methane emission rate from a discharge manhole (Shah *et al.*, 2011). However, this methodology is expected to underestimate emissions as CH<sub>4</sub> could also be emitted at other locations in the network.

### 6.4.2 Reported N<sub>2</sub>O emissions from sewer networks

Studies providing N<sub>2</sub>O emission data from sewer networks are sparse, with very few studies published to date. In 2014, Short *et al.* reported the dissolved N<sub>2</sub>O concentrations from the inlet of three WWTPs in Australia during an 8 month monitoring campaign. They found that average levels in the raw wastewater were relatively consistent among the three WWTPs monitored at around 7–10 µg N-N<sub>2</sub>O/L. Combining these results with wastewater parameters they were able to calculate presumptive per capita N<sub>2</sub>O emission factors, resulting in 1.39–1.84 g N<sub>2</sub>O/person year and 0.009–0.02 kg N-N<sub>2</sub>O/kg TN.

Another study conducted in the sewer network of the Cincinnati municipality (Fries *et al.*, 2018) reported that its wastewater collection system was a non-point source of N<sub>2</sub>O. Based on their results, they estimated approximately an average rate of 151.2 ± 326 g N<sub>2</sub>O/d for the whole city.

As the authors from both studies mentioned, all these numbers should be taken with caution as further investigations are needed to better understand the magnitude of sewer N<sub>2</sub>O emissions.

## 6.5 MITIGATION STRATEGIES APPLIED IN FULL-SCALE SYSTEMS

### 6.5.1 GHG mitigation in WWTPs

There is no standardized methodology for the establishment of N<sub>2</sub>O mitigation strategies in full-scale systems. Table 6.1 summarizes the main mitigation strategies that have been proposed or tested in full-scale wastewater treatment processes.

Testing different operational modes is regarded as one of the most effective ways to identify measures for emission mitigation. Several studies have modified the aeration intensity and/or strategy, and optimized the DO set-point and cycle duration to investigate the effect on N<sub>2</sub>O emissions in full-scale BNR processes (Castro-Barros *et al.*, 2015; Duan *et al.*, 2020; Kampschreur *et al.*, 2009a, 2009b; Mampaey *et al.*, 2016; Rodriguez-Caballero *et al.*, 2015). For instance, Mampaey *et al.* (2016) achieved a reduction in the N<sub>2</sub>O emissions of 56% when the cycles in a one-stage granular SHARON reactor were shortened by 1 h. Rodriguez-Caballero *et al.* (2015) tested different operational conditions in a full-scale SBR. They suggested an optimum control strategy for the minimization of N<sub>2</sub>O

Table 6.3 Dissolved methane concentrations and methane emission in rising mains (adapted from Liu et al. 2015b).

Country	Name	Type	Length (m)	Diameter (mm)	A/V (m <sup>-1</sup> )	HRT (h) average (min-max)	WW T (°C)	Dissolved CH <sub>4</sub> average (min-max) (mg/L)	Daily flow (m <sup>3</sup> /day)	Production (kgCH <sub>4</sub> /day)	Overall emissions (kgCO <sub>2</sub> eq/m <sup>3</sup> )	References
Australia	UC09	Rising main	828	150	26.7	2.5 (3.1-4.6)	27.7	5.3 (4.4-6.1)	200	1.1	0.11	Foley et al. (2011)
Australia	C16	Rising main	1100	300	13.3	2.6 (3.9-11.0)	22.5	15.3 (11.3-3.0)	707	9.8	0.32	Foley et al. (2011)
Australia	C16	Rising main	1100	300	13.3	2.6 (3.9-11.0)	23.5	5.2 (3.4-6.6)	707	2.6	0.11	Foley et al. (2009)
Australia	C27	Rising main	4400	525	7.6	9.1	24.6	9.1 (5.0-15.0)	2840	24.6	0.19	Liu et al. (2015b)
Australia	C27	Rising main	4400	525	7.6	9.1	20.3	7.1 (3.5-12.0)	2840	19.0	0.15	Liu et al. (2015b)
Australia	PerthB	Rising main	15 000	900	4.4	-	-	4.8	11 000	52.8	0.10	Liu et al. (2015b)
Thailand	RV	Gravity	1000	1000	-	27.9 (22-31.4)	33.3	10.1 (8.0-13.7)	-	-	-	Chaosakul et al. (2014)
Thailand	RV	Gravity	1000	1000	-	7.8 (0-12)	30.2	4.6 (0.1-11.4)	-	-	-	Chaosakul et al. (2014)
Australia	CO16 PS	Pumping station	-	-	-	-	23.5	1.5 (1.0-1.92)	707	-	-	Foley et al. (2009)
Australia	OR3 PS	Pumping station	-	-	-	-	-	0.51	2000	-	-	Liu et al. (2015b)
Spain	Radin	Rising main	2930	500	8.0	4.7 (2.9-6.8)	16.0	1.95 (1.3-2.7)	4210	8.2	0.04	Personal communication
Spain	Collet	Rising main	4800	556	3.6	12.4 (7.3-15.5)	25.4	3.0 (1.9-4.01)	2700	8.1	0.06	Personal communication



emissions based on the application of short aerobic-anoxic cycles (20-min aerobic phase and short duration of anoxic stage).

Activated sludge models have been also applied to identify potential N<sub>2</sub>O mitigation strategies in BNR systems. Ni *et al.* (2015) developed a mechanistic model utilizing the data from a two-step plug-flow reactor (Pan *et al.*, 2016) showing that the biomass specific N-loading rate is linked with the elevated N<sub>2</sub>O emissions observed in the second step of the process. Different operational conditions were tested with the model demonstrating that lower N<sub>2</sub>O emissions (<1% of the N-load) can be achieved if 30% of the total return activated sludge (RAS) stream is recirculated to the second step of the plug-flow reactor (Table 6.1). However, it is unknown whether the suggested mitigation strategy was demonstrated in the system. Similarly, Zaborowska *et al.* (2019) used multiple-pathway activated sludge modelling to investigate N<sub>2</sub>O mitigation strategies in an A<sup>2</sup>/O reactor. They showed that DO concentrations between 1 and 2 mg/L and mixed liquor recirculation rates above 500% could minimize N<sub>2</sub>O emissions and energy consumption during aeration without compromising TN removal in the studied A<sup>2</sup>/O reactor. Duan *et al.* (2020) used a multiple-pathway model to test different N<sub>2</sub>O mitigation strategies in an SBR reactor. Based on the simulation results, they modified the aeration control of the system. They showed that SND operation mode can result in 35% reduction of the N<sub>2</sub>O emissions compared to intermittent over-aeration.

Overall, the main techniques for mitigating the N<sub>2</sub>O emissions in wastewater treatment processes target (i) the reduction of the diurnal variation of NH<sub>4</sub><sup>+</sup> loads and avoidance of NH<sub>4</sub><sup>+</sup> peaks and NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub><sup>-</sup> build-up (i.e. integration of equalization tanks, recycling steps, optimization of anaerobic supernatant feeding), (ii) the increase of the MLVSS concentration to lower the specific N-loading (i.e. optimization of the RAS or SRT increase), (iii) the facilitation of complete reactions by providing sufficient electron donors (COD) during denitrification (i.e. supply of additional carbon source to ensure complete denitrification) and electron acceptors (O<sub>2</sub>) during nitrification, and (iv) the facilitation of N<sub>2</sub>O consumption during denitrification (i.e. increasing anoxic duration, lowering DO to enhance SND).

Reports on mitigation of methane from WWTPs are very scarce. Some technologies have been proposed for the removal of dissolved methane from anaerobic effluents, one for the most effective being the application of a degassing membrane (Bandara *et al.*, 2011). However, their application is very limited and no studies for their application in full-scale WWTPs have been found.

Sludge storage also contributes significantly to the fugitive methane emissions from WWTPs as digested sludge has a significant residual methane potential (Daelman *et al.*, 2012). The authors proposed the use of the ventilation air from the buffer tank as combustion air in the gas engines of the cogeneration plant, receiving the biogas produced in the digesters. This would result in less diluted methane streams going to the cogeneration plant, but this should be adapted to handle methane concentrations that exceed the lower explosive limit of methane in air.

Finally, it is important to highlight the need for good housekeeping and regular maintenance of the anaerobic digestion facilities present in WWTPs for sludge digestion, to avoid fugitive CH<sub>4</sub> emissions from these reactors.

### 6.5.2 GHG mitigation from sewers

CH<sub>4</sub> is the main GHG emitted from sewers and it is usually biogenically formed together with hydrogen sulfide under anaerobic conditions (Chapter 5). The wastewater industry uses several chemical-dosing approaches to mitigate sulfide emissions including the addition of nitrate, oxygen, ferric salts, hydroxide (pH elevation) and free nitrous acid (FNA) (Zhang *et al.*, 2008). But those can also suppress CH<sub>4</sub> formation from sewers because the methanogens are slow growers and are very sensitive to environmental conditions as compared with sulfate reducing bacteria (SRB) (Guisasola *et al.*, 2008). Also, in contrast to SRB, methanogens usually inhabit the deeper zone of sewer biofilms or sediments and are usually protected due to limited penetration of the dosed chemical. Thus, for effective control of methanogens, a higher dosage of chemicals may be needed to achieve full penetration during the

**Table 6.4** Summary of the CH<sub>4</sub> mitigation studies conducted in full-scale sewer networks.

Chemical	Dosing levels	Dosing plan	CH <sub>4</sub> reduction (%)	CH <sub>4</sub> production recovery	Reference
Nitrate	17 kg N-NO <sub>3</sub> <sup>-</sup> /ML	One shock	13	100% in 2 days	Shah <i>et al.</i> (2011)
Nitrate	50 kg N-NO <sub>3</sub> <sup>-</sup> /ML	One shock	27	100% in 2 days	Shah <i>et al.</i> (2011)
Hydroxide	pH 11.5	Shock for 6 h	97	3% in 15 days	Gutierrez <i>et al.</i> (2014)

initial dosing period, when overall bacterial activity is high. However, continuous dosing, as required for sulfide control with most chemicals, may not be necessary. Table 6.4 summarizes the mitigation studies conducted in full-scale sewer networks.

Today, the current practice of selecting chemicals and design of dosing locations/rates is still mainly based on an individual's experience (Ganigue *et al.*, 2011; Liu *et al.*, 2015a). Constant, flow-paced and profiled dosing rates are currently applied during chemical dosing, again based on experience. Instead, the approach should be based on specific features of the sewer in question. In this respect, the SeweX model (Sharma *et al.*, 2008) consists of an empowering tool in supporting decision-making. Concentrations of methane, sulfide and flows show significant temporal and spatial dynamics in sewers. The rudimentary current methods could be ineffective in methane control, resulting in over-dosing of chemicals during periods with low methane and sulfide production, and conversely underdosing during other periods.

## 6.6 CONCLUDING REMARKS

Currently, operational strategies at WWTPs do not consider the mitigation of GHG emissions. New objectives, including environmental and carbon neutrality targets, in the water industry require approaches to dynamically integrate new parameters (i.e. GHG emissions sensors, energy meters) into the process monitoring, control and decision-making.

Process-based N<sub>2</sub>O EF benchmarking is challenging due to (i) differences in the N<sub>2</sub>O generation triggered by the site-specific operational characteristics, environmental conditions and control parameters, and (ii) the sensitivity of the quantified EF to differences in monitoring strategies and duration of monitoring campaigns. The quantification of reliable annual EFs requires sampling campaigns lasting at least 1 year. Additional campaigns are required for specific groups of processes (i.e., processes treating high strength streams, biofilm technologies) that have received less attention until now.

Guidelines for N<sub>2</sub>O mitigation measures for different process groups have been developed. Further research is required to develop practical approaches to help utilities to quantify, understand and report the N<sub>2</sub>O EF and develop dynamically evolving mitigation measures based on the operational conditions. Future research can explore the possibility of coupling artificial intelligence (AI) techniques with multiple-pathway process models for full-scale applications, to facilitate the fast and adaptable online implementation of model predictive control and forecasting decision support tools.

GHG monitoring campaigns carried out in WWTPs should include the monitoring of fugitive CH<sub>4</sub> emissions, which contribute significantly to the overall plant carbon footprint. CH<sub>4</sub> emissions mainly originate from the influent, anaerobic sludge handling processes and anaerobic wastewater treatment in WWTPs. For WWTPs without anaerobic sludge handling processes, the CH<sub>4</sub> emissions can mainly be traced back to the CH<sub>4</sub> dissolved in the influent. The implementation of anaerobic sludge handling processes may contribute the most to CH<sub>4</sub> emissions in WWTPs. When anaerobic treatment is applied in WWTPs for wastewater COD removal, its CH<sub>4</sub> emissions might substantially increase the overall plant carbon footprint.

Finally, more attention should be paid to fugitive GHG emissions from sewer networks. Several studies suggest CH<sub>4</sub> emissions could be important in some parts of the sewer networks, with most of the

monitoring campaigns being conducted in pressurized sewer mains. However, very little information is reported for full-scale gravity sewers and very scarce data is available for N<sub>2</sub>O emissions from sewer networks.

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## NOMENCLATURE

A/O	Anoxic/aerobic
A <sup>2</sup> /O	Anaerobic/anoxic/aerobic
AMO	Ammonia monooxygenase
Anammox	Anaerobic ammonium oxidation
AOA	Ammonia oxidizing archaea
AOB	Ammonia oxidizing bacteria
AOR	Ammonia oxidation rate
BNR	Biological nutrient removal
CANON	Completely autotrophic nitrogen removal over nitrite
CAS	Conventional activated sludge
CO <sub>2</sub>	Carbon dioxide
COD	Chemical oxygen demand
Comammox	Complete ammonium oxidizer
CuO	Copper oxide
dGAO	Denitrifying glycogen accumulating organisms
DO	Dissolved oxygen
dPAO	Denitrifying polyphosphate accumulating organism
EF	Emission factor
FA	Free ammonia
FNA (HNO <sub>2</sub> )	Free nitrous acid
GHG	Greenhouse gas
H <sub>2</sub> S	Hydrogen sulphide
HRT	Hydraulic retention time
MLE	Modified Ludzack-Ettinger
MLVSS	Mixed liquor volatile suspended solids
N <sub>2</sub>	Nitrogen gas
N <sub>2</sub> O	Nitrous oxide

$N_2O_4$	Nitrogen tetroxide
NaR	Nitrate reductase
$NH_2OH$	Hydroxylamine
$NH_3$	Ammonia
$NH_4^+$	Ammonium
NiR	Nitrite reductase
NO	Nitric oxide
$NO_2^-$	Nitrite
$NO_3^-$	Nitrate
NOB	Nitrite oxidizing bacteria
NOH	Nitrosyl radical
NoR	Nitric oxide reductase
NoS	Nitrous oxide reductase
OD	Oxidation ditch
PN	Partial nitrification
RT-qPCR	Real time quantitative polymerase chain reaction
SBR	Sequencing batch reactor
SCENA	Short cut enhanced nutrient abatement
SP	Site-preference
WWTPs	Wastewater treatment plants