



Analysis of greenhouse gas emissions and the energy balance in a model municipal wastewater treatment plant

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ABSTRACT

Municipal wastewater treatment plants (WWTPs) with multi-stage activated sludge technology generate significant amounts of greenhouse gases (GHG) in the form of nitrous oxide N_2O , methane (CH_4) and carbon dioxide (CO_2). Although the exact magnitudes of the specific emissions are difficult to estimate, they strongly affect the energy balance of a plant. This article presents a simulation study carried out on a model municipal WWTP. The research aimed to analyse the potential for the reduction in GHG emissions through the operational optimization of some core operational parameters and its effects on the plant's energy balance. The results showed that the combined effect of optimization of the dissolved oxygen concentration in the aerobic zone, the solids retention time and the ratio of chemical oxygen demand to total nitrogen (COD:TN) in the influent may lead to a reduction in the N_2O emissions by 1,103 kg CO_2 eq/d and also a slight reduction in the CO_2 and CH_4 emissions, by 256 and 87 kg CO_2 eq/d, respectively. This was coupled with an improvement in the plant's net energy balance by 34 kW through the reduction in energy consumption for aeration of the activated sludge by 18 kW and the increased energy production from biogas by 16 kW.

Keywords: Wastewater treatment; Activated sludge; Greenhouse gases; Energy efficiency; Computer simulation

1. Introduction

Greenhouse gas (GHG) emissions from a municipal wastewater treatment plant (WWTP) are closely related to its net energy consumption. The majority of the research focuses on emissions of carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) that can reach up to 3.8 kg CO_2 eq/kg of biochemical oxygen demand (BOD) in the combined processes of aerobic and anaerobic decomposition of organic material [1]. While the mechanisms of CO_2 and CH_4 emissions have been well known and described with many mathematical models, the mechanism of N_2O

generation is still not fully understood. Although this phenomenon has been investigated for many years, as reported by [2–5] and others, its qualitative and quantitative descriptions are not sufficiently detailed. It is estimated that N_2O emissions from wastewater treatment processes may account for 26% of the total GHG emissions in the whole nexus of urban water use [6]. This comes from its high value of the 100-year global warming potential (GWP) equal to 298. However, the exact scale of this emission is difficult to assess.

In municipal WWTPs with multi-stage activated sludge reactors, N_2O is generated during the heterotrophic

denitrification and autotrophic nitrification of nitrogen present in the influent of a biological reactor [7–10]. The process of N_2O production during denitrification is well understood. It is commonly described as a four-stage reaction of nitrate (NO_3^-) reduction to gaseous nitrogen (N_2), carried on by heterotrophic bacteria and archaea, in which N_2O is one of the intermediate products. In this process, incomplete denitrification may result in increased N_2O emissions. Autotrophic nitrification is a two-stage process carried on by bacteria and archaea. In the first stage, ammonium ion (NH_4^+) is oxidized to hydroxylamine (NH_2OH), which is then further oxidized to nitrites (NO_2^-). In the second stage, NO_2^- is oxidized to NO_3^- . During this process, N_2O may be produced as a result of incomplete oxidation of NH_2OH . Although some authors state that this happens only at a very high nitrate concentration, it usually does not occur in municipal WWTPs [11,12]. At high nitrate concentrations and low oxygen availability, NO_3^- can be reduced to N_2O also by autotrophic bacteria capable of direct ammonia oxidation (AOB) with ammonia or hydrogen used as electron donors [13]. The contribution of the above processes to the overall N_2O emissions from a WWTP is unclear. Some authors state that significantly more N_2O is produced during autotrophic nitrification, especially that carried on by AOBs, than as a result of heterotrophic denitrification [6,13,14].

Three different approaches to a mathematical description of GHG emissions from wastewater treatment processes can be distinguished [15]. The first uses simple empirical models and indicators to calculate the average approximate GHG emissions for reporting and statistics. The second comprises simple static models that integrate GHG emissions from wastewater treatment and sludge processing. Such models have been developed, among others, by Shahabadi et al. [1], Monteith et al. [16] and Bridle et al. [17]. The third approach includes mechanistic models capable of dynamic simulation of GHG production during wastewater treatment and sludge processing. As none of the standard activated sludge models (e.g. ASM1, ASM2, ASM3 and ASM2d) incorporates GHG emissions, various modifications to the existing models have been proposed, e.g. by Von Schulthess and Gujer [3] or Snowling et al. [18]. An important step in this process was the publication of the activated sludge model for nitrogen (ASMN) by Hiatt and Grady [19]. The model described production of N_2O during heterotrophic denitrification but did not include the important production of N_2O during autotrophic ammonia oxidation. Thus, the ASMN is often used as a starting point in the development of

other, more complete N_2O emission models. The models which describe production of N_2O during biological nitrification and denitrification under aerobic and anaerobic conditions are presented, among others, by Ni et al. [14,20] or Mampaey et al. [21]. However, so far, even those models do not describe production of N_2O by autotrophic bacteria under aerobic conditions in full detail. In 2013, Ni et al. [22] compared four N_2O emission models with the experimental data [8,11,14,21]. The results showed that N_2O was produced as a result of both partial oxidation of hydroxylamine N_2OH and aerobic denitrification carried on by AOB. He suggested that a model of N_2O production should include the both above processes that were differently affected by the presence of oxygen in the environment. Although the production of N_2O during heterotrophic denitrification is described in good detail, the present state of knowledge does not allow to fully understand the mechanisms leading to its production by autotrophic bacteria under aerobic conditions [20]. Therefore, most research in this field focuses on understanding the mechanisms of aerobic N_2O production by autotrophs and development and validation of a reliable mathematical description of these phenomena.

2. Materials and methods

The model WWTP was patterned after the benchmark simulation model no. 2 (BSM2) presented by Jeppsson et al. [23]. Only some minor modifications have been introduced to the model for practical reasons. The influent flow to the plant was $Q = 13,300 \text{ m}^3/\text{d}$ and the plant's person equivalent (PE) was 68,052. The wastewater treatment line (Fig. 1) includes: a primary settler ($V = 665 \text{ m}^3$), a five-stage biological reactor configured as the modified Ludzack–Ettinger scheme ($V = 6,000 \text{ m}^3$) and a circular final clarifier ($V = 4,200 \text{ m}^3$). The sludge processing line comprises: a gravitational sludge thickener ($V = 300 \text{ m}^3$), an anaerobic digestion chamber ($V = 1,800 \text{ m}^3$) and the device for sludge dewatering. All zones in the biological reactor are of equal volume ($V = 1,200 \text{ m}^3$). Two of them are anoxic and three are aerobic with a fine bubble diffused aeration system and dissolved oxygen (DO) concentration set at 2.0 mg/L . The nitrate recirculation rate, from the last aerobic zone to the first anoxic zone, is set at $2Q$. The thickened sludge is subjected to mesophilic fermentation at 35°C for the period of 22 d.

Many factors may affect the magnitude of the GHG emissions from wastewater treatment. They are broadly presented by many authors [1,11,17,24–26]. The following factors have been selected for the study:

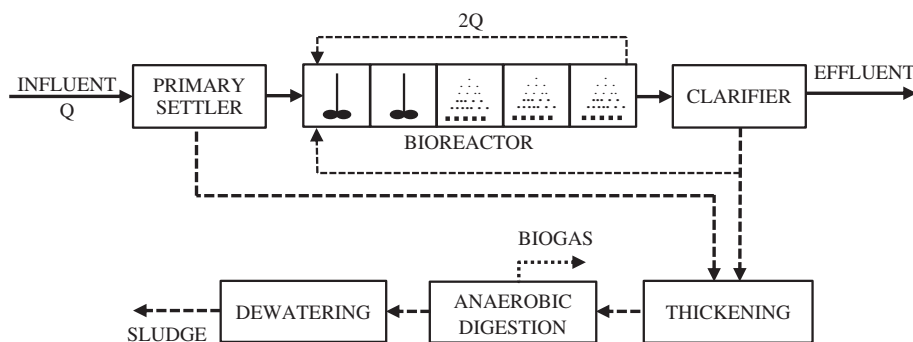


Fig. 1. Technological scheme of the studied WWTP model.

(1) oxygen conditions in the reactor; (2) process temperature; (3) biomass solids retention time (SRT); (4) availability of organic substrate in the influent represented by the ratio of chemical oxygen demand (COD) to total nitrogen (TN) in the influent wastewater. The above factors also affect the plant's net energy balance and are understood as the sum of the energy consumed for pumping, activated sludge aeration and mixing, excess sludge processing, and the heating of the sludge and the potential amount of energy which might be recovered from the methane produced during the fermentation of sewage sludge. For practical reasons, the balance does not include background and minor energy uses, indirect use of energy and energy used by the plant employees; they all do not vary significantly with the changing plant's operational parameters. The model does not consider any indirect emissions of GHGs (e.g. due to off-site energy generation or chemical production) or emission of CO_2 produced during incineration of CH_4 generated at the plant. It has also been assumed that only the portion of CH_4 that is generated during processing of sewage sludge in an anaerobic digestion chamber can be effectively captured and used as a potential energy carrier. GHGs generated in all other processes are immediately released to the atmosphere. Energy from biogas is calculated on the basis of CH_4 content only, assuming a conversion factor of 50.05 MJ/kg CH_4 . No combined heat and power installation is employed at the plant. During the study, the technological parameters were varied as follows: (1) DO concentration in the aerobic zone of the reactor in the range of $0\text{--}2.0 \text{ mg/L}$; (2) process temperature in the range of $10\text{--}20^\circ\text{C}$; (3) SRT of the biomass in the range from 13 to 43 d; (4) the ratio of COD:TN in the influent in the range of 7.5–13.3.

Simulations were carried out using the GPS-X[®] programme v.6.1 with the Model Developer module [27]. The mathematical model that was used is a mod-

ification of the "mantis2" model being a part of the "cn2iplib" library, which defines nitrification as a two-stage process with ammonia oxidized to nitrites by AOB and nitrites oxidized to nitrates by nitrite-oxidizing bacteria (NOB). The model has been supplemented with a gas emission description based on the models presented by Hiatt and Grady [19] and Ni et al. [20] incorporating NH_2OH and N_2O as new components in the mass balances. The detailed process matrices describing N_2O production by AOB and heterotrophic denitrifiers are clearly explained in Supporting Information to [14]. Emissions of CO_2 were calculated on the basis of stoichiometry where CO_2 is a product of organic material degradation according to the description proposed by Snowling et al. [18]. Energy consumption is a default variable assigned to individual processes in GPS-X[®] 6.1 and the details of its calculation are presented in [27]. The initial values of the operational parameters used during simulation are presented in Table 1 in regard to wastewater and in Table 2 in regard to GHG emissions.

3. Results

3.1. Effects of the activated sludge oxygenation level

The oxygen set point in the aeration zone of the model bioreactor was varied in the range $0\text{--}2.0 \text{ mg/L}$ with a step of 0.05 mg/L . The process temperature was maintained at 20°C . CO_2 , CH_4 and N_2O emissions from the bioreactor and from anaerobic digestion, energy consumption and production of energy from biogas generated during anaerobic digestion of sludge were simulated.

The results showed that while the gradual increase in the DO concentration in the bioreactor from approx. 0.3 to 2.0 mg/L causes the increase in energy demand for aeration from 83.5 to 108 kW , the amount of the produced CH_4 remains almost unchanged (Fig. 2(a)).

Table 1
Initial values of the operational parameters for the studied WWTP model

No.	Parameter	Unit	Value	
			Influent	Effluent
1	Influent flow rate	m ³ /d	13,300	
2	COD	mg/L	530	36.1
3	BOD ₅	mg/L	307	4.8
4	Total suspended solids	mg/L	250	9.0
5	TN	mg/L	60	14.5
6	Total phosphorus	mg/L	10	7.7
7	DO in aerobic zone	mg/L		2.0
8	Mixed liquor suspended solids (MLSS)	mg/L		3,415
9	SRT (bioreactor)	d		20.4
10	Anaerobic digestion time (at 35°C)	d		22
11	Methane production in anaerobic digestion	kg CH ₄ /d		565
12	Methane production factor	m ³ /kg BOD ₅		0.214
13	Unit energy use (per m ³ of wastewater) ^a	kWh/m ³		0.245
14	Unit energy use (per PE) ^a	kWh/PE		0.048

^aOnly direct energy consumption is included.

Table 2
Technological effects of parameter optimization in the studied WWTP model

No.	Parameter	Unit	Value	
			Before	After
1	DO concentration in aerobic zone	mg/L	2.0	1.0
2	SRT	d	20.4	17
3	COD:TN ratio	g COD/gN	8.8	10
4	CH ₄ production in anaerobic digestion	kg CH ₄ /d	564.8	590.4
5	Unit energy use (per m ³ of wastewater) ^a	kWh/m ³	0.245	0.212
6	Unit energy use (per PE) ^a	kWh/PE	0.048	0.041
7	Energy demand ^a	kW	242	224
8	Energy production from biogas	kW	346	362
9	Energy surplus	kW	104	138
10	Average production of N ₂ O	kg CO ₂ eq/d	3,641	2,538
11	Average production of CH ₄	kg CO ₂ eq/d	14,934	14,847
12	Average production of CO ₂	kg CO ₂ /d	7,921	7,665
13	Total GHG emission	kg CO ₂ eq/d	26,496	25,050

^aOnly direct energy consumption is included.

As a result, the excess energy gradually decreases from approx. 200 to 129 kW (Fig. 2(b)). Maximum production of N₂O (23.6 µg N₂O/g DS h) is observed at a DO concentration of approx. 0.25 mg/L. This matches the observations reported by Kampschreur et al. in their review article [6]. It states that the N₂O production reaches its maximum under oxygen-limiting conditions, when AOB use nitrite as the terminal electron acceptor to save oxygen for the oxygenation reaction of ammonia to hydroxylamine and that at DO concentrations below 1 mg/L N₂O production can correspond even to 10% of the TN load. Only Tallec et al.

[11] reported maximum N₂O emissions (7.1 µg N₂O/g DS h) at higher DO concentrations (1.0 mg/L) which was more typical for natural communities of AOB. This phenomenon is accompanied by an observed reduction in TN in the effluent caused by the removal of nitrogen from the system, with the N₂O released into the atmosphere.

3.2. Effects of temperature

The above simulations were repeated under modified conditions with the wastewater and ambient

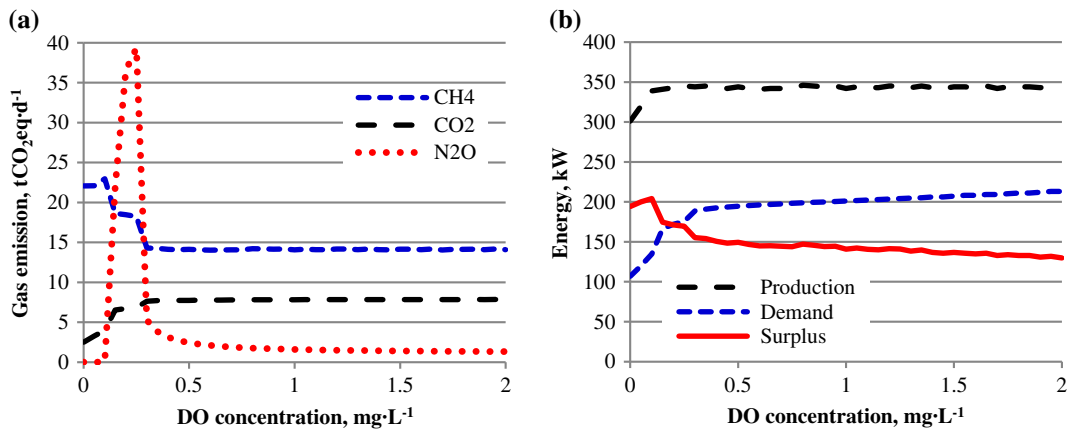


Fig. 2. The effect of the activated sludge oxygenation level on: (a) total GHG production and (b) energy production, demand and surplus.

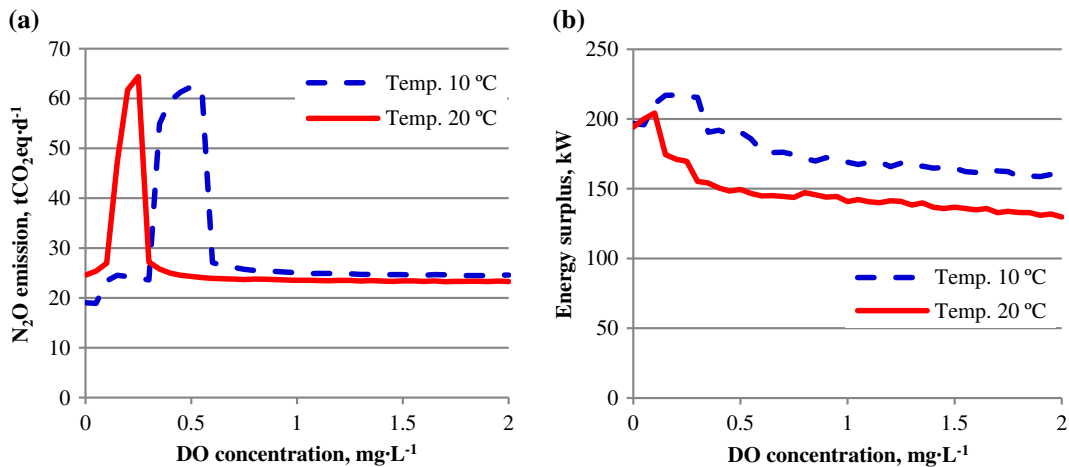


Fig. 3. The effects of the activated sludge oxygenation level and the process temperature on: (a) N₂O emissions from the bioreactor and (b) energy surplus.

temperatures set at 10 °C. The results, presented in Fig. 3, show that the decrease in the process temperature directly affects the N₂O emissions and the energy balance. Under low temperature conditions, increased N₂O emissions from the bioreactor are observed within the broad range of DO values from 0.35 to 0.60 mg/L, unlike at high temperature where it appears as a peak at 0.25 mg/L (Fig. 3(a)). At DO concentrations above 0.60 mg/L, the N₂O unit production is equal to approx. 24 µg N₂O/g DS h and is not clearly affected by the variations in process temperature. At a process temperature of 10 °C (Fig. 3(b)), surplus energy increases by approx. 19% as compared to 20 °C conditions. This is due to, on the one hand, lower energy demand for aeration from 89 at 20 °C to 76.2 kW at 10 °C, and on the other, higher methane production. The decrease in the

process temperature by 10 °C slows down the decomposition of organic matter in the reactor and increases the content of volatile organic material in the excess sludge supplied to the anaerobic digestion (54.8% at 10 °C against 51% at 20 °C). This results in higher methane production: 597 kg CH₄/d at 10 °C as compared to 560 kg CH₄/d at a temperature of 20 °C.

The reduction in the process temperature below 10 °C has no noticeable effect on the N₂O emissions, but it clearly worsens the plant's energy balance. At the ambient temperature of 0 °C, the amount of methane produced gradually reduces and the energy demand for heating the sludge in the anaerobic digestion tanks at 35 °C increases to 126 kW from approx. 84 kW at 10 °C. As a result, the surplus energy is reduced to approx. 130 kW at DO 1.0 mg/L.

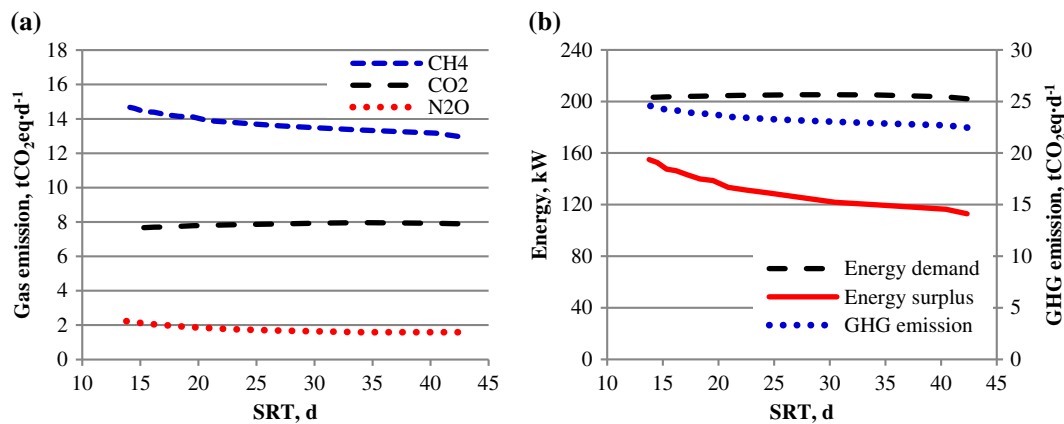


Fig. 4. The effects of the SRT variation on: (a) CO₂, CH₄ and N₂O, emissions and (b) energy demand, surplus and total GHG emission in the studied WWTP.

3.3. Effects of SRT

An overall SRT, understood as the average time the micro-organisms are in the system consisting of a bioreactor and a secondary settler, is an important operational parameter in all activated sludge systems. During the simulations, the effects of the SRT variations in the range 13–42 d, on the GHG emissions and the plant's energy balance, were observed. The DO concentration in the aerobic zone of the bioreactor was set at 1.0 mg/L and the process temperature was maintained at 20°C. The results showed that the production of CH₄ and N₂O clearly decreased with the growing SRT; it dropped from 585 kg CH₄/d and 7.5 kg N₂O/d at SRT = 13.8 d to 515 kg CH₄/d and 5.3 kg N₂O/d at SRT = 42.3 d, respectively. On the other hand, the CO₂ production was only slightly affected by the increasing SRT, growing from 7,600 to 7,900 kg CO₂/d (Fig. 4(a)). There is a clear relationship between the increase in SRT and the plant's energy balance. Although the energy demand remains almost constant at the level of approx. 203 kW, the overall energy surplus is reduced from 154.8 kW at SRT = 13.8 d to 112.9 kW at SRT = 42.3 d (Fig. 4(b)). This is due to the reduced content of biodegradable organic material in the excess sludge supplied to the anaerobic digestion process.

3.4. Effects of the availability of organic material in the influent

The processes of the heterotrophic decomposition of nitrogen compounds require the availability of readily biodegradable organic substrates. This can be expressed as a ratio of COD:TN in influent wastewater. During the simulations, the ratio of COD:TN in

the influent was varied in the range 7.5–13.3 and its effects on the GHG emissions and the energy balance were observed. The results show that the increase in the availability of organic substrate for denitrification does not have a clear effect on the plant's overall energy balance (Fig. 5(a)). The energy surplus is kept at a level of approx. 136–142 kW for the whole range of the studied COD:TN ratio. This arises from the fact that the increasing energy demand for the activated sludge aeration is compensated by the increased methane production. The surplus energy reaches its local maximum for the COD:TN ratio of 9–10. The effect of the COD:TN ratio on the GHG emissions is evident, especially for N₂O and, to a lesser extent, also for CH₄ (Fig. 5(b)). The increase in the COD:TN ratio from 7.5 to 10 causes a clear reduction in generated N₂O from 17.6 to 5.8 kg N₂O/d (from 5.2 to 1.7 t CO₂ eq/d). Its further increase has no clear effect on N₂O production.

4. Discussion

Although the BSM2 used in the research may, in some aspects, differ from the treatment schemes applied at many municipal WWTPs, the obtained results can become the basis of a more general discussion on the possibilities of reducing GHG emissions from a municipal WWTP and the improvement in its overall energy balance. In the analysis, special attention should be given to the N₂O emissions from a biological reactor because of its potential high warming coefficient and the emission of CH₄. Methane, on the one hand, contributes to an increase in overall GHG emissions, but on the other, if it is properly captured and incinerated, increases the plant's potential to achieve energy self-sufficiency.

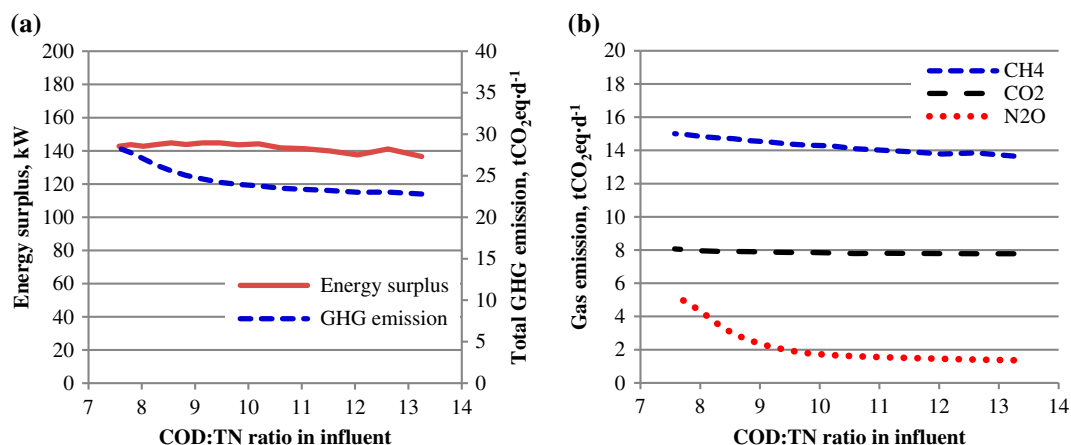


Fig. 5. The effects of the varied COD:TN ratio in the influent on: (a) energy surplus and total GHG emissions and (b) CO₂, CH₄ and N₂O emissions in the studied WWTP.

The results showed that, among the analysed factors, the DO concentration in the aerobic zone of the bioreactor plays an essential role in GHG emissions. When DO concentration in the aerobic zone was kept at 0.8 mg/L or more, the total GHG emission was maintained at almost a constant level of approx. 24 t CO₂ eq/d. At lower concentrations, a rapid increase in the amounts of N₂O and CH₄ produced in the bioreactor was observed. The increase in the DO concentration above 1.0 mg/L adversely affects the plant's energy balance through the increase in energy consumption for aeration at an almost unchanged level of the energy recovery from the biogas. This effect is observed for both the tested process temperatures, 10 and 20°C, however it should be noted that at 10°C the plant's overall energy surplus is by approx. 30 kW larger than at 20°C. Another factor that clearly affects the N₂O emissions is the availability of organic substrate for the nitrogen heterotrophic reduction process. The simulations showed that an amount of the organic substrate in the influent, described by a value of COD:TN ratio, should be equal to 10 or more. At smaller values of the COD:TN ratio, increased N₂O emissions were reported. The effect of this factor on the plant's energy balance was negligible. The biochemical activity of the activated sludge biomass strongly depends on its SRT. The simulations showed that the increase in the biomass retention time above the typical value of 16–20 d is detrimental to the plant's energy balance. This is mainly due to the increased mineralization of the waste sludge supplied to the anaerobic digestion process and the reduced production of biogas. The variations in this parameter have unnoticeable effects on GHG emissions other than the production of CH₄ in anaerobic digestion.

It was expected that the combined effect of optimization of the above parameters might bring noticeable outcomes in terms of GHG emission reductions and increased energy efficiency at the plant. The simulations were repeated with the optimized values of the analysed operational parameters (SRT = 17d, COD:TN = 10, DO = 1.0 mg/L) and compared with the results obtained from the original model (Table 2). The results showed that, after the optimization, the total production of the GHGs had been reduced by 1,446 kg CO₂ eq/d, with N₂O emissions reduced by 1,103 kg CO₂ eq/d, CH₄ emissions by 87 kg CO₂ eq/d and CO₂ emissions by 256 kg CO₂/d. This was accompanied by an increase in excess energy at the plant by approx. 34 kW. Even more benefits can be achieved if the "saved CO₂ emissions" due to the reduced purchase of non-renewable energy from outside are considered. Then, assuming an emission coefficient of 1.0 kg CO₂ eq/kWh following Moomaw et al. [28], excess energy of 816 kWh/d can save GHG emissions by approx. 816 kg CO₂ eq/d in addition to the direct emissions reductions presented above.

5. Conclusions

The simulation research carried out on the model plant showed that even simple modifications of some operational parameters at a municipal WWTP with multi-stage activated sludge technology may contribute to both a reduction in GHG emissions and an improvement in the plant's energy balance. Among the parameters that require special attention in this context are: DO concentration in the aerobic stage of the bioreactor, the SRT and the availability of organic substrate in the influent. In the specific case of the

model plant based on BSM2, the advocated values of these parameters were, respectively, 0.8–1.0 mg/L, 16–20 d and COD:TN \geq 10. This resulted in a direct reduction in the GHG emissions by 1,446 kg CO₂ eq/d and increased energy surplus of 816 kWh/d. It should however be noted that operation of an activated sludge system under low DO conditions requires effective monitoring and control of the process conditions and the effluent quality in order to avoid related problems, such as filamentous bulking, sludge mixing or a compromised quality of the effluent.

The results of the simulation study indicate significant potential for reduction in GHG emissions existing at many municipal WWTPs with multi-stage activated sludge that can be revealed by optimization of the operational parameters. However, it should be noted that the specific values of the operational parameters can differ among the plants depending on the details of the applied technologies, composition of the influent wastewater and the operational regimes. Further study is planned including model validation with the actual data derived from a medium-size municipal WWTP applying BNR technology and anaerobic digestion. Detailed and appropriately focused computer simulation research can be a valuable tool for setting the best optimization strategy for GHG emissions and energy efficiency at a specific plant.

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