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Estimation of greenhouse gas (GHG) emission from wastewater treatment plants and effect of biogas reuse on GHG mitigation

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Abstract. A comprehensive mathematical model was developed for this study to estimate on-site and off-site GHG emissions from wastewater treatment plants (WWTPs). The model was applied to three different hybrid WWTPs (S-WWTP, J-WWTP, and T-WWTP) including anaerobic, anoxic, and aerobic process, located in Seoul City, South Korea. Overall on-site and off-site GHG emissions from S-WWTP, J-WWTP, and T-WWTP were 305,253 $kgCO_2e/d$, 282,682 $kgCO_2e/d$, and 117,942 $kgCO_2e/d$, respectively. WWTP treating higher amounts of wastewater produced more on-site and off-site GHG emissions. On average, the percentage contribution of on-site and off-site emissions was 3.03% and 96.97%. The highest amount of on-site GHG emissions was generated from anoxic process and the primary on-site GHG was nitrous oxide (N_2O). Off-site GHG emissions related to electricity consumption for unit operation was much higher than that related to production of chemicals for on-site usage. Recovery and reuse of biogas significantly reduced the total GHG emissions from WWTPs. The results obtained from this study can provide basic knowledge to understand the source and amount of GHG emissions from WWTPs and strategies to establish lower GHG emitting WWTPs.

Keywords: greenhouse gas (GHG); wastewater treatment plant; on-site GHG emission; off-site GHG emission

1. Introduction

Global warming and climate change has become a world-wide environmental issue nowadays. There have been many efforts to cut down emission of greenhouse gases (GHGs), a main cause of global warming and climate change, globally and locally (Lamorena and Lee 2008, 2009, Olabisi *et al*. 2009, Hillman and Ramaswami 2010, Lamorena *et al*. 2013, Kyung *et al*. 2014). Industrial facilities such as coal-fired power plants, incineration facilities, and wastewater treatment plants (WWTPs) are known as the main sources of GHG emissions (Pacca and Horvath 2002, Astrup *et al*. 2009, Bani Shahabadi *et al*. 2009). It has been reported that WWTPs significantly contibute to the GHG emissions (US EPA 2008) because of the production of three primary GHGs, i.e., carbon

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dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O), during wastewater treatment processes at their on-site and off-site. On-site GHG emissions are generated from biochemical reaction for liquid and solid treatment and energy generation for plants by biogas and fossil fuel (Bani Shahabadi *et al*. 2009). Off-site GHG emissions are produced by the production of electricity for unit process operation as well as manufacturing and transportation of chemicals and fuels for on-site usage (Kyung *et al*. 2013). GHG emissions emitted from WWTPs accounts for 3% of global total GHG emissions (US EPA 2008). The impact of GHGs on global warming can be estimated by their global warming potential (GWP) and which of CO_2, CH_4, and N_2O are 1, 21, and 298, respectively, over a 100-years period. Because of strict regulation by international climate change prevention protocols, WWTPs will be faced with challenges of reducing of GHG emissions in the near future. Therefore, the emission of GHGs from WWTPs should be exactly estimated and reduced properly according to suitable management plan.

Under these circumstances, there have been approaches to develop a mathematical model to estimate GHG emissions resulting from on-site and off-site activities of WWTPs. However, previous models usually have overlooked CO_2 from biogenic sources under the intergovernmental panel on climate change (IPCC) protocol (Bani Shahabadi *et al*. 2010). In recent times, most attempts for the development of model have focused on the contribution of carbonaceous biochemical oxygen demand (CBOD) to GHG emissions without considering the impact of nutrient (total nitrogen: TN) removal from WWTPs. Moreover, N_2O emissions that can highly affect the variation of GHG estimation results have been neglected by models developed in previous studies due to lack of enough nutrient removal data and proper stoichiometries of biochemical reactions. Therefore, an accurate estimation of GHG emissions should be achieved by the development of novel comprehensive model.

In this study, we have (1) developed a WWTP-specific mathematical model to properly estimate on-site and off-site GHG emissions; (2) applied the model to three different WWTPs and compared the results based on type of GHG, process conditions, and WWTP capacity; and (3) predicted the amount of GHG mitigation by reusing biogas in WWTPs. This work can provide fundamental knowledge on GHG emissions from WWTPs and guideline to properly manage the WWTPs for sustainable environment.

2. Methodology

2.1 System boundary

The flow diagrams WWTP processes including aerobic, anoxic, anaerobic condition is illustrated in Fig. 1. System boundary to estimate on-site and off-site GHG emissions are represented by darker colors. Three representative WWTPs (S-WWTP, J-WWTP, and T-WWTP) in Seoul City, South Korea were chosen for the implementation of model developed in this study. The map of Seoul City and locations of the WWTPs are shown in Fig. 2. A2O process including anaerobic, anoxic, and aerobic reactor is one of the advanced hybrid system for municipal wastewater treatment and it was used for all of three WWTPs. It was the S-WWTP (2,000,000 m^3/d) that has the largest capacity for wastewater treatment among the WWTPs and followed by J-WWTP (1,710,000 m^3/d) and T-WWTP (1,110,000 m^3/d). Removal efficiency of BOD and TN in S-WWTP, J-WWTP, and T-WWTP were 91.73% and 43.44%, 91.49% and 42.65%, and 92.88% and 34.37%, respectively.

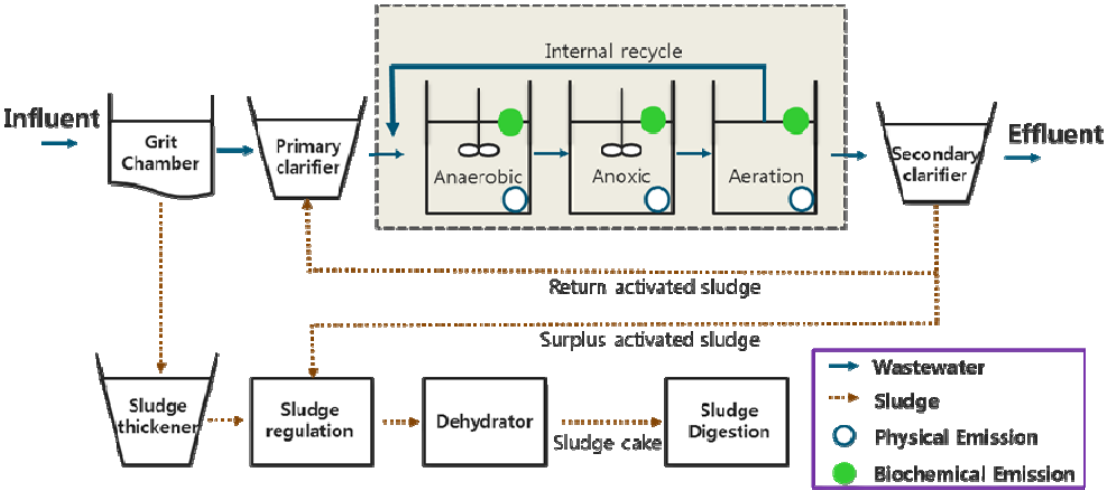

Fig. 1 Flow diagram of hybrid WWTP and system boundary for the estimation of GHG emissions

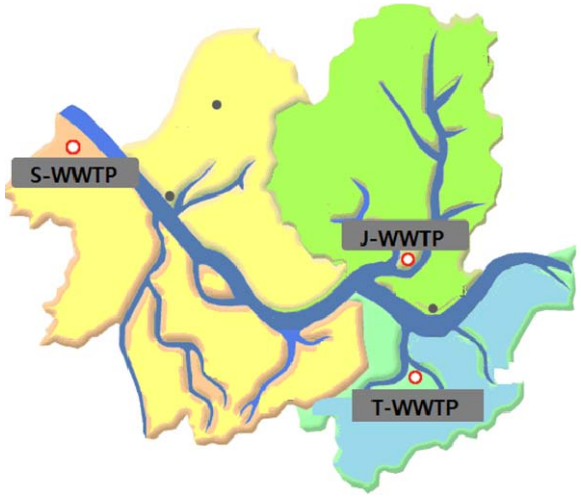

Fig. 2 The map of Seoul City and locations of the WWTPs

2.2 Estimation of GHG emissions from WWTPs

On-site GHG emissions of CO2, CH4, and N2O from biochemical reactions were separately estimated in each unit process using modified equations developed in previous studies (Keller and Hartely 2003, Tchobanoglous *et al*. 2004, IPCC 2006, US EPA 2008). The two clarifiers were assumed to be anaerobic conditions because their depth was more than 4 m, which is deeper enough to avoid oxygen contact during their operation. CO2 emissions from anaerobic, anoxic, and aerobic conditions were estimated using Eqs. (1)-(6) based on the removals of CBOD by microorganism. CO_2 emissions from anoxic and aerobic conditions were estimated by considering

denitrification and nitrification, respectively. In this study, we assumed that CH_4 can be produced only at anaerobic condition and it was estimated via Eq. (7). N_2O emissions were calculated by Eqs. (8) and (9). Different N_2O emission factors were applied to anaerobic, anoxic, and aerobic conditions due to diverse microbial activities at each condition. Denitrification and nitrification was considered to obtain N_2O emission factor for anoxic and aerobic condition, respectively (US EPA 2008). To obtain total on-site GHG emissions, estimated CH_4 and N_2O were converted to CO_2 equivalent by multiplying their GWP and then added.

$$\begin{aligned} &\Delta\text{BOD and } \Delta\text{TN} \\ &= S_{in} + S_{out} \text{ and } N_{in} + N_{out} \end{aligned} \tag{1}$$

- $CO_{2\,B}$ ($kgCO_2e/d$)

$$\begin{aligned} &\textit{Anaerobic} \\ &= EF_{\mathrm{CO_2}}^{T} \times Q \times \Delta\mathrm{BOD} \times \left(1 - 1.42 \times Y_{OBS}\right) \times 10^{-3}\, kgg^{-1} \end{aligned} \tag{2}$$

$$\begin{aligned} &\textit{Anoxic} \\ &= EF_{\mathrm{CO_2}}^{T} \times Q \times \left(\Delta\mathrm{TN} - 0.12 \times Y_{OBS} \times \Delta\mathrm{BOD}\right) \times 10^{-3}\, kgg^{-1} \end{aligned} \tag{3}$$

$$\begin{aligned} &\textit{Aerobic} \\ &= EF_{\mathrm{CO_2}}^{T} \times Q \times \Delta\mathrm{BOD} \times \left(1 - 1.42 \times Y_{OBS_n}\right) \times 10^{-3}\, kgg^{-1} \end{aligned} \tag{4}$$

$$Y_{OBS} = \frac{Y \times \left(1 + \left(f_d \times k_d \times SRT\right)\right)}{1 + \left(k_d \times SRT\right)} \tag{5}$$

$$Y_{OBS_n} = Y_{OBS} + \frac{Y_n \times \left(\left(N_{in} - N_{out}\right) - 0.12 \times Y_{OBS} \times \left(S_{in} - S_{out}\right)\right)}{1 + \left(k_{dn} \times SRT\right)} \tag{6}$$

- $CH_{4\,B}$ ($kgCO_2e/d$)

$$= GWP_{\mathrm{CH_4}} \times EF_{\mathrm{CH_4}}^{T} \times MCF \times Q \times \Delta\mathrm{BOD} \times \left(1 - 1.42 \times Y_{OBS}\right) \times 10^{-3}\, kgg^{-1} \tag{7}$$

- $N_2O_{\,B}$ ($kgCO_2e/d$)

$$= GWP_{\mathrm{N_2O}} \times EF_{\mathrm{N_2O}}^{T} \times Q \times \Delta\mathrm{TN} \times 10^{-3}\, kgg^{-1} \tag{8}$$

$$EF_{\mathrm{N_2O}}^{T} = NCF \times \left(0.16 \times Protein \times F_{industry}\right)^{-1} \tag{9}$$

During the electricity generation in power plants, GHGs can be formed via energy and fossil fuel consumption. Electricity generated from the plants is consumed for the operation of unit processes in WWTPs. We estimated off-site GHG emissions by multiplying the GHG emission factor (0.5584 $kgCO_2e/kWh$) with electricity demands at WWTPs (KEPCO 2011, Kyung *et al.* 2013) as described in Eq. (10). Off-site GHG emissions related to manufacturing of chemicals for on-site consumption were also considered in this work while that related to chemical

transportation were neglected due to lack of reliable data.

- $CO_{2\,E}$ ($kgCO_2e/d$)

$$= \sum \left(E_{unit\ process,i} EF_{Elect} \right) \tag{10}$$

To calculate the amount of reduced GHG emissions by biogas reuse, we assumed that biogas contain 80% of CH_4, and it is used to generate electricity by replacing the fossil fuel. Additionally, we assumed that all of combusted CH_4 gas is totally converted to CO_2 gas. Three scenarios were setup to investigate the effect of biogas reuse on total GHG emissions from WWTPs: (1) no biogas use; (2) 5% reuse; and (3) 10% reuse

3. Results and discussion

The total on-site GHG emissions from anaerobic, anoxic, and aerobic process at three different WWTPs are demonstrated in Fig. 3. Total on-site GHG emissions normalized by inflow wastewater at S-WWTP (4.45 gCO_2e/m^3) was the highest among the WWTPs and then followed by J-WWTP (4.33 gCO_2e/m^3) and T-WWTP (3.34 gCO_2e/m^3). This indicates that WWTP which has higher treatment capacity produces more on-site GHG emissions due mainly to higher removal mass of contaminant (BOD) and nutrients (TN). The mass of contaminant and nutrient are decided by multiplying concentration of BOD and TN (mg/L) with flow rate of wastewater (m^3/d). Because the concentration of inflow BOD and TN is similar at three different WWTPs according to geographical characteristics, total mass of contaminant and nutrient are highly influenced by treatment capacity of WWTPs. The average values of CO_2, CH_4, and N_2O emissions from three WWTPs were estimated to be 0.053, 0.528, and 3.457 gCO_2e/m^3, respectively (Table 1). The

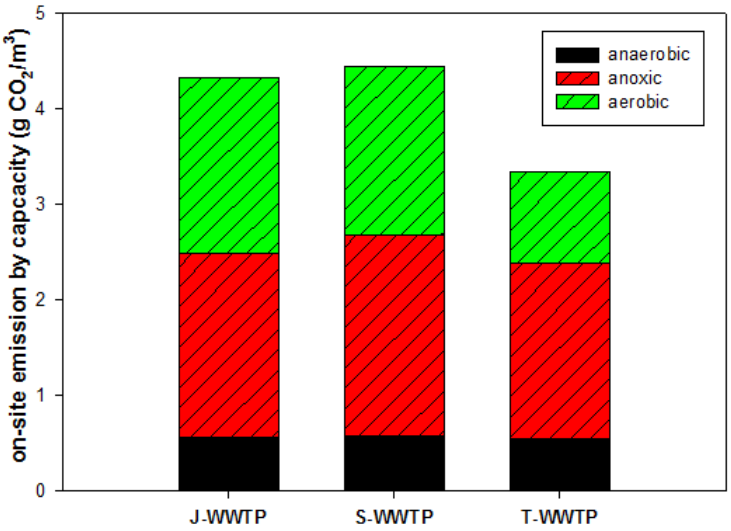

Fig. 3 The total on-site GHG emissions from anaerobic, anoxic, and aerobic process at three different WWTPs

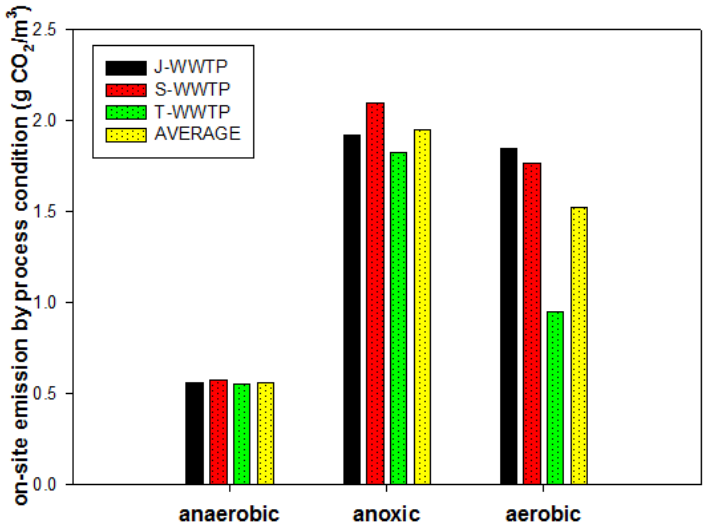

Fig. 4 On-site GHG emissions at different process conditions

Table 1 On-site emission by gas species

	CO_2	CH_4	N_2O	Total
	($g\ CO_2/m^3$)			
J-WWTP	0.0519	0.5241	3.7526	4.3285
S-WWTP	0.0542	0.5402	3.8525	4.4469
T-WWTP	0.0513	0.5185	2.7655	3.3353
Average	0.0525	0.5276	3.4568	4.0369
Percentage (%)	1.30	13.07	85.63	100

amount of N_2O was the highest and it was 6.6 and 65.8 times greater than that of CH_4 and CO_2. This indicates that N_2O is the most significant GHG contributing to on-site GHG emissions from WWTPs. During nitrification process, ammonia is oxidized to nitrite and/or nitrate in aerobic condition by microbes such as ammonium-oxidizing bacteria (AOB), ammonium-oxidizing archaer (AOA), and nitrite-oxidizing bacteria (NOB). Afterwards, nitrite and nitrate is changed to N_2O as intermediate products and further to nitrogen gas (N_2) during denitrification process at anoxic condition. This implies that perfect denitrification process is useful to significantly reduce the N_2O emissions from WWTPs. Adoption of Anammox (anaerobic ammonium oxidation) process which converts nitrate and ammonium directly into nitrogen gas or implementation of partial nitrification could be another option to mitigate the N_2O emissions (Kartal *et al*. 2007, Kampschreur *et al*. 2009, De Graaff *et al*. 2011). The generation of GHGs from anaerobic, anoxic, and aerobic conditions are estimated and results are presented in Fig. 4. Among the biochemical reactors, anoxic process mostly contributed to the on-site GHG emissions. This is because the highest amount of TN (19.93%) was removed at the process and converted to N_2O, having 310 times stronger global warming impact than CO_2. Considerable amount of GHGs were also produced from aerobic process while relatively small amount of on-site GHG emissions was

generated from anaerobic process.

Off-site GHG emissions related to electricity consumption and chemical production from three WWTPs are presented in Table 2. The percentage contribution of electricity consumption (92.56%) on total off-site GHG emissions was much bigger than that of chemical production (7.44%). This indicates that off-site GHG emissions can be significantly reduced if the electricity is less consumed with higher energy efficiency. Therefore, enhancement of energy efficiency of mechanical equipment (e.g., pumps and motor for mechanical mixing, backwashing, sludge transport, etc.) by retrofitting old one and optimizing unit operations would be helpful to remarkably mitigate off-site GHG emissions from the hybrid WWTPs. The amount of total off-site GHG emissions was the highest at S-WWTP (296,359 $kgCO_2e/d$ or 148.2 gCO_2e/m^3) and then followed by J-WWTP (225,280 $kgCO_2e/d$ or 131.7 gCO_2e/m^3) and T-WWTP (117,942 $kgCO_2e/d$ or 107.2 gCO_2e/m^3). This order was corresponded to the wastewater treatment capacity, indicating that larger treatment system demands more electricity and chemicals for process operation, thereby leading to more off-site GHG emissions.

Overall GHG emissions from S-WWTP, J-WWTP, and T-WWTP were 305,253 $kgCO_2e/d$ (152.6 gCO_2e/m^3), 282,682 $kgCO_2e/d$ (136.1 gCO_2e/m^3), and 117,942 $kgCO_2e/d$ (110.6 gCO_2e/m^3), respectively, and the portion of on-site and off-site emissions on overall was 3.03% and 96.97% on average (Table 3). This indicates that contribution of off-site GHG emissions is much higher than on-site GHG emissions for total GHG emissions from WWTPs. This is due to the high GHG emission factor for the production of electricity and chemicals related to off-site GHG emissions and their relatively high demand during the treatment process. This implies that electricity generation and chemical manufacturing for on-site consumption by alternative methods that produce lower amount of GHG is highly required to significantly reduce the off-site GHG emissions, thereby reducing overall emissions from WWTPs. The amounts of biogas generated from S-WWTP, J-WWTP, and T-WWTP were 73,743 m^3/d, 58,822 m^3/d, and 33,682 m^3/d, respectively. Total GHG emissions from three different WWTPs with and without biogas reuse are demonstrated in Fig. 5. In case of 5% biogas reuse rate for electricity generation instead of fossil fuel consumption, the reduction of the overall GHG emissions was 46.9%, for S-WWTP, 42.0% for J-WWTP, and 29.6% for T-WWTP, respectively. When 10% of biogas are reused for WWTPs, the total GHG emissions from three WWTPs reduced by 80.8% compared to the case without biogas reuse. This suggests that recovery and reuse of biogas produced in anaerobic process for energy (electricity and/or heat) generation to replace fossil fuel combustion is essential to significantly offset the total amounts of on-site GHG emissions from hybrid WWTPs (Bani Shahabadi *et al*. 2010, Yerushalmi *et al*. 2013).

Table 2 Total amounts of off-site CO2 emissions from WWTPs

	Electricity consumption	Chemical production	Total off-site emissions
	CO_2 emission ($kgCO_2/m^3$)		
J-WWTP	0.1268	0.0049	0.1317
S-WWTP	0.1347	0.0135	0.1482
T-WWTP	0.0969	0.0104	0.1072
Average	0.1194	0.0096	0.1290
Percentage (%)	92.56	7.44	100

Table 3 Total GHG emissions from WWTPs

	On-site emission	Off-site emission	Total emission
		(kg CO_2/m^3)	
J-WWTP	0.00433	0.1317	0.13607
S-WWTP	0.00445	0.1482	0.15263
T-WWTP	0.00334	0.1072	0.11056
Average	0.00404	0.1290	0.13308
Percentage (%)	3.03	96.97	100

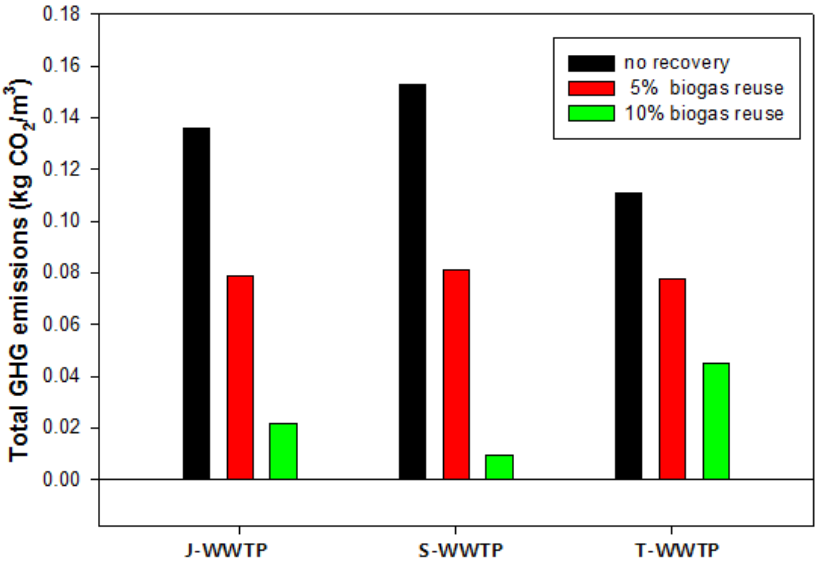

Fig. 5 Total GHG emissions with and without biogas recovery and reuse

4. Conclusions

We have quantitatively estimated on-site and off-site GHG emissions from hybrid WWTPs by using WWTP-specified model developed in this study. We also have investigated the most influential factors for GHG emissions and suggested tactics to efficiently reduce the emissions from WWTPs. The amounts of on-site and off-site GHG emissions were proportional to removed contaminants and nutrients and wastewater treatment capacity. N_2O generated from the nitrogen removal was the most influential gas for the on-site GHG emissions. Electricity consumption for the unit operation in WWTPs was the most significant factor for the off-site GHG emissions. GHG mitigation tactics based on the recovery and use of biogas for energy generation to substitute fossil fuel usage are highly recommended. Another strategy for GHG reduction is to use alternative nutrient removal process that can significantly reduce the N_2O emissions during the WWTP operation and to use sustainable methods that generate lower amount of GHGs for electricity generation and chemical production. This study can provide fundamental knowledge to understand sources and amounts of GHG emissions from WWTPs and idea to establish sustainable WWTPs

that emit lower amount of GHGs with higher water quality in the near future. The developed model can be modified and applied to other environmental infrastructure such as water treatment plants, rain water integration system, and sewer pipeline system.

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Nomenclature

$CH_{4\,B}$	theoretical–biochemical CH_4 emission (kg CO_2 e/d)	N_{in}	influent total nitrogen concentration (mg/L)
$CO_{2\,B}$	theoretical–biochemical CO_2 emission (kg CO_2/d)	N_{out}	effluent total nitrogen concentration (mg/L)
$E_{unit\ process,i}$	energy dissipation of unit process (kWh/d)	*NCF*	conversion factor of N_2O (g N_2O/person/d)
EF^T_{CH4}	emission factor of CH_4 (kg CH_4/kg BOD)	N_2O_B	theoretical–biochemical N_2O emission (kg CO_2 e/d)
EF^T_{CO2}	emission factor of CO_2 (kg CO_2/kg BOD)	*Protein*	protein intake (g protein/person/d)
EF_{Elect}	emission factor of electricity consumption (kgCO_2e/kWh)	*Q*	inflow rate of wastewater (m^3/d)
EF^T_{N2O}	emission factor of N_2O (kg N_2O/kg N)	S_{in}	influent BOD concentration (mg/L)
f_d	fraction of biomass that remains as cell debris (g VSS/g VSS)	S_{out}	effluent BOD concentration (mg/L)
$F_{industry}$	factor for non-consumed protein added to water	*SRT*	solid retention time (d)
GWP_{CH4}	global warming potential of CH_4	*Y*	yield coefficient (g VSS/g BOD)
GWP_{N4O}	global warming potential of N_2O	Y_N	biosynthesis yield coefficient for nitrifying bacteria (g VSS/g N)
k_d	endogenous decay rate for heterotrophic biomass (g VSS/g VSS/d)	Y_{OBS}	observed heterotrophic yield (g VSS/g BOD)
k_{d_n}	endogenous decay rate for nitrifying microorganisms (g VSS/g VSS/d)	Y_{OBS_n}	observed nitrifying yield (g VSS/g N)
MCF	conversion factor of CH_4		