

SIGNIFICANCE OF WASTEWATER TREATMENT TO NITROUS OXIDE EMISSION

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Abstract

The sewage flowing into the wastewater treatment plant through the sewer system, as well as transported by the slurry fleet, are subjected to treatment processes. These processes, carried out under both aerobic and anaerobic conditions, contribute to the emission of greenhouse gases. On the basis of available reports and previous research, the emission of nitrous oxide (N₂O) from wastewater was estimated at approx. 4-5% in relation to the global amount of this gas emission from anthropogenic sources. Data obtained from the operation of full-scale WWTPs show a wide range of values of the N₂O emission factors from 0.0006 to 0.045 (kgN₂O-N/kgN) [18].

The article describes possible sources of nitrous oxide emission from wastewater treatment plants and presents the basic principles of its balancing.

Keywords: greenhouse gases, nitrous oxide, municipal wastewater treatment plants

1. INTRODUCTION

Development of high-efficiency technologies improves aquatic environment. However, wastewater treatment technologies, which ensure the removal of organic compounds and nutrients, contribute to the emission of greenhouse gases (GHG), mainly: carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). In particular N₂O, which has a global warming potential 265 times greater than

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that of CO₂ [15,19] and has already been reported to be a major contributor to greenhouse gas emissions of WWTPs [36].

For this reason Wastewater Treatment Plants (WWTPs) are identified as one of the four anthropogenic source of N₂O emissions, next to agriculture, industrial acid production, and combustion. Recent research and field surveys have revealed that emissions in sewer networks and from nitrification or nitrification-denitrification processes at WWTPs, previously judged to be a minor source, may in fact result in more substantial emissions [18]. The wastewater treatment sector is assumed to be responsible for 4–5% N₂O of the total anthropogenic emissions. Data obtained from the operation of full-scale WWTPs show a wide range of values (0–14.6%) of the of the incoming nitrogen for the fraction of nitrogen that is emitted as N₂O [1,10,22,34].

The article describes possible sources of nitrous oxide emission from wastewater treatment plants and presents the basic principles of its balancing.

THE NITROGEN FORMS IN BIOLOGICAL PROCESSES

In quantitative terms, nitrogen compounds, along with carbon compounds, are the most important component of municipal wastewater. Human faeces are the main source of nitrogen in household waste. It is estimated that every person excretes 10-14 g of nitrogen daily. In municipal wastewater, nitrogen occurs in four basic forms: organic nitrogen, ammoniacal nitrogen (both in ionized form (NH₄-N) and free ammonia (NH₃-N), nitrite (NO₂-N) and nitrate (NO₃-N). Due to the particle size, organic nitrogen in wastewater can be divided into dissolved, colloidal and suspension fractions [9].

In raw wastewater, nitrogen occurs as organic nitrogen, as urea or as a result of the beginning decomposition processes in the form of ammonia. The concentration of total nitrogen in domestic wastewater varies in the range of 15-80 g N/m³, and a significant part of the nitrogen occurs in the form of ammonium nitrogen (10-15 g N-NH₄/m³) or unstable organic compounds (proteins, urea), which, as a result of deamination or hydrolysis of urea, quickly turn into N-NH₄. The content of nitrogen bound in amino acids (products of protein degradation) usually does not exceed 5 g N/m³, and the concentration of nitrogen in the form of urea can be 2-16 g N/m³. Mineral forms, nitrites and nitrates together constitute less than 1% of total nitrogen in raw wastewater, which corresponds to concentrations generally less than 0.5 g N(NO₂+NO₃)/m³ [26,30]. Characteristics of nitrogen compounds in raw wastewater are most often based on the analysis of general Kjeldahl nitrogen (TKN = organic nitrogen + ammonium nitrogen). The share of both forms depends on the time of wastewater retention in the sewage network, because this is where the hydrolysis of organic nitrogen to ammonium begins. Nitrate and nitrite nitrogen content is usually not included. For more

accurate wastewater characteristics, it is assumed that nitrogen in wastewater occurs in the forms eq.1.1[16]:

$$N_{TOT} = S_{N-NO_2} + S_{N-NO_3} + TKN = S_{N-NO_2} + S_{N-NO_3} + S_{N-NH_4} + S_{I,N} + X_{S,N} + X_{I,N} \text{ g/m}^3 \quad (1.1)$$

where:

N_{TOT} – total nitrogen;

S_{N-NO_2} , S_{N-NO_3} - nitrite and nitrate nitrogen concentration;

TKN - total Kjeldahl nitrogen;

S_{N-NH_4} - ammonium nitrogen concentration (ammonium ion and undissociated ammonia);

$S_{I,N}$ - concentration of dissolved organic nitrogen contained in non-biodegradable compounds;

$X_{I,N}$ - concentration of the bioavailable organic nitrogen contained in the total suspension;

$X_{S,N}$ - concentration of easily bioavailable organic nitrogen.

Nitrogen forms in treated wastewater have a different chemical structure than those found in raw wastewater [28]. In treated wastewater dissolved organic nitrogen can constitute a significant fraction of total nitrogen [9]. Organic nitrogen dissolved in the effluent consists of both non-decomposable forms found in the inflowing wastewater, such as purines, pyridines and pyrimidines [3], as well as products of the metabolism of activated sludge microorganisms.

In WWTPs during treatment processes, organic nitrogen is ammonified to ammonium nitrogen, which can be used to build a new cell mass or oxidized (in the nitrification process) successively to NO_2 -N and NO_3 -N. The nitrification process does not change the concentration of N_{TOT} in the wastewater, but only changes its form. Denitrification, reduction of NO_3 -N and NO_2 -N to gaseous nitrogen, makes it possible to permanently reduce the total nitrogen concentration in wastewater (Fig.1).

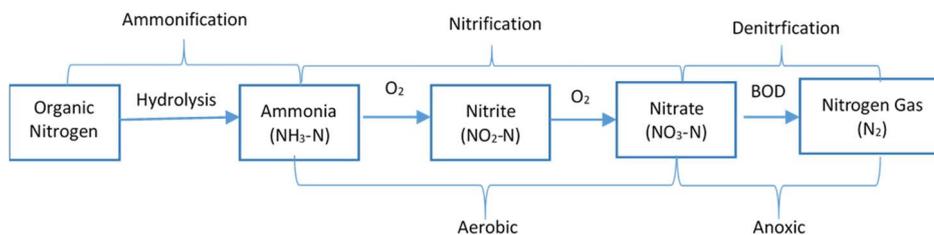


Fig.1. Nitrogen forms in treated wastewater

The removal of nitrogen from wastewater can potentially lead to the production and emission of nitrous oxide. The nitrous oxide (N_2O) emissions from wastewater treatment plants can occur as direct, and indirect emissions [17]. The

direct emissions are the processes contributing to N_2O emission during biological nitrogen removal. In conventional activated sludge plants, nitrogen is removed via nitrification and subsequent denitrification. Nitrous oxide is not only produced biologically, but also chemically in a great number of chemical reactions involving nitroxyl (HNO), hydroxylamine (NH_2OH) and nitrite (NO_2^-) [31]. Biological and chemical pathways of N_2O production in the nitrification and denitrification processes shows Figure 2.

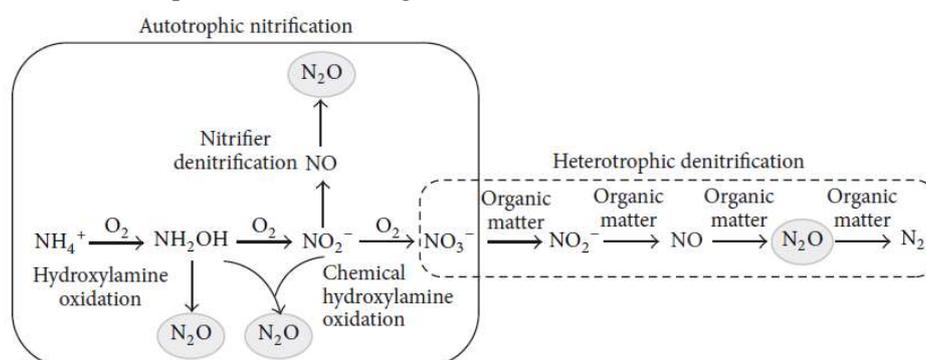


Fig. 2. Biological and chemical pathways of N_2O production in the nitrification and denitrification processes [6]

The most important operational parameters leading to N_2O emission in WWTPs are [22]:

- low dissolved oxygen concentration in the nitrification and high in denitrification stages,
- increased nitrite concentrations in both nitrification and denitrification stages,
- low COD/N ratio in the denitrification stage.

NITROUS OXIDE EMISSION IN WWTPS

As mentioned earlier, nitrous oxide is formed as a result of biological and chemical processes in the wastewater treatment plant. Biological wastewater treatment can be carried out with various technologies.

The qualitative characteristics of wastewater, and mainly the carbon to nitrogen ratio, determine the selection of processes that guarantee the achievement of the required nitrogen concentration in treated wastewater. Table 1 shows the rate of nitrogen removal from wastewater in various processes.

Table 1. The rate of nitrogen removal from wastewater in various processes [30]

| No. | Process | Oxygen demand | Organic carbon demand | Nitrogen removal rate | Biomass production |
|-----|---|-----------------------|-----------------------|-----------------------|--------------------|
| | | kgO ₂ /kgN | kg COD/kg N | kg N/m ³ d | kgd.m./kg N |
| 1 | Nitrification and denitrification | 4.6 | 7.6 | 2-8 | 3.2 |
| 2 | Short nitrification and denitrification | 2.3 | 4.6 | 1.5 | 2.0 |
| 3 | Oxygen deammonification | 2.3 | 1.5 | 4.7 | 2.0 |
| 4 | Sharon | 2.3 | 2.4 | 1.5 | 1.0 |
| 5 | Oland | 1.7 | 0 | 0.1 | 0.16 |
| 6 | Anammox | 0 | 0 | 5.1 | 0.12 |
| 7 | Anammox + Sharon | 1.9 | 0 | 0.745 | 0.3 |
| 8 | Cannon | 2.1 | 0 | 1.2-8.9 | 0.3 |

Nitrous oxide emissions from wastewater treatment plants vary due to different design and operating conditions. WWTPs with high nitrogen removal rates emit less N₂O. This is important as it indicates that no compromise is required between high quality of the treated wastewater and lower N₂O emissions [23,37].

Data obtained from the operation of full-scale WWTPs show a wide range of values (0–14.6%) of the of the incoming nitrogen for the fraction of nitrogen that is emitted as N₂O [1,10,22].

Taking into account the fact that WWTPs with nitrogen removal carried out by nitrification-denitrification processes have a median emission factor of 0.01 kg N₂O-N/kg N influent meaning that 0.6% of the inlet nitrogen is converted into N₂O. The use of nitrification - Anammox allows for a 60% reduction in oxygen demand, 100% reduction in organic carbon demand, 90% less excess sludge production and reduction of greenhouse gas emissions (CO₂ and N₂O) [25,27]. Partial nitrification-Anammox processes in both sludge line (20% of the total nitrogen load with a conversion of 0.8% into N₂O) and main stream (80% of the total nitrogen load with a conversion of 0.2% into N₂O) will signify an important decrease of the N₂O emissions.

Technologies based on the denitrification process remove N₂O with efficiencies of 75–99%. However, the mass transfer rate from the air flow to the liquid phase is limited by the low aqueous solubility of this greenhouse gas. Therefore, high hydraulic retention times (HRT) are required to achieve high N₂O removal efficiencies [6].

This is consistent with Ahn et al. [2] reports, that N₂O is primarily emitted from the aerated zones. The N₂O formed in the aerobic zones is quickly removed by aeration and mixing, and is therefore the main source of N₂O emitted from wastewater treatment systems. While N₂O, as one of the necessary intermediates in denitrification, is dissolved in the liquid and reduced to N₂ gas.

The N₂O emission factor (amount of N₂O-N emitted from nitrogen load) for full-scale WWTPS varies significantly, ranging from 0 to 25% (Table 1).

Table 2. Nitrous Oxide (N₂O) emission factors reported for several full-scale wastewater treatment plants [37]

| No. | Type of plant | N ₂ O emission (% of N-influent) | Remarks | Reference |
|-----|--|---|---|------------------------|
| 1 | activated sludge plant-primary and secondary treatment (aeration only; 4 ml d ⁻¹) | 0.035–0.05 | N ₂ O was emitted in aerated areas, low N ₂ O flux at non-aerated areas | New Hampshire, USA [8] |
| 2 | activated sludge plant | 0.001 | N ₂ O emissions increased with nitrite and nitrate concentrations | Germany [32] |
| 3 | anoxic–aerobic activated sludge plant (78 Ml d ⁻¹) | 0.001–0.04 | N ₂ O emission was dependent on COD:N | Germany [5] |
| 4 | intermittent activated sludge treatment of municipal sewerage (2.5 and 31 Ml d ⁻¹) | 0.47 (0.01) | — | France [29] |
| 5 | nitritation–anammox sludge digestion liquor treatment | 2.3 | N ₂ O emissions increased with decreasing oxygen concentration (aerated stage) and increasing nitrite concentration (anoxic stage) | Netherlands [21] |
| 6 | seven BNR plants | 0.6–25 (3.5+2.7% average) | correlation between N ₂ O emissions and nitrite accumulation was observed | Australia [13] |
| 7 | four treatment plants (completely mixed, plug-flow, membrane bioreactor) | 0–0.3 | NH ₄ -N and DO had impact on N ₂ O emission | France [14] |
| 8 | partial nitritation–anammox sequencing batch reactor (three plants, five reactors) | 0.4–0.6 | N ₂ O emissions were slightly higher than in conventional nitrogen-removal systems | Switzerland [20] |
| 9 | 12 BNR plants | 0.003–2.59 | aerobic zones contributed substantially more to N ₂ O fluxes than anoxic zones | USA [2] |
| 10 | four-stage floc-based partial nitritation and anammox process | 5.1–6.6 | high N ₂ O emissions may be partly inherent to a separate nitritation step | Belgium [11] |

GENERAL GUIDANCE TO QUANTIFY NITROUS OXIDE IN WASTEWATER TREATMENT

Nitrogen emissions from wastewater treatment plants can be estimated on the basis of actual gas measurements or on the basis of calculations. The method related to the measurement of gases requires the installation of appropriate metering and continuous monitoring, and in practice it is mainly used on a laboratory scale. The IPCC guidelines [18] are most often used as the calculation method.

In accordance with the IPCC guidelines of 2019, in order to estimate N emissions from wastewater treatment plants, the following elements (Fig.3) should be taken into account: amounts of N in wastewater entering the treatment plant (TN_{DOM}), loss or removal of N in the treatment process (either through biological conversion or sludge removal) (N_{REM}), and N content in wastewater discharged into water systems ($N_{EFFLUENT, DOM}$) [18].

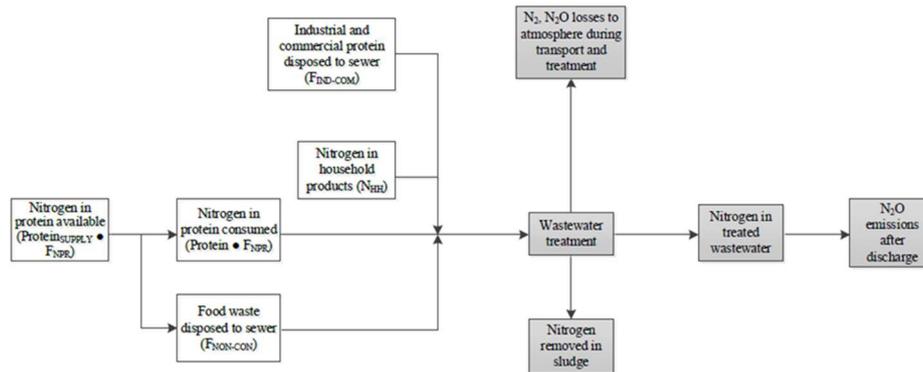


Fig.3. Nitrogen in domestic wastewater treatment [18]

The emissions from domestic wastewater treatment plants may be calculated using eq. 3.1 [18]:

$$N_2O \text{ Plants}_{DOM} = \left[\sum_{i,j} (U_i \cdot T_{ij} EF_j) \right] \cdot TN_{DOM} \cdot \frac{44}{28} \quad (3.1)$$

where:

$N_2O \text{ Plants}_{DOM}$ - N_2O emissions from domestic wastewater treatment plants in inventory year, kg N_2O /year,

TN_{DOM} - total nitrogen in domestic wastewater in inventory year, kg N/year,

U_i - fraction of population in income group in inventory year,

T_{ij} - degree of utilization of treatment/discharge pathway or system j , for each income group fraction i in inventory year,

i - income group: rural, urban high income and urban low income,

j - each treatment/discharge pathway or system,

EF_j - emission factor for treatment/discharge pathway or system j , kg N_2O -N/kg N,

44/28 - the conversion of kg N_2O -N into kg N_2O .

The indirect emissions is emissions from effluent from centralized treatment systems that has been discharged into aquatic environments. In addition to in-plant emissions from wastewater treatment (direct emissions), N_2O is emitted when the nitrogen that is released in wastewater treatment plant effluent is transformed through natural processes. This effluent-derived N_2O is considered to be the larger source of N_2O emissions associated with wastewater treatment [17,22,35]). River conditions are favorable for N_2O production [7], and numerous studies have demonstrated that rivers dominated by wastewater effluent have higher N_2O saturation values [4,24,38,31]. That emissions are dependent on the nutrient-impacted status and oxygenation level of the aquatic environment receiving the wastewater discharge. In the case of discharge to nutrient-impacted waters such as eutrophic lakes, estuaries and rivers, or locations where stagnant conditions occur, emissions can be significantly higher [18].

N_2O emissions from wastewater effluent can be calculated using eq. 3.2 [18]:

$$N_2O_{\text{EFFLUENT,DOM}} = N_{\text{EFFLUENT,DOM}} \cdot EF_{\text{EFFLUENT}} \cdot \frac{44}{28} \quad (3.2)$$

where:

$N_2O_{\text{EFFLUENT,DOM}}$ - N_2O emissions from domestic wastewater effluent in inventory year, kg N_2O /year,

$N_{\text{EFFLUENT,DOM}}$ - nitrogen in the effluent discharged to aquatic environments, kg N/year,

EF_{EFFLUENT} - emission factor for N_2O emissions from wastewater discharged to aquatic systems, kg N_2O -N/kg N,

The factor 44/28 is the conversion of kg N_2O -N into kg N_2O .

Potential sources of N_2O emissions connected with wastewater treatment are sewers networks and decentralised treatment systems of domestic wastewater (onsite sanitation). Open sewers are not a source of N_2O . Closed and underground are likely source of N_2O . However, insufficient data exist to quantify emission factors that address the variation in sewer type and operational conditions.

A septic system usually composed of a septic tank, is generally buried in the ground, and a soil dispersal system. Septic tanks, as opposed to soil dispersal, are not the source of N_2O . Gases produced in the effluent dispersal system mainly

N₂O, and CO₂ are released through the soil. Latrines are not the source of N₂O [18].

To obtain accurate results of calculations using the IPCC method, the emission factor for N₂O emissions (EF) appropriate for a given object should be determined. Research conducted by Volkova et al. [36] showed, similar to previous reports by Kampschreur et al. [22], Ahn et al. [1], and Daelman [10], significant variation in the value of the EF coefficient.

The differences demonstrated were not only between different treatment plants, but also in the same objects. It was found that high daily fluctuations in wastewater inflow contribute to a high coefficient of variation of daily N₂O emissions. It has been shown that a reliable method of estimating the N₂O load from a wastewater treatment plant is a method based on the average annual efficiency of nitrogen removal, and not the constant value of the emission factor assumed in IPCC as 3.2 g N₂O-N/PE (for countries with a highly developed wastewater treatment system). The German estimate of this emission factor from wastewater treatment processes, not taking into account the inflow of industrial wastewater, is 7.0 g N₂O/person · year [35], while in the United States 4.0 g N₂O/person · year [33]. For WWTPs with a TN removal degree lower than 70%, a maximum EF_{N₂O} was 1.4%, for TN removal degrees higher than 93%, the minimum EF was set at 0.03% [36].

CONCLUSIONS

High-efficiency wastewater treatment requires effective nitrogen removal to protect the receiving waters from eutrophication. The processes leading to the removal of nitrogen compounds from wastewater result in the emission of N₂O to the atmosphere. Currently, in order to reduce N₂O emissions related to wastewater treatment, research is being carried out on recovering N₂O from wastewater as an energy resource. High-strength wastewater treatment is more favorable for N₂O recovery due to the high energy potential, established nitrification approaches, and significant carbon/aeration savings [12].

It should be noted that additionally wastewater treatment plants must be adapted to the appropriate waste management generated in the technological process of wastewater treatment. These processes contribute to the emission of greenhouse gases. These emissions come from three different sources - energy consumption at different stages, depositing sewage sludge in the environment and releasing gases into the atmosphere from incineration, drying and anaerobic digestion processes.

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