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Seasonality of nitrous oxide emissions at six full-scale wastewater treatment plants

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ABSTRACT

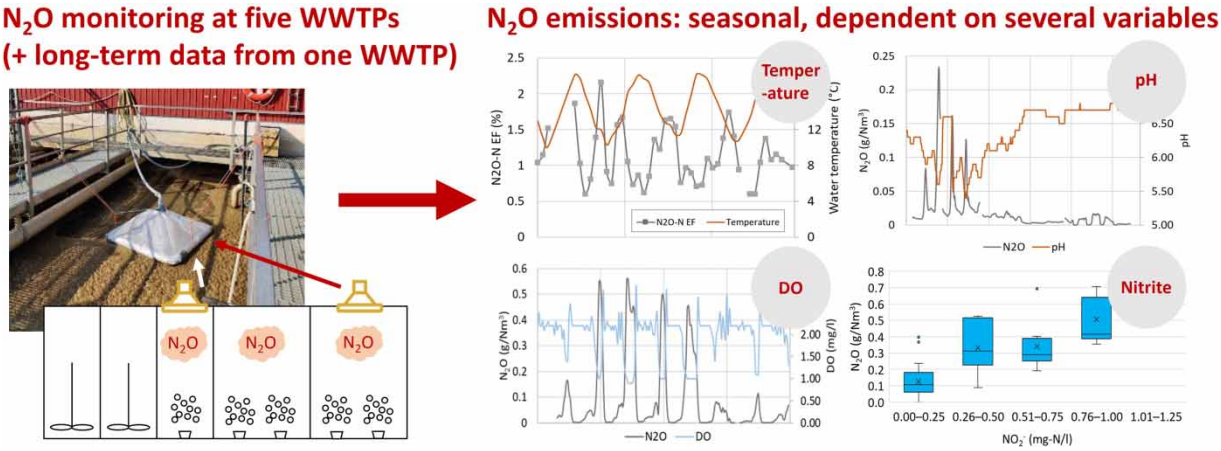
Nitrous oxide (N_2O) is an ozone-depleting greenhouse gas that contributes significantly to the carbon footprint of a wastewater treatment plant (WWTP). Plant-specific measurement campaigns are required to reliably quantify the emission level that has been found to significantly vary between WWTPs. In this study, the N_2O emissions were quantified from five full-scale WWTPs during 4- to 19-day measurement campaigns conducted under both cold period conditions (water temperature below 12 °C) and warm period conditions (water temperature from 12 to 20 °C). The measurement data were studied alongside long-term monitoring data from a sixth WWTP. The calculated emission factors (EFs) varied from near 0 to 1.8% relative to the influent total nitrogen load. The results confirmed a significant seasonality of N_2O emissions as well as a notable variation between WWTPs in the emission level, which a single fixed EF cannot represent. Wastewater temperature was one explanatory factor for the emission seasonality. Both low and high emissions were measured from denitrifying–nitrifying activated sludge (AS) processes, while the emissions from only nitrifying AS processes were consistently high. Nitrite (NO_2^-) at the end of the aerobic zones of the AS process was linked to the variability in N_2O emissions during the cold period.

Key words: activated sludge process, continuous off-gas monitoring, emission factor, full-scale wastewater treatment plant, greenhouse gas, nitrous oxide

HIGHLIGHTS

- N_2O emissions exhibit seasonal variability that can be linked to wastewater temperature.
- Fixed emission factors are highly unrepresentative of the variability in N_2O emissions between treatment plants.
- N_2O emissions are higher in only nitrifying processes than in nitrifying–denitrifying processes.
- Process disturbances may significantly affect the N_2O emission level during short-term monitoring.

GRAPHICAL ABSTRACT



INTRODUCTION

The main focus of wastewater treatment has been to meet the effluent requirements specified by the environmental legislation while operating the treatment process cost-effectively. Recently, ambitious goals for climate change mitigation have been set at the country, municipality, and water utility levels (Pijuan & Zhao 2022). As a result, water utilities are increasingly incorporating climate change mitigation into their strategies and plant operations (Pijuan & Zhao 2022).

In wastewater treatment, direct nitrous oxide (N_2O) emissions from the treatment process can contribute over 60% to the total greenhouse gas emissions of a municipal wastewater treatment plant (WWTP) (Daelman *et al.* 2013; Kosonen *et al.* 2016; Maktabifard *et al.* 2022a). N_2O is produced during nitrification and denitrification as a part of biological nitrogen removal (Kampschreur *et al.* 2009). It is an ozone-depleting greenhouse gas with a global warming potential 300 times that of carbon dioxide (CO_2) (IPCC 2019). N_2O emissions exhibit a complex multivariate dependency on wastewater treatment process variables, such as dissolved oxygen (DO) level (Kampschreur *et al.* 2008), nutrient accumulation (Tallec *et al.* 2006), pH (Su *et al.* 2019), system shocks (Burgess *et al.* 2002), and the microbial community in the activated sludge (AS) (Gruber *et al.* 2021b).

High variation in the N_2O emission level has been measured at full-scale WWTPs around the world. For example, an emission factor (EF) range of 0.01–1.8% relative to influent total nitrogen load was measured by Ahn *et al.* (2010) in the US. The EFs measured by Gruber *et al.* (2020) varied between 1.0 and 2.4% at Swiss WWTPs. The high variability of N_2O emissions makes it difficult to represent the emission level using fixed EFs (Cadwallader & VanBriesen 2017; Gruber *et al.* 2020). Moreover, applying fixed EFs in carbon footprint analysis or life cycle assessment (LCA) (e.g. by Liao *et al.* 2020; Maktabifard *et al.* 2022a) likely leads to an unreliable estimation of the contribution of N_2O emissions. Fixed EFs may also erroneously harmonize the N_2O emissions to a similar level for each WWTP irrespective of their treatment process configuration and operational conditions. Thus, plant-specific N_2O monitoring is needed to reliably quantify the emission level (Vasilaki *et al.* 2019).

The focus of this paper was on studying how the N_2O emission level varies seasonally at Finnish WWTPs by conducting relatively short-term N_2O monitoring at several full-scale WWTPs under summer and winter conditions. Even though there is a consensus on long-term monitoring being recommendable (Ribera-Guardia *et al.* 2019; Vasilaki *et al.* 2019; Gruber *et al.* 2021a), the primary objective of this study was to assess the feasibility of fixed EFs in N_2O emission estimation rather than develop new shorter-term seasonal EFs for Finnish WWTPs. Therefore, the measured N_2O emission levels were critically compared to the fixed EF proposed by the Intergovernmental Panel on Climate Change (IPCC) (2019). Additionally, available process data were analyzed to explain the variability in N_2O emissions.

METHODS

N_2O measurements were carried out at five full-scale AS plants (plants A–E) in Finland. Long-term N_2O monitoring data were received from a sixth Finnish WWTP (plant F) to support the study and plan the N_2O monitoring at plants A–E. Plants A–D are outdoor plants, whereas plants E–F have been built underground. Table 1 introduces the size of the plants in population equivalents (PE) and the requirements for nitrification or nitrogen removal set by the environmental permit for each plant.

The treatment process at each treatment plant begins with screening, grit removal, and primary clarification. The pre-treatment is followed by an AS process consisting of bioreactors and secondary clarifiers. The bioreactors are divided into several zones. The reactors typically have a denitrification–nitrification configuration with anoxic zones in the beginning, followed by aerobic zones. However, some of the plants operate them for part of the year as only nitrifying as they need to switch the denitrifying anoxic zones to aerobic zones to ensure there is enough aerobic treatment volume for the completion of nitrification even at low temperatures. The plants are especially prone to omit denitrification during the cold period, if their environmental permit requires only a sufficient nitrification rate but not total nitrogen removal, such as plants B and D (Table 1), as they are typically designed with more limited treatment capacity than WWTPs where total nitrogen removal is required. The sludge age is adjusted according to the wastewater temperature, and pH is controlled with alkali addition. Phosphorus is precipitated chemically at each plant. Additional details of the treatment process and the used chemicals are included in Table 1.

Measurement campaigns of 4–19 days were conducted at plants A–E to measure the direct N_2O emissions. The long-term N_2O measurement data from plant F supported the planning of the measurements, as the plant was assumed to be a good

Table 1 | Details of the treatment process at plants A–F

	Size of the plant (PE)	Requirement for yearly average nitrification/nitrogen removal rate in the environmental permit	Nitrogen removal process	BOD_7/N in the influent to the AS process	Alkali and phosphorus removal chemicals
Plant A	20,000	50% nitrogen removal (above 12 °C)	AS process (4 bioreactors with 2 zones)	2.4	Calcium hydroxide, ferric sulphate
Plant B	228,000	80% nitrification rate	AS process (4 bioreactors with 8 zones)	2.1	Calcium and sodium carbonate, ferrous sulphate
Plant C	14,000	70% nitrogen removal	AS process (2 bioreactors with 6 zones)	3.6	Calcium hydroxide, ferric chloride
Plant D	350,000	90% nitrification rate	AS process (8 bioreactors with 5 zones)	1.2	Calcium hydroxide, ferric sulphate
Plant E	315,000	75% nitrogen removal	AS process (4 bioreactors with 6 zones)	3.4	Calcium carbonate, ferrous sulphate
Plant F	1,100,000	80% nitrogen removal	AS process (9 bioreactors with 5 zones), denitrifying postfiltration	2.5	Calcium hydroxide, ferrous sulphate

representation of the typical conditions and treatment process of Finnish full-scale WWTPs. The emission level at plant F exhibits distinctive seasonality, and thus the emission monitoring at plants A–E was conducted primarily with one measurement campaign in the warm period and one campaign in the cold period. A threshold water temperature of 12 °C was applied to separate warm and cold period conditions. Generally, the average water temperature has been detected to stay below 12 ± 1 °C for more than 6 months of the year at Finnish WWTPs (Kruglova *et al.* 2014). Additionally, the current Government Decree on Urban Wastewater Treatment 888/2006 does not require total nitrogen removal from WWTPs when the water temperature is below 12 °C (Ministry of the Environment Finland 2006). The water temperature at the plants varied from 12 to 20 °C during the warm period conditions and from 7 to 12 °C during the cold period conditions, respectively. Table 2 details the length and measurement locations of each measurement campaign. The nitrification rate was high (90–100%) during all measurements.

The N_2O measurements at plants A–E focused only on the aerobic zones, as they have been found to emit the majority of the N_2O emissions at plant F in comparison to the anoxic zones. For example, Maktabifard *et al.* (2022b) studied the dominant N_2O emission pathways at plant F with a process model and found that the anoxic zones produce only around 7% of the

Table 2 | The length and measurement locations of the N_2O measurements at plants A–F

	Warm period conditions (Water temperature above 12 °C)		Cold period conditions (Water temperature below 12 °C)	
	Length of measurements	Measurement locations	Length of measurements	Measurement locations
Plant A	*Not measured*		17 days in Dec 2022	Beginning and end of aerobic zone[a]
Plant B	11 days in Jun 2022	Second and last aerobic zone[a]	15 days in Apr 2022	First and last aerobic zone[b]
Plant C	4 days in Jun-Jul 2022	End of aerobic zone[a]	16 days in Jan 2023	Third and last aerobic zone[b]
Plant D	17–19 days in Sep and Nov 2022	First and last aerobic zone[a]	*Not measured*	
Plant E	11 days in May-Jun 2022	First and last aerobic zone[a]	15 days in Mar-Apr 2022	Last aerobic zone[a]
Plant F	Continuous	Exhaust air pipe[a]	Continuous	Exhaust air pipe[a]

[a]Denitrifying–nitrifying AS process.
[b]Nitrifying AS process.

N_2O emissions, as the majority (93%) of the emissions originates from the aerobic zones. In addition, only a low contribution has been found from secondary clarifiers to the total N_2O emissions (Mikola *et al.* 2014). The process configuration and conditions as well as the wastewater quality at plants A–E were similar to plant F. The goal of the monitoring campaigns was to measure the first and last aerobic zones based on the assumed emission pattern within the zones (Blomberg *et al.* 2018; Maktabifard *et al.* 2022b). As the focus of the monitoring at plants A–E was on the first and last aerobic zones and the emissions for the middle aerobic zones were linearly interpolated, there are a few essential conditions for the validity of this approximation: (1) The majority of N_2O emissions is produced during nitrification, (2) nitrification proceeds gradually in the aerobic zones of the plug flow reactors, and (3) the aeration control should aim at maximizing the nitrification rate.

The measurement setup (Figure 1) consisted of an off-gas hood and a Gasmet GT5000 Terra FTIR gas analyzer. The stripping N_2O was collected by placing the off-gas hood to float in one aerobic zone at a time. The hood was connected to a pipe that led the off-gas to the gas analyzer. The analyzer utilizes Fourier transform infrared (FTIR) spectroscopy technology to quantify the concentration of N_2O in the off-gas. The measurement was continuous with a measurement interval of one minute. Measurement breaks occurred due to the calibration of the analyzer every 24 h and occasionally due to the shutdown of the computer storing the results.

Plant F measures the N_2O emissions from the exhaust air pipe, as it is an underground plant, and all the off-gas from the treatment process is conducted to the open air via the pipe. The off-gas is analyzed with an FTIR-based Gasmet Continuous Emissions Monitoring System (CEMS) II. Measurement data from 2019 to 2022 were used in this study. Periods with atypical emission levels were defined by the plant operators, and these data were excluded from the analysis to consider only the typical seasonality of the emission level at plant F.

The measurement data were used to calculate N_2O-nitrogen (N_2O-N) EFs by relating the total N_2O-N load from the AS process ($L_{N2O\text{-}N}$ as kg-N) to the influent total nitrogen load (L_N as kg-N) (Equation (1)).

$$N_2O - N\ EF\ (\%) = \frac{L_{N_2O-N}}{L_N} * 100 \tag{1}$$

The off-gas N_2O concentrations for the unmeasured aerobic zones were estimated based on the measured zones with linear regression analysis. If only one zone was measured, the same N_2O concentration was assumed for all aerobic zones. The plants analyze the water quality, such as influent total nitrogen, with flow proportional, 24-h composite samples which are collected biweekly, weekly, or monthly.

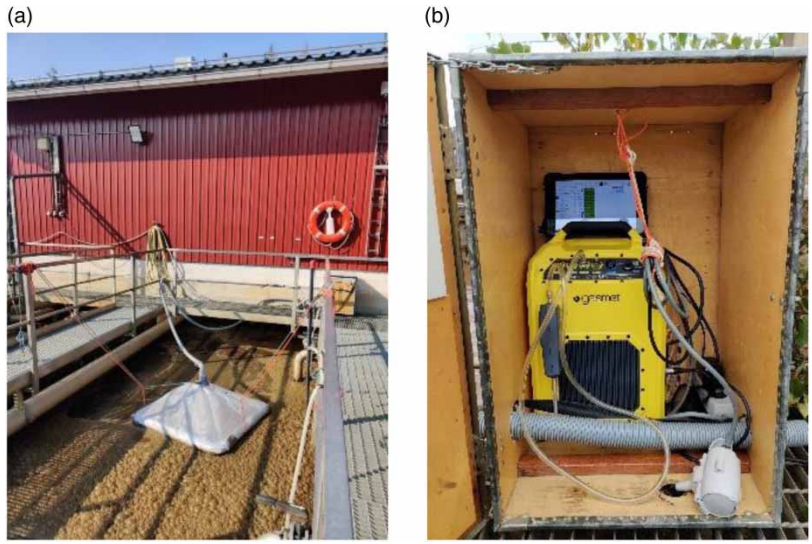


Figure 1 | The measurement setup consisted of an off-gas hood (a) and an FTIR-based gas analyzer (b) for the N_2O measurement campaigns at plants A–E.

Relevant process data were collected to support the analysis and find explanatory factors for the variability in the measured emission levels. To study the variability in the loading of the AS processes, the specific total nitrogen loading in the aerobic treatment volume (kg-N/kg-MLSS/d) was calculated. The specific loading relates the incoming total nitrogen load to the AS process to the mass of mixed liquor suspended solids (MLSS) in the aerobic zones. Additionally, dissolved nitrite (NO_2^-) was analyzed from grab samples near the off-gas hood to study its correlation to N_2O emissions. The standard for the NO_2^- analysis was SFS 3029:1976.

RESULTS AND DISCUSSION

The EFs calculated for plants A–E as well as the off-gas N_2O concentrations in the last aerobic zone (Table 3) varied significantly, both between the plants and between two measurement campaigns conducted at the same plant. The EFs relative to influent total nitrogen varied from 0.04 to 1.0% in the warm period and from 0.2 to 1.8% in the cold period. Similarly, EFs were computed relative to the removed total nitrogen load to study how nitrogen conversion could affect the emitted N_2O. However, these EFs were close to the EFs related to the influent total nitrogen load, as the plants generally achieved relatively high total nitrogen removal (around 60–80%). The only notable exception was the EF from the cold period monitoring at plant B, as the EF relative to the influent total nitrogen load was 1.3%, whereas the EF related to the removed total nitrogen load was 28%. This was due to the low total nitrogen removal rate (around 7%), as there is no requirement for total nitrogen removal in the environmental permit of plant B. Thus, the study only considers the EFs relative to the influent total nitrogen load as they are more commonly used and comparable to other studies.

The seasonality of the N_2O emissions at plants A–E is supported by the long-term data from plant F, where the emission level has a distinctive seasonal pattern. Typically, periods of low and high emissions at plant F have a duration of several months. Figure 2 includes a data series of the monthly average EFs at plant F from 2019 to 2022, excluding data points of atypical emissions levels due to long-term disturbance in the treatment process. At plant F, warm temperatures and low emissions typically occur from May to November (Figure 2). This observation was utilized to plan the monitoring periods at plants A–E (Table 2) to similarly assess the emission seasonality.

A comparison to reported EFs from measurements at other full-scale WWTPs and the fixed EF by IPCC (2019) (Table 4) revealed a similar emission level as was measured from plants A to F (Table 3). The IPCC factor is close to the highest measured EFs, but it is clearly an overestimation of the EFs below 0.5% that were measured especially during the warm period (above 12 °C). Over year-long measurements by Daelman *et al.* (2015) and Gruber *et al.* (2020) resulted in a high emission level of around 1–3%, while the EFs from shorter measurement campaigns by Ahn *et al.* (2010), Mikola *et al.* (2014), and Valkova *et al.* (2021) had a wide range from close to 0% to above 2%.

The measured EFs were generally higher during the cold period than the warm period (Figure 3), which is a similar finding to that made by Gruber *et al.* (2020). During the cold period, the highest EFs of 1.3–1.8% were measured, while low EFs (0.04–0.09%) were measured only during the warm period. By contrast, plant A had a low emission level (EF 0.2%) during the cold period, whereas plants B and F produced high N_2O emissions (EF 1.0%) also during the warm period.

Table 3 | The N_2O-N EFs and the off-gas N_2O concentration range in the last aerobic zone at plants A–F

	Warm period (Water temperature above 12 °C)		Cold period (Water temperature below 12 °C)	
	N_2O-N EF (% of influent total nitrogen load)	Off-gas N_2O concentration range (min.–max.) in the last aerobic zone (g/Nm^3)	N_2O-N EF (% of influent total nitrogen load)	Off-gas N_2O concentration range (min.–max.) in the last aerobic zone (g/Nm^3)
Plant A	–	–	0.2	0.00–0.23
Plant B	1.0	0.00–0.13	1.3	0.05–0.59
Plant C	0.04	0.00–0.01	1.8	0.09–0.48
Plant D	0.08–0.4[a]	EF 0.08: 0.02–0.11 EF 0.4: 0.09–0.56	–	–
Plant E	0.09	0.00–0.09	1.7	0.15–0.78
Plant F (2019–2022)	1.0	*No data*	1.3	*No data*

[a]EFs from two measurement campaigns.

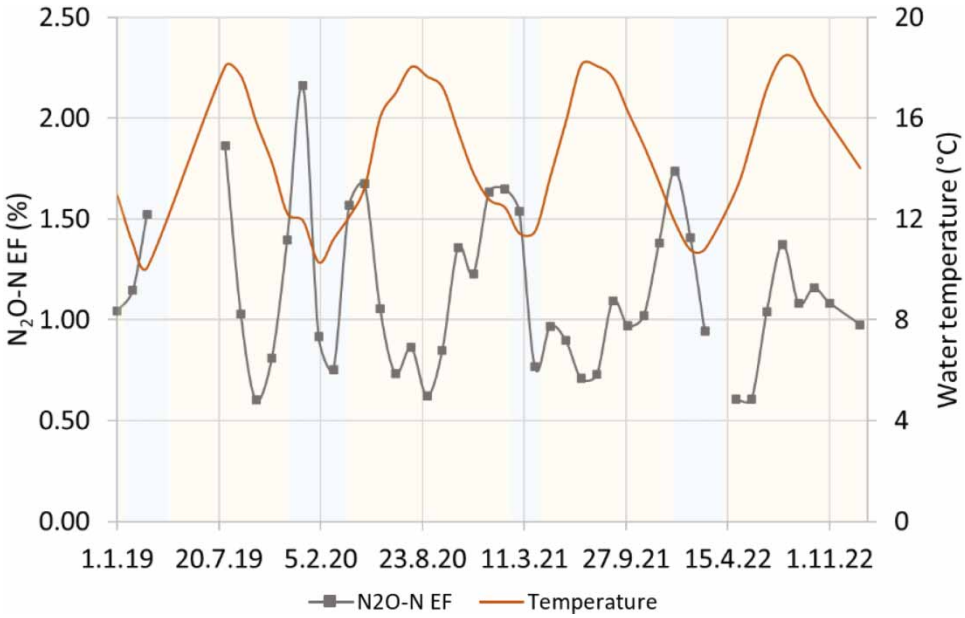


Figure 2 | The monthly average EFs (dark grey squares) at plant F and the water temperature in 2019–2022. Cold periods are marked with light blue columns and warm periods with light yellow columns.

Table 4 | Reference EFs from full-scale WWTPs and the fixed EF by IPCC (2019)

Reference	N_2O-N EF (% of influent total nitrogen load)
Ahn *et al.* (2010)	0.01–1.8
Mikola *et al.* (2014)	0.02–2.6
Daelman *et al.* (2015)	2.8
Gruber *et al.* (2020)	1.0–2.4
Valkova *et al.* (2021)	0.002–1.5
IPCC (2019)	1.6

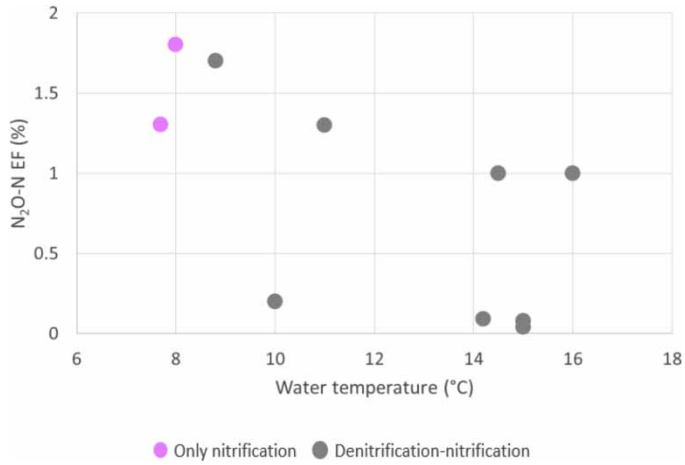


Figure 3 | The relation between the N_2O-N EFs and the water temperature (Pearson correlation coefficient $r = -0.64$ and coefficient of determination $r^2 = 0.42$) at plants A–F.

Two of the highest EFs (1.3 and 1.8%) were measured from processes that were only nitrifying. This indicates that only nitrifying processes could be more prone to produce higher N_2O emissions than processes that are both nitrifying and denitrifying. This finding is supported by the N_2O measurement data from 10 Austrian WWTPs by Valkova *et al.* (2021), who concluded that nitrification was consistently the main pathway for N_2O emissions at all plants. Moreover, Valkova *et al.* (2021) found the low dissolved N_2O concentrations in the denitrifying anoxic zones to indicate that the stepwise reduction of nitrate (NO_3^-) to nitrogen gas (N_2) was complete during denitrification, and there was no accumulation of the N_2O intermediate. Even though the anoxic zones were not directly monitored at plants A–E, we concluded that N_2O was not produced during heterotrophic denitrification in the anoxic zones due to the consistently low N_2O emissions at the beginning of the aerobic zones (following the anoxic zones). Therefore, the minimum and maximum off-gas N_2O concentrations in the last aerobic zones (Table 3) represent well the emission variability between plants A and E.

The short-term measurements are more prone to be impacted by disturbances in the operation of the treatment process than long-term monitoring. For example, a defect in the calcium hydroxide dosing system occurred at plant A at the start of the measurements, which caused a decreased pH (below 6.0) that seemed to cause a higher emission level (off-gas N_2O around 0.1–0.2 g/Nm^3) than the low emissions (off-gas N_2O around 0.02 g/Nm^3) measured after the pH had normalized back to near neutral (Figure 4(a)). At plant D, two measurement campaigns were conducted in similar conditions, but an insufficient aeration capacity (75% of the capacity was available due to a defective aeration compressor) during the first measurement caused the DO to drop from the setpoint of 2.5 to around 1 mg/l in the aerobic zones. During the suboptimal DO level, the off-gas N_2O peaked from near 0 to 0.4–0.6 g/Nm^3 in the last aerobic zone (Figure 4(b)). This was found to contribute to the significantly higher emission level (EF 0.4%) compared to the second measurement campaign (EF 0.08%) when the DO level was consistently around 2.0 mg/l. Therefore, the EF 0.08% was decided to be more representative of the emission level at the plant and the EF 0.4% and other measurement data from the first measurement campaign were excluded from the analyses. To avoid the uncertainty caused by operation disturbances, longer-term measurements are recommendable. Additionally, long-term measurements would help to determine the lengths of the periods of high and low emission levels.

The dissolved NO_2^- concentration was analyzed from grab samples near the measurement location to study the correlation between NO_2^- and N_2O. For example, Gruber *et al.* (2020) and Kuokkanen *et al.* (2021) have reported a link between NO_2^- accumulation and increased N_2O emissions. The NO_2^- concentration was compared directly to the measured off-gas N_2O concentration during the hour the grab sample was collected. The studied data were collected from plants A to E. The most distinct correlation was found between NO_2^- and N_2O at the end of the aerobic zone of the AS bioreactors. However, a strong positive Pearson correlation ($r = 0.83$) was found only with the data points ($n = 71$) from the cold period measurements (Figure 5(a)), while there was no correlation with the data points ($n = 24$) from the warm period measurements (Figure 5(b)). The NO_2^- concentrations were high (0.5–1.0 mg-N/l) in some cases in the first aerobic zones as well, but only small N_2O emissions were measured in these zones. There are several possible explanations for the observed correlation in the last aerobic zone during the cold period. The diurnal load variation causes fluctuations in the NO_2^- concentrations when nitrification is ongoing. When the capacity and conditions of the AS process support nitrification well (in this case,

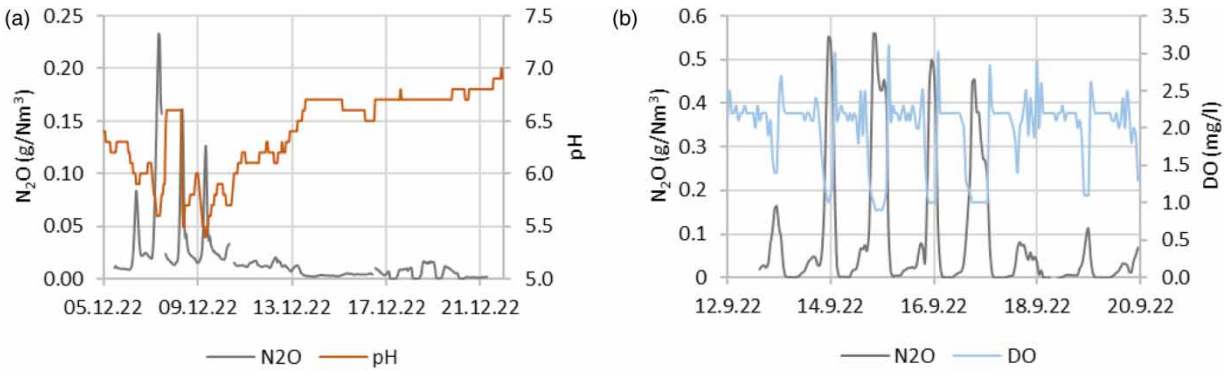


Figure 4 | Increased N_2O emissions were measured during a defect in calcium hydroxide dosing (a) and during low DO levels due to insufficient aeration capacity (b).

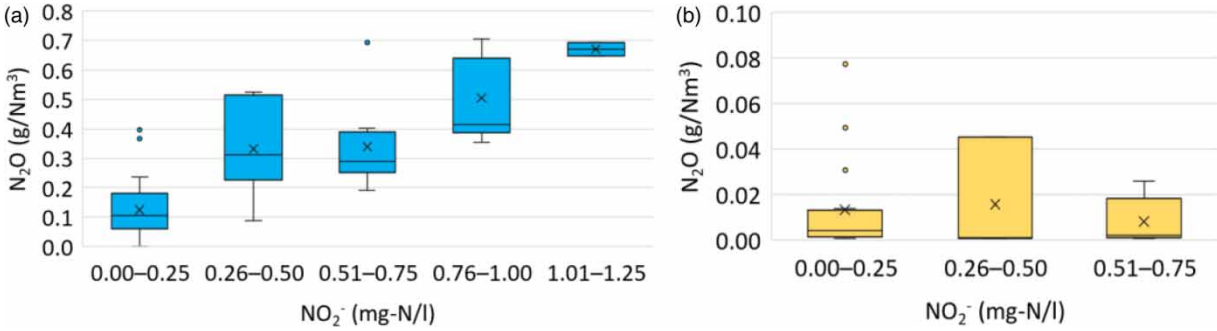


Figure 5 | The relation between off-gas N_2O concentration and dissolved NO_2^- in the last aerobic zone during the cold period (a) and warm period (b) monitoring campaigns at plants A–E.

during the warm period), the NO_2^- concentrations in the last aerobic zone stay below 0.8 mg-N/l. Increased concentrations in this zone might be a sign of increased N_2O emissions, even if NO_2^- is not a direct cause for the emissions. We speculate it could rather be an indicator of nitrifier denitrification being triggered by (1) lower activity of nitrite-oxidizing bacteria (NOB) or (2) lower activity of heterotrophic denitrification inside the flocs in the aerobic zones and the resulting accumulation of NO_2^- and N_2O.

The results indicate that specific nitrogen loading (kg-N/kg-MLSS/d) in the aerobic treatment volume did not explain the high variation in the N_2O-N EFs, as no correlation was found between the variables. Similarly, the nitrogen removal rate did not link to the N_2O emissions at plants A–F. Plants achieving over 80% nitrogen removal produced both low (EF 0.04–0.09%) and high (EF 1.7%) emissions. On the other hand, low EFs of 0.08 and 0.2% were measured with nitrogen removal of 40 and 60%, respectively. Moreover, the BOD_7/N ratio in the influent to the AS process (Table 1) varied between 1.2 and 3.6, but it did not correlate with the EFs. We assumed that a low BOD_7/N ratio could result in increased N_2O emissions if there was not enough biodegradable organic matter for the completion of heterotrophic denitrification. The hypothesis was supported by the study by Zhang *et al.* (2012), who found that a sequencing batch reactor (SBR) treating influent wastewater with a BOD_5/N ratio of 0.7 produced higher emissions than an SBR treating influent wastewater with a BOD_5/N ratio of 2.9. Surprisingly, a low EF of 0.08% was measured at plant D even when the BOD_7/N ratio was very low (1.2).

CONCLUSIONS

The N_2O-N EFs from six Finnish full-scale WWTPs showed significant seasonality in emissions, with the EFs varying from near 0% to almost 2%. The seasonality could be partly due to the variation in wastewater temperature. High N_2O emissions were typically measured at low-temperature water (below 12 °C), while the emission level could be very low under warm period conditions (12–20 °C). High emissions were measured from AS processes with a nitrification configuration, whereas both low and high emissions were measured from denitrifying–nitrifying AS processes. The dissolved NO_2^- at the end of the aerobic zones of the AS bioreactors was found to be a good indicator of the N_2O emission variability during the cold period conditions. On the other hand, the nitrogen removal rate, BOD_7/N ratio, or specific nitrogen loading could not explain the variation in the measured emissions.

The EFs are in line with reported EFs from other full-scale WWTPs, and the high variability in the EFs highlights the unrepresentativeness of the fixed EF suggested by the IPCC. Furthermore, the EFs calculated from short-term measurement data depict well the N_2O emission seasonality but can be uncertain if disturbances to the normal operation of the WWTP occur during the measurements. Therefore, longer-term measurements are recommended to study the root causes of the emissions and determine the length of low and high emission levels.

DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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