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Critical review on end-of-pipe technologies for nitrous oxide removal as part of a novel comprehensive concept for greenhouse gas emission mitigation at wastewater treatment plants

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ABSTRACT

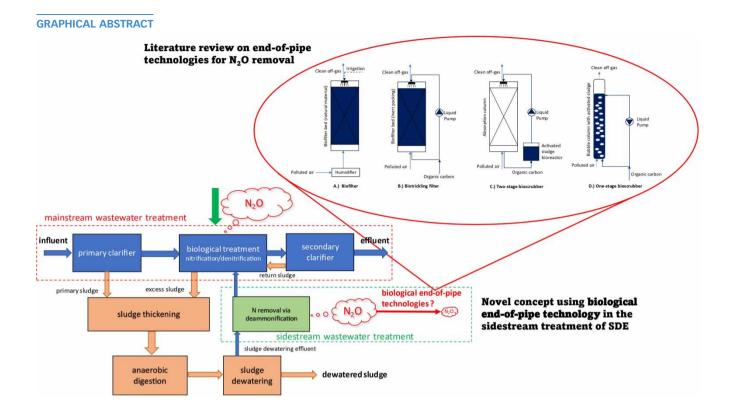
The greenhouse gas nitrous oxide, a byproduct of the biological nitrogen removal in wastewater treatment plants (WWTPs), strongly impacts the carbon footprint of the wastewater sector due to its high global warming potential. Although plant design and operational process optimization can reduce N_2O generation and emission, complete avoidance is not possible according to the current knowledge. While physical-chemical end-of-pipe technologies are available for the removal of nitrous oxide in industrial off-gases, a feasible treatment for the exhaust air of WWTPs still needs to be established. This paper critically reviews the currently available treatment technologies, with particular focus on biological systems, as bioscrubbers. The review indicated that implementing N_2O removal technologies is more feasible in sidestream than in the mainstream wastewater treatment. This is primarily due to the smaller area required for exhaust air collection, lower air flowrates, and higher N_2O concentrations observed in the sidestream. Therefore, the proposed concept focuses on sidestream application and is based on the following two main pillars: (i) introducing sidestream deammonification of the sludge dewatering effluent to deplete the N_2O emission factor in the mainstream biological stage and (ii) treating the N_2O -rich exhaust air from the deammonification process using a denitrifying bioscrubber with wastewater as an organic carbon source.

Key words: bioscrubber, deammonification, nitrous oxide, off-gas treatment, sidestream treatment, wastewater treatment

HIGHLIGHTS

- Established physical-chemical N₂O end-of-pipe air treatment technologies show limitations when implemented at WWTPs.
- Denitrifying bioscrubbers are a feasible option for treating N₂O-rich exhaust air from sidestream treatment of WWTPs.
- Implementation of sidestream deammonification and treatment of exhaust air in the sidesstream can significantly reduce the overall N₂O emission of a WWTP.

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1. INTRODUCTION

The greenhouse gas (GHG) nitrous oxide (N₂O) is formed in wastewater treatment plants (WWTPs) as a byproduct of biological nitrogen removal during nitrification and denitrification, and it is partially emitted into the air by aeration essential for biological wastewater treatment. Due to its high global warming potential (273 g CO₂e/gN₂O, IPCC 2023), N₂O often represents a major source of GHG emission from WWTPs (e.g. Marinelli *et al.* 2021).

The estimated N_2O emission from municipal WWTPs in the European Union (EU 27) contributes 7,683 kt $CO_2e/year$, accounting for about 0.25% of the overall GHG emission at the European level (EEA 2023). Despite the relatively low impact on GHG emissions at a global level, N_2O mitigation strategies are required to ensure a sustainable wastewater treatment sector, as also addressed by the recast urban wastewater treatment directive (EU 2024/3019).

Two main N₂O generation pathways during the autotrophic nitrification process have been reported in literature: (i) the hydroxylamine (NH₂OH) oxidation and (ii) the nitrifier denitrification. In the first pathway, N₂O formation is probably attributed to the accumulation of transient NH₂OH within the first nitrification step during the oxidation of ammonium (NH₄⁺) to nitrite (NO₂⁻) by ammonia-oxidizing bacteria (AOB), where N₂O generation as a byproduct increases at higher NH₄⁺ oxidation rate (e.g. Ahn *et al.* 2010; Law *et al.* 2012; Ribera-Guardia & Pijuan 2017). Nitrifier denitrification might be predominant under limited dissolved oxygen (DO) conditions whereby AOB reduces a part of the produced NO₂⁻ to nitric oxide (NO) and N₂O, using NH₂OH oxidation as the electron donor (e.g., Peng *et al.* 2014; Tallec *et al.* 2006; Ye *et al.* 2022).

On the other hand, during complete heterotrophic denitrification, N_2O is formed as an obligate intermediate within the reduction steps of nitrate (NO_3^-) to N_2 . In this process, N_2O is reduced to N_2 by nitrous oxide reductase. Therefore, the heterotrophic denitrification pathway is both a source and sink for N_2O in WWTPs. As a matter of fact, Conthe *et al.* (2019) clearly showed that heterotrophic denitrifiers can reduce more N_2O to N_2 than the amount of N_2O produced during the denitrification cascade. This pattern is particularly pronounced when there is an ample supply of easily degradable organic matter. Thus, the overall N_2O emission of an activated sludge tank is the result of the interplay between nitrification and denitrification as source and sink of N_2O .

Two main point sources of N_2O emissions in a WWTP can be distinguished: (i) the aerated activated sludge tank in the mainstream, where N_2O concentrations in the exhaust air usually range from 1 to 400 ppmv (Valkova *et al.* 2021) and the

air flowrate is high; (ii) the sidestream treatment of the effluent of anaerobic sludge dewatering effluent (SDE), e.g. by deam-monification, where N_2O concentrations in the exhaust air can reach up to 3,000 ppmv (Kosonen *et al.* 2016; Mampaey *et al.* 2016) but the air flowrate is much lower.

SDE exhibits very high NH₄⁺ concentrations between 500 and 1,500 mg N/L compared to municipal wastewater concentration (Metcalf & Eddy 2014). The deammonification process, which is a shortcut nitrogen removal pathway bypassing conventional nitrification and heterotrophic denitrification processes, can be applied in sidestream treatment to achieve autotrophic nitrogen removal of the SDE (Lackner *et al.* 2014; Gao & Xiang 2021). It combines two key biological processes: partial nitritation and anaerobic ammonium oxidization (anammox) (Lackner *et al.* 2014; Gao & Xiang 2021), as depicted in Equation (1) and (2) (Lotti *et al.* 2014).

Nitritation process:

$$NH_4^+ + 1.5 O_2 \leftrightarrow NO_2^- + 2 H^+ + H_2O$$
 (1)

Anammox process:

$$\begin{array}{l} NH_4^+ + 1.146 \ NO_2^- + 0.071 \ HCO_3^- + 0.057 H^+ \leftrightarrow \\ 0.986 \ N_2 + 0.161 \ NO_3^- + 2.002 \ H_2O + 0.071 \ CH_{1.74}O_{0.31}N_{0.2} \end{array} \tag{2}$$

In the first step, due to the alkalinity limitation in the SDE, only about half of the NH_4^+ is oxidized to NO_2^- by AOB. Controlled operating conditions (e.g. limited aeration) are maintained to inhibit the complete oxidation of NO_2^- to NO_3^- by nitrite-oxidizing bacteria (NOB). In the second step, the residual NH_4^+ and the produced NO_2^- are converted to N_2 and NO_3^- by autotrophic anammox bacteria without the need of an external carbon source as an electron donor, unlike the heterotrophic denitrification pathway. The two processes can take place in two separate reactors (two-stage configuration) or in one-stage configuration operated with intermittent aeration.

Process-related N₂O emissions from mainstream activated sludge tanks vary widely, ranging from 0.0025 to 5.6% (average 0.87%) of the influent nitrogen load, depending on operational conditions and measurement methodologies (Kampschreur et al. 2009; Vasilaki et al. 2019). The N₂O emissions are strongly influenced by various process parameters. NH₄⁺ and NO₂⁻ concentrations above 1 mg N/L have been identified as the main triggers of N₂O generation by AOB (Kampschreur et al. 2009; Law et al. 2012). The level of aeration in the treatment process affects DO availability and microbial activity, directly impacting N₂O production and reduction rates. The direct impact of water temperature on N₂O generation pathways remains unclear, however its indirect impact – such as promoting NO₂⁻ accumulation during or after cold seasons due to adapted aeration strategy and/or a shift in the nitrifiers population – have been reported (Gruber et al. 2021). Optimal pH value is also essential to minimize N₂O production (Foley et al. 2010). A further impacting factor is the COD and nitrogen loading pattern entering the biological stage. Nitrogen loading peaks leading to fluctuation in the NH₄⁺ effluent concentration, were shown to increase N₂O generation and emissions. The availability of organic carbon sources (e.g., wastewater organics, external carbon dosing) impacts denitrification rates and the risk of N₂O accumulation. Limited availability of carbon source can result in incomplete denitrification leading to N₂O accumulation and emission (Daelman et al. 2013; Guo et al. 2017; Ribera-Guardia & Pijuan 2017).

Furthermore, during the planning and upgrading of WWTPs, ensuring a high sludge age (sludge retention time) is essential to establish reserve nitrification capacity that can buffer nitrogen loading peaks without adversely affecting effluent concentrations. Recommended strategies to reduce N₂O emissions in activated sludge tanks include preventing NH₄⁺ concentration gradients, ensuring flexible aerobic/anoxic operation, employing intermittent aeration and simultaneous denitrification, and avoiding excessive aeration (Desloover *et al.* 2012; Valkova *et al.* 2021; Xie *et al.* 2023).

High NH₄⁺ and NO₂⁻ concentrations occurring in the sidestream treatment of SDE have been identified in both laboratory and large-scale research works as the triggers for N₂O production, especially in the partial nitritation step of the deammonification process (Law *et al.* 2012; Mampaey *et al.* 2016). According to the literature, the N₂O emission factor for the sidestream deammonification process with suspended biomass and one-stage process configuration ranges between 0.6% (Joss *et al.* 2009) and 2% of the incoming N load (Castro-Barros *et al.* 2015), which is higher than the emission factors reported for mainstream nitrification/denitrification processes. This is also confirmed by Song *et al.* (2024), who report in their review for one-stage deammonification an average N₂O emission factor of 1.25% (range: 0.4–2%) referred to nitrogen

influent. One-stage deammonification exhibits lower emissions than the two-stage configuration because in the one-stage option nitrite accumulation is limited. Optimization of process operation contributes to reducing the emission factor to levels within the lower range reported in the literature.

To summarize, scientific works so far confirm that design and operational process optimizations can reduce but not eliminate N_2O emissions (Duan *et al.* 2020). To enable further mitigation, end-of-pipe treatment technologies may be applied to remove residual N_2O from the exhaust air emitted from aerated tanks. A fundamental prerequisite for this is the encapsulation of tanks to collect the N_2O -rich exhaust air streams. While this can be achieved relatively easily for sidestream treatment units, it presents an economical effort for the large surface areas typical of mainstream treatment tanks. In addition, any chosen treatment technology must be suitable for operating under water vapor saturation, high oxygen concentrations, and at ambient temperature and pressure. Moreover, the low and fluctuating concentrations of N_2O in the exhaust air can significantly limit removal efficiency (RE).

Currently, there is no established technology to remove N_2O emission either in the mainstream or in the sidestream biological treatment steps. This highlights the need to identify suitable technologies for N_2O mitigation and to evaluate how they could be practically implemented within WWTP operations. The aim of this review work is therefore (i) to provide an overview of the existing end-of-pipe technologies for N_2O emission control, with focus on those that have been already investigated for N_2O emission mitigation at WWTPs and (ii) to propose and critically assess a novel comprehensive N_2O mitigation approach involving the integration of the deammonification processes for SDE in the sidestream, combined with the biological treatment of the exhaust air of this stage.

Sidestream treatment of ammonium-rich SDE offers several advantages in reducing overall N_2O emissions from WWTPs. If it is considered that the deammonification process reduces the NH_4^+ load of SDE up to 90% (Lackner *et al.* 2014) and that the load in the sidestream can make up 20% of the incoming nitrogen load of the plant (Baumgartner *et al.* 2022), a substantial decrease in the nitrogen entering the mainstream biological process is achieved. The lower nitrogen load to be oxidized in the mainstream creates advantageous operational conditions to deplete the N_2O emission in this stage. Furthermore, enhanced biological nitrogen removal in the WWTP contributes to decrease N_2O formation and emissions in the receiving water bodies.

However, a disadvantage of the sidestream SDE treatment using deammonification is the intensive production and emission of N_2O during partial nitritation. To avoid this additional N_2O emission, the innovative approach proposed in this work also includes the collection of the exhaust air from the deammonification tank for subsequent treatment with a suitable end-of-pipe technology. Air extraction and N_2O removal in the sidestream are technically simpler and more cost-effective than in the mainstream. The novelty of the concept also relies in shifting the treatment of N_2O -rich exhaust air from the mainstream aeration tank to the sidestream treatment of SDE. However, operating the deammonification process in the sidestream remains challenging due to issues like biomass retention and aeration control (Lackner *et al.* 2014). These considerations will be thoroughly discussed within the proposed concept.

2. STATE OF THE ART END-OF-PIPE TECHNOLOGIES FOR N2O REMOVAL FROM EXHAUST AIR

A diagram categorizing available end-of-pipe technologies for N_2O removal in exhaust air streams is shown in Figure 1. Physical-chemical air treatment technologies as catalytic-based processes are widely applied in the chemical industry to reduce the environmental impact of N_2O emissions. However, these technologies have hardly been implemented to treat exhaust air from municipal WWTPs. This is attributed to the high oxygen (O_2) levels, the low and fluctuating N_2O concentrations, and the lack of the necessary pressures and temperatures of the exhaust air, which increases energy demand and costs consistently (Frutos *et al.* 2018).

On the other hand, biological treatment technologies as biofilters are employed in WWTP for odor control. Nevertheless, their application to treat N_2O -rich airflows is limited, despite of the several advantages. This is likely due to the challenges associated with achieving extensive and stable N_2O denitrification as the main biological removal pathway.

Removing N_2O from exhaust air in the mainstream wastewater line of WWTPs requires the management of large air flowrates with low, fluctuating N_2O concentrations across extensive aerated tank surfaces, leading to technical efforts and increased investment and operating costs. Sidestream treatment of SDE (e.g. via deammonification), on the other hand, is more practical due to its smaller tank footprint to be covered, higher N_2O concentrations, and lower air flowrates. Thus, focusing on sidestream exhaust air offers an advantage for N_2O mitigation efforts. These aspects have been considered in

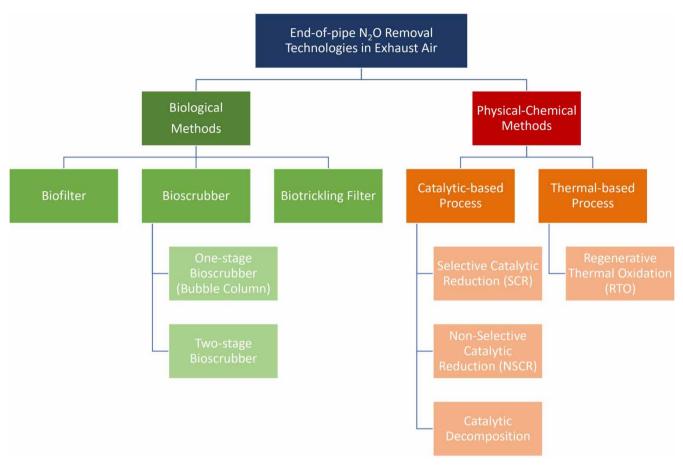


Figure 1 | Categorization of end-of-pipe technologies for N₂O removal in exhaust air streams.

the evaluation of the individual technologies within the review to determine which options are suitable and how they could be effectively integrated into the configuration of a WWTP.

2.1. Physical-chemical end-of-pipe treatment processes

Advancements in catalyst development have facilitated the application of effective physical–chemical technologies to reduce N_2O emissions especially in nitric and adipic acid manufacturing processes. These technologies offer the dual advantages of short empty bed residence times (EBRTs) and rapid start-up, along with small space requirements. Successfully implemented and operated in practice, they are widely adopted for treating large off-gas streams with high N_2O concentrations (Kennes & Veiga 2010; Estrada *et al.* 2012; Frutos *et al.* 2018).

Catalytic and thermal methods are commonly used to remove N_2O in industrial off-gas streams as shown in Figure 2. These methods operate at high pressures (up to 4 bar), temperatures (up to 1,000 °C), and low oxygen levels (<4% vol.). When required, preheating systems or oxygen-reducing agents are employed to ensure effective abatement (Shimizu *et al.* 2000). While these methods achieve high N_2O removal efficiencies (up to 100%), their drawbacks include high energy demands, unburned fuel emissions, fossil CO_2 generation, and hazardous waste from spent catalysts (Heck 1999). These factors limited the application of these technologies at WWTPs up to date.

2.1.1. Catalytic-based treatments

Catalytic decomposition directly converts N_2O into nitrogen and oxygen at temperatures between 300 and 500 °C without requiring a reductant dosage (Alves *et al.* 2022). This process is typically catalyzed by noble metals, perovskite-type oxides, or spinel-structured oxides (Flores-Lasluisa *et al.* 2022). In a study by Haber *et al.* (2004), the optimal RE achieved for N_2O using this technology at the pilot scale was 100% at 375 °C using Rh/ Na_2O /Al₂O₃ catalyst under a gas hourly

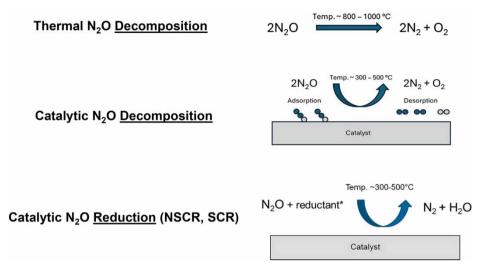


Figure 2 | Physical-chemical N₂O treatment paths adapted from Galle et al. (2001).

space velocity (GHSV, defined as volumetric flowrate of inlet gas per unit volume of catalyst bed) of 4,000 h^{-1} (EBRT = 0.9 s) with 10,000 ppmv N₂O diluted in nitrogen.

Catalytic-based treatments can be also applied in reductive processes, divided into two main categories of selective catalytic reduction (SCR) and non-selective catalytic reduction (NSCR).

SCR is widely used for NO_x and N_2O reduction involving injection of a reductant, typically ammonia (NH_3), into the offgas, where it reacts with N_2O in the presence of catalysts such as noble metals, metal oxides, or ion-exchanged zeolites to produce nitrogen (N_2) and H_2O (Alves *et al.* 2022). Studies using Fe/ZSM-5 demonstrated RE of up to 80% with GHSV of 18,000 h^{-1} (EBRT = 0.2 s) under optimal conditions for an industrial off-gas at temperatures between 300 and 500 °C with N_2O concentration of 500 ppmv (Centi & Vazzana 1999). In other study by Wang *et al.* (2018), Fe/SSZ-13 zeolite catalysts were investigated for the SCR of 540 ppmv N_2O by NH_3 dosing. Under optimal conditions, the Fe/SSZ-13 catalysts attained over 90% RE at temperatures around 500 °C. However, their performance may decline in WWTP-like conditions due to higher O_2 levels of the exhaust air.

NSCR was initially developed for the removal of nitrogen oxides (NO_x). It can effectively reduce N_2O via thermolysis and reducing agents such as hydrogen (H_2) or natural gas, reducing nitrogen oxides to nitrogen and water (Marnellos *et al.* 2004). One-stage systems operate at O_2 levels below 3% vol., while higher O_2 concentrations require a two-stage setup. Typical ignition temperatures range from 300 to 480 °C, with Pd/Al_2O_3 as the primary catalyst. Pilot studies achieved 70–90% RE for N_2O concentrations of 900–1,500 ppmv in GHSV of 60,000 h⁻¹ (EBRT = 0.06 s) (Pérez-Ramírez *et al.* 2003). Excess heat from this exothermic process (>730 °C for highly concentrated N_2O -laden off-gas) is often used for on-site steam generation, thus improving the energy balance of this application.

Full-scale applications at WWTPs have not been reported so far. In Denmark a catalytic decomposition process was operated at pilot scale at the WWTP Ejby Mølle in the time period of 2023–2024. Detailed information about the catalyst type used (manufacturer: NACAT company) could not be found. The process operates at temperatures between 420 and 470 °C. High removal efficiencies of 70 to 80% were achieved at high N_2O concentrations in the sidestream SDE treatment (approximately 1,000 ppmv). However, further investigations at HCR Syd WWTPs revealed that the presence of CH_4 and H_2S in the exhaust air can severely poison the catalyst. At a third Danish WWTP (Tårnby), measurements were conducted in the mainstream wastewater line at lower N_2O concentrations (5–20 ppmv), revealing that RE varied with inlet concentration. Approximately 15% removal was observed at the lower end of the concentration range, while efficiencies increased up to 80% at higher N_2O levels (Uri-Carreño 2024). However, the overall energy balance, expected investment and operating costs, and long-term process stability still require thorough evaluation.

2.1.2. Thermal-based treatment

Regenerative thermal oxidation (RTO) is a non-catalytic process for decomposition of N_2O in industrial off-gas streams. Incoming N_2O -rich air is preheated using the heat recovered from the treated gas, often via ceramic-based regenerative

heat exchangers. The preheated gas enters a combustion chamber, where temperatures of 800–1,000 °C thermally decompose N₂O into N₂ and O₂ (Shimizu *et al.* 2000; Galle *et al.* 2001).

RTO systems achieve N₂O destruction efficiency up to 99%. They are highly scalable and energy-efficient, though they require high capital investment and regular maintenance. Also, precise temperature control is essential to avoid incomplete decomposition or by-products like NO_x and dioxins. NO_x can become an issue at high N₂O concentrations, requiring downstream SCR. Further operational issues can occur in the presence of hydrochloric acid (HCl) or sulfur dioxide (SO₂), though these substances are typically absent in WWTP exhaust air (Galle *et al.* 2001; Kobler *et al.* 2022). Considering all of these aspects, their application in WWTPs is limited due to the high energy demand for achieving the required temperatures and for the exhaust air pre-treatment (e.g., particle removal) (Heck 1999).

Recent pilot-scale research at a Swiss WWTP (Bern) with airflows of 200–400 m³/h demonstrated the feasibility of RTO to treat N₂O-rich exhaust air from the covered aerated tanks of the mainstream and of the sidestream wastewater treatment lines. Operating at over 950 °C, the system achieved 85% N₂O removal with 88% heat recovery degree, the latter could be improved by better insulation (Kobler *et al.* 2022). The propane consumption for heating combustion chamber ranged between 0.005 and 0.009 kg/Nm³ of exhaust air. This RTO system treated approximately 450 m³ of inlet gas per m³ of combustion chamber per hour, corresponding to an EBRT of 8 s. The technology has recently been scaled up.

2.2. Biological end-of-pipe treatment processes

Over the past four decades, biological methods have been widely investigated at lab, pilot, and industrial scales to control industrial gas pollutants such as hydrogen sulfide (H_2S), NH_3 , NO_x , volatile organic compounds (VOCs), odors, and recently N_2O . These methods are cost-effective because operating at ambient conditions they require less energy as physical-chemical processes. In addition, they do not produce hazardous end-products, and do not need extra fuels, reducing agents, or expensive catalysts to operate (Barbusinski *et al.* 2017).

In these systems, heterotrophic denitrification plays a key role as degradation pathway for N_2O . Effective N_2O removal requires efficient transfer of N_2O from the gas phase to the liquid phase and the presence of biodegradable organic carbon. Organic carbon is essential for the biological reduction of N_2O to N_2 by heterotrophic bacteria and creating anoxic conditions necessary for denitrification (reductant function). High oxygen levels in the exhaust air from WWTP can hinder the process.

Biological air treatment methods (Figure 3) include biofilter (a), biotrickling filter (b), two-stage bioscrubber (c) and one-stage bioscrubber (d).

Table 1 compares the biological methods in different research works based on N_2O concentrations, EBRT, volumetric load in absorber, volumetric load in biological reactor, packing material, carbon source, and RE. These technologies are discussed in detail in the following sections.

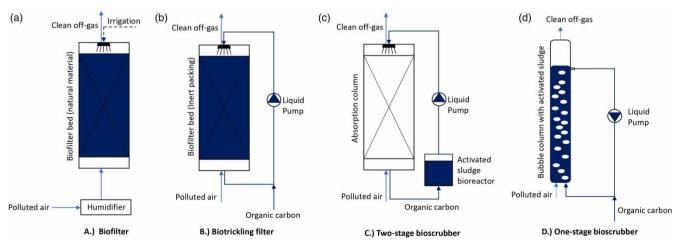


Figure 3 | Scheme of (a) biofilter; (b) biotrickling filter; (c) two-stage bioscrubber; (d) one-stage bioscrubber; biological process takes place in the blue-coloured volumes.

Table 1 | Summary of researches on biological end-of-pipe technologies for N₂O removal, including a comparison of key process characteristics

Reference	Technology	N_2 O concentration in inlet gas stream [ppmv]	EBRT ^a [min]	Hydraulic retention time ^b [min]	Packing material	Specific surface area of packing material [m²/m³]	Carbon source for denitrification –	Experiment duration [d]	N₂O RE [%]
Kühn & Brückner (1994)	Biofilter	512 in air	10	n.a. ^c	Heather peat and bark mulch humus	-	No carbon source added	13	20–30
Akdeniz <i>et al.</i> (2011)	Biofilter	0.43 in air	0.02-0.08	n.a.	Pine nugget, lava rocks	-	No carbon source added	180	0.7
Hood <i>et al</i> . (2011)	Biofilter	0.43–0.58 in air	0.12	n.a.	Compost and woodchips	-	No carbon source added	240	14–18
Yoon <i>et al</i> . (2017)	Biotrickling filter	100–200 in air and nitrogen	8–80	181	Polyurethan foam cubes	820	Raw wastewater, synthetic wastewater	22	4–80
Yoon <i>et al</i> . (2019)	Biotrickling filter	100 in air	8–80	181	Polyurethan foam cubes	820	Synthetic wastewater	25	28–52
Ramezani & Zamir (2022)	Biotrickling filter	500–4,150 in nitrogen	20	100	Polypropylene pall rings	480	Glucose, phenol	130	28–71
Han <i>et al</i> . (2023)	Biotrickling filter	4.8–96 in air ^d	2.4–4.8	167	Polyurethan foam cubes	-	Raw wastewater	256	43–58
Frutos <i>et al</i> . (2015)	2-stage bioscrubber	104 in air	3	120–1,740	A ^e : Polyurethan foam cubes, B ^f : Kaldnes rings	A: -, B: 950	Methanol	95	6–40
Frutos <i>et al.</i> (2016)	2-stage bioscrubber	95–104 in air	3–80	120-1,740	A: Kaldnes rings B: Polyurethan foam cubes	A: 950, B: -	Synthetic wastewater	140	8–92
Frutos <i>et al.</i> (2017a)	1-stage bioscrubber	3,560 in nitrogen	17	n.a.	n.a.	n.a.	Methanol, glycerol, Ac-HAc	180	81–91
Frutos <i>et al.</i> (2017b)	1-stage bioscrubber	3,520–3,560 in nitrogen	17	n.a.	n.a.	n.a.	Methanol	180	57–87
Valipoor <i>et al.</i> (2025)	1-stage bioscrubber	600 in air	51	77	n.a.	n.a.	wastewater and activated sludge mixture	300	60-85

^aEmpty bed residence time of the air in all one-stage technologies and for absorption column in two-stage bioscrubber.

^bHydraulic retention time of liquid in all one-stage technologies and for biological stage of two-stage bioscrubber.

cn.a. = not applicable.

^dExhaust air of activated sludge tank in WWTP.

^ePacking material of absorption column.

^fPacking material of biological stage.

2.2.1. Biofilter

A biofilter is a fixed-bed biological reactor, designed for the removal of gaseous substances rather than solid particles. In a biofilter, microorganisms are attached to a solid medium that also provides nutrients and organic carbon for denitrification. To achieve high mass transfer and degradation of N₂O, biofilters use filling materials with high specific surface areas, such as composted bark mulch, wood chips, lava rocks, gravel, activated carbon, or other adsorbents. These materials must support microbial growth with properties like porosity, stability, moisture retention, and pH buffering capacity. Optimal temperatures for microorganisms in this process range from 5 to 40 °C. The filter material should be replaced every 2–5 years to maintain pollutant degradation efficiency (Akdeniz *et al.* 2011). For N₂O denitrification, the bed material must either supply biodegradable organic carbon or an external source must be introduced, usually through a liquid that is pumped, sprayed, or dripped.

There are limited studies on denitrifying biofilters for N_2O removal. Some pilot studies focused on treating exhaust air from pig farms. For example, Akdeniz *et al.* (2011), used pine nuggets and lava rock, achieving a low RE (0.7%) at an inlet N_2O concentration of 0.43 ppmv, EBRT of 5 s and at a relative humidity of 90%. In another study, Hood *et al.* (2011) used compost and wood chips, obtaining a RE of 14–18% with an inlet N_2O concentration of 0.43–0.58 ppmv and EBRT of 7 s. Low degradation in both studies was attributed to low N_2O levels, oxygen presence, and short residence times.

Kühn & Brückner (1994) investigated biofilters with varied bed materials and oxygen levels at lab scale. They found bark mulch as best option which could reduce N_2O at concentrations of 512 ppmv, even after operational pauses. However, some materials required pH buffering to prevent acidification. N_2O reduction efficiency ranged from 10 to 30%, depending on the material. The study highlighted challenges with fluctuating N_2O levels and high oxygen content in biofilters, suggesting bioscrubbers as a better option to manage unstable conditions.

Biofilters could be further developed to a cost-effective option for treating N_2O -rich air at stable loading conditions. However, they require significant space and maintenance. Loading and process conditions as oxygen concentration, humidity, pH value, and COD availability cannot be easily controlled within the bed.

2.2.2. Biotrickling filter

Biotrickling filters have emerged as an alternative technology addressing the limitations of conventional biofilters, such as the ageing of packing materials and non-uniform air distribution. These systems are characterized by an aqueous phase trickling over the filter bed, made of inert natural or synthetic material. This setup provides the nutrients necessary for microbial growth, enhances process control, and increases RE, particularly for air streams with high flowrates and pollutant concentrations (Rybarczyk *et al.* 2019). The operation of biotrickling filters relies on the establishment of biofilms on the packing material, usually seeded with sewage sludge, where pollutants like VOCs, odorous compounds, and N₂O are first dissolved in the aqueous phase and subsequently biodegraded in the biofilm (Mudliar *et al.* 2010). The rate limiting step in this process is the mass transfer of pollutants from gas to the liquid phase, particularly for pollutants with dimensionless Henry constant (Conc.gas/Conc.liquid) of below 1. The Henry's constant for N₂O is approximately 1.6 (Sander 2015). The mass transfer can be improved by increasing the specific surface area and contact time (Waweru *et al.* 2000).

Biotrickling filters face operational challenges such as excessive biomass growth, which can lead to clogging of the filter bed. Clogging reduces the efficiency of pollutant removal by limiting airflow and liquid flow through the medium, and raising maintenance costs (Barbusinski *et al.* 2017). Oxygen-rich conditions in the filter bed also inhibit the denitrification process, significantly reducing the capacity of the system to remove N_2O .

Yoon et al. (2017) developed a self-sustaining biofiltration system at lab scale, integrating a biotrickling filter packed with polyurethane foam cubes ($820 \text{ m}^2/\text{m}^3$) and using pre-screened municipal wastewater as trickling medium for gas-to-liquid transfer and biological N_2O reduction with an anaerobic activated sludge tank. This means that the first step is a biotrickling filter, where the mass transfer from gas to liquid occurs, and partial reduction of N_2O takes place. With self-sustaining, it is meant that the effluent of the biotrickling filter is recirculated to the anaerobic section of an activated sludge tank, where the degradation of residual N_2O – not fully degraded due to suboptimal anoxic conditions in the biotrickling filter – takes place. This system achieved removal efficiencies ranging from 4 to 80%, with EBRTs between 8 and 80 min and N_2O concentration varying between 100 and 200 ppmv. Nevertheless, the presence of oxygen compromised the denitrification process, diminishing overall performance. Building on this, Yoon et al. (2019) tested a full-scale biotrickling filter system at a municipal WWTP under realistic operational conditions. By configuring multiple biotrickling filters in series, the system established oxygen gradients within biofilms, which allowed for effective N_2O reduction. This arrangement achieved an RE of 52.5% and an

elimination capacity of $0.402 \text{ g N}_2\text{O}/(\text{m}^3.\text{h})$ with EBRT of 16 min, even in oxygen-rich conditions, demonstrating its potential for large-scale application.

Ramezani & Zamir (2022) investigated an anoxic biotrickling filter in lab scale for N_2O removal under varying organic carbon-to-nitrogen (C:N) ratios in the liquid phase. Using a synthetic gas stream with a high N_2O concentration (2,300 ppmv) diluted in N_2 and a C:N ratio of 1.5 (achieved by addition of phenol as carbon source), the system reached a 71% RE with trickling rate of 20 mL/min, and EBRT of 20 min. However, the experimental conditions are not transferable to municipal WWTPs due to the high oxygen concentrations present in the exhaust air.

Recently, Han *et al.* (2023) evaluated the potential of biotrickling filter, packed with polyurethane foams and fed by wastewater as trickling medium, as an end-of-pipe treatment for N_2O emissions from mainstream of WWTP. Over 165 days, the pilot-scale system achieved an average RE of $57.9 \pm 29.1\%$ for exhaust air containing N_2O concentrations of 16.6 ± 19.5 ppmv with trickling rate of 2.4 L/min. These results confirm the potential of biotrickling filtration in mitigating low and varying concentration N_2O emissions from the mainstream tanks. Also, the system achieved higher removal efficiencies when N_2O emissions were higher.

Overall, biotrickling filters represent a promising approach for mitigating N_2O emissions, particularly in scenarios with stable pollutant concentrations. However, challenges such as providing anoxic condition, surplus biomass management, and variability in exhaust air composition from municipal WWTPs necessitate further refinement and optimization.

2.2.3. Two-stage bioscrubber

A two-stage bioscrubber consists of an absorption column coupled with a biological reactor. In the absorption column, pollutants such as N₂O are transferred from the gas phase into a liquid phase, typically water, which serves as a washing liquid. This process is facilitated by a packing material with a high specific surface area, ensuring prolonged retention time for the liquid and maximizing its contact with the gas. The rate of absorption depends on the gas solubility and the concentration gradient. In the subsequent stage, the N₂O-laden liquid is pumped into a biological reactor, where it is denitrified. The biological reactor can employ either a fixed-bed biofilm system or a suspended growth system, such as an activated sludge tank. The treated effluent is then recirculated as washing liquid to the absorption column, enabling a continuous treatment loop (Mudliar *et al.* 2010; Barbusinski *et al.* 2017). To improve the RE in two-stage bioscrubbers, several strategies have been suggested. These include optimizing liquid recirculation rates, extending contact time to enhance the mass transfer and microbial activity, and employing packing materials with high surface-area-to-volume ratios. Additionally, monitoring and adjusting operational parameters such as pH value, and nutrients availability can significantly improve microbial performance (Barbusinski *et al.* 2017).

Similar to biofilters and biotrickling filters, sufficient amounts of readily degradable organic carbon (e.g. mechanically pretreated municipal wastewater) must be available in the biological reactor to deplete O_2 and ensure complete denitrification of N_2O to molecular nitrogen (Frutos *et al.* 2017a).

Complete denitrification also requires that the biological reactor has a sufficient turnover rate of heterotrophic bacteria to carry out the process. This means that, depending on the load of O_2 and N_2O to be treated, either flocs or biofilm are required (Frutos *et al.* 2016).

One advantage of a two-stage bioscrubber over a biotrickling filter is that the recirculated water is biologically treated, which results in fewer problems with clogging of the absorption column. Working with treated wastewater can also increase the N_2O transfer efficiency into the liquid. The separate absorption and biodegradation processes in this configuration provides better control over microbial activity and process conditions, such as pH value and DO enhancing N_2O reduction. Disadvantages are a longer start-up time because of the growth of microorganisms in the biological reactor and higher operational costs due to the need to recirculate and treat large volumes of liquid, including energy costs for pumping and potential costs for chemicals and substrate to maintain biological activity (Frutos *et al.* 2015; Barbusinski *et al.* 2017).

Frutos *et al.* (2015) examined a two-stage bioscrubber system for treating N_2O -laden air, using methanol as a carbon source and electron donor. This system combined a 2-L packed-bed absorption column with a counter-current scrubbing liquid of mineral salt solution and a 3-L anoxic stirred tank reactor containing immobilized denitrifying biomass. Methanol was added to deplete O_2 creating anoxic condition and allow the reduction of N_2O to N_2 . The packed absorption column operated at a EBRT of 3 min, achieving N_2O RE ranging from 6 to 40% for N_2O concentrations of 104 \pm 12 ppmv, depending on the liquid recycling velocity (1–8 m/h).

Frutos *et al.* (2016) explored in a further research work the use of municipal wastewater as a carbon source and electron donor in the biological stage to reduce operating costs. In this setup, the same absorption column was paired with an anoxic packed-bed bioreactor (3 or 7.5 L), both filled with polyurethane foam to support microbial immobilization. The performance of N_2O abatement was assessed at varying liquid recycling velocities (1–8 m/h) and air EBRTs (3, 6, 12, 18, 40, and 80 min). At an EBRT of 3 min and the highest liquid velocity, N_2O removal efficiencies of $36 \pm 3\%$ were achieved, along with $91 \pm 1\%$ removal of total organic carbon. Increasing the EBRT to 40 min significantly improved N_2O removal, reaching up to 92% transfer in the liquid phase. However, overall system performance was limited by the low denitrification activity in the fixed-bed reactor and the low liquid-to-air-flow ratio controlling the N_2O load transported with the recycled liquid. Hydraulic retention time in the biological stage was also found to be a relevant factor.

It is also crucial to monitor and adjust pH value, temperature, and nutrient levels to optimize process conditions for microbial activity. Moreover, uniform air distribution throughout the absorption column is necessary for effective mass transfer. Also, exploring packing materials that provide higher mass transfer efficiency would be beneficial to improve the efficiency.

In summary, the performance of two-stage bioscrubbers for N_2O removal mostly depends on optimizing gas-liquid mass transfer through liquid-to-air flowrate ratio and EBRT in the absorption column. Counter-current operation enhances transfer efficiency of the absorption column, while the use of activated sludge in the biological reactor can reduce costs by providing a robust microbial community for denitrification and O_2 depletion and accelerating system start-up. Bioscrubbers are easier to control in operation than biofilters and biotrickling filters when the N_2O concentration varies significantly and showed higher RE in the reviewed studies.

Despite these advantages, challenges remain. Two-stage bioscrubbers are prone to the risk of clogging in the biological stage in the case a fix-bed reactor and municipal wastewater are used. However, there is no clear evidence because the studies reported above used synthetic wastewater without suspended solids. Additionally, using treated wastewater for the absorption column operation could be problematic in the long term because residual pollutants might lead to the growth of biofilm on the packed material. The duration of the laboratory experiments documented in the literature is too short to evaluate this aspect.

2.2.4. One-stage bioscrubber

In a one-stage bioscrubber, the transfer of gaseous pollutants to the liquid phase and their biodegradation by suspended microorganism occurs simultaneously in the same unit (e.g., bubble column with activated sludge). In this configuration, the gas stream to be treated is introduced in the form of bubbles at the bottom of the bioreactor, which is filled with activated sludge flocs, and the transfer of O₂ and N₂O occurs at the surface of the bubbles. A mixture of wastewater and activated sludge can be continuously fed to deplete DO level and provides denitrification of N₂O. In this case, the throughput rate of the influent wastewater-activated sludge mixture, pumped from an external activated sludge tank (e.g. activated sludge tank of the mainstream treatment in WWTP), should be adjusted relative to the air flowrate to establish an anoxic-redox potential within the reactor. The advantages of this configuration include its simple design and operation, the efficient gasliquid transfer facilitated by the mixing effect of rising bubbles, and high concentration gradient at the bubble surface due to the continuous depletion of N₂O in liquid through bacterial activity. It is also noteworthy that their design is compact in comparison to that of two-stage bioscrubbers. Additionally, they have nearly no risk of clogging since they do not rely on a fixed-bed material that can become blocked by biomass growth. However, aerator performance declines over time and requires regular cleaning to maintain efficiency. This treatment configuration often results in lower operational costs and greater flexibility in handling different wastewater compositions (Revah & Morgan-Sagastume 2005).

In two studies by Frutos *et al.* (2017a, b), one-stage bioscrubbers have been investigated for the reduction of industrial N_2O emissions, utilizing methanol, glycerol and a mixture of sodium acetate-acetic as a carbon source and electron donor for the heterotrophic bacteria. The authors investigated whether a bubble column bioscrubber is feasible to treat N_2O -rich off-gas streams of nitric acid production plants, characterized by low O_2 levels (1–4%) and high N_2O concentrations around 3,500 ppmv. They achieved a removal degree of 57–91% depending on the carbon source used. The investigated approach aimed at concurrently producing value-added bioproducts (e.g. biopolymers) while continuously removing N_2O , employing a biorefinery approach. This approach seeks to enhance the cost-effectiveness of N_2O reduction biotechnologies (Frutos *et al.* 2017a).

Valipoor et al. (2025) developed and investigated a one-stage bioscrubber at lab scale in integration with a pilot-scale WWTP. The column had a height of 1.8 m and an inner diameter of 7.4 cm. Synthetic air containing 600 ppmv of N_2O

was introduced at an inlet gas flowrate of 9 L/h, while the liquid feed, consisting of a municipal wastewater and activated sludge mixture pumped out of the anaerobic section of the activated sludge tank of the pilot WWTP, was supplied at a rate of 6 L/h. The effluent from the bioscrubber was directed back to anaerobic section of the activated sludge tank of the pilot-scale WWTP. Using this setup, an N_2O RE of 60–85% was achieved.

In summary, despite certain operational challenges, this technology seems to be promising for integration into WWTPs, offering the potential for effective implementation with low capital and operational costs.

2.3. Conclusion of the literature study

As discussed, the implementation of physical-chemical methods to treat N_2O -rich exhaust air streams is possible, but the energy costs of these methods are higher compared to biological methods, due to the high temperature and pressure, and pre-treatment process requirements. The use of catalysts is also associated with significant costs. Additionally, due to the high oxygen concentration in the exhaust air of WWTPs, multi-stage processes would be necessary, which further increase capital and operation costs. On the other hand, physical-chemical technologies have much lower space requirements than biological ones and provide higher removal efficiencies. In contrast to biological processes, which may require up to 80 min of EBRT to achieve acceptable removal efficiencies, physical-chemical technologies can attain comparable performance within seconds.

Biofiltration is a highly reliable and cost-effective method for odor removal at WWTPs. However, biofilters are less suitable for N_2O removal because they are typically dimensioned based on fixed sizes, making the process operation difficult to control. Another issue is the strong aerobic milieu during operation, which significantly affects the N_2O RE. Optimization in design and operation may solve some of these problems, however their large footprint, which generates additional costs, and relatively low RE continue to pose significant limitations. Therefore, implementing biofilters for N_2O removal in WWTPs is for now not advisable.

Bioscrubbers and biotrickling filters require moderate investments upfront and operational expenses related to liquid management (pumps) and maintenance.

Biotrickling filters for N_2O treatment have drawbacks including potential clogging from biofilm growth, risk of non-uniform load distribution in the filter bed, sensitivity to fluctuating N_2O concentration levels, and complex operation requiring specialized expertise. They can also generate odors if not managed properly and require significant installation space for trickling liquid management.

Bioscrubbers seem the most promising option to be implemented at WWTPs, as they are better controllable in operation and can cope with N_2O fluctuations in load and concentration. However, implementing an anoxic fixed-bed reactor operated with municipal wastewater in the biological stage can be challenging, due to clogging and short-circuit flows, that lead to reduced removal rate, operational efforts and increased costs. These issues can be mitigated by using flocculant activated sludge, which is largely available at WWTPs.

As a consequence, one-stage denitrifying bioscrubbers in form of a bubble column would be most applicable for implementation in municipal WWTPs, considering the simple construction and operation. Large-scale implementation also depends on the activity of the activated sludge and the availability of COD.

Although the denitrifying bioscrubbers show a potential feasibility for N_2O removal in WWTPs, their application seems to be more suitable in the sidestream treatment of SDE than in the mainstream treatment tanks. Sidestream treatment is favorable due to several advantages: exhaust air collection is simpler and more cost efficient due to smaller tank footprint, N_2O concentrations are higher and less variable, and the flowrate of exhaust air is lower. Additionally, treating the exhaust air of sidestream treatment requires less COD, to provide anoxic conditions in the scrubber, which can be supplied using a small portion of the inflow COD to the WWTP. A possible integration of a one-stage bioscrubber in a municipal WWTP will be assessed in the following section.

3. A NOVEL COMPREHENSIVE CONCEPT TO REDUCE N2O EMISSIONS AT WWTPS

Based on the literature study, we propose a novel comprehensive concept to reduce N_2O emission at WWTPs with anaerobic sludge digestion, beyond the operational and design suggestions made by Valkova *et al.* (2021) and Duan *et al.* (2020). The concept, which considers the specific interactions between mainstream and sidestream wastewater treatment processes, integrates (i) sidestream deammonification of SDE to minimize N_2O emissions in the mainstream biological stage and (ii) utilizes a denitrifying bioscrubber to treat the N_2O -rich exhaust air from the deammonification tank. The bioscrubber can be either

implemented as a dedicated reactor or could be integrated in the mainstream biological stage in an anaerobic or anoxic tank tank upstream the aerated biological stage. The treatment concept is graphically displayed in Figure 4.

The introduction of the autotrophic nitrogen removal in the sidestream aims to provide process conditions in the mainstream wastewater treatment that reduces N_2O production by AOB while keeping the N_2O -sink potential of heterotrophic denitrification unvaried (COD availability is unchanged because not required by deammonification). While the nitrogen load that can be denitrified in the mainstream remains unvaried, the nitrogen load to be nitrified in the mainstream is significantly reduced. In other terms, the ratio between the N_2O amount potentially removed by denitrification and the N_2O amount potentially produced by nitrification increases, potentially leading to a net decrease of the N_2O emission factor in the mainstream. This hypothesis is supported by the findings of Valkova *et al.* (2021), who observed that the estimated N_2O emission factor of activated sludge tanks referred to Kjeldahl nitrogen in the influent was found to be lower at plants and in temporal frames exhibiting an increasing ratio between denitrified and oxidized nitrogen.

In addition, the higher nitrogen removal achieved at the plant implementing deammonification provides both improved water quality (lower nitrogen load discharge with the effluent) and lower N_2O emissions in the receiving water bodies.

The application of an alternative biological treatment, such as nitrification/denitrification or nitritation/denitritation, which rely on heterotrophic pathways and consume organic carbon, instead of deammonification would limit the potential for mitigating N_2O emissions in the mainstream. This is because both pathways require a portion of the influent COD for the sidestream treatment.

It is also important to emphasize that this implementation offers further operational benefits for the plant. It reduces electricity consumption for aeration, as the oxygen transfer efficiency (α -factor) is higher in SDE than in mainstream wastewater (Baumgartner *et al.* 2022). The oxygen demand for deammonification compared to nitrification/denitrification or nitritation/denitritation is approximately the same when the oxygen consumption for the aerobic processes and the oxygen gain through denitrification or denitritation are considered. The total net oxygen demand for the conversion of 1 g NH₄-N to N₂-N via side-stream SDE treatment and redirecting the effluent in the mainstream biological treatment for the three pathways is almost the same (1.5 g O₂/g N_{removed}) (see detailed information in Supplemental Information, Chapter SI3). The contribution of denitrification and denitritation on overall net oxygen demand is often disregarded in literature when comparing the three aforementioned biological nitrogen removal options.

Despite being a globally established treatment technology (Lackner *et al.* 2014; Wang *et al.* 2022), deammonification still presents certain operational challenges. Anammox bacteria exhibit high sensitivity to environmental perturbations and are characterized by slow growth kinetics, necessitating stringent control of reactor conditions and effective biomass retention strategies to maintain stable process performance. Controlling operating conditions (such as free ammonia, free nitrous acid, pH value, and DO), is also essential for suppressing NOB activity, which would otherwise reduce nitrite availability

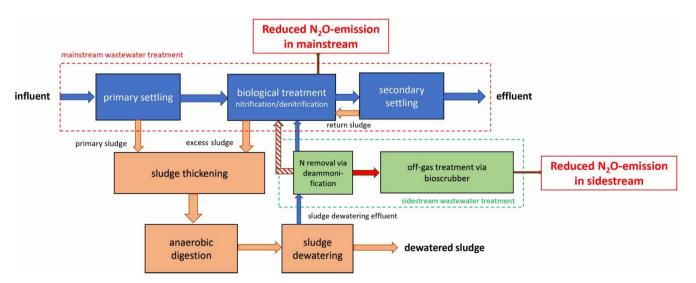


Figure 4 | Scheme of the proposed concept to deplete N_2O emissions at WWTPs with anaerobic sludge digestion. The red dashed arrow represents the option to treat the exhaust air in EBPR or in an upstream pre-denitrification tank.

for anammox bacteria (i.e. Izadi *et al.* 2021; Wang *et al.* 2022). These operational challenges must be carefully considered when assessing the advantages and limitations of the proposed N_2O mitigation concept, particularly the risk of short-term failure in the sidestream process. Such failures can lead to a sudden increase in nitrogen load to the mainstream treatment, potentially resulting in temporary non-compliance with effluent quality standards. Table 2 depicts the N_2O emission estimated on COD and nitrogen mass balances for a conventional model WWTP (treatment capacity >100,000 PE) with anaerobic sludge digestion prior and after the implementation of the proposed concept. The PE specific influent COD and N load were set to 120 g $O_2/(PE.d)$ and 9 g N/(PE.d) respectively, reflecting the fact that in bigger agglomerations the share of commercial/industrial wastewater impacts the N-to-COD ratio (Nowak *et al.* 1996; Lindtner & Zessner 2003). COD and nitrogen removal in the primary clarifier was assumed to be 30 and 10% respectively (DWA 2016). The influent N load to the biological stage in mainstream incl. SDE return flow (1.07 g N/(PE.d)) results in 9.2 g N/(PE.d). Detailed information on the calculations and assumptions of the assessment is reported in Supplemental Information (Chapter SI1).

In the novel concept the SDE is treated in a deammonification tank, and the nitrogen load is assumed to be reduced by 85% to 0.16 g N/(PE.d), according to reviewed full-scale data (Lackner *et al.* 2014). This decreases the N influent load in the biological stage to 8.2 g N/(PE.d). Various reactor configurations for deammonification have been successfully implemented in practice, including suspended growth systems (e.g. Wett *et al.* 2007) and fixed-film processes such as Moving Bed Biofilm Reactors (e.g. Veuillet *et al.* 2013). Any of these configurations could be considered for the proposed model plant. For this preliminary mass balance–level assessment of the N₂O mitigation concept, selecting a specific reactor configuration is not required.

The nitrogen in excess sludge was assumed 1.8 g N/(PE.d) based on the estimated excess sludge production of 29 g COD/ (PE.d) and typical N sludge content of 0.06 kg N pro kg COD measured at municipal WWTPs (Nowak *et al.* 1996). Sludge production in the activated sludge process was estimated over the COD mass balance applying a percentage of 38% of the COD removed, which is typical at plants operated at 14 days sludge age (DWA 2016, see Supplemental Information). The effluent of the mainstream biological treatment was assumed 1.6 gN/(PE.d) for the conventional WWTP and 0.7 with the

Table 2 | Comparison of COD and nitrogen mass balances at a conventional WWTP with activated sludge tank and anaerobic sludge digestion

Nomenclature	Conventional WWTP	WWTP with the novel concept	
COD mass balance			
Influent mainstream biological treatment after primary settling	84 g COD/(PE.d)	84 g COD/(PE.d)	
Effluent mainstream biological treatment	7 g COD/(PE.d)	7 g COD/(PE.d)	
COD excess sludge	29 g COD/(PE.d)	29 g COD/(PE.d)	
COD removal WWTP	94%	94%	
Nitrogen mass balances			
Influent mainstream biological treatment after primary settling and incl. SDE ^a	9.2 gN/(PE.d)	8.2 gN/(PE.d)	
Effluent mainstream biological treatment	1.6 gN/(PE.d) 0 gNH ₄ -N/(PE.d)	0.7 gN/(PE.d) 0 gNH ₄ -N/(PE.d)	
Nitrogen removal WWTP	82%	92%	
Nitrogen in excess sludge	1.8 gN/(PE.d)	1.8 gN/(PE.d)	
Nitrogen nitrified in mainstream treatment	7.4 gN/(PE.d)	6.5 gN/(PE.d)	
Nitrogen denitrified in mainstream treatment	5.8 gN/(PE.d)	5.8 gN/(PE.d)	
Ratio $N_{denitrified}/N_{nitrified}$ mainstream treatment	0.78	0.89	
N ₂ O-N emission mainstream treatment	$0.037~{\rm gN_2O\text{-}N/(PE.d)^b}$	$0.008~{\rm gN_2O\text{-}N/(PE.d)^c}$	
N ₂ O-N emission sidestream treatment	_	$0.014~{\rm gN_2O\text{-}N/(PE.d)^d}$	
N_2O -N emission sidestream treatment after exhaust air treatment	_	$0.004 \text{ gN}_2\text{O-N/(PE.d)}$	
N ₂ O-N emission WWTP	$0.037~\mathrm{gN_2O\text{-}N/(PE.d)}$	$0.012~\mathrm{gN_2O\text{-}N/(PE.d)}$	

^aN removal in sidestream via deammonification = 85%

 $^{{}^{}b}\text{EF} = 0.4\%$ of $N_{influent}$

 $[^]c\text{EF} = 0.15\%$ of $N_{influent}$

dEF = 1.3% of Ninfluent

^{100,000} PE; influent 120 g COD/(PE.d) and 9 g N/(PE.d) prior and after the implementation of the proposed concept to deplete N₂O emissions.

novel concept respectively, reflecting an overall N removal degree of 82 and 92% respectively. With these values, a mass balance was drawn up and the nitrogen nitrified, and nitrogen denitrified in the mainstream biological treatment were calculated for both models (Equations SI1 and SI2 in Supplemental Information). Based on the ratio $N_{denitrified}/N_{nitrified}$ and the correlation model by Valkova *et al.* (2021), N_2O -N emission from the mainstream biological treatment was estimated by applying an emission factor of 0.004 kg N_2O -N per kg N of nitrogen in the influent to the mainstream biological step. In the scenario with sidestream deammonification the N_2O emission factor in the mainstream biological treatment was reduced to 0.001 kg N_2O -N/kg $N_{influent}$. This represents a worst-case assumption considering that the model by Valkova *et al.* (2021) suggests a lower emission factor of 0.01%. N_2O -N emissions from the sidestream deammonification process were estimated based on the influent nitrogen load in the SDE, using an assumed emission factor of 0.013 kg N_2O -N/kg $N_{influent}$, reflecting an average derived from values reported in the literature.

The novelty of the concept also relies in shifting the treatment of N_2O -rich exhaust air from the mainstream aeration tank to the sidestream treatment of SDE. The deammonification process in the sidestream, where substantial amounts of N_2O are released, offers more suitable technical conditions to apply an exhaust air N_2O removal technology, in terms of smaller surface to be covered for air collection and air flowrate to be treated.

Our review of the currently available end-of-pipe technology indicates that a denitrifying one-stage bioscrubber relying on activated sludge and municipal wastewater as a COD source can represent a feasible option. Assuming a N_2O removal degree of 70% in the bioscrubber, the reduction in the total N_2O emissions of the WWTP is estimated to 0.037 minus 0.012 = 0.025 g N_2O -N/(PE.d), corresponding to a reduction of ~70% compared to a conventional configuration (Table 2).

The assumption of an average N_2O removal degree of 70% in the bioscrubber is based on experimental results by Frutos *et al.* (2017a, b) and by Valipoor *et al.* (2025). In the latter, a one-stage lab scale denitrifying bioscrubber has been operated with activated sludge and real municipal wastewater for over one year achieving RE of 60–85%. The N_2O concentration in the treated synthetic air ranged from 600 to 5,000 ppmv, which can be expected in sidestream deammonification exhaust air. A few percent of the COD of the wastewater are required to ensure denitrifying conditions and N_2O removal. It is expected that in a deeper column the N_2O removal degree can be improved due to the longer contact time of the bubbles. Further details and results will be presented by the authors in an upcoming publication.

The application of the bioscrubber as a bubble column opens the perspective of performing the exhaust air treatment directly in the mainstream biological treatment, introducing the exhaust air of the deammonification stage into the anaerobic tank for enhanced biological phosphorus removal (EBPR) or in a pre-denitrification tank. A similar approach was proposed by Trautmann *et al.* (2017), with the difference that they considered only the sidestream treatment and suggested to introduce the exhaust air of the nitritation stage of a two-stage deammonification process into the anoxic anammox stage.

A matter of concern is if the introduction of the exhaust air in these tanks could negatively affect the anaerobic and anoxic biological processes. The comparison of the airflows to be treated with reactor volumes helps assessing this potential risk.

The PE specific aeration demand for the partial nitritation at 30 °C and 1.5 mg O_2/L in the deammonification tank was estimated to be \sim 2 g $O_2/(PE.d)$ (Supplemental Information, chapter SI2). Considering that 1 Nm³ of air contains 300 g of O_2 and an O_2 utilization degree of 5% per meter of water depth, which is typical for fine-bubble aerators, the O_2 input in a 5 m deep tank can be calculated with 75 g $O_2/Nm³$ of air. To determine how much aeration air is needed for the process, O_2 demand is divided by the O_2 input leading to a PE specific aeration airflow of 0.03 Nm³ air/(PE.d) or 30 L/(PE.d). Based on the laboratory experiments with bubble columns by Frutos *et al.* (2017b) and Valipoor *et al.* (2025), volume specific air flowrate was calculated to 75 and 28 L air per L of bubble column volume per day respectively. Using the volume specific air flowrate and the amount of air required for aeration the PE specific volume requirement of the bubble column can be estimated to 0.5–1 L/PE. This corresponds to only \sim 10% of the volume of a EBPR tank or \sim 2.5% of a pre-denitrification tank, according to DWA (2016) design guidelines.

Additionally, the biological uptake of the O_2 introduced with the exhaust air in the EBPR tank (~ 1.6 g O_2 /(PE.d)) would imply a COD consumption of approximately 3% of the influent wastewater to ensure DO depletion through microbial respiration. Since this is much lower than the amount of readily degradable COD usually presents in the influent of a biological stage (DWA 2016), the COD availability for EBPR and/or denitrification process is still provided.

The electricity consumption for introducing the exhaust air into a EBPR or pre-denitrification can be calculated to ~ 1.1 Wh/(PE.d) or 0.4 kWh/(PE.a) assuming an aeration efficiency in operation of 2 kg O_2 /kWh (Baumgartner *et al.* 2022). This is negligible compared to the average overall electricity consumption of WWTP in this size category of 30–35 kWh/(PE.a) (Ganora *et al.* 2019).

Summarizing, theoretical evaluations indicate the potential feasibility of treating sidestream exhaust air within the main-stream treatment tanks. However, further research is needed to prove the technical feasibility and operational applicability of this approach under full-scale conditions. In the end, the estimated reduction of N₂O emissions in CO₂ equivalents achievable with the proposed approach was calculated based on a Global Warming Potential value of 273 g CO₂e/gN₂O (IPCC 2023). The new concept would produce about 5 kg CO₂e/(PE.a) less than a conventional WWTP, which represents a reduction of approximately 20% when assuming a total CO₂-footprint of 25 kg CO₂e/PE.a for the operation of a yet CO₂-optimized Austrian WWTP (Parravicini *et al.* 2016). The assessment accounted for both N₂O removal and the additional electricity consumption associated with the bioscrubbing process.

4. CONCLUSIONS

In municipal WWTPs, biological nitrogen removal via nitrification and denitrification leads to the formation and emission of the GHG N_2O . While process optimization and plant design can help reduce N_2O production and emissions, complete avoidance is not possible in practice. Therefore, end-of-pipe technologies are necessary as additional mitigation strategies.

The literature review indicated that currently available exhaust air treatment technologies for N_2O removal, both physical-chemical and biological, face significant limitations when applied at WWTPs. These challenges arise from technical constraints, economic considerations, and the specific characteristics of the exhaust air of WWTPs. Moreover, implementation often requires covering extensive tank surfaces to enable air collection and treatment. The review identified sidestream treatment processes as more suitable for the application of end-of-pipe technologies due to their compact foot-print, higher and less variable N_2O concentrations, and reduced exhaust air flowrate.

Based on these findings, a novel approach combining sidestream deammonification with biological N_2O removal from exhaust air of this unit has been proposed. This strategy offers a promising pathway for plant-wide N_2O mitigation, particularly in large WWTPs with anaerobic sludge digestion. In addition to direct emission mitigation in the sidestream, this strategy may indirectly reduce N_2O formation in the mainstream biological treatment by altering the influent nitrogen-to-COD ratio, which can decrease N_2O accumulation and emission.

Among the technologies reviewed, biological end-of-pipe technologies based on heterotrophic denitrification represents a feasible and cost-effective option for N_2O removal at WWTPs. This exhaust air treatment could be implemented in a dedicated one-stage denitrifying bioscrubber configured as a bubble column, or alternatively, by introducing the exhaust air stream into an EBPR tank or an upstream denitrification tank. The latter option, however, requires further investigation to assess its technical feasibility and operational performance under full-scale conditions.

Although sidestream deammonification is a well-established and widely implemented technology, its operation remains challenging and requires careful process control. Process disturbances in the sidestream could adversely impact mainstream operations and effluent quality, potentially resulting in regulatory non-compliance. These challenges must be critically addressed to ensure the robustness and viability of the proposed N₂O mitigation strategy.

Furthurmore, the concept of treating sidestream exhaust air in a denitrifying one-stage bioscrubber is particularly suitable for facilities already employing sidestream treatment. In such cases, it enables targeted reduction of N_2O emissions from the sidestream without further mitigating effect on mainstream emissions.

While alternative sidestream nitrogen removal methods, such as ammonia stripping processes, may avoid N_2O generation, this study has focused exclusively on biological end-of-pipe N_2O mitigation strategies. Future evaluations of these alternatives should incorporate comprehensive carbon footprint analyses to enable transparent comparisons with biological approaches.

In conclusion, end-of-pipe technologies to remove N_2O in exhaust air streams have the potential to enhance N_2O reduction. Nevertheless, further investigation is warranted to ascertain their feasibility in WWTPs. A detailed evaluation of the practical challenges and operational efficiency of these methods is essential to support their widespread implementation and achieve a sustainable reduction in N_2O emissions in WWTPs.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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