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Review

A decade of nitrous oxide (N_2O) monitoring in full-scale wastewater treatment processes: A critical review

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ABSTRACT

Direct nitrous oxide (N_2O) emissions during the biological nitrogen removal (BNR) processes can significantly increase the carbon footprint of wastewater treatment plant (WWTP) operations. Recent onsite measurement of N_2O emissions at WWTPs have been used as an alternative to the controversial theoretical methods for the N_2O calculation. The full-scale N_2O monitoring campaigns help to expand our knowledge on the N_2O production pathways and the triggering operational conditions of processes. The accurate N_2O monitoring could help to find better process control solutions to mitigate N_2O emissions of wastewater treatment systems. However, quantifying the emissions and understanding the long-term behaviour of N_2O fluxes in WWTPs remains challenging and costly.

A review of the recent full-scale N_2O monitoring campaigns is conducted. The analysis covers the quantification and mitigation of emissions for different process groups, focusing on techniques that have been applied for the identification of dominant N_2O pathways and triggering operational conditions, techniques using operational data and N_2O data to identify mitigation measures and mechanistic modelling. The analysis of various studies showed that there are still difficulties in the comparison of N_2O emissions and the development of emission factor (EF) databases; the N_2O fluxes reported in literature vary significantly even among groups of similar processes. The results indicated that the duration of the monitoring campaigns can impact the EF range. Most N_2O monitoring campaigns lasting less than one month, have reported N_2O EFs less than 0.3% of the N-load, whereas studies lasting over a year have a median EF equal to 1.7% of the N-load. The findings of the current study indicate that complex feature extraction and multivariate data mining methods can efficiently convert wastewater operational and N_2O data into information, determine complex relationships within the available datasets and boost the long-term understanding of the N_2O fluxes behaviour. The acquisition of reliable full-scale N_2O monitoring data is significant for the calibration and validation of the mechanistic models -describing the N_2O emission generation in WWTPs. They can be combined with the multivariate tools to further enhance the interpretation of the complicated full-scale N_2O emission patterns. Finally, a gap between the identification of effective N_2O mitigation strategies and their actual implementation within the monitoring and control of WWTPs has been identified. This study concludes that there is a further need for i) long-term N_2O monitoring studies, ii) development of data-driven methodological approaches for the analysis of WWTP operational and N_2O data, and iii) better understanding of the trade-offs among N_2O emissions, energy consumption and system performance to support the optimization of the WWTPs operation.

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Abbreviations

amoA	Ammonia monooxygenase
Anammox	Anoxic Ammonia Oxidation
ANN	Artificial Neural Networks
A/O	Anaerobic-Oxic
A^2/O	Anaerobic-Anoxic-Oxic
AOA	Ammonia Oxidizing Achaea
AOB	Ammonia Oxidizing Bacteria
AOR	Ammonia Oxidation Rate
AS	Activated Sludge
ASM	Activated Sludge Models
BNR	Biological Nitrogen Removal
BSM2	Benchmark Simulation Model 2
CAS	Conventional Activated Sludge
CO_2	Carbon Dioxide
COD	Chemical Oxygen Demand
DO	Dissolved Oxygen
EF	Emission Factor
GC	Gas Chromatography
GHG	Greenhouse Gas
HAO	Hydroxylamine Oxidoreductase
HRT	Hydraulic Retention Time
ICA	Independent Component Analysis
LCA	Life cycle assessment
MDS	Multidimensional Scaling
MLE	Modified Ludzack-Ettinger
MLSS	Mixed Liquor Suspended Solids
N	Nitrogen
N_2O	Nitrous Oxide
N_2OR	Nitrous Oxide Reductase
NH_2OH	Hydroxylamine
NH_3	Ammonia
NH_4^+-N	Ammonium Nitrogen
NO	Nitric Oxide
NO_2^--N	Nitrite Nitrogen
NO_3^--N	Nitrate Nitrogen
NOB	Nitrite Oxidizing Bacteria
NOR	Nitric Oxide Reductase
nosZ	Nitrous Oxide Reductase
OD	Oxidation Ditch
PCA	Principal Component Analysis
PE	Population Equivalent
PF	Plug Flow
PLS	Partial Least Squares
r-A^2/O	Reversed A^2/O
qPCR	Real-time Polymerase Chain Reaction
RAS	Return Activated Sludge
Redox	Reduction-Oxidation
RT-qPCR	Reverse Transcription Polymerase Chain Reaction
SBR	Sequencing Batch Reactor
SP	Site Preference
SRT	Sludge Retention Time
SVM	Support Vector Machine
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
WWTP	Wastewater Treatment Plant

1. Introduction

Wastewater treatment plants (WWTPs) produce substantial anthropogenic nitrous oxide (N_2O) amounts (Law et al., 2012). N_2O emissions from wastewater treatment processes have increased by 44% from 1990 to 2014 (US EPA, 2016). Moreover, they are responsible for up to 26% of the greenhouse gas (GHG) emissions of the whole water supply chain (including drinking water supply,

wastewater collection and treatment, effluent discharge, sludge processing and disposal) (Lane et al., 2015). Therefore, the evaluation of the environmental impact of WWTPs is gaining increasing attention worldwide (Guo et al., 2017; Harris et al., 2015; Law et al., 2012; Mannina et al., 2016; Massara et al., 2017). N_2O has an approximately 300 times higher GHG effect than CO_2 (IPCC, 2013). Compared to other unregulated combined halocarbon emissions, reducing the anthropogenic N_2O emissions in the atmosphere plays the most significant role in preventing the ozone layer depletion (Daniel et al., 2010; Portmann et al., 2012).

Stricter environmental regulations are being imposed on WWTPs, while the need to control the energy consumption costs, pushes the plant managers towards the deployment of more energy and cost efficient operational strategies (Batstone et al., 2015; Ghoneim et al., 2016). The integration of additional sustainability metrics (e.g. GHG emissions, resource recovery) for the evaluation of the WWTP performance is also gaining attention (Flores-Alsina et al., 2014; Cornejo et al., 2016). Recent studies have revealed that the direct N_2O emissions of biological processes in WWTPs can increase the operational carbon footprint by ~78% (Daelman et al., 2013a) or even ~83% (Desloover et al., 2011). Moreover, the new technology adoption in WWTPs requires the consideration of trade-offs between direct and indirect GHG emissions to ensure that it will not result in increase of the overall carbon footprint. Since the GHG emissions have a major contribution to the total environmental impact of WWTPs, they should be considered as part of the decision-making process for the improvement of the technological and operational plant performance (Sun et al., 2017b; Frutos et al., 2018; Conthe et al., 2018). Therefore, research has been conducted in the past years to identify and suggest strategies leading to N_2O mitigation during the biological nutrient removal (BNR) processes (Desloover et al., 2012; Duan et al., 2017; Law et al., 2012). Different operating parameters, such as dissolved oxygen (DO), pH, ammonium (NH_4^+) and nitrite (NO_2^-) concentration, configuration types and environmental conditions (e.g. temperature), can affect the N_2O production in WWTPs (Desloover et al., 2012; Kampschreur et al., 2009b; Massara et al., 2017). The exact triggering operational and environmental conditions that govern the N_2O generation are still under investigation by researchers and operators (Wan et al., 2019). Additionally, the exact mechanisms that determine the N_2O generation are not fully understood, thus hindering the establishment of mitigation measures (Duan et al., 2017). Consequently, the long-term dynamics of N_2O emissions in full-scale WWTPs cannot be fully explained, even for conventional processes (e.g. plug-flow (PF) reactors) (Daelman et al., 2015).

Different data sampling techniques and analytical tools have been employed in existing full-scale N_2O studies. The objectives of the current review paper are to: i) evaluate recent findings from N_2O monitoring campaigns in terms of N_2O EFs, dominant pathways and mitigation measures for different groups of full-scale configurations and reactor types, ii) examine the discrepancies among the methods applied in different monitoring campaigns, and iii) evaluate the data provided by studies investigating N_2O emissions in WWTPs. The current study critically assesses how the findings of the research on N_2O emissions have been extrapolated in different full-scale process groups. Moreover, this work evaluates the methods that have been applied at full-scale systems for the N_2O quantification, control and mitigation. Finally, the research gaps that need to be addressed in future studies are highlighted.

2. The impetus for quantifying the N_2O emissions in WWTPs

2.1. EF benchmarks

EF quantification in conventional and advanced wastewater treatment processes is essential to assess and reduce the resulting environmental impact (Foley et al., 2015). Current methods for the theoretical calculation of N_2O EFs at WWTPs rely on fixed (Palut and Canziani, 2007) or country-specific EFs and similar oversimplified approaches (Singh and Maurya, 2016). These methods underestimate the actual emissions and are considered unreliable (Cadwallader and VanBriesen, 2017), since they are not representative for different process configurations, operational and environmental conditions. Therefore, a main target of the studies analysed in this paper was to: i) develop a N_2O emission database from different mainstream (Ahn et al., 2010b, 2010a)/sidestream (Kampschreur et al., 2008; Weissenbacher et al., 2010) processes and innovative configurations (Desloover et al., 2011), and ii) identify N_2O EFs from processes located in previously unreported regions with different environmental conditions (Wang et al., 2011).

2.2. N_2O pathways and triggering operational conditions

The majority of the full-scale N_2O monitoring campaigns were driven by the need to expand the knowledge on the N_2O generation in wastewater treatment systems. Several studies have highlighted that it is important to understand the dominant N_2O generation pathways in full-scale processes and investigate the effect of specific operational conditions triggering specific enzymatic reactions linked with elevated EFs in full-scale biological systems (Bollon et al., 2016a; Pan et al., 2016; Wang et al., 2016b).

The N_2O production during wastewater treatment involves several microbiological reactions during both autotrophic and heterotrophic processes that require either aerobic or anoxic conditions. Three main biological pathways have been identified in BNR systems; hydroxylamine (NH_2OH) oxidation, nitrifier denitrification and heterotrophic denitrification (Kampschreur et al., 2009b). The NH_2OH oxidation pathway is mainly catalysed by the autotrophic ammonia oxidizing bacteria (AOB) and ammonia-oxidizing archaea (AOA). However, several studies have shown that the contribution of AOA to N_2O emissions in wastewater is expected to be low (Hooper, 1968; Ritchie and Nicholas, 1972; Law et al., 2012). Caranto et al. (2016) have recently showed that N_2O can also be the main product of anaerobic NH_2OH oxidation catalysed by the cytochrome P460 in *N. europaea*. The latter can be considered as evidence of biological N_2O generation under limited DO and high NH_3 concentrations (i.e. Law et al., 2012), or as potential explanation for the high N_2O emissions observed during the transition from aerobic to anoxic conditions. White and Lehnert (2016) have also suggested that N_2O can be directly produced during the NH_2OH oxidation (mediated by the NH_2OH oxidoreductase (HAO) enzyme) under aerobic conditions, whereas the NO_2^- detected can result as by-product of the nitric oxide (NO) oxidation. The AOB can also reduce NO_2^- to NO (by the aid of *nirk*) and, subsequently, NO to N_2O (catalysed by *norB*) mainly under oxygen-limiting conditions via the other nitrification-related pathway (nitrifier denitrification) (Poth and Focht, 1985). During denitrification, the heterotrophic denitrifiers are responsible for the reduction of NO_3^-/NO_2^- to nitrogen gas (N_2). N_2O is an intermediate of denitrification (Schulthess and Gujer, 1996). With the NO reductase (NOR) as catalyst, NO is reduced to N_2O (Hochstein and Tomlinson, 1988). NO can also result as by-product of the incomplete NH_2OH oxidation and then serve as a substrate during denitrification (Hooper and Terry, 1979). If the denitrification process continues undisturbedly, N_2O is reduced to N_2 in the final denitrification step (catalyzed by the N_2O reductase (N_2OR)). Consequently, the heterotrophic denitrification process can act either as a sink or as a source of N_2O (Robertson and Tiedje, 1987). Under elevated NH_2OH and NO_2^- concentrations, abiotic yet biologically-

driven N_2O pathways can also constitute important contributors to the N_2O emissions (Soler-Jofra et al., 2016; Terada et al., 2017; Harper et al., 2015). Inside nitritation reactors for example, the abiotic-biotic pathway of nitrosation is possible; NO_2^- can react with the biologically produced NH_2OH and form N_2O as end-product (Zhu-Barker et al., 2015).

Based on the existing knowledge on the N_2O production pathways, recent reviews on N_2O emissions from wastewater treatment processes have concluded that the key operational variables responsible for the N_2O generation include but are not limited to the following: i) low DO, NO_2^- or free nitrous acid (HNO_2) accumulation and changes in the NH_4^+ concentration in the nitrifying zones, ii) limitation of organic substrate (i.e. low chemical oxygen demand to N (COD:N) ratio) as well as NO_2^- accumulation in the denitrifying zones, iii) alternation of anoxic/aerobic conditions, and iv) abrupt changes in the processes and system shocks (Duan et al., 2017; Guo et al., 2017; Law et al., 2012; Massara et al., 2017).

Therefore, N_2O emissions can occur because of diverse contributing factors and enzymatic reactions. However, these parameters and reactions can occur simultaneously, dynamically and beyond operators' control in full-scale systems, whereas small changes (e.g. DO changes) can significantly affect the N_2O formation. Previous studies on full-scale monitoring campaigns intended to: i) identify the most important operating conditions (e.g. aeration rate, DO, NO_2^- concentration, pH, etc.) and correlate them with the N_2O generation (Brotto et al., 2015; Rodriguez-Caballero et al., 2014), ii) reveal the effects of seasonal variations on the N_2O formation (Yan et al., 2014), and iii) identify the key pathways for the N_2O production (Wang et al., 2016b).

2.3. N_2O mitigation strategies

Another key objective of the N_2O monitoring campaigns performed in the past years, particularly significant for the WWTP operators, was the development of operational strategies for the minimization of the emissions (Desloover et al., 2012). Therefore, several authors have suggested N_2O mitigating measures based on the findings of full-scale N_2O monitoring campaigns (i.e. Chen et al., 2016; Mampaey et al., 2016; Pan et al., 2016; Wang et al., 2016b). The proposed strategies to control N_2O emissions, are analysed in the following sections.

3. EF estimation using full-scale N_2O monitoring data

This section emphasizes the need to increase the comparability amongst different studies that report N_2O emissions. Moreover, it investigates potential trends in the EFs for certain groups of processes and summarises the data requirements for the EF assessment in WWTPs. In cases where the EFs were reported with respect to units other than the influent total nitrogen (TN) or the influent NH_4^+ content, appropriate conversions were made where possible (see the supplementary material A – Table A2).

Fig. 1 shows the percentage of past full-scale N_2O monitoring campaigns with reference to the treatment configurations applied each time. All processes considered in Fig. 1 are given in the supplementary material B (Tables B1 and B2). Distinct mainstream process configurations include, modified Ludzack-Ettinger (MLE) reactors, conventional activated sludge (CAS) systems (only aerobic reactors), A^2/O (anaerobic/anoxic/aerobic) processes and A/O (anoxic/aerobic) reactors. Additionally, oxidation ditch (OD) reactor types and sequence batch reactor (SBR) types have been considered as distinct process groups. Sidestream processes that include partial-nitritation reactors, 1-step and 2-step partial-nitritation-anammox configurations) are considered a distinct process group in Fig. 1. The processes that do not belong to the aforementioned

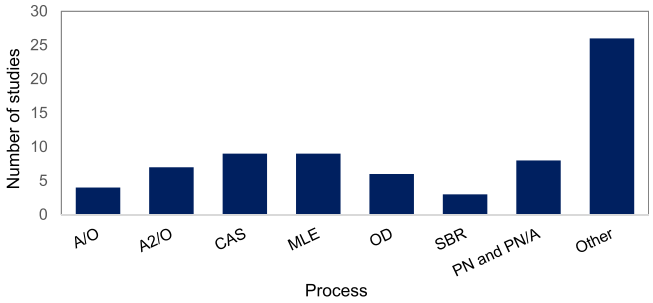

Fig. 1. The number of monitoring campaigns reported in literature per process group; A/O: Anoxic/oxic reactor, A^2/O: anaerobic-anoxic-oxic reactor, CAS: conventional activated sludge, MLE: Modified Ludzack-Ettinger reactor, OD: oxidation ditch, SBR: sequencing batch reactor, PN and PN/A: partial-nitritation and partial-nitritation-anammox (1 and 2-stage).

process groups are categorized separately (i.e. Baresel et al., 2016; Mello et al., 2013; Wang et al., 2016a). Details for all the processes are provided in the supplementary material.

Most of the process-focused monitoring campaigns include, CAS systems (~12%), (MLE) configurations (~12%), A^2/O configurations (~10%), oxidation ditches (ODs) (~8%) and sidestream partial-nitritation reactors or anammox systems (~11%) (Fig. 1). Overall, these studies refer to a wide range of configurations (i.e. Aboobakar et al., 2013; Brotto et al., 2015; Castro-Barros et al., 2015; Sun et al., 2017a) that have been monitored mainly for short periods (i.e. Ahn et al., 2010b; Bellandi et al., 2018; Foley et al., 2010; Pan et al., 2016) with varying methodology (e.g. different gaseous sampling and analytical measurement protocols) (Daelman et al., 2015; Ren et al., 2013).

EF comparability limitations and benchmarking amongst the various processes will be discussed in the following sections. However, as a general remark, the identification of potential emission patterns and the EF classification for specific groups of processes is still challenging, mainly due to differences in monitoring strategies, operational conditions and length of monitoring periods among the existing studies. Additionally, there is still little real-field data regarding N_2O emissions for several conventional and advanced biological processes (e.g. trickling filters, denitrifying packed bed reactors, biofilm or hybrid partial-nitritation anammox systems, etc.).

3.1. EF of secondary and sidestream treatment processes

The N_2O emissions of the full-scale wastewater treatment processes reported in past studies vary significantly; e.g. ranging from 0.0025% of the TN-load for a mainstream MLE reactor (Spinelli et al., 2018) to 5.6% of the TN-load for a mainstream aerobic/anoxic settling SBR reactor (Sun et al., 2013). Overall, the potential of N_2O emissions from sidestream reactors (ranging from 0.17% to 5.1% of the influent N-load – supplementary material B, Table B1) is considered higher compared to the mainstream BNR processes. The latter is mainly because the nitritation/nitrification occurring during sidestream treatment is linked with higher ammonia oxidation rate (AOR) and NO_2^- accumulation (Desloover et al., 2011; Gustavsson and la Cour Jansen, 2011; Kampschreur et al., 2008). Fig. 2 shows boxplots of the observed EFs (with respect to the influent N-load) based on the treatment step. The width of the shaded area surrounding the boxplots represents the data kernel density distribution of the EFs. Specific information for the mainstream and sidestream technologies included in Fig. 2 can be found in the supplementary material (Tables A1, B1 and B2). Average N_2O emissions for the studied mainstream processes is equal to ~0.87%

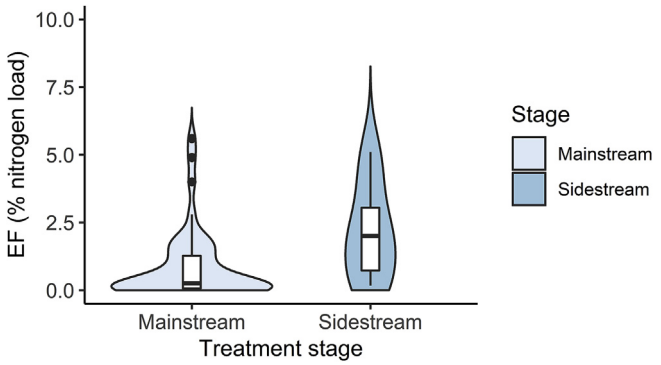

Fig. 2. Boxplots of the of the reported EFs with respect to the stage of the treatment processes (i.e. mainstream or sidestream) using violin plot outlines. The rectangles represent the interquartile range. The median is denoted by the black horizontal line dividing the box in two parts. The dots represent the values exceeding 1.5 times the interquartile range. The upper and lower whiskers stand for values higher or lower 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 shaded area represents the proportion of the data located there.

of the N-load, whereas the majority of the quantified EFs are below 0.27% of the influent N-load according to Fig. 2. On the other hand, Fig. 2 shows that N_2O EFs resulting from the treatment of the anaerobic digestion supernatant (sidestream process) are highly concentrated just below the median (2% of the N-load). On average, ~2.1% of the N-load is emitted as N_2O in sidestream processes (supplementary material B, Table B2). According to a life cycle assessment (LCA) study quantifying the direct GHG emissions for a WWTP in Austria, the sidestream DEMON process contributed by over 90% to the total direct N_2O emissions compared to the mainstream BNR (Schaubroeck et al., 2015). However, examples of full-scale sidestream Anammox processes with EFs lower than 1% exist in the literature and demonstrate that the configuration and efficient operational strategies can mitigate a significant amount of the N_2O produced (Joss et al., 2009; Weissenbacher et al., 2010).

Fig. 3 shows the EF boxplots of various processes applied in WWTPs. In total 51 systems were considered in Fig. 3 (supplementary material, Tables A1 and B2). A general remark is that the N_2O EF for the majority of the different process groups shown in Fig. 3 varies from 0.01 to 2% of the N-load. Discrepancies in the emission loads are observed in the majority of the different process groups and can be partially attributed to the different site-specific operational characteristics and control parameters. This indicates

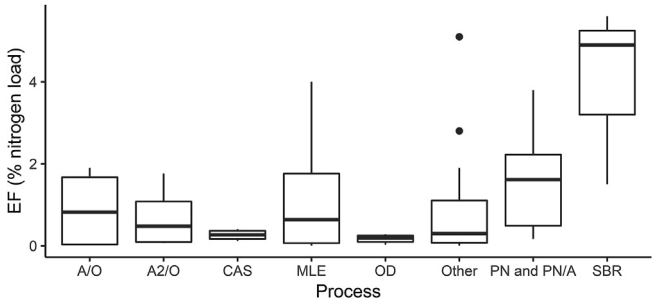

Fig. 3. Boxplots visualizing the EF range for the different groups of mainstream processes. The rectangles represent the interquartile range. The median is denoted by the black horizontal line dividing the box in two parts. The dots represent values exceeding 1.5 times the interquartile range. The upper 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).

that apart from the reactor configuration, emission fluxes depend also on the operational/environmental conditions and preferred enzymatic pathways (Wan et al., 2019).

Mainstream SBRs are generally associated with higher N_2O emissions compared to the other process groups. EFs range between 2% of the influent TKN-load for an SBR operating under aerated feeding, aerobic, settling and decanting sequences (Foley et al., 2010) 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). High N_2O fluxes in SBRs are attributed to sudden changes in the concentrations of NH_4^+ and NO_2^- within the cycle (compared to other configurations) or to accumulated dissolved N_2O during anoxic settling and decanting in the subsequent aerobic phase (Pijuan et al., 2014).

OD reactor types have been linked with relatively low N_2O emissions (average equal to 0.14% of the N-load), probably due to the strong dilution of the reactor concentrations (very high recycling rates) and less sensitivity to system shocks. One exception is the study of Daelman et al. (2015) who monitored a covered anaerobic/anoxic/oxic plug-flow reactor followed by two parallel Carrousel reactors for 1 year and found that the system had EF equal to 2.8% of the N-load. The authors argued that in the Carrousel reactor, the surface aerators led to zones with limited oxygen concentration to allow for complete nitrification (leading to NO_2^- accumulation), whereas the anoxic zones were also limited to allow for complete denitrification.

CAS systems shown in Fig. 3 consist of aerobic reactors (1-step feed or multiple step-feed) without dedicated anoxic zones for denitrification. They are characterised by average EF equal to 0.27% of the N-load, whereas the NH_4^+ removal ranges between 38% and 53%. Peak loads and recirculation of the anaerobic supernatant can be responsible for the N_2O fluxes observed in CAS systems, whereas high aeration rates have been reported, enhancing N_2O stripping (Chen et al., 2016).

MLE configurations have a median EF equal to 0.857% of the N-load. MLE processes with high EFs (up to 4% of the N-load) have been reported by Foley et al. (2010). Low EFs in MLE configurations (i.e. 0.003%, 0.065%) have been observed in reactors with diluted influent concentrations due to groundwater infiltration (Bellandi et al., 2018), nitrification efficiency less than 73% (Ahn et al., 2010b) and low TN removal (~59%) due to COD/TN < 1.9 (Spinelli et al., 2018). Low EF in MLE reactors ranging from 0.003% to 0.065% of the NH_4^+ load have been also reported in the studies of Caivano et al. (2017) and Bellandi et al. (2018); however, conversion of these EF to % N-load was not possible and were not included in Fig. 3.

N_2O emission fluxes in A^2/O configurations are relatively low in the majority of the studies, with median equal to 0.1% of the N-load. One exception is the study of Wang et al. (2016b); they monitored an A^2/O reactor once per month for 1 year and showed that the EF varied from 0.1 to 3.4% of the N-load between different months. The DO concentration and operating conditions varied significantly in the reactor (i.e. DO ranged from 0.6 to 6.8 mg L^{-1}).

Limited N_2O monitoring studies exist in full-scale sidestream processes. One-stage granular anammox reactors have an average EF of 1.1% of the N-load. The same two-stage suspended biomass partial-nitritation and anammox process has been monitored in two studies (Kampschreur et al., 2008; Mampaey et al., 2016). In these studies, the average EF in the partial-nitritation SHARON reactor was ~2.8% of the N-load and was elevated compared to full-scale one-stage anammox reactors. N_2O fluxes quantification, in lab and pilot-scale single-stage granular anammox reactors have shown EFs ranging from 0.1 to 12.19% of N-load (Wan et al., 2019). Therefore, more studies are required to establish reliable ranges of EFs in sidestream processes.

Differences in the reported N_2O fluxes are also observed in studies that apply similar configurations and operational conditions (supplementary material B, Tables B1 and B2). For example, Kampschreur et al. (2008) and Mampaey et al. (2016) monitored the N_2O emissions in the same two-step SHARON-Anammox reactor system and observed EFs that were equal to 1.7% and 3.8% of the N-load, respectively. Apart from slightly different DO setpoints (2 mg L^{-1} in the work of Mampaey et al. (2016) and 2.5 mg L^{-1} in the work of Kampschreur et al. (2008), the operational conditions (i.e. temperature, influent N-load, system treatment efficiency, hydraulic retention time (HRT), etc.; Table B1) during the two monitoring periods were quite similar. The main identified difference was the increased anoxic liquid N_2O formation during the anaerobic period of the partial-nitritation reactor in the study of Mampaey et al. (2016). In terms of monitoring protocols, Kampschreur et al. (2008) collected grab-samples for approximately 3 days, whereas continuous gas monitoring for 21 days was conducted by Mampaey et al. (2016). Moreover, the air infiltration in the covered reactor due to negative pressure was not considered in the study of Kampschreur et al. (2008).

The aforementioned examples emphasize the difficulty in the comparison of GHG emissions among various studies and development of EF databases for process groups. The benchmarking of GHG emissions of different plants can be hampered even if the same monitoring protocol is applied to monitor processes belonging to the same group. There are also cases where emissions have been measured according to different analytical procedures within the same system; fact that adds further difficulty in the comparisons. The relatively short monitoring periods and varying monitoring strategies can also influence the comparability and accuracy of the reported EFs. This will be discussed in detail in the following sections.

3.2. Duration of monitoring campaigns and seasonality

Seasonal environmental variabilities, such as temperature, can influence the bacterial community structure in WWTPs (Flowers et al., 2013). The N_2O formation and emissions during the BNR are expected to have temporal variations. Temperature can significantly affect the AOB specific growth rate during nitrification (Van Hulle et al., 2010). The higher temperature also decreases the N_2O solubility, thus intensifying the N_2O stripping to the atmosphere (Reino et al., 2017). On the other hand, Adouani et al. (2015) observed that the N_2O emissions increased up to 13%, 40% and 82% of the TN-removed at temperatures equal to 20 °C, 10 °C and 5 °C, respectively, in a batch reactor fed with synthetic wastewater. The latter was attributed to the increased sensitivity of the N_2O 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 also impact on the enzymatic reactions and affect the emissions. Vasilaki et al. (2018) observed peaks of N_2O emissions coinciding with precipitation events, at low temperatures, in an OD during a 15-month monitoring campaign. However, further investigation is required to understand potential seasonal effects on the N_2O emissions.

The monitoring periods of the full-scale campaigns for all the processes considered in this analysis are summarized in supplementary material B, Tables B1 and B2. Fig. 4 shows the EF for mainstream technologies based on the length of the monitoring period. Only studies that have reported EFs in terms of the influent N-load have been considered (supplementary material A - Table A1). The monitoring campaigns have been categorized into 3 distinct groups, based on their duration, i) short-term campaigns performed in a limited period of time (less than 1 month), ii) medium-term monitoring campaigns that last longer than one month but have not captured all the temperature ranges observed in the system, and ii) long-term monitoring campaigns that last at least 1 year. Both continuous and discontinuous monitoring studies have been included in the analysis. In the discontinuous N_2O monitoring studies, the gaseous N_2O fluxes have been grab-sampled (i.e. via gas-bags, closed chambers etc.) and subsequently quantified using analytical methods in the lab (offline monitoring) or intermittently sampled (i.e. with floating chambers for 1–2 days/month) but quantified continuously on-site (i.e. via GHG analyzers) (online monitoring). Most discontinuous monitoring campaigns had monthly or bi-monthly sampling frequency. The only exception is the study of Ahn et al. (2010b) who monitored several systems in only two distinct seasons (warmest and coldest temperatures). Discontinuous studies with sampling extending over 1-month period, have been categorized as medium-term or long-term based on the duration of the study (less or more than 1 year). In continuous monitoring campaigns N_2O fluxes have been collected continuously (i.e. via chambers) and quantified online, on-site (i.e. via GHG analysers).

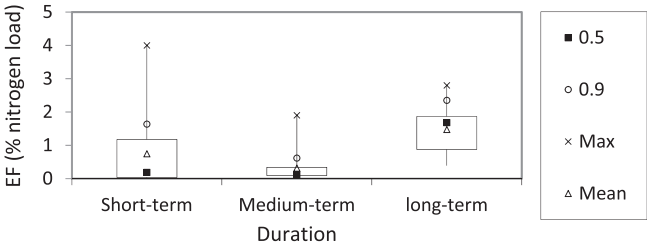

Fig. 4. EF values with respect to the length of the monitoring period for mainstream treatment technologies.

About 30% of the EFs shown in Fig. 4 are based on monitoring periods lasting less than two days (Foley et al., 2010; Filali et al., 2013; Samuelsson et al., 2018). Annual EF variation has been investigated discontinuously (monthly or bimonthly sampling frequency) in approximately 10% of these systems (i.e. Sun et al., 2015; Wang et al., 2016b), whereas long-term (≥1 year) continuous monitoring campaigns have been performed only in two of the examined works (Daelman et al., 2015; Kosonen et al., 2016). SBRs have been excluded in order to avoid further biases in the results, since the reported average N_2O emissions are significantly higher than the average EF of other mainstream N-removal configurations. Sidestream technologies have been also excluded because they have not been monitored long-term to examine seasonal effects. The average EF is equal to 0.8% (median 0.2%) and 0.3% (median 0.1%) of the N-load for the systems monitored short-term and medium-term (but without capturing the whole spectrum of seasonality effects), respectively. The studies investigating seasonal trends of N_2O emissions reported an average EF of 1.5% (median 1.7%) of the N-load.

Daelman et al. (2013b) demonstrated that short-term campaigns, in the system investigated, are likely to produce unreliable EF estimates independently of the monitoring approach. Additionally, the authors found that short-term campaigns have a high probability to underestimate actual emissions. According to Fig. 4, the highest gaseous N_2O loads belong to long-term continuous or discontinuous monitoring campaigns.

Long-term and medium-term campaigns have also shown a high variability of the reported N_2O emissions. Amongst the examined studies, Daelman et al. (2015) implemented the longest continuous real-field campaign that reinforced the existence of seasonal emission variability. Seasonality is also supported by the findings of several other studies (Brotto et al., 2015; Sun et al., 2013;

Yan et al., 2014). Bollon et al. (2016b) and Bollon et al. (2016a) studied a sidestream nitrifying and post-denitrifying biofiltration system, respectively, by performing two monitoring campaigns; one in summer at 22.5 °C and one in winter at temperatures lower than 14 °C. Their results indicated a significant seasonal variation of the N_2O formation. The EF of the nitrifying filters was equal to 2.3% of the NH_4^+ removed during the summer campaign, and 4.9% of the NH_4^+ removed during the winter campaign. The dissolved N_2O concentration in the post-denitrifying biofilter effluent was equal to 1.3% in summer and 0.2% in winter with respect to the NO_3^- uptake.

Short-term monitoring periods are likely to miss underlying seasonal variations in the N_2O formation (or be affected by short-term process perturbations), and, consequently, complicate the direct cross-comparisons between different studies and their findings. For instance, the monitoring of an A^2/O process for nine months (grab-samples taken once per month) led to an average EF equal to 0.08% of the influent TN (Yan et al., 2014), whereas a similar A^2/O process monitored for two days (by taking grab-samples) presented an EF of 0.85% of the influent TKN (Foley et al., 2010). Wang et al. (2016b) showed that the EF from a PF A^2/O reactor was characterized by significant seasonality and varied from 0.01% to 3.5% of the influent TN; within the range of EFs reported in the studies by Yan et al. (2014) and Foley et al. (2010). It can be concluded that the EF differences between similar configurations shown in Figs. 2 and 3 are strongly affected by the seasonality of the emissions.

3.3. Monitoring and sampling methods

Several authors have highlighted that the sampling methodology can influence the EF quantification (Daelman et al., 2013b; Aboobakar et al., 2013; Wang et al., 2016a; Kosonen et al., 2016; Hwang et al., 2016). For instance, several sources of uncertainty in chamber techniques similar to the techniques applied in the wastewater sector, have been identified in GHG monitoring campaigns of running waters (Duchemin et al., 1999; Lorke et al., 2015; Matthews et al., 2003; Vachon et al., 2010). A detailed analysis of the potential uncertainties related to the different sampling methods was out of the scope of this review. However, it is important to underline that further research is needed to reveal the influence of different sampling methods and facilitate the benchmarking of EF values for different groups of processes.

This section focuses on the sampling method (continuous and discontinuous). The main characteristics of the sampling strategies are shown in supplementary material B, Table B1 and B2.

Fig. 5 illustrates the boxplot of the average EF of mainstream processes for cases of continuous gaseous monitoring using a gas analyser versus the boxplot for studies with intermittent sampling campaigns. Only medium-term and long-term studies have been considered in the analysis (supplementary material A – Table A1). Mainstream processes monitored discontinuously exhibited an average EF of 0.44% of the N-load (median EF was 0.2% of the N-load), whereas processes monitored continuously with gas analysers had an average EF equal to 1.2% of the N-load (median EF is 1.1%). The majority of the process monitored intermittently (once or twice per month) have collected grab-samples of N_2O fluxes (supplementary material B, Table B2). Offline grab sampling is often characterised by time limitations; usually the sampling occurs during WWTP operating times and provides discrete measurements (e.g. Wang et al., 2011) that are unable to capture the whole spectrum of diurnal variabilities (Daelman et al., 2015; Wang et al., 2016a). Additionally, the temporal variability of N_2O emissions is highly dynamic (i.e. Daelman et al., 2015) and strongly affected by operational conditions. Therefore, low-frequency, long-term sampling might not capture adequately the whole range of N_2O emissions induced by short-term changes in operational conditions and pollutant concentrations. Overall, the differences can be attributed to the case-specific nature of the EF, as well as to the restrictions concerning the duration and frequency of the discontinuous campaigns (difficulty in collecting grab-samples for longer periods - whole days, night time, weekends, etc.).

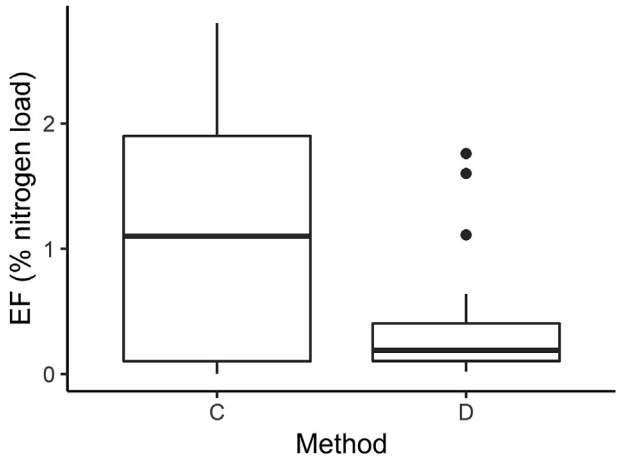

Fig. 5. Boxplots of the average EF with respect to the method of gaseous sampling for medium-term and long-term studies (D: discontinuous monitoring, C: continuous monitoring) for mainstream treatment processes. The rectangles represent the interquartile range. The median is represented by the black horizontal line dividing the box in 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).

3.4. Towards benchmarking of EFs: progress and limitations

The amount of quantified emissions is highly affected by a variety of parameters (e.g. process type, WWTP characteristics, monitoring strategy, duration of monitoring campaign, etc.). Therefore, estimating the N_2O EFs in WWTPs with either offline or online monitoring campaigns remains challenging. In this section, efforts have been focused on the classification of EFs for different groups of processes based on the NH_4^+ removal efficiency and the influent flow-rate. Additionally, an- N_2O monitoring framework for the development of comparable EFs for the wastewater sector is also discussed.

Fig. 6 shows the EF (with respect to the influent N-content) for mainstream processes and the achieved NH_4^+ removal efficiency (%). The different processes are represented by different colours. A detailed list of the examined studies is provided in the supplementary material B (Table B2). Triangles show that seasonal effects have been investigated in the respective process, whereas circles represent short-term studies that have not investigated seasonality. The size of the data points represents the size of the WWTP. No specific trend was identified between the observed EFs and NH_4^+ removal for the different mainstream processes. The N_2O emissions for most of the processes in smaller WWTPs (influent flow-rate<200,000 m^3 d^{-1}) were less than 0.5% of the N-load, independently on the process type and nitrification efficiency. In addition, most of the processes with N_2O emissions less than 0.1% of the N-load referred to studies performing short-term and medium-term N_2O monitoring campaigns.

In-depth comparisons require more details on configurations, control strategies and operational conditions. Currently, there are still no specific guidelines to standardize the reporting of

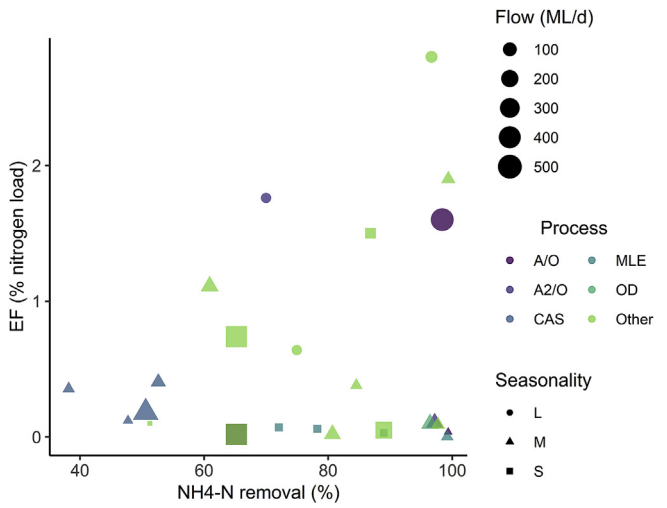

Fig. 6. EF of the mainstream technologies with respect to the achieved NH_4^+ removal. The different colours represent different processes, whereas the different shapes differentiate short-term, medium-term and long-term studies. The size of the data points depicts the size of the WWTP in terms of influent flow-rate. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

operational, process and monitoring strategy information from existing studies. The supplementary material B summarizes process-based information from past full-scale N_2O monitoring campaigns. Overall, ~70% of the mainstream studies have reported the EFs in terms of N-load. Additionally, influent and effluent NH_4^+ concentrations are available for ~35% of the systems analysed. Information on the water temperature during the monitoring campaign has not been provided for half of the studies. Limited studies provided information on the control strategy of the system (i.e. DO set-point) and other operational parameters of the processes (i.e. HRT, SRT). Given the variability of EFs amongst similar process groups (Figs. 2–6), the identification of EF patterns needs to consider process-specific operational and environmental information.

The monitoring strategies require sampling protocols that are case-specific (e.g. the choice of appropriate sampling locations). Specific protocols for the design of monitoring strategies can be found in the study of van Loosdrecht et al. (2016). Elemental mass balances can also be used to confirm the validity of the measurements (Castro-Barros et al., 2015; Mampaey et al., 2016) independently of the applied monitoring protocol. As shown in supplementary material B, Table B2, grab-sampling the gaseous fluxes often lacks the acquisition of weekend or night-time samples, thus failing to depict the diurnal variability of the emissions. Moreover, short-term monitoring studies are frequently unable to accurately capture the temporal N_2O dynamics (Daelman et al., 2015; Kosonen et al., 2016). Daelman et al. (2013b) concluded that the accurate quantification of the average N_2O emissions requires long-term online or grab-sampling monitoring campaigns that consider the seasonal variations of temperature.

The analysis of historical process and plant data can be also useful, linking the emissions with specific and reoccurring operational and environmental conditions (e.g. dry vs wet weather, temperature) for short-term and long-term monitoring campaigns. Relationships established during short-term monitoring campaigns can be linked with the periodic operational and environmental process conditions and cannot be generalized to understand the long-term N_2O dynamics (Vasilaki et al., 2018). The latter is important due to the restrictions on the duration of the monitoring campaigns resulting from the entailed costs. It is also essential to identify and report process perturbations that can affect the N_2O emissions even on a long-term basis (Vasilaki et al., 2018).

The current analysis shows that N_2O fluxes must be reported together with the configuration type, the seasonal operating conditions (e.g. pH, temperature, influent TN, effluent TN, PE, wastewater volume, COD, mixed liquor suspended solids (MLSS), SRT, HRT, recycle ratios, etc.).

4. N_2O monitoring campaigns and N_2O dynamics

Several methods that have been applied in N_2O monitoring campaigns can increase the understanding of the N_2O generation in full-scale processes. These include: i) techniques for the translation of WWTP operational and N_2O data into information (e.g. graphical representation of variables, feature extraction techniques, multivariate analysis, etc.), ii) mechanistic models simulating the N_2O dynamics, and iii) techniques for unveiling the relative contribution of different pathways to the emissions (e.g. isotopic analysis, real-time polymerase chain reaction (qPCR) etc.).

The effect of several parameters that are significant for N_2O generation (i.e. DO, COD/N, pH, temperature) has been extensively investigated (Kampschreur et al., 2009b; Law et al., 2012; Massara et al., 2017) based on lab-scale, pilot scale and full-scale mainstream and sidestream processes. This section aims to complement these studies; the main findings of the full-scale technologies are categorized for different process groups focusing on the techniques that have been applied and their contribution to the understanding of the behaviour of N_2O emissions.

4.1. Overview of the techniques

4.1.1. Techniques translating WWTP operational and N_2O data into information

More than 40 physical, physico-chemical and biochemical variables (e.g. temperature, flow-rates, reduction-oxidation (redox) potential, DO, N-compounds and organic matter concentrations, alkalinity, etc.) can be monitored online to evaluate process performance (Vanrolleghem and Lee, 2003). Online monitored variables when combined with laboratory analyses can provide useful insight into the N_2O patterns and behaviour. Techniques that have been applied to translate WWTP data into information in full-scale N_2O monitoring campaigns include: i) graphical representations and simple feature extraction methods and ii) statistical analysis and data mining methods.

In most of the studies, the online and laboratory data utilization has been limited to the investigation and graphical representation of the significant parameters' profiles (i.e. DO, NO_2^-, NH_4^+, aeration flow-rate) in combination with the response of the N_2O emission behaviour. Additionally, descriptive statistics (i.e. central tendency, dispersion, position, etc.) of the process variables and N_2O emissions is commonly analysed and reported.

Correlation analysis and linear multivariate regression models are the main statistical techniques that have been used to reveal the N_2O emissions' dependencies with operational variables in full-scale systems (i.e. Brotto et al., 2015; Bollon et al., 2016a; Aboobakar et al., 2013). Dimensionality reduction techniques (e.g. principal component analysis (PCA), independent component analysis (ICA)), clustering (e.g. hierarchical, k-means), linear and non-linear supervised learning techniques (e.g. partial least squares (PLS), artificial neural networks (ANN) and support vector machines (SVM)) are also powerful tools utilised to transform the WWTP data into knowledge (Haimi et al., 2013; Corominas et al., 2018). However, advanced information extraction methods have rarely been used to analyse data from N_2O monitoring campaigns.

Recently, Sun et al. (2017a,b) constructed a back-propagation ANN to simulate N_2O emissions in an anaerobic-oxic (A/O) process; thus demonstrating the feasibility and simplicity of predicting N_2O emissions with data-driven models.

4.1.2. Experimental studies in full-scale systems – modification of operational conditions

Several N_2O monitoring campaigns in full-scale sidestream processes have tested different operational conditions to investigate their impact on the emissions (Castro-Barros et al., 2015; Mampaey et al., 2016). The majority of the studies have focused on inducing changes in the duration and flow-rate of aeration compared to the baseline control strategy of the examined reactors.

4.1.3. Techniques unveiling the relative contribution of different pathways to the emissions

Isotopic and molecular biology analysis are emerging techniques that can provide insights into the N_2O generation pathways. Molecular biology methods (e.g. quantitative reverse transcription polymerase chain reaction RT-qPCR, FISH, etc.) can quantify the microbiological structure driving the N-cycle and the bacterial population able to reduce N_2O at WWTPs under various environmental and operational conditions (Castellano-Hinojosa et al., 2018; Song et al., 2014). Isotope techniques have only recently been implemented at full-scale systems to distinguish the respective contribution among the N_2O pathways and increase the understanding of the pathways that are responsible for the N_2O formation (Townsend-Small et al., 2011; Tumendelger et al., 2014). A recent critical evaluation of natural abundance and labelled isotopes for N_2O studies can be found in the study of Duan et al. (2017).

4.1.4. Mechanistic models

The mechanistic models are a popular tool for the prediction of the N_2O generation and emission during the BNR in WWTPs. Based on different assumptions, a variety of one- and multiple-pathway models have been suggested. Their structure is based either on the widely accepted ASM layout (as suggested by Henze et al., 1987, 2000), or on the more recent electron carrier concept that describes the N_2O production via the mechanism of the relevant complex oxidation-reduction reactions taking place during wastewater treatment (e.g. Ni et al., 2014). All models, though, consider the effect of changing operational parameters (e.g. DO, NO_2^- levels, aeration regime, etc.) on the N_2O generation. A comprehensive evaluation of the different modelling approaches, underlying assumptions, kinetics, stoichiometric parameters, calibration and validation procedures of several single-pathway and two-pathway AOB models, heterotrophic denitrification pathway models, and integrated N_2O models describing all three major microbiological pathways is provided by Ni and Yuan (2015). The authors have provided guidelines for the selection of the most appropriate modelling approach under different DO and NO_2^- concentrations based on the structural assumptions of the models. The debate on the model that best describes and decouples the major N_2O formation pathways is still ongoing with several extensions and variations of the original approaches developed recently (Ding et al., 2017; Domingo-Félez and Smets, 2016; Massara et al., 2018).

4.2. Process-based insights based on the applied techniques

Robust documentation of the dominant pathways among the different process configurations is still missing (Ma et al., 2017). This section discusses correlations between N_2O emissions and operational variables and dominant N_2O pathways that have been identified for different full-scale process groups. Table 1 provides a summary of the dominant N_2O pathways that have been reported for different wastewater treatment processes based on the techniques that have been applied in the monitoring campaigns. Studies that have not discussed possible N_2O pathways have not been considered.

Overall, the majority of the studies investigating the N_2O dominant pathways in mainstream full-scale systems with descriptive statistics and visual inspection (i.e. via univariate/bivariate graphs) of significant variables (e.g. NH_4^+, DO concentrations, influent flow-rate and N_2O emissions) have not considered NH_2OH to be a significant pathway for N_2O generation, regardless the configuration (Table 1).

Specifically, in 1-step feed A/O and A^2/O configurations and processes with anoxic/aerobic alternations and plug-flow pattern, studies have observed: i) spatial N_2O peaks in the transitions from anoxic to aerobic zones (i.e. Rodriguez-Caballero et al., 2014; Sun et al., 2017a) under low DO concentrations (<1 mg L^{-1}), ii) temporal increase in the N_2O emissions that coincide with increases in the NO_2^- and NH_4^+ concentrations (i.e. peak loads) under oxygen-limiting conditions (Wang et al., 2011, 2016b), and iii) N_2O emission peaks coinciding with elevated NO_2^-concentrations in the aerobic zones (Sun et al., 2017a). Therefore, the nitrifier denitrification pathway has been suggested to be dominant. This is also supported by Wang et al. (2016b) who studied the relative abundancy of the AOB and the denitrifying bacteria under different seasonal conditions for an A^2/O process with a plug-flow pattern (DO 0.6–6.8 mg L^{-1} and N-concentration up to 30 mg L^{-1}) to provide insights on the N_2O generation pathways. The authors quantified the expression of functional genes harboring the NH_3 monooxygenase (*amoA*) (i.e. enzyme catalyzing the first step of nitrification) for the AOB, as well as of the *nosZ* harboring the N_2O reductase (i.e. enzyme catalyzing the reduction of N_2O to N_2) for the denitrifiers by RT-qPCR. Wang et al. (2016b) also applied correlation analysis; the N_2O emissions (ranging from 0.01 to 3.4% of the TN-load) were mainly dependent on the NO_2^- concentrations as well as on the relative AOB abundances, hence indicating that nitrifier denitrification was the dominant pathway in the aerobic zones of the reactor. On the contrary, the obtained results revealed that the emissions were not affected by the relative abundancies of the denitrifiers. However, it must be noted that the functional gene levels are not always representative of the activity of the corresponding enzyme (Henderson et al., 2010). Similarly, Castellano-Hinojosa et al. (2018) quantified the 16SrRNA, *amoA* and *nosZ* genes of the total bacterial and archaeal population in three full-scale predenitrification-nitrification systems. They employed multivariate analysis (i.e. non-metric multidimensional scaling (MDS) and similarity analysis based on Euclidean distance, to identify the environmental variables that are best linked to the patterns of community structure via BIO-ENV procedure) to link the bacterial structure with the N_2O emissions and environmental/operational variables. They found a strong positive correlation between the AOB and the emissions in the anoxic compartments, where the N_2O release was higher. Therefore, the authors suggested nitrifier denitrification as the dominant pathway. On the other hand, the emissions were negatively correlated with the AOA abundance and the N_2O reducers. It was concluded that the elevated NO_2^- concentrations, the low temperatures and short SRTs mainly influenced the abundance of the bacterial community encoding the *nosZ* gene and contributed to the N_2O accumulation.

NH_2OH oxidation has not been considered as a dominant N_2O pathway in the majority of A^2/O and A/O process groups. However, Toyoda et al. (2011) applied site-preference (SP) isotopic analysis in a A^2/O configuration and found that the NH_2OH oxidation and nitrifier denitrification pathways contributed almost equally to the N_2O formation in the beginning of the aerobic tank. The DO concentrations in the reactor, though, were not reported. Additionally,

Table 1
Main findings of past studies that result in the identification of the most contributive N_2O production pathway (where possible). a: Visualization of significant profiles & descriptive analysis, b: Modified operation mode, c: Statistical analysis and data mining, d: Mechanistic model development, e: Isotopic analysis, f: real-time qPCR.

Source	Process	NH_2OH oxidation	Nitrifier denitrification	Heterotrophic denitrification
Castro-Barros et al., 2015[a, b]	One-stage PNA granular Sidestream	N_2O emissions elevated during shifts from low to high aeration (NH_4^+ accumulation, high AOR) → main pathway	Not a main pathway	N_2O emissions elevated during shifts from low to high aeration (NH_4^+ accumulation, high AOR) → potential contributor
Mampaey et al., 2016[a, b]	One-stage SHARON granular Reactor Sidestream	Not a main pathway	Presence of NH_2OH in anoxic periods; lower DO resulting in increased N_2O emissions	N_2O formation during anoxic periods under the presence of NO_2^- & small amounts of organic substrate
Kampschreur et al., 2008[a]	Two-reactor partial-nitritation-anammox process Sidestream	Excluded during anoxic conditions in the nitritation reactor (despite significant N_2O formation)	• Anammox reactor: absence of O_2 • Nitritation reactor: emissions not affected by the influent composition (therefore C/N ratio)	Not a main pathway
Stenström et al., 2014[a]	Nitrification-denitrification SBR, Sidestream	Not discussed	Considerable N_2O formation under $DO = 0.5\ mg\ L^{-1}$ & NO_2^- accumulation ($>20\ mg\ L^{-1}$)	N_2O accumulation during denitrification (under conditions of low COD:N and high NO_2^-); quickly stripped off to the atmosphere as soon as aeration resumed
Wang et al., 2016b[a, c, f]	A^2/O with plug-flow pattern	Not a dominant pathway (low NH_4^+ concentrations)	• Coexistence of NO_2^-, NH_4^+ & O_2-limiting conditions • Correlation between NO_2^- and N_2O emissions • Strong responses between NO and N2O emissions and the relative abundance of AOB.	Not a dominant pathway (no peaks observed after anoxic zones)
Wang et al., 2011[a]	A^2/O	Not a main pathway	• Rapidly increased N_2O emissions due to DO limitation ($DO < 2.5\ mg\ L^{-1}$); maximum N_2O emission at $DO = 0.75\ mg\ L^{-1}$ • Increase in NO_2^- concentration (from 0.2 to 0.6 $mg\ L^{-1}$) during nitrification leading to increase in N2O fluxes	Not a main pathway
Toyoda et al., 2011[a, f]	A^2/O	SP Isotopic analysis: • ~50% contribution in the beginning of aerobic tank	SP Isotopic analysis: • ~50% contribution in the beginning of aerobic tank • Dominant pathway from middle to the end of aerobic tank	N_2O was produced during denitrification
Aboobakar et al., 2013[a, c]	A/O plug-flow reactor	Not dominant pathway	Considered dominant in zones with $DO < 1.5\ mg\ L^{-1}$	Considered dominant in zones with depleted NH_4^+, DO fluctuations, NO_3^- availability & lack of NO_2^-
Sun et al., 2017a[a]	A/O	Higher DO: certain NO_2^- amount potentially utilized to oxidize NH_3 to NH_2OH, thus leading to the N_2O production	Low-DO condition (i.e. $<1\ mg\ L^{-1}$) usually observed at the beginning of the oxic zone	Not a main pathway
Kosonen et al., 2016[a, c] & Blomberg et al., 2018[d]	A/O bioreactor	Only this AOB pathway modelled due to the existing DO & NO_2^- conditions, N_2O production mainly in the aerated zones, N_2O consumption in the anoxic zones → main pathway	• Increasing the number of nitrifying zones resulting in higher overall N_2O emissions (N_2O production possibly via nitrifier denitrification) • Given that anoxic-aerobic volume controlled by the NH_4^+ concentration, unclear if increased emissions caused by increased NH_4^+ concentration or increased number of nitrifying zones	Not main pathway
Pan et al. (2016) & Ni et al., (2015)[a, d]	2 step-feeding, anoxic/oxic/anoxic/oxic plug-flow reactor	N_2O emissions increasing with the AOR increase (2nd step of the plug-flow reactor)	N_2O emissions increasing with the AOR increase (2nd step of the plug-flow reactor)	Not a main pathway
Rodriguez-Caballero et al.2014[a, b]	Anoxic/oxic/short anoxic/oxic plug-flow reactor	Not a main pathway	• N_2O peaks when transitioning from anoxic to aerobic conditions; O_2 limitation considered as enhancing the activation of the nitrifier denitrification pathway • N_2O emissions increasing with potential shock loads; the AOB likely to activate their denitrification pathway after shock loads of toxic compounds	Not a main pathway
Castellano-Hinojosa et al., 2018[c, e]	Two sequential bioreactors (anoxic and aerated)	Not a main pathway	• Strong positive correlation between AOB abundance & N_2O emission; hence, more possible pathway under anoxic conditions • $0.5 < DO < 1\ mg\ L^{-1}$: enough O_2 provided to the AOB for the oxygenation of NH_3 to NH_2OH but not	Not a main pathway

(*continued on next page*)

Table 1 (*continued*)

Source	Process	NH_2OH oxidation	Nitrifier denitrification	Heterotrophic denitrification
			for aerobic respiration; NO_2^- potentially used as alternative electron acceptor to complete nitrification	
Tumendelger et al., 2014[a, f]	CAS	SP Isotopic analysis: • Up to 90% contribution at DO ~2.5 mg L^{-1} • ~50% contribution at DO ~1.5 mg L^{-1}	SP Isotopic analysis: • Dominated at DO < 1.5 mg L^{-1} • ~50% contribution at DO ~1.5 mg L^{-1}	Not discussed
Daelman et al., 2015[a, c]	Carrousel reactor	Carrousel: emissions coinciding with aerated periods (AOR governed by DO); the relationship between the AOR & the N_2O production usually explained by referring to the NH2OH pathway; however, not considered dominant	• Carrousel: emissions correlated with the NO2-concentration peaks • Prevalence of low-DO zones	Carrousel: reactor lacking sufficient anoxic space to allow the completion of denitrification
Ni et al., 2013[d]	OD with surface aerators	Main pathway since high NH_4^+ concentrations were observed without simultaneous NO_2^- increase in the aerated zones/phases	Not a main pathway	Not a main pathway
Ni et al., 2013[d]	Feeding and aeration (90 min)/settling (35 min)/ decanting (55 min) SBR	Main pathway since high NH_4^+ concentrations were observed without simultaneous NO_2^- increase in the aerated zones/phases	Not a main pathway	Not a main pathway
Sun et al., 2013[a, c]	Feeding (synchronous aeration)/aeration/settling/ decanting SBR (1 h each)	Not a main pathway	• Low DO during nitrification significantly affecting the N2O production → main pathway	Correlation between N2O emission and influent COD/N → contributor
Rodriguez-Caballero et al., 2015[a, b]	Reaction phase (~130 min)/ settling (~65min) and decanting (~65 min) SBR (anoxic/aerobic alternations − 3 cycle types)	Not a main pathway	• Certain NO_2^- accumulation under aerobic conditions • N_2O generation continuing after aeration stop	N_2O generation continuing after aeration stop
Wang et al., 2016a [a, c]	Full-scale biological aerated filter (BAF) for secondary nitrification	Low influent NH_3 concentration (<6 mg L^{-1}) → not a main pathway	• Significant linear correlation between N_2O & NO EFs in different seasons • Nitrifier denitrification suggested as possible pathway in accordance with the fact that the influent NO_2^- found as key factor regarding the N_2O & NO production	• Significant linear correlation between N_2O & NO EFs in different seasons • Minor possibility of heterotrophic denitrification contribution during the denitrification of NO to N_2O
Bollon et al., 2016b[a]	Nitrifying biofiltration (Biostyr® filters)	Not discussed	Not discussed	• Rapid increase in the net N_2O production rate at BOD:N < 3 • Intensity increased with the duration of carbon-limiting conditions

Blomberg et al. (2018) developed an ASM3-type NH_2OH-heterotrophic denitrification N_2O model with a k_La-based approach for the N_2O stripping (k_La_{N2O}: mass transfer coefficient for N_2O). The model was developed for the full-scale underground WWTP of Viikinmäki (Kosonen et al., 2016) that is divided into six zones (i.e. one anoxic pre-denitrifying zone, two alternating switch zones, three aerated nitrifying zones). High DO concentrations in the aerated zones (i.e. 1.5–3.8 mg L^{-1}) and low NO_2^- concentrations (i.e. 0.1 and 0.7 mg L^{-1}) were noted and the model adequately fitted the observed dissolved N_2O profiles. However, under the applied stripping modelling, the model overestimated the EF; hence showing that the stripping modelling approach must be improved.

Ni et al. (2015) considered all N_2O production pathways in a two-step plug-flow type reactor (anoxic/aerobic/anoxic/aerobic), in an attempt to explain the difference between the EFs of each step (1st step: 0.7% of influent-N, 2nd step: 3.5% of influent-N) using real data obtained from the study of Pan et al. (2016), for a full-scale step-feed plug-flow reactor. The N_2O production was mainly attributed to the heterotrophic denitrification taking place in the anoxic zone of the 2nd step that was receiving 70% less biomass compared to the 1st step (Table 1). This model has been successfully applied for the explanation of the observed EF difference and identification of dominant pathways since it (i) includes all the possible production pathways, (ii) has considered the design and operating features of the WWTP, and (iii) was calibrated/validated using data from the plant operation.

It must be noted that, in an anoxic-aerobic plug-flow reactor, the application of zone-based stepwise multiple regression showed that the effect of the N-load, DO and temperature on the N_2O emissions varied within the reactor (Aboobakar et al., 2013). Therefore, the dominant pathways can potentially vary in the aforementioned studies based on the location of the sampling among the different studies.

In the OD reactors N_2O fluxes have been mainly linked with: i) stripping of the dissolved N_2O that is generated in the anoxic zones (Sun et al., 2015; Yan et al., 2014) and ii) NO_2^- accumulation in the low-DO zones, thus indicating nitrifier denitrification and heterotrophic denitrification (Daelman et al., 2015) as dominant pathways. For instance, in an OD reactor, strong positive correlation (Pearson's coefficient) was identified between daily N_2O emissions and daily NO_2^- peaks (0.7) (487 days monitoring campaign) (Daelman et al., 2015). The authors proposed to use NO_2^- peaks as a diagnostic method for the prediction of N_2O peaks. In the same system, Vasilaki et al. (2018) applied changepoint detection techniques combined with hierarchical k-means clustering and PCA, to reveal the N_2O emission patterns, generation pathways and identified changes in the N_2O fluxes. The study concluded that the N_2O dependencies with other operational variables (i.e. NH_4^+, NO_3^-, DO) are dynamic and affected by the seasonal variations. The preferred N_2O pathways were also found to be dependent on time and operational conditions.

Additionally, a full-scale OD was modelled by Ni et al. (2013) considering the NH_2OH oxidation pathway (via the electron carriers approach, assuming the NH_2OH/NO model and no inhibition

of AOB NO reduction by DO) and the heterotrophic denitrification pathway (based on Hiatt and Grady (2008) and the electron competition between denitrification steps). Although the operational control of the OD is not explicitly described (i.e. aeration/DO set-points), more than 90% of the N_2O emissions were observed in aerated zones with DO > 2 mg L^{-1}. The model was calibrated using 3-day data from an intensive monitoring campaign and validated based on 1-day data with different influent conditions. The developed model linked the higher N_2O generation with the NH_4^+ concentration peaks (up to ~9 mg L^{-1} in the calibration and ~4 mg L^{-1} in the validation dataset) within the aerated zones suggesting the NH_2OH oxidation pathway as dominant. However, in the study of Ni et al. (2013), the respective contribution among the two AOB pathways was not explored whereas short-term data were used to validate the model. The AOB denitrification model developed by Mampaey et al. (2013) was applied using the same dataset from the OD for calibration and validation purposes (Spérandio et al., 2016). The model could adequately follow the trends of the N_2O behaviour after calibration of the anoxic reduction function (high value of 0.63 was required). NO_2^- accumulation and the resulting nitrifier denitrification contribution to the emissions cannot be excluded in full-scale WWTPs. Future enhanced model versions must consider this fact.

Overall, as shown in section 3, N_2O fluxes in CAS systems are generally low. Tumendelger et al. (2014) applied SP isotopic analysis and observed that the NH_2OH oxidation pathway was responsible for up to 90% of the N_2O formation under high DO (~2.5 mg L^{-1} at the middle and end of the aerobic tank) in a conventional AS system (>44.2% of NH_4^+ was nitrified and removed in gaseous form probably due to unintentional zones with low DO). Nitrifier denitrification and NH_2OH oxidation were almost equally contributing to DO levels around 1.5 mg L^1, whereas nitrifier denitrification dominated at DOs below 1.5 mg L^{-1}.

In sidestream reactors, elevated N_2O emissions during shifts from low to high aeration (NH_4^+ accumulation, high AOR), have been attributed to the NH_2OH pathway (Castro-Barros et al., 2015). Aeration intensity and profiles have been determined as significant control parameters for the N_2O generation (Harris et al., 2015; Rathnayake et al., 2015). The N_2O dynamics under different aeration intensities is likely to depend on the reactor configuration. For example, Mampaey et al. (2016) and Stenström et al. (2014) observed higher emissions when lower DO was applied in a partial nitritation-anammox system and a sidestream nitrification-denitrification SBR, respectively. On the other hand, Kampschreur et al. (2009a) could not identify a relationship between the N_2O increase and the higher aeration flow-rate during a prolonged aeration experiment in a single-stage nitritation-anammox reactor. Hence, the influence of the aeration regime on the N_2O generation is variable and depends on the reactor configuration. In Anammox reactors, N_2O formation during anoxic periods has been mainly attributed to the nitrifier denitrification pathway and partially to heterotrophic denitrification (e.g. under conditions of limited substrate provision; Castro-Barros et al., 2015; Mampaey et al., 2016). However, a recent study performed by Ma et al. (2017) demonstrated that N_2O formation via NH_2OH oxidation can also occur at low DO (~1 mg L^{-1}) probably catalysed by cytochrome P460. This finding contradicts previous experimental and model-based works according to which the NH_2OH oxidation pathway dominates solely at higher DO concentrations (e.g. Brotto et al., 2015) and is linked with the AOR (Peng et al., 2014).

Studies investigating dominant N_2O pathways for several groups of full-scale processes are still missing (Table 1) and further research is required for a robust mapping of the dominant pathways in different process groups. As a general remark, in most processes with elevated N_2O emissions (>0.85% of the N-load), independently of the configuration, elevated NO_2^- concentrations were also observed (Daelman et al., 2015; Foley et al., 2010; Rodriguez-Caballero et al., 2015; Wang et al., 2016b).

4.3. Limitations and future research

Several authors have underlined the difficulties in determining the respective contribution of each N_2O generation pathway during full-scale monitoring campaigns (Aboobakar et al., 2013; Wang et al., 2016b). None of the techniques analysed, in the previous sections can be applied standalone to explain the ambiguities surrounding the mechanisms and operational conditions that enhance the N_2O formation during wastewater treatment. This section, summarizes the main limitations of the techniques applied to explain N_2O fluxes behaviour in wastewater systems and describes how these techniques (combined) can maximize the outcome of future monitoring campaigns.

Two major drawbacks have been identified when studies rely exclusively on simple descriptive statistics and graphical representations of operational variables to provide insights on the N_2O emissions behaviour. Firstly, this approach considers independently several parameters that affect the N_2O generation. Thus, it becomes difficult to quantify the combined effect of several variables in full-scale systems via simple univariate or bi-variate graphical representations. For instance, Castro-Barros et al. (2015) observed higher N_2O emissions and formation in the transition from the anoxic to the aerated periods in a one-stage granular partial nitritation-anammox reactor. However, this increase cannot be solely attributed to the DO change, since the AOR, the NH_4^+ and the NO_2^- concentrations were also elevated during the transition. Secondly, graphs representing the behaviour of process variables in relation to the N_2O emissions usually cover only a limited period (often shorter than the monitoring campaign duration) or visualize average data in the majority of the reported studies. There are limitations in the effective visualization and dependencies in the extraction of information from long-term temporal multivariate datasets (i.e. overcrowded and cluttered visualisation) (Shurkhovetskyy et al., 2018). For example, Aboobakar et al. (2013) monitored the N_2O emissions of a PF reactor for 56 days; they reported the diurnal profile of the normalised average emissions, the average NH_4^+ diurnal profile and the average daily DO concentration- throughout the duration of the monitoring campaign.

The N_2O emission behaviour is characterised by temporal variations. Additionally, for a specific process, the dependencies of the emissions with operational variables are also expected to fluctuate under different environmental/operational conditions based on the preferred N_2O production pathway. Vasilaki et al. (2018) showed that the dependencies between N_2O emissions and operational variables fluctuated in a Carrousel reactor that was monitored for 15 months. Therefore, employment of advanced visualisation techniques capturing the dynamic behaviour of the operational variables from the whole duration of the monitoring campaigns (e.g. data abstraction, principal component-based analysis, clustering; Shurkhovetskyy et al., 2018; Aigner et al., 2008) can facilitate an accurate and deep understanding of the long-term N_2O behaviour in both sidestream and mainstream treatment processes. There is an interchangeable link among operational/environmental conditions, N_2O production pathways and emission rates. The combination of data mining methods can also be applied to identify distinct, different patterns and operational conditions in order to determine the respective contribution among the N_2O production pathways. It can reveal the links and relationships among the conditions triggering the N_2O emissions, the dominant pathways and the emission rates. However, few data-driven monitoring and control approaches have been validated in full-scale applications in

the wastewater sector (Haimi et al., 2013). Additionally, there is still little guidance for the selection of the most appropriate techniques for particular wastewater applications (Hadjimichael et al., 2016). Multivariate statistical analysis techniques have only recently been applied to translate data from the monitoring campaigns into useful information regarding the N_2O production (Vasilaki et al., 2018). Hence, structured approaches and data-driven extraction techniques need to be developed to process the incoming data from WWTPs (Corominas et al., 2018) and acquire information concerning the N_2O emission patterns.

Given that previously unreported pathways (Harris et al., 2015) or alternative conditions under which the already known pathways are activated have been recently identified in literature, isotopic and molecular biology analysis can be applied to provide insights into the N_2O generation pathways. In a recent review on the isotopic methods for the identification of the respective contributions of the different N_2O pathways, Duan et al. (2017) concluded, though, that there are still uncertainties regarding the accuracy of the SP methods (i.e. not standard SP signature values, unknown N_2O production pathways, etc.). Therefore, the authors suggest to complement the isotopic methods with other approaches, such as the mRNA-based transcription analysis (Ishii et al., 2014).

Mechanistic models considering all the possible N_2O production pathways are powerful tools to describe the operation of full-scale WWTPs, unveil the most contributive N_2O emission generation pathways and guide towards mitigation measures. However, there are still several challenges in the practical application, calibration and validation of mechanistic N_2O models in full-scale systems. Parameter uncertainty still plays a significant role in explicitly differentiating the contribution of different N_2O pathways via modelling studies; for instance, different AOB pathway models (i.e. Spérandio et al., 2016) and models with different contributions of denitrification N_2O-producing pathways (i.e. Domingo-Félez et al., 2017) have been adequately fitted to describe the N_2O emissions behaviour in the same systems. Inclusion of all major N_2O production pathways results in complex and overparameterized models impairing reliable calibration and validation. Additionally, short-term calibration and validation of models under specific operational conditions (i.e. dry weather) limits their accuracy when the system varies significantly (Guo and Vanrolleghem, 2014).

Future research can also explore the possibility of coupling sophisticated statistical tools (e.g. multivariate statistics, machine learning algorithms) with multiple-pathway mechanistic models for full-scale applications, and, thus, facilitate the fast and adaptable online implementation of model predictive control and forecasting decision support tools. The co-application of machine learning and mechanistic models in serial or parallel configurations have already been demonstrated in the wastewater sector. For instance, studies have shown that artificial neural network (ANN) models trained with the residuals of ASM-type models can enhance the prediction and generalization capabilities of the ASM models for highly daily variable influent pollutant concentrations and flow rates that are usually observed at WWTPs (Fang et al., 2010; Keskitalo and Leiviskä, 2015). Additionally, detailed simulations of variables that are not conventionally monitored in WWTPs (i.e. NO_2^-, NO) can be derived from the computationally universal mechanistic models and used together with raw process data to train the machine learning models. Trained machine learning models are very efficient for online monitoring and provision of decision support, and are widely applied in the industry (Wang et al., 2018). For example, a support vector machine regression (SVM) model was trained with the simulation results of an ASM2d model for a full-scale A^2/O reactor in the study of Zhang et al.(2014) to develop a flexible multi-objective optimization tool. The SVM model linked the operating parameter data sets from the ASM2d simulations with performance indexes considering cost and pollutant concentration aspects. The development of layer based on machine learning techniques can be integrated in a universal complex N_2O model to facilitate individual process parameters calibration. Multivariate statistics and pattern recognition algorithms can be applied to the variables monitored online in WWTPs to differentiate operational conditions (i.e. based on seasonal influent composition variations, different process rates affected by environmental conditions, system shocks, etc.) and guide towards different calibration requirements within the same process. Finally, multivariate statistics can be applied to identify and isolate complex relationships between system variables and guide towards process-specific simplified modelling approaches. Such integrated practical tools can help plant operators design effective mitigation strategies.

Quantifying the contribution of the N_2O production pathways in addition to the triggering mechanisms in biological processes remains a challenge and still requires extensive research (Guo et al., 2017). As a general remark, standardization and reporting of long-term N_2O monitoring campaigns, along with combined multivariate analyses of the provided data and mechanistic model development are required to increase the understanding and effectively control the N_2O emissions at WWTPs.

5. Monitoring campaigns and mitigation strategies

Mitigation measures have been developed and proposed mainly as outcome of studies that targeted at the: i) testing of different aeration/feeding control strategies in full-scale sidestream technologies (e.g. Castro-Barros et al., 2015; Mampaey et al., 2016; Rodriguez-Caballero et al., 2015), ii) development of mechanistic models simulating changing operational conditions (e.g. Ni et al., 2015), and iii) establishment of non-linear regression models (e.g. ANNs) to predict the behaviour of N_2O emissions (e.g. Sun et al., 2017a).

5.1. Mitigation measures and full-scale monitoring campaigns

Table 2 summarises the main N_2O mitigation strategies that have been proposed for full-scale systems along with the methodological approaches that facilitated the identification of these measures. As shown in Table 2, there is no standardised methodology for the establishment of N_2O mitigation strategies in full-scale systems.

Several studies have modified the aeration intensity, DO and cycle duration to investigate the effect on N_2O emissions within full-scale systems (Castro-Barros et al., 2015; Kampschreur et al., 2009a; Mampaey et al., 2016; Rodriguez-Caballero et al., 2015). For instance, Mampaey et al. (2016) achieved a reduction in the N_2O emissions by 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 have suggested an optimum control strategy for the minimisation of N_2O emissions based on the application of short aerobic-anoxic cycles (20-min aerobic phase and short duration of anoxic stage). Therefore, testing different operational modes is regarded as one of the most effective ways to identify measures for the emission mitigation (Table 2).

Ni et al. (2015) developed a mechanistic model utilising the data from a two-step PF reactor (Pan et al., 2016) showing that the biomass specific N-loading rate was responsible for the elevated N_2O emissions observed in the 2nd step of the process. Different operational conditions were tested with the model demonstrating that lower N_2O emissions (<1% of the N-load) can be achieved if 30% of the total return activated sludge (RAS) stream is recirculated

Table 2
Methods and main findings of studies resulting in mitigation measures.

Source	Process	Method	Main findings	Mitigation Measures
Castro-Barros et al. (2015)	One-stage PNA granular Sidestream	• Calculation of dissolved N_2O based on Mampaey et al. (2016) • Modified operation mode: Prolonged anoxic & aeration periods • Visualization of significant profiles (i.e. DO) & descriptive analysis	• Smoother aeration transitions during normal reactor operation connected with lower N_2O emissions; comparison with experiments • Prolonged anoxic periods leading to increased N_2O emissions	• Optimize the aeration regime • Ensure smooth shifts in the aeration pattern • Opt for short aeration intervals
Mampaey et al. (2016)	One-stage SHARON granular Reactor Sidestream	• Modified operation mode: prolonged anoxic & aeration periods, lower DO experiments, shorter SBR cycles • Calculation of dissolved N_2O based on Mampaey et al. (2016) • Visualization of significant profiles (i.e. DO) & descriptive analysis	• Nitritation reactor: N_2O formation higher during anoxic periods • Splitting the anoxic period: average anoxic N_2O formation rate decreased • Shorter cycles reducing the N_2O EF by 56% at the expense of higher NO_3^- concentrations	• Preferably operate under shorter cycles • Apply continuous aeration in nitritation reactor; this requiring optimization • Alternatively operate under lower DO setpoint
Kampschreur et al., 2009a	One-stage PNA granular	• Modify operation: varying aeration rate • Visualization of significant profiles & descriptive analysis	• Over-aeration significantly impacting on N2O emissions	• Ensure sufficient aeration control
Kampschreur et al. (2008)	Two-reactor partial-nitritation-anammox process Sidestream	• Visualization of significant profiles (N-compounds) & descriptive analysis	• Nitritation reactor: N_2O accumulation during the non-aerated phase • Anammox reactor: NO_2^- accumulation potentially increasing N_2O emissions	• Avoid anoxic phases in nitritation reactor (i.e. smaller reactors to ensure sufficient HRT) • Control the aeration in the nitritation reactor • Operate a one-reactor nitritation-anammox system; potentially emitting less N_2O due to limited NO_2^- accumulation
Ahn et al., 2010a,b	Multiple processes (i.e. MLE, step-feed BNR, OD)	• Multiple linear regression for several processes	• Investigate possible links between WWTP operating conditions & N_2O emission fluxes • Aerobic zones: N_2O fluxes correlated with location-specific pH, AS mixed liquor temperature, DO, NH_4^+ & NO_2^- concentrations & interactive combinations • Anoxic zones: N_2O fluxes correlated with location-specific sCOD, pH, AS mixed-liquor temperature, DO, NO_2^- & NO_3^- concentrations & interactive combinations	• BNR processes: Avoid high NH_4^+ & NO_2^- concentrations, DO & transients • Aerobic processes: avoid incomplete/intermittent nitrification & over-aeration • Rely on more uniform spatial DO profiles to promote SND • Minimize peak N-flow (flow equalization)
Ni et al. (2013)	OD with surface aerators & Feeding and aeration (90 min)/settling (35 min)/decanting (55 min) SBR	• Mechanistic model development	• Modelling of two full-scale municipal WWTPs (i.e. an OD & an SBR) • OD: decrease in the NH_4^+ concentration without simultaneous NO_2^- increase in the aerated zones • SBR: NH_4^+ accumulation leading to a high AOR during the aerobic SBR phases &, finally, to the increased production of intermediates (e.g. NH_2OH)	• Use the developed model to accurately simulate the emissions from the surface aerator zone in OD WWTPs, thus potentially correcting the N2O emission underestimation in full-scale WWTPs where the floating chamber method is not valid
Li et al., 2016	Reversed A2/O and OD	• Observations & literature	• N_2O generated & emitted more in summer than in winter • Microbial population & aeration strategy as key factors of N_2O generation & emission	• Avoid incomplete/intermittent nitrification & over-aeration during the aerobic processes to achieve lower N_2O emissions • Apply uniform spatial DO profiles to promote SND that probably leads to less N_2O emissions • Perform flow equalization to control the peaking factor of the influent N-loading to the AS • Ensure a sufficiently long SRT to prevent NO_2^- accumulation during nitrification • Avoid the COD limitation of the denitrification process by minimizing the pre-sedimentation of organic carbon in the influent & dosing additional organic carbon
Pan et al. (2016) &	2 step-feeding, anoxic/aerobic/anoxic/aerobic plug-flow reactor	• Mechanistic model development	• Step-feeding resulting in incomplete denitrification & affecting the AOR in	• Decrease the N_2O EF to the lowest value of <1% if 30% of the total RAS returns to the 2nd step

(*continued on next page*)

Table 2 (*continued*)

Source	Process	Method	Main findings	Mitigation Measures
Ni et al. (2015)			nitrification, hence increasing the total N_2O emission	
Wang et al. (2016b)	A^2/O with plug-flow pattern	• Investigation of AOB abundances	• N_2O emitted mainly from the oxic zone, with the emitting levels increasing greatly from the beginning of the oxic zone towards the zone end • NO_2^- accumulation directly triggering N_2O production • Both diurnal & seasonal N_2O emission levels fluctuating strongly • Other factors influencing the N_2O emission: low DO/temperature	• Increase DO availability for both AOB & NOB • Improve the AOB living conditions • Apply a step-stage aeration mode with varying aeration intensities (location-specific emission patterns for a plug-flow process) • Ensure a 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^2/O tank working volume
Sun et al. (2013)	Feeding (synchronous aeration)/aeration/settling/decanting (1 h each) SBR	• Visualization of significant profiles (DO & N_2O) & descriptive analysis • Multiple linear regression analysis to investigate relationship of N_2O emissions & environmental factors • Bimonthly sampling to examine changes in the relationship between N_2O emissions &environmental factors (long-term: 12 months)	• N_2O flux from different treatment units/periods following a descending order: feeding period, aeration period, settling period, swirl grit tank, decanting period & wastewater distribution tank • Feeding & aeration periods accounting for >99% of N_2O emissions • Low DO during nitrification majorly influencing N_2O production	• Increase the aeration rate during the feeding period & decrease it to a proper level for nitrification in the aerobic stage • Supply external carbon source during denitrification/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
Rodriguez-Caballero et al. (2015)	Reaction phase (~130 min)/settling (~65min) and decanting (~65 min) SBR (anoxic/aerobic alternations – 3 cycle types) SBR	• During the experimental campaign, 3 different cycle configurations implemented as part of the normal SBR operation • Testing of a modified cycle configuration for the N_2O mitigation • Observe dissolved & gaseous N_2O profiles vs time	• N_2O emissions accounting for >60% of the total carbon footprint of the WWTP • Cycles with long aerated phases showing the largest N_2O emissions, with a consequent increase in the carbon footprint • Transient NH_4^+ & NO_2^- concentrations & transition from anoxic to aerobic possibly involved in the increased N_2O production	• Apply intermittent aeration to reduce the NO_2^- accumulation • Adopt a cycle configuration with short aerated periods • Allow the system to consume N_2O through denitrification
Spinelli et al. (2018)	MLE	• Event-based sensitivity analysis • Box-plots of diurnal behaviour of significant variables & N_2O	• Lower COD:N resulting in higher N_2O emissions due to disturbed denitrification • Daily N_2O peaks occurring under conditions of higher aeration flow-rate (more intense stripping)	• Equalization of the influent flow-rate
Townsend-Small et al. (2011)	MLE	• Isotopic composition of N_2O	• Both nitrification & denitrification contributing to the N_2O emissions within the same WWTP • BNR significantly increasing urban N_2O emissions	• Apply engineering processes for the selection of bacteria capable of reducing NO_3^- without releasing significant N_2O amounts
Castellano-Hinojosa et al. (2018)	Two sequential bioreactors (anoxic and oxic)	• Simultaneously link the abundance of AOB, AOA & N_2O-reducers with the changes of the operational/environmental variables	• N_2O emissions strongly correlated with increased abundances of AOB & lower counts of N_2O-reducers • Unlikely significant contribution of AOA to N_2O generation since their abundance correlated negatively to N_2O emissions • AOB abundance favoured by higher NO_3^- & NO_2^- concentrations in the AS	• Avoid NO_2^- accumulation, low temperatures & excess DO in the anoxic bioreactors to enable complete heterotrophic denitrification & hinder nitrifier denitrification
Sun et al. (2017a,b)	Biological tank with an anoxic & an oxic zone A/O	• Construction & performance evaluation of BP-ANN model	• DO having a significant influence on the N_2O production • BP-ANN model suitable for the prediction of N_2O emissions in other WWTPs with different configurations (e.g. A^2/O, SBR & nitrification-anammox), if influent/environmental parameters & N_2O emission data can be investigated through full-, pilot- or lab-scale experiments	• Apply proper control of DO during both nitrification & denitrification • Apply the BP-ANN model as a convenient & effective method for the prediction of N_2O emissions in an A/O WWTP
Blomberg et al. (2018) based on Kosonen et al. (2016)	A/O bioreactor	• Mechanistic model development	• Model describing the full-scale underground WWTP of Viikinmäki • AOB pathways: only NH_2OH oxidation included due to the dynamic & relatively high DO concentrations (1.5 –3.8 mg L^{-1}) in the aerated zones & the low NO_2^- concentrations (0.1 & 0.7 mg L^{-1})	• Improve the stripping modelling approach • Consider the nitrifier denitrification contribution in future model versions

Table 2 (*continued*)

Source	Process	Method	Main findings	Mitigation Measures
			• N_2O production mainly in the aerated zones, minor N_2O consumption & minor stripping effect in the anoxic zones • Applied stripping model: EF overestimation	
Chen et al. (2016)	CAS	• Fugacity model • Lab-scale in situ experiments	• Compared to other parameters (e.g. sludge concentration/retention time), the adjustment of the aeration rate effectively mitigated the GHG emission in the AS without significantly affecting the treated water quality • N_2O as main contributor to the total GHG emission (i.e. 57–91% of total GHG emission) • Lowering the aeration rate in the AS by 75% enabled decreasing the mass flux of N_2O by up to 53% • Most important benefit of changing the aeration rate: lower energy consumption during the WWTP operation (fractional contribution of pumping to the total emission from the WWTP = 46–93% within the range of the aeration rate tested)	• Reduce the aeration rate
Ribeiro et al., 2017	Extended aeration CAS	• Different aeration rates tested	• Nitrification as the main driving force behind N_2O emission peaks • Air flow-rate variations possibly influencing the N_2O emissions; high N_2O emissions under conditions of over-aeration or incomplete nitrification along with NO_2^- accumulation	• Add an anoxic zone & recirculation to a non-BNR system for nitrification; otherwise, high N_2O emissions expected in case of increased DO • Control the DO; dynamic changes in DO concentrations reported as being responsible for N_2O emission peaks in SND BNR systems • Control the TN (denitrification) • Avoid the concurrence of decreased DO & NO_2^- accumulation

to the 2nd step of the PF reactor (Table 2). However, it is unknown whether the suggested mitigation strategy was actually demonstrated in the system. Ahn et al. (2010b) identified dependencies between the WWTP operating conditions and the N_2O emissions via multiple linear regression. According to their findings, intermittent aeration or over-aeration must be avoided in aerobic reactors. Castellano-Hinojosa et al. (2018) linked the population of AOB, AOA and N_2O-reducers with the changes in the operational and environmental variables in 4 conventional AS systems with pre-denitrification zones. They observed that N_2O emissions mainly occurred due to incomplete denitrification, thus underlining the importance of ensuring the completion of the process for the emission mitigation. Overall, the main techniques for mitigating the N_2O emissions in wastewater treatment processes include: i) the application of the optimal aeration intensity and DO concentration, ii) preventing NH_4^+ concentration peaks (e.g. via equalisation tanks), iii) the avoidance of NO_2^- accumulation through proper control, and iv) the supply of additional carbon source (when required) to ensure complete denitrification in the anoxic reactors (Table 2).

However, studies applying and evaluating mitigation measures for long-term applications are still missing. Additionally, there is a gap between the data coming from the monitoring campaigns and their processing in order to establish a mitigation strategy. Moreover, several monitoring campaigns do not conclude on the development of mitigation strategies (Filali et al., 2013; Stenström et al., 2014; Yan et al., 2014). The long-term implementation and evaluation of the proposed mitigation strategies is still an issue. Therefore, the establishment of standardised methodological approaches for the identification of N_2O mitigation strategies is required.

5.2. N_2O mitigation strategies: progress and limitations

As shown in section 5.1, statistical and/or mechanistic modelling as well as observatory analysis of the N_2O emissions' behaviour have been commonly used either as standalone or in combined analyses for the development of mitigation strategies. However, the suggested mitigation schemes have not yet reached commercial applications at full-scale wastewater treatment processes. There is a gap between the identification of appropriate N_2O mitigation measures and their integration into the control of WWTPs. Future studies shall focus on the development, implementation and integration of the mitigation strategies into the existing control strategy of wastewater treatment processes. Special attention must be paid to trade-offs between GHG emissions, energy consumption, system performance and compliance with the legislative requirements in order to support evidence-based multi-objective optimisation of the WWTPs operation.

The investigation of direct GHG emissions at full-scale wastewater systems is important for the minimisation of the environmental footprint of WWTPs and the integration of the sustainability dimension into wastewater treatment process control.

5.3. Integrating the N_2O emission monitoring into the WWTP operation

Multivariable and multi-objective approaches have been proposed for the optimisation of the WWTPs performance to enable the correlation of energy consumption or operational costs with system performance (Qiao and Zhang, 2018; Zhang et al., 2014). Sweetapple et al. (2014) used a modified version of the benchmark simulation model 2 (BSM2) (Jeppsson et al., 2006) to identify

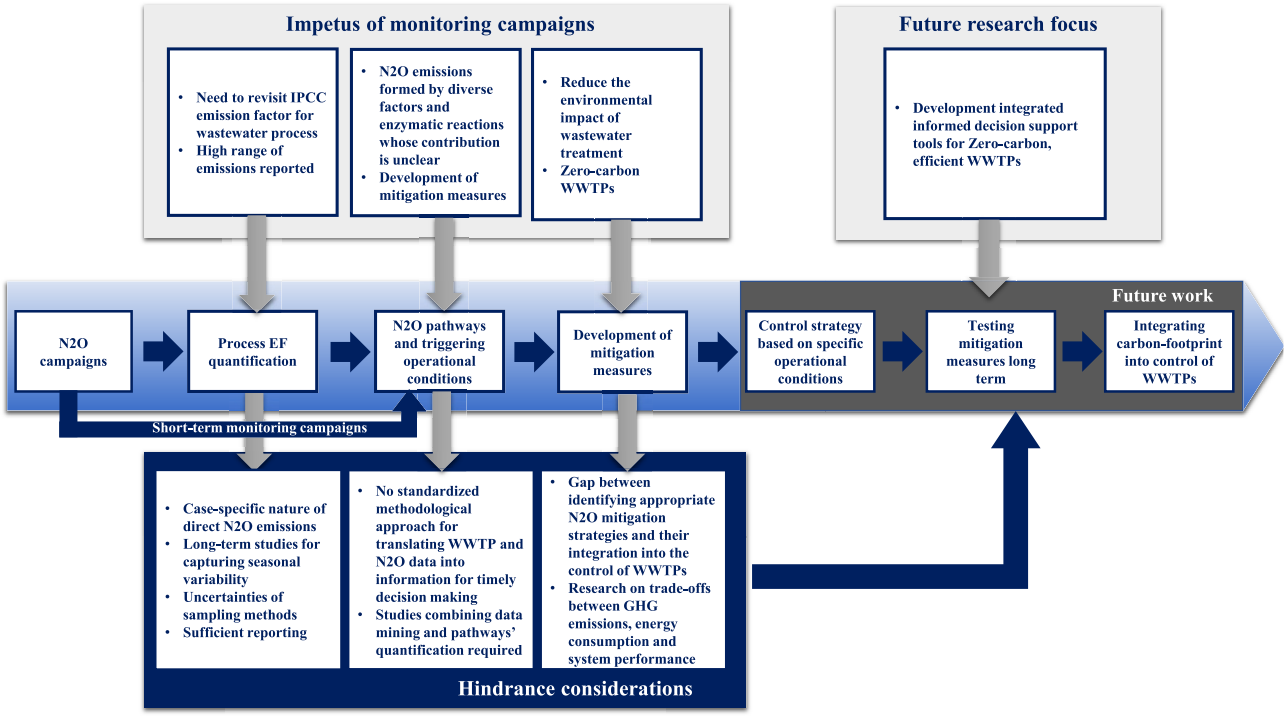

Fig. 7. Monitoring N_2O emissions in full-scale wastewater treatment systems - research priorities.

control strategies for the simultaneous minimisation of GHG emissions, operational costs and pollutant loads. The BSM2 models have also been combined with LCA to evaluate the sustainability of different operating strategies (Flores-Alsina et al., 2010; Arnell et al., 2017). However, studies utilising long-term real-field WWTP data are still scarce.

Fig. 7 summarises the research priorities in terms of real-field N_2O monitoring campaigns that can act as foundation for the integration of N_2O emissions into WWTP monitoring and control. Long-term monitoring campaigns that capture the seasonal variability, studies on the uncertainties of the sampling strategies and reporting of operational, environmental and sampling data can ensure the robustness and comparability of the monitoring results. The development of methodological approaches for the translation of WWTP data into information can facilitate the understanding of the N_2O emission behaviour and relationship with different operational conditions. Combination of mechanistic models and/or data mining techniques with methods for the N_2O quantification can validate the models and provide insights into the dominant pathways under changing operational conditions. Finally, research studies implementing and translating the N_2O mitigation measures into control strategies are essential.

6. Conclusions

A number of full-scale N_2O monitoring campaigns in WWTP were studied. The processes were classified so that the ranges of N_2O EFs, dominant N_2O pathways and triggering operational conditions, and the mitigation measures for different process groups could be set. The key conclusions of the current review are:

- There is a wide range of EFs within similar groups of wastewater treatment processes. The emission factor ranges between 2% and 5.6% of the influent N-load in mainstream SBR, while OD reactor types, exhibit have a low EF, ~0.14% of the N-load. Long-term continuous or discontinuous monitoring campaigns are characterised by higher EFs compared to short-term campaigns. The studies investigating the seasonal behaviour of N_2O emissions have an average EF equal to 1.7% of the N-load, whereas monitoring campaigns lasting less than a month have an average EF of 0.7%. Long-term campaigns show a high variability in the N_2O emissions. There is no specific EF correlation with the NH_4^+ removal in the mainstream processes or in specific groups of processes. Most of the processes in smaller WWTPs (i.e. flow-rate<200,000 $m^3\,d^{-1}$) had EFs less than 0.5% of the N-load, independently of the process type and nitrification efficiency. This study concluded that efficient operational strategies can mitigate the generated N_2O for different configurations and groups of processes.
- It is difficult to compare the results of the N_2O monitoring campaigns because of: i) the differences in the duration of the monitoring campaigns (e.g. short-term campaigns ignoring seasonal variations), ii) the uncertainty in the gaseous sampling methods and analytical measurements, and iii) the insufficient reporting regarding the reactor control strategy, operational and environmental conditions, etc.
- Simple feature extraction and graphical representations of selected process variables and N_2O emissions are employed to explain the N_2O triggering mechanisms. Given that a combination of several parameters affects the N_2O generation, multivariate statistical analysis techniques can be a useful alternative for analysing data and understanding the N_2O emission behaviour. The combined application of mechanistic models and statistical techniques can lead to better design of mitigation strategies.
- Isotopic and molecular biology analyses are emerging techniques that can qualitatively and quantitatively assess the N_2O generation pathways. Data mining methods can be deployed to identify patterns of operational conditions and N_2O emissions.

This can be complemented with techniques for the determination of the N_2O production pathways.

- Studies testing and validating the long-term full-scale N_2O mitigation measures are still missing.
- Future research should focus on: i) long-term N_2O monitoring campaigns, ii) the uncertainties of different sampling protocols, iii) the application of data mining approaches, machine learning and mechanistic models for the development of effective and adaptive models that will be integrated into WWTP operation and control, and iv) the development, implementation and integration of the mitigation strategies into the existing control strategies of WWTPs.

Declaration of interests

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

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Appendix A. Supplementary data

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References

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

Adouani, N., Limousy, L., Lendormi, T., Sire, O., 2015. N2O and NO emissions during wastewater denitrification step: influence of temperature on the biological process. In: Comptes Rendus Chim., International Chemical Engineering Congress (ICEC) 2013: from Fundamentals to Applied Chemistry and Biochemistry, vol. 18, pp. 15–22. https://doi.org/10.1016/j.crci.2014.11.005.

Ahn, J.H., Kim, S., Park, H., Katehis, D., Pagilla, K., Chandran, K., 2010. Spatial and temporal variability in atmospheric nitrous oxide generation and emission from full-scale biological nitrogen removal and non-BNR processes. Water Environ. Res. 82, 2362–2372. https://doi.org/10.2175/106143010X12681059116897.

Ahn, J.H., Kim, S., Park, H., Rahm, B., Pagilla, K., Chandran, K., 2010. N2O emissions from activated sludge processes, 2008–2009: results of a national monitoring survey in the United States. Environ. Sci. Technol. 44, 4505–4511. https://doi.org/10.1021/es903845y.

Aigner, W., Miksch, S., Müller, W., Schumann, H., Tominski, C., 2008. Visual methods for analyzing time-oriented data. IEEE Trans. Vis. Comput. Graph. 14, 47–60. https://doi.org/10.1109/TVCG.2007.70415.

Arnell, M., Rahmberg, M., Oliveira, F., Jeppsson, U., 2017. Multi-objective performance assessment of wastewater treatment plants combining plant-wide process models and life cycle assessment. J. Water Clim. Change 8, 715–729. https://doi.org/10.2166/wcc.2017.179.

Baresel, C., Andersson, S., Yang, J., Andersen, M.H., 2016. Comparison of nitrous oxide (N2O) emissions calculations at a Swedish wastewater treatment plant based on water concentrations versus off-gas concentrations. Adv. Clim. Change Res. 7, 185–191. Including special topic on atmospheric black carbon and its effects on cryosphere. https://doi.org/10.1016/j.accre.2016.09.001.

Batstone, D.J., Hülsen, T., Mehta, C.M., Keller, J., 2015. Platforms for energy and nutrient recovery from domestic wastewater: a review. Chemosphere 140, 2–11.

Bellandi, G., Porro, J., Senesi, E., Caretti, C., Caffaz, S., Weijers, S., Nopens, I., Gori, R., 2018. Multi-point monitoring of nitrous oxide emissions in three full-scale conventional activated sludge tanks in Europe. Water Sci. Technol. 77, 880–890. https://doi.org/10.2166/wst.2017.560.

Blomberg, K., Kosse, P., Mikola, A., Kuokkanen, A., Fred, T., Heinonen, M., Mulas, M., Lübken, M., Wichern, M., Vahala, R., 2018. Development of an extended ASM3 model for predicting the nitrous oxide emissions in a full-scale wastewater treatment plant. Environ. Sci. Technol. 52, 5803–5811. https://doi.org/10.1021/acs.est.8b00386.

Bollon, J., Filali, A., Fayolle, Y., Guerin, S., Rocher, V., Gillot, S., 2016a. Full-scale post denitrifying biofilters: sinks of dissolved N2O? Sci. Total Environ. 563–564, 320–328. https://doi.org/10.1016/j.scitotenv.2016.03.237.

Bollon, J., Filali, A., Fayolle, Y., Guerin, S., Rocher, V., Gillot, S., 2016b. N2O emissions from full-scale nitrifying biofilters. Water Res. 102, 41–51. https://doi.org/10.1016/j.watres.2016.05.091.

Brotto, A.C., Kligerman, D.C., Andrade, S.A., Ribeiro, R.P., Oliveira, J.L.M., Chandran, K., Mello, W.Z. de, 2015. Factors controlling nitrous oxide emissions from a full-scale activated sludge system in the tropics. Environ. Sci. Pollut. Res. 22, 11840–11849. https://doi.org/10.1007/s11356-015-4467-x.

Cadwallader, A., VanBriesen, J.M., 2017. Incorporating uncertainty into future estimates of nitrous oxide emissions from wastewater treatment. J. Environ. Eng. 143, 04017029. https://doi.org/10.1061/(ASCE)EE.1943-7870.0001231.

Caivano, M., Bellandi, G., Mancini, I.M., Masi, S., Brienza, R., Panariello, S., Gori, R., Caniani, D., 2017. Monitoring the aeration efficiency and carbon footprint of a medium-sized WWTP: experimental results on oxidation tank and aerobic digester. Environ. Technol. 38, 629–638. https://doi.org/10.1080/09593330.2016.1205150.

Caranto, J.D., Vilbert, A.C., Lancaster, K.M., 2016. Nitrosomonas europaea cytochrome P460 is a direct link between nitrification and nitrous oxide emission. Proc. Natl. Acad. Sci. 113, 14704–14709. https://doi.org/10.1073/pnas.1611051113.

Castellano-Hinojosa, A., Maza-Márquez, P., Melero-Rubio, Y., González-López, J., Rodelas, B., 2018. Linking nitrous oxide emissions to population dynamics of nitrifying and denitrifying prokaryotes in four full-scale wastewater treatment plants. Chemosphere 200, 57–66. https://doi.org/10.1016/j.chemosphere.2018.02.102.

Castro-Barros, C.M., Daelman, M.R.J., Mampaey, K.E., van Loosdrecht, M.C.M., Volcke, E.I.P., 2015. Effect of aeration regime on N2O emission from partial nitritation-anammox in a full-scale granular sludge reactor. Water Res. 68, 793–803. https://doi.org/10.1016/j.watres.2014.10.056.

Chen, W.-H., Yang, J.-H., Yuan, C.-S., Yang, Y.-H., 2016. Toward better understanding and feasibility of controlling greenhouse gas emissions from treatment of industrial wastewater with activated sludge. Environ. Sci. Pollut. Res. 23, 20449–20461. https://doi.org/10.1007/s11356-016-7183-2.

Conthe, M., Parchen, C., Stouten, G., Kleerebezem, R., van Loosdrecht, M.C.M., 2018. O2 versus N2O respiration in a continuous microbial enrichment. Appl. Microbiol. Biotechnol. 102, 8943–8950. https://doi.org/10.1007/s00253-018-9247-3.

Cornejo, P.K., Zhang, Q., Mihelcic, J.R., 2016. How does scale of implementation impact the environmental sustainability of wastewater treatment integrated with resource recovery? Environ. Sci. Technol. 50, 6680–6689. https://doi.org/10.1021/acs.est.5b05055.

Corominas, L., Garrido-Baserba, M., Villez, K., Olsson, G., Cortés, U., Poch, M., 2018. Transforming data into knowledge for improved wastewater treatment operation: a critical review of techniques. Environ. Model. Softw. 106, 89–103. Special Issue on Environmental Data Science. Applications to Air quality and Water cycle. https://doi.org/10.1016/j.envsoft.2017.11.023.

Daelman, M.R., De Baets, B., van Loosdrecht, M.C., Volcke, E.I., 2013. Influence of sampling strategies on the estimated nitrous oxide emission from wastewater treatment plants. Water Res. 47, 3120–3130.

Daelman, M.R.J., van Voorthuizen, E.M., Van Dongen, L., Volcke, E.I.P., Van Loosdrecht, M.C.M., 2013. Methane and nitrous oxide emissions from municipal wastewater treatment–results from a long-term study. Water Sci. Technol. 67, 2350–2355.

Daelman, M.R.J., van Voorthuizen, E.M., van Dongen, U.G.J.M., Volcke, E.I.P., van Loosdrecht, M.C.M., 2015. Seasonal and diurnal variability of N2O emissions from a full-scale municipal wastewater treatment plant. Sci. Total Environ. 536, 1–11. https://doi.org/10.1016/j.scitotenv.2015.06.122.

Daniel, J.S., Fleming, E.L., Portmann, R.W., Velders, G.J.M., Jackman, C.H., Ravishankara, A.R., 2010. Options to accelerate ozone recovery: ozone and climate benefits. Atmos. Chem. Phys. 10, 7697–7707. https://doi.org/10.5194/acp-10-7697-2010.

Desloover, J., De Clippeleir, H., Boeckx, P., Du Laing, G., Colsen, J., Verstraete, W., Vlaeminck, S.E., 2011. Floc-based sequential partial nitritation and anammox at full scale with contrasting N2O emissions. Water Res. 45, 2811–2821. https://doi.org/10.1016/j.watres.2011.02.028.

Desloover, J., Vlaeminck, S.E., Clauwaert, P., Verstraete, W., Boon, N., 2012. Strategies to mitigate N2O emissions from biological nitrogen removal systems. Curr. Opin. Biotechnol. 23, 474–482. https://doi.org/10.1016/j.copbio.2011.12.030.

Ding, X., Zhao, J., Hu, B., Li, X., Ge, G., Gao, K., Chen, Y., 2017. Mathematical modeling of nitrous oxide (N2O) production in anaerobic/anoxic/oxic processes: improvements to published N2O models. Chem. Eng. J. 325, 386–395. https://doi.org/10.1016/j.cej.2017.05.082.

Domingo-Félez, C., Pellicer-Nàcher, C., Petersen, M.S., Jensen, M.M., Plósz, B.G., Smets, B.F., 2017. Heterotrophs are key contributors to nitrous oxide production in activated sludge under low C-to-N ratios during nitrification-Batch experiments and modeling. Biotechnol. Bioeng. 114, 132–140. https://doi.org/10.1002/bit.26062.

Domingo-Félez, C., Smets, F.B., 2016. A consilience model to describe N 2 O production during biological N removal. Environ. Sci. Water Res. Technol. 2, 923–930. https://doi.org/10.1039/C6EW00179C.

Duan, H., Ye, L., Erler, D., Ni, B.-J., Yuan, Z., 2017. Quantifying nitrous oxide production pathways in wastewater treatment systems using isotope technology – a critical review. Water Res. 122, 96–113. https://doi.org/10.1016/j.watres.2017.05.054.

Duchemin, E., Lucotte, M., Canuel, R., 1999. Comparison of static chamber and thin

boundary layer equation methods for measuring greenhouse gas emissions from large water bodies§. Environ. Sci. Technol. 33, 350–357. https://doi.org/10.1021/es9800840.

Fang, F., Ni, B.-J., Xie, W.-M., Sheng, G.-P., Liu, S.-G., Tong, Z.-H., Yu, H.-Q., 2010. An integrated dynamic model for simulating a full-scale municipal wastewater treatment plant under fluctuating conditions. Chem. Eng. J. 160, 522–529. https://doi.org/10.1016/j.cej.2010.03.063.

Filali, A., Fayolle, Y., Peu, P., Philippe, L., Nauleau, F., Gillot, S., 2013. Aeration control in a full-scale activated sludge wastewater treatment plant: impact on performances, energy consumption and N2O emission. In: Presented at the 11ème Conférence IWA sur l'instrumentation, le contrôle et l'automatisation. ICA2013, p. 4 pp.

Flores-Alsina, X., Arnell, M., Amerlinck, Y., Corominas, L., Gernaey, K.V., Guo, L., Lindblom, E., Nopens, I., Porro, J., Shaw, A., Snip, L., Vanrolleghem, P.A., Jeppsson, U., 2014. Balancing effluent quality, economic cost and greenhouse gas emissions during the evaluation of (plant-wide) control/operational strategies in WWTPs. Sci. Total Environ. 466–467, 616–624. https://doi.org/10.1016/j.scitotenv.2013.07.046.

Flores-Alsina, X., Gallego, A., Feijoo, G., Rodriguez-Roda, I., 2010. Multiple-objective evaluation of wastewater treatment plant control alternatives. J. Environ. Manag. 91, 1193–1201. https://doi.org/10.1016/j.jenvman.2010.01.009.

Flowers, J.J., Cadkin, T.A., McMahon, K.D., 2013. Seasonal bacterial community dynamics in a full-scale enhanced biological phosphorus removal plant. Water Res. 47, 7019–7031. https://doi.org/10.1016/j.watres.2013.07.054.

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

Foley, J., Yuan, Z., Keller, J., Senante, E., Chandran, K., Willis, J., Shah, A., van Loosdrecht, M.C.M., van Voorthuizen, E., 2015. N2O and CH4 Emission from Wastewater Collection and Treatment Systems: State of the Science Report and Technical Report. IWA Publishing.

Frutos, O.D., Quijano, G., Aizpuru, A., Muñoz, R., 2018. A state-of-the-art review on nitrous oxide control from waste treatment and industrial sources. Biotechnol. Adv. 36, 1025–1037. https://doi.org/10.1016/j.biotechadv.2018.03.004.

Ghoneim, W.A.M., Helal, A.A., Wahab, M.A., 2016. Minimizing energy consumption in wastewater treatment plants. In: Renewable Energies for Developing Countries (REDEC), 2016 3rd International Conference on. IEEE, pp. 1–8.

Guo, G., Wang, Y., Hao, T., Wu, D., Chen, G.-H., 2017. Enzymatic nitrous oxide emissions from wastewater treatment. Front. Environ. Sci. Eng. 12, 10. https://doi.org/10.1007/s11783-018-1021-3.

Guo, L., Vanrolleghem, P.A., 2014. Calibration and validation of an activated sludge model for greenhouse gases no. 1 (ASMG1): prediction of temperature-dependent N<Subscript>2</Subscript>O emission dynamics. Bioproc. Biosyst. Eng. 37, 151–163. https://doi.org/10.1007/s00449-013-0978-3.

Gustavsson, D.J.I., la Cour Jansen, J., 2011. Dynamics of nitrogen oxides emission from a full-scale sludge liquor treatment plant with nitritation. Water Sci. Technol. J. Int. Assoc. Water Pollut. Res. 63, 2838–2845.

Hadjimichael, A., Comas, J., Corominas, L., 2016. Do machine learning methods used in data mining enhance the potential of decision support systems? A review for the urban water sector. AI Commun. 29, 747–756. https://doi.org/10.3233/AIC-160714.

Haimi, H., Mulas, M., Corona, F., Vahala, R., 2013. Data-derived soft-sensors for biological wastewater treatment plants: an overview. Environ. Model. Softw 47, 88–107. https://doi.org/10.1016/j.envsoft.2013.05.009.

Harper, W.F., Takeuchi, Y., Riya, S., Hosomi, M., Terada, A., 2015. Novel abiotic reactions increase nitrous oxide production during partial nitrification: modeling and experiments. Chem. Eng. J. 281, 1017–1023. https://doi.org/10.1016/j.cej.2015.06.109.

Harris, E., Joss, A., Emmenegger, L., Kipf, M., Wolf, B., Mohn, J., Wunderlin, P., 2015. Isotopic evidence for nitrous oxide production pathways in a partial nitritation-anammox reactor. Water Res. 83, 258–270. https://doi.org/10.1016/j.watres.2015.06.040.

Henderson, S.L., Dandie, C.E., Patten, C.L., Zebarth, B.J., Burton, D.L., Trevors, J.T., Goyer, C., 2010. Changes in denitrifier abundance, denitrification gene mRNA levels, nitrous oxide emissions, and denitrification in anoxic soil microcosms amended with glucose and plant residues. Appl. Environ. Microbiol. 76, 2155–2164. https://doi.org/10.1128/AEM.02993-09.

Henze, M., Grady Jr., C.L., Gujer, W., Marais, G., Matsuo, T., 1987. Activated Sludge Model No. 1. IAWQ Scientific and Technical Report No. 1 IAWQ, Lond. UK.

Henze, M., Gujer, W., Mino, T., Van Loosdrecht, M.C.M., 2000. Activated Sludge Models ASM1, ASM2, ASM2d and ASM3. IWA publishing.

Hiatt, W.C., Grady, C.P.L., 2008. An Updated Process Model for Carbon Oxidation, Nitrification, and Denitrification [WWW Document]. https://doi.org/10.2175/106143008X304776.

Hochstein, L.I., Tomlinson, G.A., 1988. The enzymes associated with denitrification. Annu. Rev. Microbiol. 42, 231–261. https://doi.org/10.1146/annurev.mi.42.100188.001311.

Hooper, A.B., 1968. A nitrite-reducing enzyme from Nitrosomonas europaea Preliminary characterization with hydroxylamine as electron donor. Biochim. Biophys. Acta BBA - Bioenerg. 162, 49–65. https://doi.org/10.1016/0005-2728(68)90213-2.

Hooper, A.B., Terry, K.R., 1979. Hydroxylamine oxidoreductase of Nitrosomonas: production of nitric oxide from hydroxylamine. Biochim. Biophys. Acta Enzymol. 571, 12–20. https://doi.org/10.1016/0005-2744(79)90220-1.

IPCC, 2013. The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, USA.

Ishii, S., Song, Y., Rathnayake, L., Tumendelger, A., Satoh, H., Toyoda, S., Yoshida, N., Okabe, S., 2014. Identification of key nitrous oxide production pathways in aerobic partial nitrifying granules. Environ. Microbiol. 16, 3168–3180. https://doi.org/10.1111/1462-2920.12458.

Jeppsson, U., Rosen, C., Alex, J., Copp, J., Gernaey, K.V., Pons, M.N., Vanrolleghem, P.A., 2006. Towards a benchmark simulation model for plant-wide control strategy performance evaluation of WWTPs. Water Sci. Technol. J. Int. Assoc. Water Pollut. Res. 53, 287–295.

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

Kampschreur, M.J., Poldermans, R., Kleerebezem, R., van der Star, W.R.L., Haarhuis, R., Abma, W.R., Jetten, M.S.M., van Loosdrecht, M.C.M., 2009a. Emission of nitrous oxide and nitric oxide from a full-scale single-stage nitritation-anammox reactor. Water Sci. Technol. 60, 3211–3217. https://doi.org/10.2166/wst.2009.608.

Kampschreur, M.J., Temmink, H., Kleerebezem, R., Jetten, M.S.M., van Loosdrecht, M.C.M., 2009b. Nitrous oxide emission during wastewater treatment. Water Res. 43, 4093–4103. https://doi.org/10.1016/j.watres.2009.03.001.

Kampschreur, M.J., van der Star, W.R.L., Wielders, H.A., Mulder, J.W., Jetten, M.S.M., van Loosdrecht, M.C.M., 2008. Dynamics of nitric oxide and nitrous oxide emission during full-scale reject water treatment. Water Res. 42, 812–826. https://doi.org/10.1016/j.watres.2007.08.022.

Keskitalo, J., Leiviskä, K., 2015. Artificial neural network ensembles in hybrid modelling of activated sludge plant. In: Angelov, P., Atanassov, K.T., Doukovska, L., Hadjiski, M., Jotsov, V., Kacprzyk, J., Kasabov, N., Sotirov, S., Szmidt, E., Zadrożny, S. (Eds.), Intelligent Systems'2014, Advances in Intelligent Systems and Computing. Springer International Publishing, pp. 683–694.

Kosonen, H., Heinonen, M., Mikola, A., Haimi, H., Mulas, M., Corona, F., Vahala, R., 2016. Nitrous oxide production at a fully covered wastewater treatment plant: results of a long-term online monitoring campaign. Environ. Sci. Technol. 50, 5547–5554. https://doi.org/10.1021/acs.est.5b04466.

Lane, J.L., de Haas, D.W., Lant, P.A., 2015. The diverse environmental burden of city-scale urban water systems. Water Res. 81, 398–415. https://doi.org/10.1016/j.watres.2015.03.005.

Law, Y., Ye, L., Pan, Y., Yuan, Z., 2012. Nitrous oxide emissions from wastewater treatment processes. Phil. Trans. R. Soc. B 367, 1265–1277. https://doi.org/10.1098/rstb.2011.0317.

Li, H., Peng, D., Liu, W., Wei, J., Wang, Z., Wang, B., 2016. N_2O generation and emission from two biological nitrogen removal plants in China. Desalination Water Treat 57, 11800–11806. https://doi.org/10.1080/19443994.2015.1046145.

Lorke, A., Bodmer, P., Noss, C., Alshboul, Z., Koschorreck, M., Somlai-Haase, C., Bastviken, D., Flury, S., Mcginnis, D.F., Maeck, A., Müller, D., Premke, K., 2015. Technical note: drifting versus anchored flux chambers for measuring greenhouse gas emissions from running waters. Biogeosciences 12, 7013–7024.

Ma, C., Jensen, M.M., Smets, B.F., Thamdrup, B., 2017. Pathways and controls of N2O production in nitritation–anammox biomass. Environ. Sci. Technol. 51, 8981–8991. https://doi.org/10.1021/acs.est.7b01225.

Mampaey, K.E., Beuckels, B., Kampschreur, M.J., Kleerebezem, R., van Loosdrecht, M.C. m., Volcke, E.I. p., 2013. Modelling nitrous and nitric oxide emissions by autotrophic ammonia-oxidizing bacteria. Environ. Technol. 34, 1555–1566. https://doi.org/10.1080/09593330.2012.758666.

Mampaey, K.E., De Kreuk, M.K., van Dongen, U.G.J.M., van Loosdrecht, M.C.M., Volcke, E.I.P., 2016. Identifying N2O formation and emissions from a full-scale partial nitritation reactor. Water Res. 88, 575–585. https://doi.org/10.1016/j.watres.2015.10.047.

Mannina, G., Ekama, G., Caniani, D., Cosenza, A., Esposito, G., Gori, R., Garrido-Baserba, M., Rosso, D., Olsson, G., 2016. Greenhouse gases from wastewater treatment — a review of modelling tools. Sci. Total Environ. 551–552, 254–270. https://doi.org/10.1016/j.scitotenv.2016.01.163.

Massara, T.M., Malamis, S., Guisasola, A., Baeza, J.A., Noutsopoulos, C., Katsou, E., 2017. A review on nitrous oxide (N 2 O) emissions during biological nutrient removal from municipal wastewater and sludge reject water. Sci. Total Environ. 596, 106–123.

Massara, T.M., Solís, B., Guisasola, A., Katsou, E., Baeza, J.A., 2018. Development of an ASM2d-N2O model to describe nitrous oxide emissions in municipal WWTPs under dynamic conditions. Chem. Eng. J. 335, 185–196.

Matthews, C.J.D., StLouis, V.L., Hesslein, R.H., 2003. Comparison of three techniques used to measure diffusive gas exchange from sheltered aquatic surfaces. Environ. Sci. Technol. 37, 772–780. https://doi.org/10.1021/es0205838.

Mello, W.Z. de, Ribeiro, R.P., Brotto, A.C., Kligerman, D.C., Piccoli, A. de S., Oliveira, J.L.M., 2013. Nitrous oxide emissions from an intermittent aeration activated sludge system of an urban wastewater treatment plant. Quím. Nova 36, 16–20. https://doi.org/10.1590/S0100-40422013000100004.

Ni, B.-J., Yuan, Z., 2015. Recent advances in mathematical modeling of nitrous oxides emissions from wastewater treatment processes. Water Res. 87, 336–346. https://doi.org/10.1016/j.watres.2015.09.049.

Ni, B.-J., Pan, Y., van den Akker, B., Ye, L., Yuan, Z., 2015. Full-scale modeling explaining large spatial variations of nitrous oxide fluxes in a step-feed plug-flow wastewater treatment reactor. Environ. Sci. Technol. 49, 9176–9184. https://doi.org/10.1021/acs.est.5b02038.

Ni, B.-J., Peng, L., Law, Y., Guo, J., Yuan, Z., 2014. Modeling of nitrous oxide

production by autotrophic ammonia-oxidizing bacteria with multiple production pathways. Environ. Sci. Technol. 48, 3916–3924. https://doi.org/10.1021/es405592h.
Ni, B.-J., Ye, L., Law, Y., Byers, C., Yuan, Z., 2013. Mathematical modeling of nitrous oxide (N2O) emissions from full-scale wastewater treatment plants. Environ. Sci. Technol. 47, 7795–7803. https://doi.org/10.1021/es4005398.
Palut, M.P.J., Canziani, O.F., 2007. Contribution of working group II to the fourth assessment report of the intergovernmental panel on climate change. Cambridge University Press.
Pan, Y., van den Akker, B., Ye, L., Ni, B.-J., Watts, S., Reid, K., Yuan, Z., 2016. Unravelling the spatial variation of nitrous oxide emissions from a step-feed plug-flow full scale wastewater treatment plant. Sci. Rep. 6. https://doi.org/10.1038/srep20792.
Peng, L., Ni, B.-J., Erler, D., Ye, L., Yuan, Z., 2014. The effect of dissolved oxygen on N2O production by ammonia-oxidizing bacteria in an enriched nitrifying sludge. Water Res. 66, 12–21. https://doi.org/10.1016/j.watres.2014.08.009.
Pijuan, M., Torà, J., Rodríguez-Caballero, A., César, E., Carrera, J., Pérez, J., 2014. Effect of process parameters and operational mode on nitrous oxide emissions from a nitritation reactor treating reject wastewater. Water Res. 49, 23–33. https://doi.org/10.1016/j.watres.2013.11.009.
Portmann, R.W., Daniel, J.S., Ravishankara, A.R., 2012. Stratospheric ozone depletion due to nitrous oxide: influences of other gases. Phil. Trans. R. Soc. B 367, 1256–1264. https://doi.org/10.1098/rstb.2011.0377.
Poth, M., Focht, D.D., 1985. 15N kinetic analysis of N2O production by nitrosomonas europaea: an examination of nitrifier denitrification. Appl. Environ. Microbiol. 49, 1134–1141.
Qiao, J., Zhang, W., 2018. Dynamic multi-objective optimization control for wastewater treatment process. Neural Comput. Appl. 29, 1261–1271. https://doi.org/10.1007/s00521-016-2642-8.
Rathnayake, R.M., Oshiki, M., Ishii, S., Segawa, T., Satoh, H., Okabe, S., 2015. Effects of dissolved oxygen and pH on nitrous oxide production rates in autotrophic partial nitrification granules. Bioresour. Technol. 197, 15–22.
Reino, C., van Loosdrecht, M.C.M., Carrera, J., Pérez, J., 2017. Effect of temperature on N2O emissions from a highly enriched nitrifying granular sludge performing partial nitritation of a low-strength wastewater. Chemosphere 185, 336–343. https://doi.org/10.1016/j.chemosphere.2017.07.017.
Ren, Y. g., Wang, J. h., Li, H. f., Zhang, J., Qi, P. y., Hu, Z., 2013. Nitrous oxide and methane emissions from different treatment processes in full-scale municipal wastewater treatment plants. Environ. Technol. 34, 2917–2927. https://doi.org/10.1080/09593330.2012.696717.
Ribeiro, R.P., Bueno, R.F., Piveli, R.P., Kligerman, D.C., de Mello, W.Z., Oliveira, J.L.M., 2017. The response of nitrous oxide emissions to different operating conditions in activated sludge wastewater treatment plants in Southeastern Brazil. Water Sci. Technol 76, 2337–2349. https://doi.org/10.2166/wst.2017.399.
Ritchie, G. a. F., Nicholas, D.J.D., 1972. Identification of the sources of nitrous oxide produced by oxidative and reductive processes in Nitrosomonas europaea. Biochem. J. 126, 1181–1191. https://doi.org/10.1042/bj1261181.
Robertson, G.P., Tiedje, J.M., 1987. Nitrous oxide sources in aerobic soils: nitrification, denitrification and other biological processes. Soil Biol. Biochem. 19, 187–193. https://doi.org/10.1016/0038-0717(87)90080-0.
Rodriguez-Caballero, A., Aymerich, I., Marques, R., Poch, M., Pijuan, M., 2015. Minimizing N2O emissions and carbon footprint on a full-scale activated sludge sequencing batch reactor. Water Res. 71, 1–10. https://doi.org/10.1016/j.watres.2014.12.032.
Rodriguez-Caballero, A., Aymerich, I., Poch, M., Pijuan, M., 2014. Evaluation of process conditions triggering emissions of green-house gases from a biological wastewater treatment system. Sci. Total Environ. 493, 384–391. https://doi.org/10.1016/j.scitotenv.2014.06.015.
Samuelsson, J., Delre, A., Tumlin, S., Hadi, S., Offerle, B., Scheutz, C., 2018. Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Water Res. 131, 299–309. https://doi.org/10.1016/j.watres.2017.12.018.
Schaubroeck, T., De Clippeleir, H., Weissenbacher, N., Dewulf, J., Boeckx, P., Vlaeminck, S.E., Wett, B., 2015. Environmental sustainability of an energy self-sufficient sewage treatment plant: improvements through DEMON and co-digestion. Water Res. 74, 166–179. https://doi.org/10.1016/j.watres.2015.02.013.
Schulthess, R.V., Gujer, W., 1996. Release of nitrous oxide (N2O) from denitrifying activated sludge: verification and application of a mathematical model. Water Res. 30, 521–530. https://doi.org/10.1016/0043-1354(95)00204-9.
Shurkhovetskyy, G., Andrienko, N., Andrienko, G., Fuchs, G., 2018. Data abstraction for visualizing large time series. Comput. Graph. Forum 37, 125–144. https://doi.org/10.1111/cgf.13237.
Singh, D.P., Maurya, N.S., 2016. A comparative study of the various methodologies for estimation of green house gas emission from wastewater treatment systems (A review). Orient. J. Chem. 32, 1373–1380.
Soler-Jofra, A., Stevens, B., Hoekstra, M., Picioreanu, C., Sorokin, D., van Loosdrecht, M.C.M., Pérez, J., 2016. Importance of abiotic hydroxylamine conversion on nitrous oxide emissions during nitritation of reject water. Chem. Eng. J. 287, 720–726. https://doi.org/10.1016/j.cej.2015.11.073.
Song, K., Suenaga, T., Hamamoto, A., Satou, K., Riya, S., Hosomi, M., Terada, A., 2014. Abundance, transcription levels and phylogeny of bacteria capable of nitrous oxide reduction in a municipal wastewater treatment plant. J. Biosci. Bioeng. 118, 289–297. https://doi.org/10.1016/j.jbiosc.2014.02.028.
Spérandio, M., Pocquet, M., Guo, L., Ni, B.-J., Vanrolleghem, P.A., Yuan, Z., 2016. Evaluation of different nitrous oxide production models with four continuous long-term wastewater treatment process data series. Bioproc. Biosyst. Eng. 39, 493–510. https://doi.org/10.1007/s00449-015-1532-2.
Spinelli, M., Eusebi, A.L., Vasilaki, V., Katsou, E., Frison, N., Cingolani, D., Fatone, F., 2018. Critical analyses of nitrous oxide emissions in a full scale activated sludge system treating low carbon-to-nitrogen ratio wastewater. J. Clean. Prod. 190, 517–524. https://doi.org/10.1016/j.jclepro.2018.04.178.
Stenström, F., Tjus, K., Jansen, J. la C., 2014. Oxygen-induced dynamics of nitrous oxide in water and off-gas during the treatment of digester supernatant. Water Sci. Technol. 69, 84–91. https://doi.org/10.2166/wst.2013.558.
Sun, S., Bao, Z., Li, R., Sun, D., Geng, H., Huang, X., Lin, J., Zhang, P., Ma, R., Fang, L., Zhang, X., Zhao, X., 2017. Reduction and prediction of N2O emission from an Anoxic/Oxic wastewater treatment plant upon DO control and model simulation. Bioresour. Technol. 244, 800–809. https://doi.org/10.1016/j.biortech.2017.08.054.
Sun, S., Bao, Z., Sun, D., 2015. Study on emission characteristics and reduction strategy of nitrous oxide during wastewater treatment by different processes. Environ. Sci. Pollut. Res. 22, 4222–4229. https://doi.org/10.1007/s11356-014-3654-5.
Sun, S., Cheng, X., Sun, D., 2013. Emission of N2O from a full-scale sequencing batch reactor wastewater treatment plant: characteristics and influencing factors. Int. Biodeterior. Biodegrad. 85, 545–549. https://doi.org/10.1016/j.ibiod.2013.03.034.
Sun, Y., Guan, Y., Pan, M., Zhan, X., Hu, Z., Wu, G., 2017. Enhanced biological nitrogen removal and N2O emission characteristics of the intermittent aeration activated sludge process. Rev. Environ. Sci. Biotechnol. 16, 761–780. https://doi.org/10.1007/s11157-017-9444-z.
Sweetapple, C., Fu, G., Butler, D., 2014. Multi-objective optimisation of wastewater treatment plant control to reduce greenhouse gas emissions. Water Res. 55, 52–62. https://doi.org/10.1016/j.watres.2014.02.018.
Terada, A., Sugawara, S., Hojo, K., Takeuchi, Y., Riya, S., Harper, W.F., Yamamoto, T., Kuroiwa, M., Isobe, K., Katsuyama, C., Suwa, Y., Koba, K., Hosomi, M., 2017. Hybrid nitrous oxide production from a partial nitrifying bioreactor: hydroxylamine interactions with nitrite. Environ. Sci. Technol. 51, 2748–2756. https://doi.org/10.1021/acs.est.6b05521.
Townsend-Small, A., Pataki, D.E., Tseng, L.Y., Tsai, C.-Y., Rosso, D., 2011. Nitrous oxide emissions from wastewater treatment and water reclamation plants in southern California. J. Environ. Qual. 40, 1542–1550. https://doi.org/10.2134/jeq2011.0059.
Toyoda, S., Suzuki, Y., Hattori, S., Yamada, K., Fujii, A., Yoshida, N., Kouno, R., Murayama, K., Shiomi, H., 2011. Isotopomer analysis of production and consumption mechanisms of N2O and CH4 in an advanced wastewater treatment system. Environ. Sci. Technol. 45, 917–922. https://doi.org/10.1021/es102985u.
Tumendelger, A., Toyoda, S., Yoshida, N., 2014. Isotopic analysis of N2O produced in a conventional wastewater treatment system operated under different aeration conditions. Rapid Commun. Mass Spectrom. 28, 1883–1892. https://doi.org/10.1002/rcm.6973.
US EPA, O., 2016. U.S. Greenhouse Gas Inventory Report: 1990-2014 [WWW Document] accessed 1.25.17. https://www.epa.gov/ghgemissions/us-greenhouse-gas-inventory-report-1990-2014.
Vachon, D., Prairie, Y.T., Cole, J.J., 2010. The relationship between near-surface turbulence and gas transfer velocity in freshwater systems and its implications for floating chamber measurements of gas exchange. Limnol. Oceanogr. 55, 1723–1732. https://doi.org/10.4319/lo.2010.55.4.1723.
Van Hulle, S.W.H., Vandeweyer, H.J.P., Meesschaert, B.D., Vanrolleghem, P.A., Dejans, P., Dumoulin, A., 2010. Engineering aspects and practical application of autotrophic nitrogen removal from nitrogen rich streams. Chem. Eng. J. 162, 1–20. https://doi.org/10.1016/j.cej.2010.05.037.
van Loosdrecht, M.C., Nielsen, P.H., Lopez-Vazquez, C.M., Brdjanovic, D., 2016. Experimental Methods in Wastewater Treatment. IWA Publishing.
Vanrolleghem, P.A., Lee, D.S., 2003. On-line monitoring equipment for wastewater treatment processes: state of the art. Water Sci. Technol. J. Int. Assoc. Water Pollut. Res. 47, 1–34.
Vasilaki, V., Volcke, E.I.P., Nandi, A.K., van Loosdrecht, M.C.M., Katsou, E., 2018. Relating N2O emissions during biological nitrogen removal with operating conditions using multivariate statistical techniques. Water Res. 140, 387–402. https://doi.org/10.1016/j.watres.2018.04.052.
Wan, X., Baeten, J.E., Volcke, E.I.P., 2019. Effect of operating conditions on N2O emissions from one-stage partial nitritation-anammox reactors. Biochem. Eng. J. 143, 24–33. https://doi.org/10.1016/j.bej.2018.12.004.
Wang, J., Ma, Y., Zhang, L., Gao, R.X., Wu, D., 2018. Deep learning for smart manufacturing: methods and applications. J. Manuf. Syst. Special Issue Smart Manuf. 48, 144–156. https://doi.org/10.1016/j.jmsy.2018.01.003.
Wang, Jinhe, Zhang, J., Wang, Jing, Qi, P., Ren, Y., Hu, Z., 2011. Nitrous oxide emissions from a typical northern Chinese municipal wastewater treatment plant. Desalination Water Treat. 32, 145–152. https://doi.org/10.5004/dwt.2011.2691.
Wang, Y., Fang, H., Zhou, D., Han, H., Chen, J., 2016a. Characterization of nitrous oxide and nitric oxide emissions from a full-scale biological aerated filter for secondary nitrification. Chem. Eng. J. 299, 304–313. https://doi.org/10.1016/j.cej.2016.04.050.
Wang, Y., Lin, X., Zhou, D., Ye, L., Han, H., Song, C., 2016b. Nitric oxide and nitrous oxide emissions from a full-scale activated sludge anaerobic/anoxic/oxic process. Chem. Eng. J. 289, 330–340. https://doi.org/10.1016/j.cej.2015.12.074.
Weissenbacher, N., Takacs, I., Murthy, S., Fuerhacker, M., Wett, B., 2010. Gaseous nitrogen and carbon emissions from a full-scale deammonification plant. Water Environ. Res. 82, 169–175. https://doi.org/10.2175/106143009X447867.

White, C.J., Lehnert, N., 2016. Is there a pathway for N2O production from hydroxylamine oxidoreductase in ammonia-oxidizing bacteria? Proc. Natl. Acad. Sci. 113, 14474–14476. https://doi.org/10.1073/pnas.1617953114.
Yan, X., Li, L., Liu, J., 2014. Characteristics of greenhouse gas emission in three full-scale wastewater treatment processes. J. Environ. Sci. 26, 256–263. https://doi.org/10.1016/S1001-0742(13)60429-5.
Zhang, R., Xie, W.-M., Yu, H.-Q., Li, W.-W., 2014. Optimizing municipal wastewater treatment plants using an improved multi-objective optimization method. Bioresour. Technol. 157, 161–165. https://doi.org/10.1016/j.biortech.2014.01.103.
Zhu-Barker, X., Cavazos, A.R., Ostrom, N.E., Horwath, W.R., Glass, J.B., 2015. The importance of abiotic reactions for nitrous oxide production. Biogeochemistry 126, 251–267. https://doi.org/10.1007/s10533-015-0166-4.