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GREENHOUSE GAS EMISSIONS FROM BIOLOGICAL FIXED-FILM REACTORS FOR ADVANCED WASTEWATER TREATMENT

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Abstract:

Municipal wastewater treatment plants (WWTPs) provide a high removal of organic carbon (BOD_5, COD) and nutrients (nitrogen and phosphorus) from wastewater. However, its environmental footprint in terms of greenhouse gas (GHG) emissions remains underexplored. The biological WW treatment results in direct emissions of GHG such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) thus contributing to global warming. Hence, the understanding and estimation of these emissions is essential to tackle this challenge. The GHGs emitted depend upon the treatment technology employed. The present study evaluates the direct greenhouse gas emissions generated during advanced treatment of urban wastewater in submerged biofilm reactors using data from author's pilot plant experiments.

1. INTRODUCTION

Municipal wastewater treatment plants (WWTPs) provide a high removal of organic carbon (BOD_5, COD) and nutrients (nitrogen and phosphorus) from wastewater. However, its environmental footprint in terms of greenhouse gas (GHG) emissions remains underexplored (Tong et al., 2024). The biological WW treatment results in direct emissions of GHG such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) thus contributing to global warming. Hence, the under-standding and estimation of these emissions is essential to tackle this challenge (Gupta & Singh, 2012).

Direct CO_2 emissions from degradation of wastewater organics are considered a carbon-neutral process in GHG accounting because they are of biogenic origin and are part of the natural carbon cycle "atmosphere – plant photosynthesis – animals – humans – WW treatment – back to atmosphere" thus being rapidly reabsorbed or sequestered. As such, it is not considered to be part of the global warming problem. Still calculation of the biogenic carbon dioxide has been conducted by some wastewater treatment plants (McGuckin et al., 2013).

On the other hand, non-CO_2 GHG emissions (N_2O and CH_4) are recognized as significant due to their much higher global warming potentials (GWP) than CO_2. Therefore, even relatively small amounts can result in a significant carbon footprint. The latest estimation of the GWP factors for a 100 year horizon according to the IPCC Sixth Assessment Report AR6 of 2021 (IPCC, 2021) are given in **Table 1**. The previous IPCC estimates in the Fourth Assessment Report AR4 of 2007 and Fifth Assessment Report AR5 of 2014 respectively are also used sometimes for inventory and reporting purposes (GHG Protocol, 2024).

Using the European database EDGAR (Monforti Ferrario et al., 2019) it is estimated that N_2O and CH_4 emissions from wastewater treatment and discharge have doubled during 1970–2015, accounting for 9.6 % of global non-CO_2 GHG emissions in 2015 (Hua et al., 2022). According to the United Nation Framework Convention on Climate Change (UNFCCC), in 2018 N_2O and CH_4 emissions from wastewater treatment and discharge in industrialized countries contributed 2.6 % and 3.6

% to the total CO_2 eq emissions, respectively (Ranieri et al., 2023).

TABLE 1. IPCC GWP values of GHGs produced in WWTPs relative to CO_2

Gas	Chemical formula	GWP values for 100 year time horizon		
		AR4	AR5	AR6
Carbon dioxide	CO_2	1	1	1
Methane (non-fossil origin)	CH_4	25	28	27.0
Methane (fossil origin)	CH_4	-	30	29.8
Nitrous oxide	N_2O	298	265	273

At the same time, the criteria for environmental quality are becoming stricter. On 12 May 2021 an EU Action Plan was adopted titled "Towards Zero Pollution for Air, Water and Soil". It sets particular targets for 2030 to reduce pollution at source in comparison to the current situation. This is a regulatory challenge to the conventional techniques for urban wastewater treatment given the climate change with its trends of warming.

The greenhouse gases emitted from a WWTP depend upon two main factors: treatment process and electricity consumption. A large number of WWTPs in various countries use fossil energy (coal, gas, oil) for power generation. These therefore contribute considerably to the overall GHG production (Gupta and Singh, 2012). In this study, however, only direct emissions from the biological processes of WW treatment itself are considered.

The extent of CH_4 production depends primarily on the quantity of degradable organic material in the wastewater and the type of treatment system. Aerobic treatment may produce limited CH_4 from anaerobic pockets or due to reduced removal of organics with the sludge during primary treatment. Plants with nutrient removal processes are also sources of CH_4 (IPCC, 2019). Higher CH_4 emissions from the aeration tank are suspected of being residual CH_4 from the sewers that was not completely stripped at the inlet works (McGuckin et al., 2013). During anaerobic process, the BOD_5 of wastewater is either incorporated into biomass or converted to CO_2 and CH_4. A fraction of biomass is further converted to CO_2 and CH_4 via endogenous respiration.

Direct emissions of N_2O may be generated during both nitrification and denitrification processes in the plant. Nitrification is an aerobic process converting ammonia and other nitrogen compounds into nitrite (NO_2^-) and nitrate (NO_3^-) by various types of autotrophic bacteria. Complete removal of nitrogen after nitrification is achieved through denitrification under anoxic conditions (without free oxygen) for conversion of nitrate into nitrogen gas (N_2) by heterotrophic bacteria using organic matter as electron donors. Nitrous oxide is an intermediate product of denitrification when there are suboptimal concentrations of carbon (C) and nitrogen substrate and presence of dissolved O_2 in the anoxic zone coming with the recirculated effluent of the aerobic reactor. N_2O may also be generated as a by-product of nitrification when the influent contains high concentrations of BOD_5 and ammonium, and the aeration is insufficient. The main biochemical reactions responsible for production of N_2O are (Sun et al., 2017):

$$NH_{4^+} \rightarrow NH_2OH \rightarrow NO_{2^-} \rightarrow NO \rightarrow N_2O \qquad (1)$$

$$NO_{3^-} \rightarrow NO_{2^-} \rightarrow NO \rightarrow N_2O \rightarrow N_2 \qquad (2)$$

The objectives of the present study are: (i) to evaluate the direct GHG emissions from submerged biological fixed-film reactors for carbon and nitrogen removal using data from author's pilot plant experiments; (ii) to identify practical ways for reducing GHG release in order to contribute to global warming mitigation.

2. MATERIALS AND METHODS

2. 1. EXPERIMENTAL DATA USED

Direct emissions of CH_4 and N_2O are calculated using data from author's pilot plant experiments implemented at an urban WWTP on two submerged biofilm reactors (SBRs) for advanced wastewater treatment (Vatralova, 2015). Submerged fixed-film reactors are an alternative to the widely used suspended growth activated sludge facilities and combine depth filtration of undissolved substances with biodegradation of organic matter and particular nitrogen compounds in a pre-treated wastewater. In this study, the first one is anoxic and receives a primary settled sewage at a hydraulic load of 1,0 m^3/m^2h to serve as a carbon source for denitrification of recirculated effluent of the second reactor.

The latter is a biological aerated filter (BAF) for biodegradable organic carbon removal (expressed as BOD_5) and nitrification. The recirculation ratio varies from 0 to 3 (1 part primary settled influent to 3 parts BAF effluent). The process data (average values) are given in **Table 2**. In **Table 2** the value of BOD_5 retained in the anoxic filter as sludge when r = 0 (without recirculation) is 30 % of the influent BOD_5 coming from the primary settled sewage. This percentage is used to approximately estimate the sludge retained in the anoxic filter during the next set of experiments (with r = 1÷3).

TABLE 2. BOD_5 and total inorganic N values.

r	Q_s	Q_d	v_f	BOD_5		Total inorganic
				Influent	Retained as sludge	Influent
-	dm^3/min	dm^3/d	m/h	g/d	g/d	g/d
0	0,3	432	1,0	83,64	24,88	15,72
1	0,3	432	2,0	74,39	22.32	13,48
2	0,3	432	3,0	77,76	23,32	14,64
3	0,3	432	4,0	96,64	28,99	15,47

r-recirculation ratio; v_f – filtration velocity (hydraulic load)

2. 2. GHG EMISSIONS ESTIMATION METHODOLOGY

The methodology of this study for estimation of GHG emissions is based on the IPCC Guidelines for National Greenhouse Gas Inventories of 2006 revised in 2019 (IPCC, 2019). The production of N_2O and CH_4 emissions is estimated on the basis of the organic carbon (expressed as BOD_5) and total inorganic nitrogen contents in the influent to the experimental reactors, and the extent, to which they generate GHG, expressed by particular emission factors (EFs) (Eq. 3 and Eq. 4). CH_4 and N_2O emission factors vary considerably among WWTPs using different technologies. Here they are calculated using published data from similar processes of carbon and nutrient removal in full-scale activated sludge WWTPs as very limited information about biofilm reactors could be found in literature. Using available values from suspended growth reactors in the present study is justified because the processes are identical and the microorganisms involved in the fixed-film reactors are similar to the ones present in activated sludge reactor systems (after Gonçalves, 2007). Similar conclusion is found in an investigation of a full-scale biological aerated filter (BAF) (Wang et al., 2016) placed as a tertiary stage for additional nitrification of the effluent from an activated sludge WWTP for BOD_5 and nitrogen removal. The on-line monitoring of the N_2O emissions from the BAF implemented over 12 months proved that the N_2O emission factors were comparable with the mainstream activated sludge system.

$$CH_4 = (BOD_{5.infl} - S) \times EF_{CH4} \qquad (3)$$

where S = BOD_5 removed in the anoxic fixed-film reactor in the form of sludge and EF_{CH4} = 0,0017 CH_4 per kg influent BOD_5 – calculated emission factor as explained below.

CH_4 emission factor is calculated as a mean value using data reported in two sources: (i) (Rodriguez-Caballero et al., 2014) – EF_1 = 0,0004, and (ii) (Masuda et al., 2018) – EF_2 = 0,0038 and EF_3 = 0,00083.

$$N_2O = N_{infl} \times EF_{N_2O-N} \times 44/28\ , \qquad (4)$$

where EF_{N2O-N} = 0,00157 kg N_2O-N per kg influent total inorganic N – calculated emission factor as explained below; 44/28 – the conversion factor from kg N_2O-N to kg N_2O.

N_2O-N emission factor is calculated as a mean value of seven emission factors chosen from sources reporting similar treatment processes as in the present study as given in the following **Table 3**. To compare the effect between different gases, their carbon dioxide equivalents (CO_2 eq) are calculated by using the GWP of each gas as a multiplier taken from AR6 in **Table 1**. The assessment period is 1 year.

TABLE 3. N_2O emission factors in full-scale domestic wastewater treatment plants.

Type of treatment process	References	N_2O emission factor (kg N_2O-N/kg N)
Anoxix-aerobic (An-A) for Carbon and Biological Nitrogen Removal (CBNR)	Rodriguez-Caballero et al. (2014)	0.00116
An-A for CBNR	Masuda et al. (2018)	0.00124
An-A for CBNR	Masuda et al. (2018)	0.00461
Plug-flow 1 for CBNR	Ahn et al. (2010)	0,00261
Plug-flow 2 for CBNR	Ahn et al. (2010)	0,00057
Modified Ludzack-Ettinger (MLE) 1 for CBNR	Ahn et al. (2010)	0,00045
MLE 2 for CBNR	Ahn et al. (2010)	0,00038

3. RESULTS AND DISCUSSION

The GHGs are comprised of emissions related to the processes in the pilot plant system of two submerged fixed film reactors. The calculations are given in **Table 4**.

TABLE 4. GHG emissions from the SBRs system for advanced WW treatment**.**

r	Infl. BOD$_5$ - S	Infl. N	EF$_{CH4}$	EF$_{N2O-N}$	Emissions CH_4		Emissions N_2O		Total direct emissions	
-	kg/yr	kg/yr	kg CH_4 per kg BOD$_5$	kg N_2O-N per kg N$_{infl}$	kg CH_4 per yr	kg $CO_{2,}$eq per yr	kg N_2O per yr	kg CO_2eq per yr	kg $CO_{2,}$eq per yr	g CO_2eq per m^3 WW
0	21,447	5,738	0,0017	0,00157	0,036	0,972	0,0142	3,877	4,849	30,8
1	19,006	4,920	0,0017	0,00157	0,032	0,864	0,0121	3,303	4,167	26,4
2	19,871	5,344	0,0017	0,00157	0,034	0,918	0,0132	3,604	4,522	28,7
3	24,692	5,647	0,0017	0,00157	0,042	1,134	0,0139	3,795	4,929	31,3

The results show that the direct GHG emissions from the experimental submerged biological filters for carbon and nitrogen removal as a second stage of a WWTP are of the same magnitude, within the lower values of the range, as the estimates for biological film processes in a recent investigation (Hua et al., 2022) being in the range 35,8 ÷182 g CO_2eq per m^3 of WW treated. The individual gas results show the same trend. The estimates here are between 20,9 ÷ 24,6 g CO_2eq per m^3 of WW treated for N_2O and between 5,5÷7,2 g CO_2eq for CH_4 emissions while those reported in (Hua et al., 2022) are 35,2÷133,9 g CO_2eq for N_2O and 0,6÷48,1 g CO_2eq for CH_4. The direct emissions of N_2O expressed in CO_2eq dominate compared to CH_4, which coincides with the recent findings in (Hua et al., 2022) and (Tong et al, 2024).

It is necessary to emphasize again that the calculations here are with EFs from selected activated sludge processes relevant to the experimental study. Hence, the results could not be very precise having in mind the conclusion in (Sabba et al., 2017) that the N_2O emissions from nitrifying and denitrifying biofilms and their mechanisms in many cases may be higher and are more complex compared to suspended growth systems. This is due to the more intensive processes in the fixed film because of the higher concentration of biomass. The IPCC methodology used for GHG emission assessment is also based on EFs for general activated sludge technologies. This is obviously due to the insufficient data about real GHG emissions from biofilm reactors.

Optimizing treatment technology in order to reduce N_2O emissions is a challenge. High nitrogen removal degree of WWTP helps to lower the N_2O emissions. Biogenic N_2O production and emission during nitrification-denitrification can be reduced by optimizing process conditions, e.g. by providing sufficient nitrification reserve capacity; through introduction of intelligent aeration system for proper aeration because low dissolved oxygen may drive N_2O generation; by ensuring enough organic carbon availability for denitrification in response to influent fluctuations.

4. CONCLUSIONS

The widely used IPCC methodology is based on influent nitrogen and BOD_5 values, and emission factors for very general groups of treatment technologies based on suspended growth reactors. Hence, there is a need of more detailed technology classification and a customized approach in the selection of suitable EFs according to the research purpose.

In the present study, most of the direct GHG emissions from advanced WW treatment in experimental submerged biological filters are attributed to N_2O as compared to CH_4. A number of measures to reduce GHG emission at a WWTP could be implemented. However, biogenic N_2O production and emission during nitrification-denitrification cannot be completely avoided.

The results presented give a good idea of the magnitude of the GHG emissions from submerged fixed film reactors for advanced wastewater treatment and can be used as a basis for further developments in this field when there are no real measurements.

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