N_2O emission in full-scale wastewater treatment: Proposing a refined monitoring strategy

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Abstract

Nitrous oxide (N_2O) emissions from wastewater treatment contribute significantly to greenhouse gas emissions. They have been shown to exhibit a strong seasonal and daily profile in previously conducted monitoring campaigns. However, only two year-long online monitoring campaigns have been published to date. Based on three monitoring campaigns on three full-scale wastewater treatment plants (WWTPs) with different activated sludge configurations, each of which lasted at least one year, we propose a refined monitoring strategy for long-term emission monitoring with multiple flux chambers on open tanks. Our monitoring campaigns confirm that the N_2O emissions exhibited a strong seasonal profile and were substantial on all three plants (1 - 2.4% of the total nitrogen load). These results confirm that N_2O is the most important greenhouse gas emission from wastewater treatment. The temporal variation was more distinct than the spatial variation within aeration tanks. Nevertheless, multiple monitoring spots along a single lane are crucial to assess representative emission factors in flow-through systems. Sequencing batch reactor systems were shown to exhibit comparable emissions within one reactor but significant variation between parallel reactors. The results indicate that considerable emission differences between lanes are to be expected in cases of inhomogeneous loading and discontinuous feeding. For example, N_2O emission could be shown to depend on the amount of treated reject water: lanes without emitted less than 1% of the influent load, while parallel lanes emitted around 3%. In case of inhomogeneous loading, monitoring of multiple lanes is required. Our study enables robust planning of monitoring campaigns on WWTPs with open tanks. Extensive full-scale emission monitoring campaigns are important as a basis for reliable decisions about reducing the climate impact of wastewater treatment. More specifically, such data sets help us to define general emission factors for wastewater treatment plants and to construct and critically evaluate N_2O emission models.

1. Introduction

Nitrous oxide (N_2O) emissions have a strong impact on the environment. N_2O has a global warming potential 300 times greater than that of carbon dioxide and is emitted in significant quantities across the globe (Stocker et al., 2013). Additionally, it is considered the most important ozone-depleting substance of anthropogenic origin (Ravishankara et al., 2009). Wastewater treatment (WWT), in particular biological nitrogen removal, has been shown to produce N_2O (Hanaki et al., 1992). Even if only small amounts of the influent nitrogen load are emitted as N_2O, it can represent the dominant greenhouse gas emission of a wastewater treatment plant (WWTP): Two year-long monitoring campaigns, at Kralingseveer in the Netherlands (Daelman et al., 2015) and Viikinmäki in Finland (Kosonen et al., 2016), found that N_2O constituted the majority (78% and 86%) of the total greenhouse gas emissions of the WWTP (Daelman et al., 2013b; Kosonen et al., 2016). In these studies, 2–3% of the influent nitrogen load was emitted as N_2O.

The IPCC guidelines, which were updated in 2019, suggest an emission factor of 1.6% of the total nitrogen load. The value is an average of current short-term and long-term monitoring campaigns (IPCC, 2019). Based on the new value it can be expected that N_2O from wastewater contributes significantly more to the total N_2O emissions than previously assumed 3.1% for Europe in 2017 (EEA, 2017). For almost all of the countries, the calculations were based on the previously used, significantly lower emission factor of 0.035% (Czepiel et al., 1995; EEA, 2017).

No quantitative model currently explains both long-term and short-term N_2O emission dynamics. Although long-term monitoring studies confirm significant emission variation on a yearly scale, no modelling study covering more than a few months has been published (Ni and Yuan, 2015). Without significant advances in long-term emissions modelling, monitoring

63 campaigns remain necessary to determine emission factors and provide a solid data basis for
64 validating the current emission models (Vasilaki et al., 2019).

65 Emission factors have been reported ranging from 0.01% to 25% of the incoming nitrogen load
66 (Ahn et al., 2010; Kampschreur et al., 2009). Although emissions vary significantly between
67 treatment plants and processes, the wide range of emission factors assessed can at least
68 partly be attributed to differences in monitoring strategies and methods (Kosonen et al., 2016).
69 A consensus on a generally accepted emission factor has yet to be reached. Because
70 emissions have been found to vary greatly over the year as well as over single days, a
71 representative quantification of N_2O emissions requires long-term online measurement, which
72 is currently rarely applied (Kosonen et al., 2016). Preferably, measurement campaigns should
73 cover the complete annual temperature range of the incoming wastewater at a time resolution
74 of <30 minutes (Daelman et al., 2013a).

75 In both published long-term monitoring campaigns, fully covered WWTPs were monitored by
76 measuring the N_2O off-gas concentration in the centrally collected off-gas (Daelman et al.,
77 2015; Kosonen et al., 2016). The application of this monitoring approach is only possible on
78 fully covered WWTP. To date, no studies have been published of monitoring campaigns over
79 a year or longer on WWTPs with open tanks. Hence, guiding principles for long-term monitoring
80 on open tanks are not yet available. Since many WWTPs in industrialized countries are
81 equipped with open tanks, long-term online monitoring campaigns on uncovered WWTPs are
82 crucial to establish a representative N_2O emission factor for greenhouse gas reporting (Pan et
83 al., 2016).

84 N_2O emissions from open tanks are typically collected with flux chambers (Chandran et al.,
85 2016). Another possible approach to monitor the emissions are the application of a
86 measurement for dissolved N2O via i) Clark-Type electrodes (Marques et al., 2014) or ii)
87 stripping of N2O from a sludge sample (Mampaey et al., 2015) combined with an air stripping

model (Marques et al., 2014). Due to the potentially high spatial heterogeneity of N_2O emissions within a treatment process, measurements at various spots on a WWTP are required to arrive at a representative measurement of the N_2O emission factor for an entire WWTP (Aboobakar et al., 2013; Pan et al., 2016). For this purpose, a flux chamber based monitoring approach is more appropriate than a dissolved N_2O measurement based monitoring, due to i) significantly higher costs for additional point measurements and ii) that quantifying stripping efficiency with sufficient accuracy is not a simple task at full-scale. N_2O emissions from activated sludge lanes with open reactors operated in parallel have not yet been compared, although this is an important step in assessing plant-wide emission factors.

The present study proposes a refined strategy to quantify N_2O emission factors for biological treatment in open tanks, based on long-term high-resolution monitoring campaigns on three municipal WWTPs in Switzerland with three different activated sludge configurations. At each plant, multiple spots were monitored at high resolution (10-15 min) over more than one year. A decision tree is proposed for planning the number of sampling points required by the type of treatment plant and operational choices (e.g. feeding regimes). All monitoring data and operational data series are made available at high resolution in the supplementary data.

2. Method

2.1. Field sites

N_2O emissions were monitored in three Swiss municipal WWTPs operated with different activated sludge process configurations: a conventional activated sludge process (CAS), an alternatingly fed and intermittently aerated activated sludge process (A/I), and a sequencing batch reactor activated sludge process (SBR). Key numbers and general information on the WWTPs are summarized in Table 1.

Table 1 Features and key numbers of the monitored WWTP. **A/I**: alternating / intermitting feed plant; **CAS**: conventional activated sludge; **CH**: Switzerland; **SBR**: sequencing batch reactor. **PE**: Person equivalent.

Process	CAS	A/I	SBR
Location	Altenrhein (CH)	Lucerne (CH)	Uster (CH)
Wastewater design load (m^3/d)	30,000	90,000	15,000
Biological treatment design load (PE)	70,000	250,000	40,000
Pretreatment	Screening Grit Primary settler	Screening Grit Primary settler	Screening Grit Primary settler
Lanes with segrated sludges	3 (divided into 2 sublanes)	6 (divided into 2 sublanes)	6
Aerobic sludge age (days)	Winter: 12 Summer: 10	Winter: 14 Summer: 10	Winter: 12 Summer: 8
Dissolved oxygen concentration (mgO_2/l)	2	2	2
Nitrogen removal rate (%)	65	75	40
Nitrification discharge limit ($mgNH_4^+$-N/l)	2	2	2

CAS plant: The nitrogen load of 125,000 PE exceeds the COD load, since the WWTP treats the sludge containing reject water of other plants in the region. After pretreatment, most of the wastewater is treated with a conventional activated sludge process on three lanes. During the winter season, the anoxic zone is aerated completely, and denitrification is reduced. Additionally, eight fixed-bed biofilm reactors (Biostyr®) are operated in parallel to treat the rest of the wastewater. These are equipped with a lower anoxic zone and an aerobic zone above

this. During low influent flow, only two regularly switched fixed biofilm reactors are operated. On average, the fixed-bed system treats 30% of the wastewater.

Reject water was bypassed around one activated sludge lane at Altenrhein WWTP over a period of two months (March 2016 to May 2016). The experiment was then repeated one year later for three months (December 2016 to March 2017). In the experiments, reject water was dosed after the primary clarifier instead of before it. The surplus reject water was shared in a ratio of two to one between the remaining activated sludge lanes and the fixed-bed reactor. The reject water supply to the fixed-bed and the activated sludge process was controlled and monitored separately by flow measurement (Endress+Hauser, Promag P). Emissions were monitored on both lanes with and without reject water, with three floating gas hoods per lane.

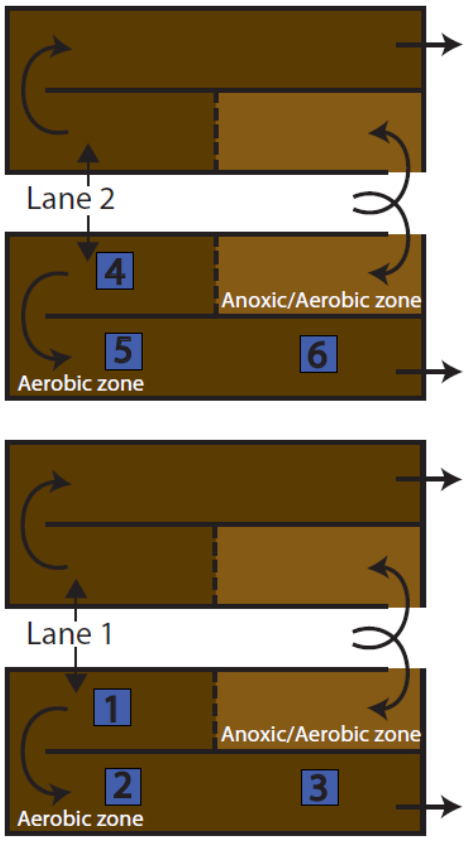

Figure 1 Monitoring setup of the Altenrhein WWTP monitoring campaign

AI plant: The biological treatment comprises six lanes each consisting of two reactors of 5,500 m^3. One of the reactors in each lane is always filling. In the middle of a cycle, the influent is directed to the other reactor. During the filling, the reactors are operated in plug-flow mode, starting with an anoxic phase followed by an aerobic treatment (main-aeration). After filling, the reactors are operated in batch mode. However, the reactors cannot be assumed to be fully mixed, since longitudinal mixing is limited due to the rectangular shape of the reactors. The cycle time is at least 1.5 hours in summer and 2 hours in winter and includes a 1 hour filling time and a variable aeration time, depending on the NH_4^+ concentration. Subsequently, one post-aeration reactor per lane (6 × 1,600 m^3) allows the nitrification discharge limit of 2 mgNH_4^+-N/L to be safely met. Anoxic cycles are shortened or canceled at low temperatures during the winter season to enable sufficient nitrification. During anoxic periods, the activated sludge is mixed at intervals by pulses of coarse bubble aeration, and fine bubble aeration is used to aerate the mixed liquor.

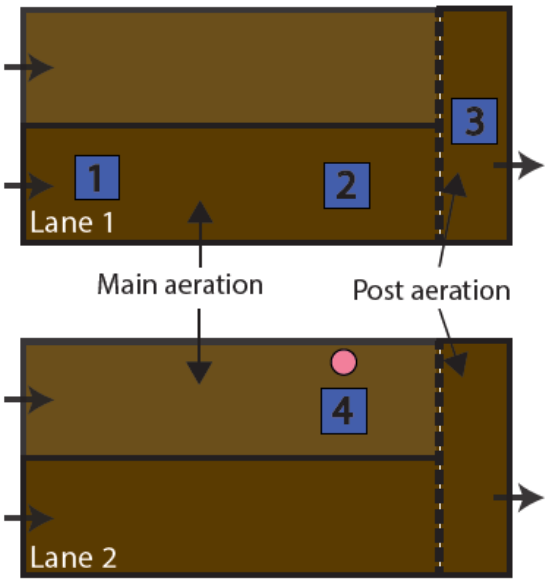

Figure 2 Monitoring setup of the Lucerne WWTP monitoring campaign

SBR plant: A sequencing batch reactor system with activated sludge was monitored at Uster WWTP (Uster, Switzerland). Most SBR cycles last about 6 hours and exhibit a fixed sequence of operational phases: 30 minutes to 1 hour of feeding, about 3 hours of reaction phase (anoxic, aerobic), 1.5 hours settling phase, and 0.5 hours decanting. The reaction phase ends as soon as the target NH_4^+ concentration is reached. During the winter season, an anoxic phase is skipped completely, and denitrification is reduced. In the summer season, the anoxic phase is set to a maximum of 25 minutes. Depending on the influent load, the anoxic phase can be skipped also in summer.

Figure 3 Monitoring setup of the Uster WWTP monitoring campaign

For each plant, the plant layout is shown in the supporting information (SI) in Figures S1, S2, and S3.

2.2. N_2O measurement and monitoring

An adapted version of the flux chamber method was used for the assessment of the gaseous emissions from the open tanks (Chandran et al., 2016). Gas emissions were collected with four to nine flux floating chambers per sampling campaign, each covering an area of 1 m^2 (Section A2 in the SI). In contrast to the standard method, an open outlet tube, with a diameter of 50 mm and length of 500 mm, was installed on top of the flux chambers as an off-gas pipe (Chandran et al., 2016). A small share of the off-gas (1 l/min) was diverted through a tube with

a diameter of 5 mm from the off-gas pipe's base to the central measuring unit. The gas was dehumidified by cooling (JCP SL, JCT, Wiener Neustadt, AUT). The N_2O concentrations were measured with a non-dispersive infra-red sensor (X-stream, Emerson, St. Louis MO, US). The device measures at a controlled, constant temperature of 60°C. Measurements were spatially resolved with an automated valve system preceding the dehumidifier and the analyzer. This enables the measurement of the off-gas composition sequentially at several spots within the WWTP, as in the procedure presented by Pan et al. (2016). The off-gas sampling location was changed every 4 to 6 minutes. The first 2 minutes after switching were ignored to account for purging of the connection hose, which had a maximum dead volume of 1.2 liters. The N_2O measuring range spanned from 0 ppm to 1000 ppm. The detection limit was 1 ppm. The devices were serviced and calibrated according to the manufacturer's instructions.

The Altenrhein WWTP was monitored with six to seven flux chambers: two activated sludge lanes, with three flux chambers each according to the scheme in Figure A2 (SI) and one to three on biofilm fixed-bed reactors. Each hood was monitored for 5 minutes, resulting in a measurement at each spot every 30 minutes. The off-gas was monitored for 16 months (from December 2015 to March 2017). Daily influent nitrogen loads were obtained with flow proportional composite samples for nitrogen (Hach, LCK338) measured by the plant operators twice a week.

The N_2O emissions of two out of six A/I-reactors were monitored at Lucerne WWTP for 19 months between March 2014 and September 2015. Four flux chambers were installed according to the scheme in figure S1 (SI): three on one lane and one on a separate lane. Each hood was monitored for 5 minutes, leading to a sample at each spot every 15 minutes. Daily influent nitrogen loads were obtained with flow proportional composite samples for nitrogen (Hach, LCK338) measured by the plant operators twice a week.

At the Uster WWTP, the emissions of all six SBR reactors were monitored between March 2018 and April 2019. On each reactor a flux chamber was installed. On one reactor (SBR 3), three chambers were installed to resolve the spatial variation of the emissions. Each hood was monitored for 3 minutes, resulting in a measurement at each spot every 20 minutes. Daily influent nitrogen loads were obtained with flow proportional composite samples for nitrogen (Hach, LCK338) measured by the plant operators twice a week.

The positioning of the hoods on the reactors are visualized in Figures 1-3. For each plant, a data set with the monitoring data, operational data and standard wastewater indices is provided in SI. The respective resolution was chosen according to the frequency of the gas measurement.

2.3. Calculation of N_2O emissions

The air flow supplied to the reactor was estimated via the blower speed recorded at intervals of 1 minute. Emissions were calculated in 1 minute intervals for the aerated phases only by linearly interpolating the measured N_2O concentrations. The N_2O net flux was calculated according to the formula provided by Aboobakar et al. (2013): the air flow supplied to each reactor section equipped with a flux chamber and the respective N_2O concentration was calculated according to Equation 1 (Aboobakar et al., 2013). Monthly and yearly average values of the nitrogen load, measured in 24 h composite samples, were used to calculate the N_2O emissions relative to the influent nitrogen load. For lanes with inhomogeneous influent distribution amongst lanes (Altenrhein WWTP, Uster WWTP), assumptions on the respective loads were made according to composite samples taken by the operators.

$$F = C * Qair \cdot \left(\frac{AH}{AT}\right), \quad (1)$$

where F is the net N_2O flux [kg N_2O-N/min], C is the measured N_2O concentration in the off-gas [kg N_2O-N/m^3], Qair is airflow supplied by the blower [m^3/min], AH is the number of air

212 blowers attributed to a specific flux chamber [-], and AT is the total number of air blowers in
213 the reactor [-].

3. Results and Discussion

3.1. Significant emission variation on a yearly scale

The measured N_2O emissions from the monitored activated sludge configurations all exhibit a similar strong seasonal pattern when plotted as a function of time (Figure 4). Emissions occurred mainly in one half-year (December–May), and they were very low in the other half-year (June–November). Daelman et al. (2015) also reported a seasonal emission pattern with similarly high differences on a year (Daelman et al., 2015). Emissions at Kralingseveer showed a very similar temperature variation, but their yearly peak shifted towards the temperature rise between March and June. Overall, the period of the emission peaks in this study were more distinct and shorter during the campaigns in this study than at the Kralingseveer WWTP. The seasonal emission patterns assessed by Kosonen et al. (2016) at the Viikinmäki WWTP were not as pronounced as in the other studies, which could be linked to the higher wastewater temperatures (20–40°C) (Kosonen et al., 2016). Additionally, the wastewater temperatures at Viikinmäki WWTP do not exhibit the seasonal profile typical of WWTPs connected to combined sewer systems in the temperate climate zone. The emission variation monitored in this study confirms the necessity of long-term (≥ 1 year) monitoring for the assessment of emission factors (Daelman et al., 2013a).

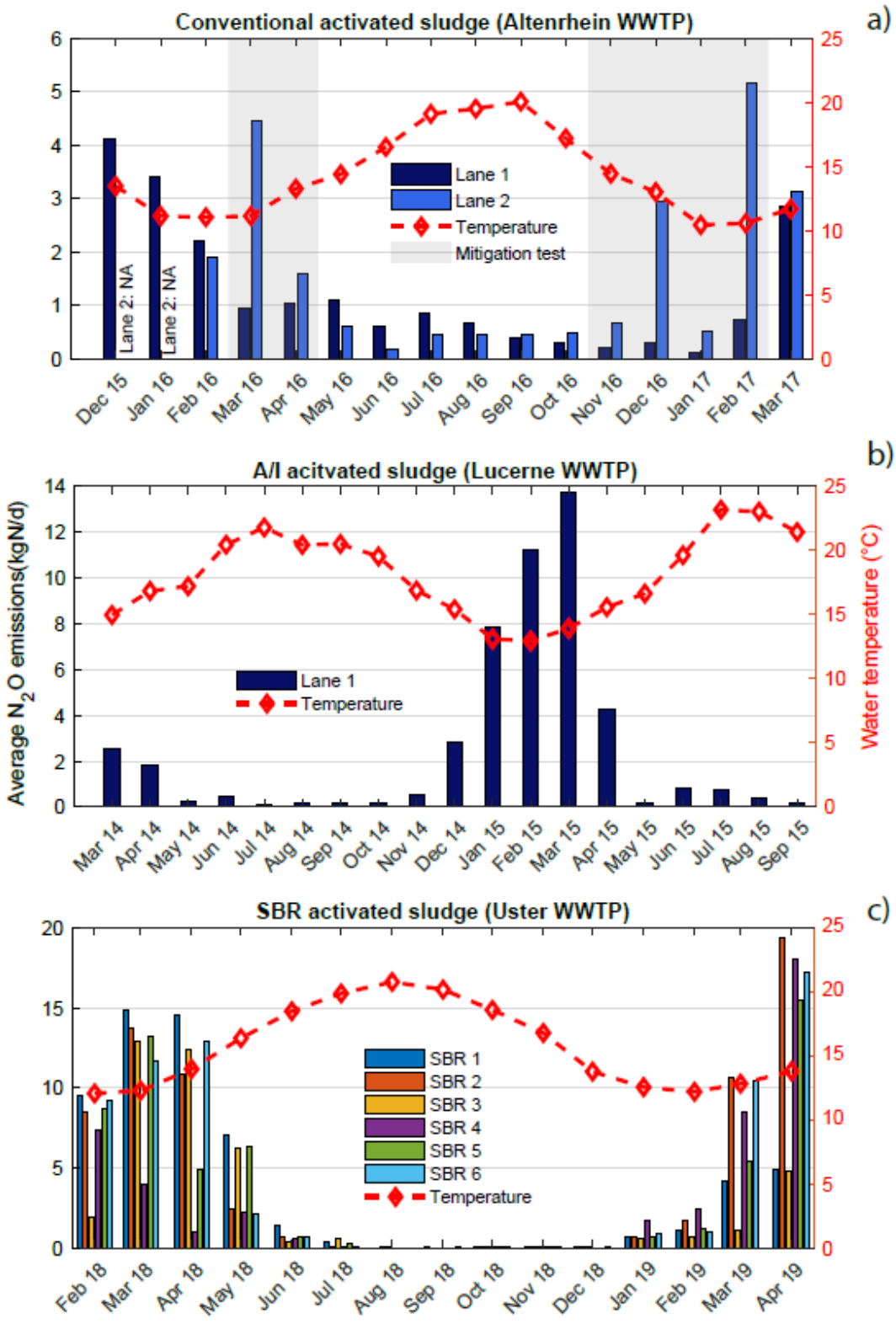

Figure 4 a) Monthly mean N_2O emissions of the activated sludge lane 1 and the fixed-bed reactor, and wastewater temperatures at Altenrhein WWTP. Grey background indicates times when reject water was omitted from the feed to lane. The activated sludge lanes 2 and 3 and the fixed bed reactors treated all the reject water during this period. In Dec 15, Jan 16 lane 2 was not monitored b) Monthly mean N_2O emissions and wastewater temperatures at Lucerne WWTP c) Monthly mean N_2O emissions and wastewater temperatures at Lucerne WWTP

A potential cause for the high emission variation could be failure of nitrification. In the Lucerne and the Uster campaign, nitrite concentrations peaked with N_2O emissions, which could be expected according to literature (Colliver and Stephenson, 2000). However, in the Lucerne campaign nitrite peaked also in summer while emissions were low. Another potential cause is the reduced denitrification capacity of the WWTP at cold temperatures (Conthe et al., 2018). In the Uster campaign, the dramatic emission variation among the reactors clearly speaks against this. Linear and non-linear regression models show that the seasonal variation of the Altenrhein and the Lucerne campaign cannot be explained with the available operational data (SI). We conclude that at least one important driver governing the high emission variation in the yearly course has not yet been identified. The necessary information to solve this challenges may be extracted from newer data types, such as microbiological data (Stein, 2018), inorganic carbon measurements (Peng et al., 2016), and concentration of isotopomers (Ostrom and Ostrom, 2017). For example, the measurement of microbial diversity and gene, protein, and metabolites expression has been applied successfully to explain variations in lab-scale measurements of N_2O emissions (Ge et al., 2018; Perez-Garcia et al., 2014).

The yearly average N_2O emission factor from the reactors monitored at the three WWTPs are summarized in Table 2.The SBR system at the Uster WWTP exhibited the highest emissions with 2.4%. The emission values of each reactor are shown in the SI (section A4). The A/I system at Lucerne WWTP was shown to have the lowest emissions with 1%. The average emission factor assessed in this study is 1.7% of the total incoming nitrogen load. The high standard deviations of the emissions reflect the strong emission variability (Table 2). In general, the emissions are in the same order of magnitude as the values assessed in the other two long-term studies published, where N_2O emission factors of 2.8% were found by Daelman et al. (2015) (Kralingseveer WWTP) and 1.9% by Kosonen et al. (2016) (Viikinmäki WWTP).

Table 2 Yearly average emissions and standard deveations assessed in the monitoring campaigns; **L**: lane number. **CAS**: conventional activated sludge plant of Altenrhein; **A/I**: alternating / intermitting plant of Lucerne; **SBR**: sequencing batch reactor of Uster, all six lanes monitored separately.

WWTP	Yearly average emissions		Standard deviation
	kgN_2O-N/year	% of total nitrogen load	kgN_2O-N/year
CAS, L1 CAS, L2	537 749	1.6 2.0	701 795
A/I, L1	1204	1.0	1737
SBR, L1 to L6 SBR, average	447-1172 805	 2.4	824-1059 976

Considering all published long-term monitoring campaigns, the average emission factor is 1.9%, which is in the same range as the value suggested by the IPCC guidelines (1.6%). However, more long-term campaigns are required given the site-specific variation of observed emission factors as well as the current lack of understanding differences amongst plant types (IPCC, 2019). Assuming the long-term monitoring campaign's emission factor, the contribution of wastewater treatment to the total N_2O emissions would increase from 4% to 19% for Switzerland (SI).

The emission factors monitored in this study confirm the relevance of N_2O to the greenhouse gas balance of WWTP: it is quantitatively more important than the indirect emissions caused by WWTPs' electricity consumption. An N_2O emission factor of 2% causes 40% higher emissions than a WWTP powered exclusively by coal (Larsen, 2015; Wunderlin et al., 2013). Hence, the N_2O emission factors assessed in our study confirm that N_2O is the most important GHG source from WWTP.

3.2. Low spatial emission variation

At all plants, abrupt changes from low emissions to high emissions were observed to occur over a few days. For example, at Lucerne WWTP, a 7 month low emission period was terminated by an emission rise from <0.2% to 4% within 4 days at the end of December 2014

277 (Figure 5a). Similarly, dramatic changes in N_2O emissions were observed on a daily basis at
278 Uster WWTP, as shown in Figure 5b. The rapid dynamic highlights confirm the previously
279 discussed importance of online monitoring approaches with sufficient temporal resolution
280 (Daelman et al., 2013a). Typical daily N_2O emission profiles are shown in the SI (A3 Daily
281 variation).

282 In the same periods, the spatial emission distribution remained relatively stable. In the
283 monitoring campaigns of this study, the spatial emission variation was generally not as
284 pronounced as the temporal variation within one lane. However, systematic differences were
285 also seen in this study in the A/I and the CAS reactors at Lucerne (Figure 5a) and Altenrhein
286 WWTP. In contrast, all three sampling points in one SBR reactor at Uster WWTP exhibited
287 comparable N_2O concentrations (Figure 5b).

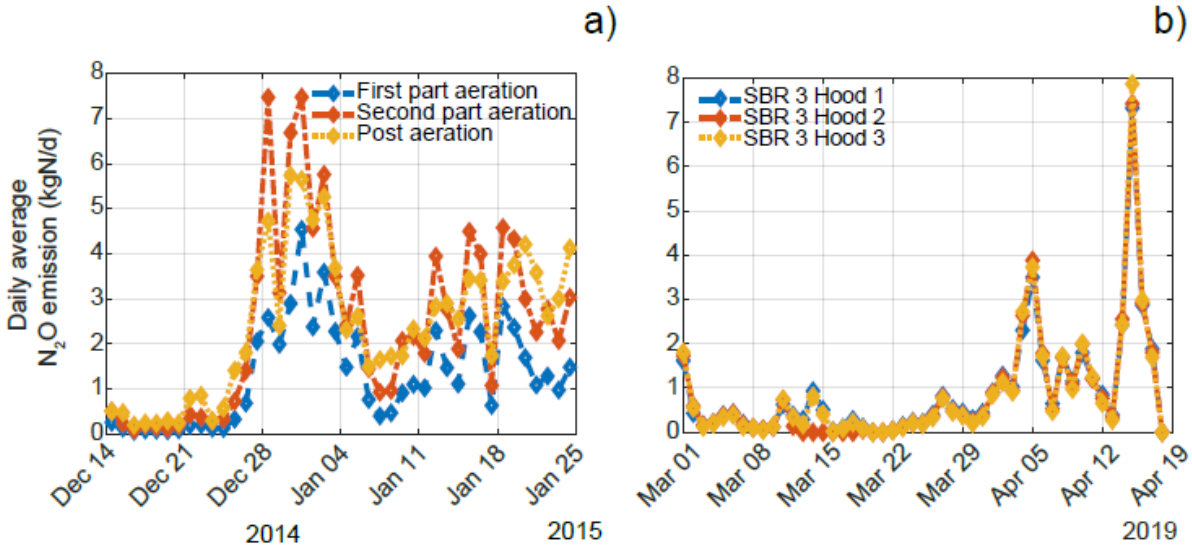

Figure 5 Spatial distribution of N_2O emissions within one lane as daily average at a) CAS plant (Lucerne WWTP) and b) SBR plant (Uster WWTP).

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289 Aboobakar et al. (2013) found high spatial variation (0.01-0.1%) in a CAS process; however,
290 this was paired with a low overall emission factor of 0.1% and a short-term emission period
291 (Aboobakar et al., 2013). For a two-step activated sludge process, the spatial distribution was
292 shown to be substantial (0.6-3.5%), with significantly higher emissions in the second step (Pan

et al., 2016). This is to be expected, since nitrification occurs predominantly in the second step. It is hypothesized that multiple measuring points are required if substrate gradients or different aeration strategies are applied along a lane. This is typically the case for lanes featuring plug-flow characteristics. Multiple measurement points are not required in a single SBR reactor.

3.3. How many lanes to monitor?

The emissions from parallel lanes in both plants with continuous feeding (CAS, A/I) exhibited strong variations from day to day, but these were reproducible across lanes even though these were operated with completely separated activated sludges (Figure 6a). In cases of uneven substrate load distribution between lanes, emissions can be significantly different. Two mitigation test phases between March 16 and April 16 and between November 16 and March 17 at Altenrhein WWTP provide an extreme example: During these phases, lane 2 treated part of the reject water from the sludge treatment, but lane 1 did not. During the second period, N_2O emissions from lane 2 increased dramatically to values around 6 kg N_2O-N/d. Emissions from lane 1 remained low at roughly 1 kg N_2O-N/d. The resulting emissions relative to the influent load for lane 1 and lane 2 were 0.7% and 2.7% during the first experiment and 0.5% and 3.1% during the second experiment. The increase in N_2O emissions of 80% was clearly higher than the increase in nitrogen load of 30%.

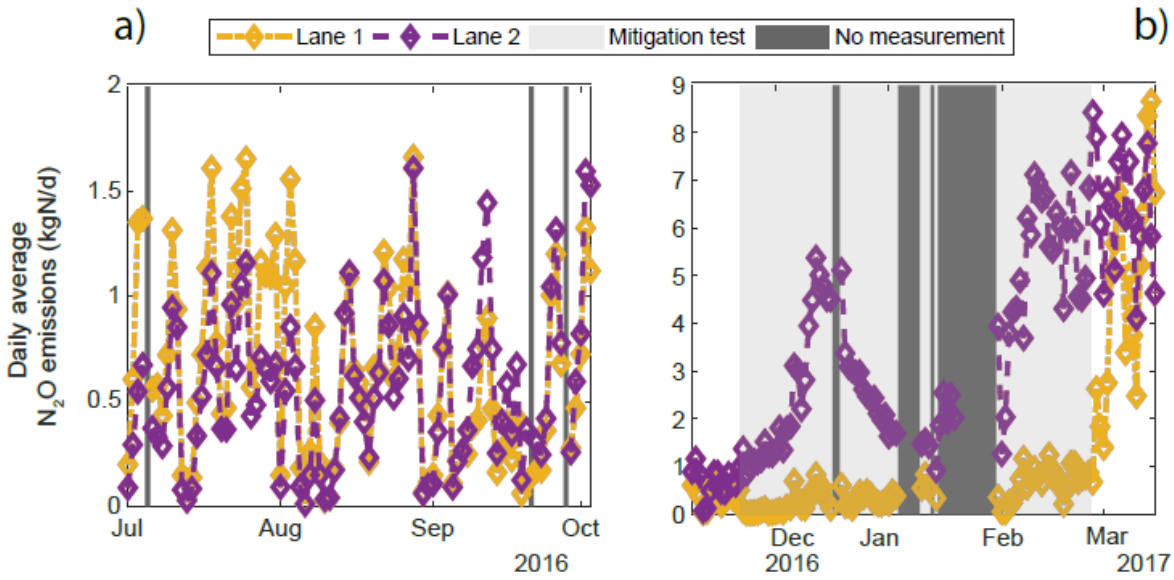

Figure 6 N_2O emissions of two parallel activated sludge lanes operated with separated sludge lanes at Altenrhein WWTP during a) the same influent composition and b) different influent composition: activated sludge lane 1 was operated without reject waters in the feed; activated sludge lane 2 was fed with wastewater containing reject water.

310

311 It is expected that inhomogeneous influent load can be caused not only by WWTP-internal
312 recycles but also other factors, such as incomplete mixing in the sewer system or uneven
313 preferential flows in the influent distribution. Key performance indicators of the WWTP, such
314 as oxygen requirement and effluent concentration, are expected to be indicative of
315 inhomogeneous influent loads.

316 Emission from discontinuously fed reactors may exhibit different emission profiles, as was
317 seen in the monitored SBR system (Figure 4c). The monthly average emissions of all reactors
318 differed substantially during the whole monitoring campaign. Based on the finding from parallel
319 CAS lanes (Figure 6a), it is expected that parallel-fed SBR reactors also exhibit parallel N_2O
320 emissions. It still needs to be confirmed that monitoring a restricted number of reactors is
321 sufficiently representative in this case.

3.4. Refined monitoring strategy

The monitoring campaigns described above allow a refined monitoring strategy to be derived for assessing N_2O emission factors and for generating data for identification and validation of full-scale GHG emission models.

Both emission factors and models should be based on data series of at least a 1 year duration, since seasonal emission patterns can be very conspicuous (Figure 4; (Daelman et al., 2015; Kosonen et al., 2016). Given the short-term variability, a sampling frequency higher or equal to twice per hour for each point of measurement is required (Daelman et al., 2013a). For a flux-chamber-based monitoring study of open reactors, the choice of the number and the positioning of flux chambers is crucial to assessing a representative emission factor. A decision tree for such a representative monitoring campaign is provided in Figure 7.

To design a sampling campaign, the following criteria are considered crucial:

a) **Homogenous influent and continuous feeding of all lanes**: All lanes fed simultaneously and performing comparably (i.e. supplied with comparable amounts of air and exhibiting similar soluble O_2 concentration and effluent concentrations) can be assumed to exhibit comparable N_2O emission dynamics (Figure 6a). Inhomogeneous influent may result from WWTP-internal recycles (e.g. supernatant from digesters; Figure 6b) or from incomplete mixing of the influent sewage. At most full-scale sites, it is easier to identify feed heterogeneity by comparing key performance indicators among lanes than by sampling the influent of different lanes separately. Also, in cases of intermittent feeding not occurring simultaneously for different lanes, separate monitoring is required due to variations in the distribution of the daily incoming load.
b) **Mixing conditions**: reactors that are completely stirred during aeration do not require multiple off-gas measuring points (Figure 5b); this is mostly the case for SBR reactors.

c) **Cascaded lanes:** Lanes featuring spatial concentration gradients require an adequate number of measuring points. This is also the case if the density of aeration units varies along a lane: each section requires one measuring point. A minimum of three measuring points (e.g. after 1/6 of the reactor length, in the middle and at 5/6) is suitable when a lane exhibits no subdivision into cascades and aerators are evenly distributed (Figure 5a).

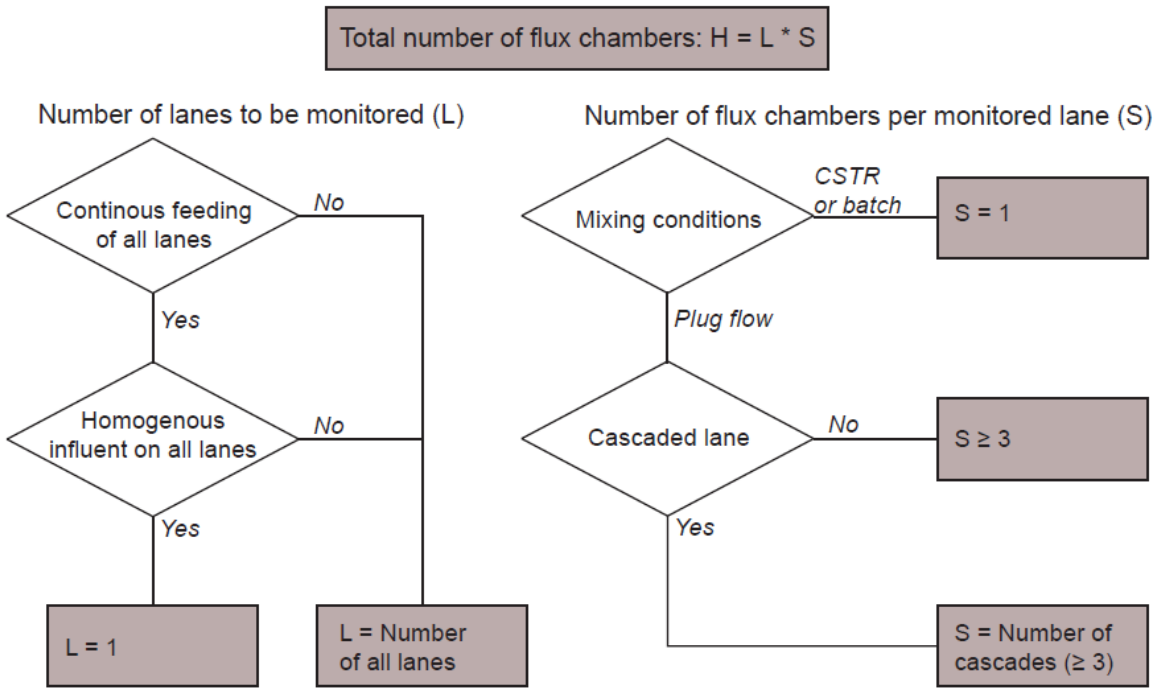

Figure 7 Decisision tree for the selection of the number of flux chambers and the placement on a particular lane. H = total number of flux chambers, L = number of lanes to be monitored, S = number of flux chambers per monitored lane

3.5. Outlook

Given a) that short and long-term N_2O emission variations are conspicuous, and b) that no model is available that can describe and quantify all sources of N2O emissions , it is advisable to plan monitoring campaigns with some redundancy, such as in the number of measuring points. Nevertheless, it must be acknowledged that the uncertainty of a monitoring campaign is not only dependent on the number and positioning of the measuring points but also on such

359 other factors as the long-term accuracy of the measuring device and emission changes from
360 year to year (Figure 4b).

361 Reliable monitoring strategies applicable to a range of treatment schemes are deemed of great
362 importance to improving understanding of the mechanisms relevant to N_2O emissions in
363 WWTPs (Vasilaki et al., 2019). Since the majority of WWTPs are composed of open reactors,
364 a flux-chamber-based measurement approach is required. The present paper therefore
365 contributes to a critical evaluation of this off-gas sampling method. A significant simplification
366 of the required monitoring effort is expected only once all the important parameters influencing
367 N_2O emission have been identified and a reliable model is available. At present, we consider
368 the number of publicly available data sets describing long-term variations in GHG emissions
369 too low to achieve this. Until then, model identification and validation are expected to be
370 feasible only with reliable and representative long-term monitoring data. Consequently, further
371 long-term monitoring campaigns are necessary to establish a broadly supported emission
372 factor for N_2O from wastewater treatment (Vasilaki et al., 2019).

373 The separate treatment of reject water was shown to be an efficient measure for reducing N_2O
374 emissions (Figure 6b). In the Altenrhein WWTP, about 80% of the emissions could be reduced,
375 so the emission increase during winter could be avoided almost completely. The fact that this
376 resulted from reducing the influent nitrogen load by only 30% shows that quantitative modelling
377 cannot be based on simple linear correlation with standard parameters. This promising result
378 also confirms the practical value of a better quantitative understanding of these emissions,
379 since solutions for significantly reducing them are feasible in at least some cases.

4. Conclusions

- This study confirms the high N_2O emission factors suggested in recent literature for three different activated sludge process configurations: N_2O is the most important GHG caused by wastewater treatment.
- The temporal emission variation (short- and long-term) was shown to be significant for all process configurations, while the spatial emissions variation along a single lane is only significant in flow through systems with plug-flow characteristics. Here, multiple monitoring spots are required per lane.
- On WWTPs with inhomogeneous loading or discontinuous feeding of different lanes, monitoring of all or at least representative lanes is required to calculate emission factors reliably.
- A separate reject water treatment is a promising strategy to mitigate N_2O emissions from biological treatment.

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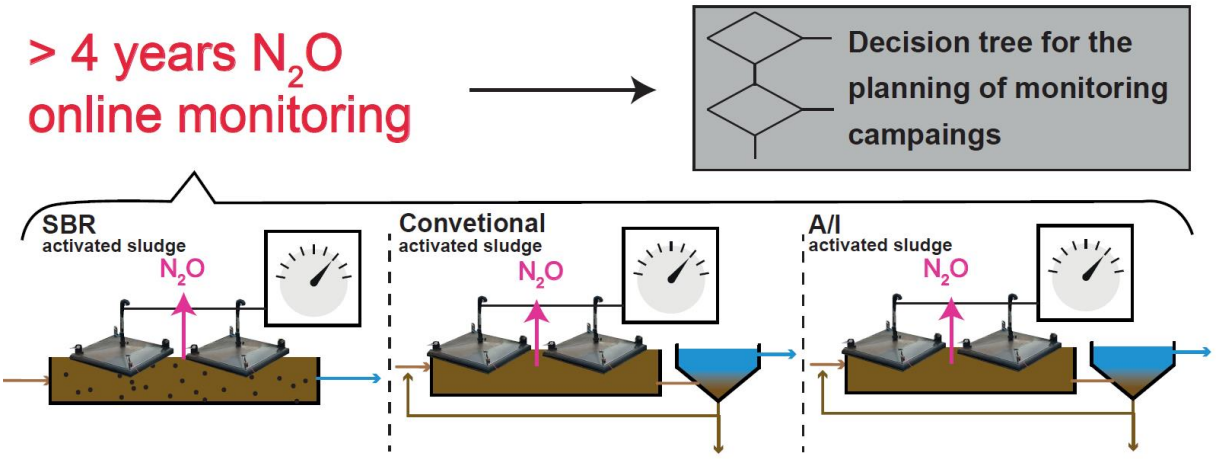

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