Aerosol and Air Quality Research, 17: 2608–2623, 2017

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2017.08.0276

Technical Note

Diurnal Variation of Greenhouse Gas Emission from Petrochemical Wastewater Treatment Processes Using *In-situ* Continuous Monitoring System and the Associated Effect on Emission Factor Estimation

Wen-Bin Yang[1], Chung-Shin Yuan[1*], Wei-Hsiang Chen[1], Ying-Hsien Yang[2], Chung-Hsuang Hung[3]

[1] *Institute of Environmental Engineering, National Sun Yat-sen University, Kaohsiung 80424, Taiwan*
[2] *Department of Environmental Resources Management, Chia Nan University of Pharmacy and Science, Tainan 71710, Taiwan*
[3] *Department of Safety, Health and Environmental Engineering, National Kaohsiung First University of Science and Technology, Kaohsiung 81164, Taiwan*

ABSTRACT

The temporal variation of greenhouse gas (GHG) emission in a petrochemical wastewater treatment plant (WWTP) was investigated in this study. Two approaches including an *in-situ* continuous monitoring and a typical grab sampling methods were also compared. The *in-situ* continuous monitoring method provided more detailed information regarding the temporal variations of GHG concentrations. A sufficient sampling frequency (e.g., once every 6 hours) for the grab sampling method is required to effectively resolve the diurnal variations of GHG concentrations. This study highlights significant diurnal variations of GHG concentrations in different wastewater treatment units. Only with proper and reliable sampling and analytical methods, it becomes possible to correctly identify the characteristics of GHG emissions and to develop strategies to curtail the GHG emissions from such an important source in response to regulatory measures and international treaties. This study revealed that N_2O was the dominant species responsible for GHG emissions in the WWTP and the emission factors of CH_4 and N_2O were higher in the equalization tank and final sedimentation tank compared to other units. We further compared the GHG emission factors of this study with other literatures, showing that the GHG emission factors were lower than those measured in Netherlands, Australia, and IPCC, but similar to those measured in Japan.

Keywords: Greenhouse gas; Wastewater treatment; *In-situ* continuous measurement; Grab sampling method; Diurnal variation; Emission factor.

INTRODUCTION

Greenhouse gas (GHG) emissions by anthropogenic activities such as production and use of fossil fuels and agricultural and industrial activities have considerably increased the GHG concentrations in the atmosphere (El-Fadel and Massoud, 2001; Fangueiro *et al.*, 2010; Daelman *et al.*, 2012; Daelman *et al.*, 2013; Muangthai *et al.*, 2016). Of many sources, wastewater treatment plants (WWTPs) represent one important source of GHGs, particularly methane (CH_4) and nitrous oxide (N_2O) (IPCC, 2006a; Amina *et al.*, 2013; Huang and Tan, 2014). While the GHG emissions from WWTPs may be less significant compared to those from energy sectors or solid waste disposal sites, WWTPs are common components in these facilities and their impacts ought to be discussed before determining the importance of this particular GHG source. Although the GHG emission fluxes of wastewater treatment plant were higher than those of solid waste disposal, the amounts of GHG emission from wastewater treatment plants were relatively low due to their limited emission area compared to other GHG sources. Previous study reported that the wastewater treatment and discharge account for 9% of the main source of entire greenhouse gas emissions. The emission of carbon dioxide (CO_2) in WWTPs is typically not considered given its biogenic origin (Shahabadi *et al.*, 2009). As the GHG emissions in WWTPs are affected by source water characteristics, industrial WWTPs are reported to be more important for producing CO_2, CH_4, and N_2O in the fields (Kishida *et al.*, 2004). The GHG emission was further influenced by the compound's physicochemical

* Corresponding author.
E-mail address: ycsngi@mail.nsysu.edu.tw

properties and treatment technologies of interest (Amina *et al.*, 2013). Due to the significant environmental impacts by climate change attributable to global warming, there is an increasing need to comprehensively understand the GHG emission from industrial WWTPs and to provide detailed and correct information for developing mitigation approaches.

Wastewater treatment is the fifth largest source for the anthropogenic CH_4 emissions in the atmosphere, contributing approximately 9% of total CH_4 emission in 2000 (USEPA, 2006; Jarosław *et al.*, 2016). The combined emissions in the U.S., China, India, and Indonesia accounted for 49% of the global CH_4 emission from wastewater treatment. For N_2O, wastewater treatment represents the sixth largest contributor for anthropogenic emissions in the atmosphere (approximately 3% of total N_2O emission). The combined emissions from wastewater treatment in the U.S., China, India, and Indonesia contributed to approximately 50% of total N_2O emission in the atmosphere. The CH_4 and N_2O emissions from treatment of wastewater are expected to grow by approximately 20% and 13% between 2005 and 2020, respectively (Diksha and Santosh, 2012).

As CH_4, and N_2O represent two species of major concern in WWTPs, their formation and emission characteristics are different. Biological degradation of organic and inorganic matters in wastewater is the main process that forms GHGs in WWTPs (El-Fadel and Massoud, 2001; Shahabadi *et al.*, 2010; Casey, 2011; Dimoula *et al.*, 2016). Methane is mainly produced from anaerobic biological processes in WWTPs, accounting for 3–19% of global anthropogenic CH_4 emission. N_2O is the intermediate present in aerobic nitrification or anoxic denitrification processes widely used in industrial WWTPs. Three percent of the anthropogenic N_2O emission to the atmosphere was associated with the formation in WWTPs (IPCC, 2006b; Diksha and Santosh, 2012; Rajab *et al.*, 2012; Tolkou and Zouboulis, 2012; Lin *et al.*, 2015). The Intergovernmental Panel on Climate Change (IPCC) determined the global warming potentials of GHGs based on the radiative efficiencies and lifetimes of GHGs in the atmosphere (IPCC, 2006b). For example, the GWPs of CH_4 and N_2O for 100-year time horizon are 28 and 265, respectively, as the GWP of CO_2 for the same time horizon equals to unity (IPCC, 2014a).

In addition to treatment technologies, activities including energy consumption in operation and sludge treatment and disposal contribute to additional GHG emission in WWTPs (Keller and Hartley, 2003; Foley *et al.*, 2008; Ren *et al.*, 2013; Xu *et al.*, 2014). With water quality and operation variations, the campaign to manage the GHG emission from WWTPs becomes complicated and challenging. The typical grab sampling method (i.e., the United States Environmental Protection Agency's TO-17 Standard Method) collects GHG samples at certain time intervals followed by laboratory analysis, possibly insufficient to describe the temporal variation of GHG formation and emission in WWTPs if the sampling frequency is inadequate. The objective of this study was to test an *in-situ* measurement that continuously monitors the GHG emission, avoid misestimating the GHG production in WWTPs and their emission factors. The results were compared with those acquired by using the USEPA's TO-17 Standard Method for consistency. The data were applied to estimate the mass fluxes and emission factors, quantifying the extents of errors that could be caused by using different monitoring approaches. The novelty was to indicate the importance of accurate measurement of GHG emission in WWTPs by analyzing the emissions continuously or with sufficient frequencies, providing reliable information for the following management approaches.

MATERIALS AND METHODS

Wastewater Treatment Plant

A petrochemical WWTP in southern Taiwan was selected as the study site. The WWTP is located in Kaohsiung City of Southern Taiwan, which has been an important hub for the national industrial development in Taiwan. The extents of air pollutions and GHG emissions from industrial complexes in these areas have been well known by the public (Yang *et al.*, 2014). The WWTP of interest treats the wastewater generated by Renwu and Dashe petrochemical complexes, as the contributions from these two industrial complexes accounted for more than 80% of total GHG emission in Kaohsiung City (Yang *et al.*, 2014).

The treatment technologies in the selected WWTP consist of two equalization tank in parallel (The hydraulic retention time (HRT) is 4–9 hours), two primary sedimentation tanks in parallel (HRT = 1–4 hours), two aeration tanks in parallel (HRT = 4–8 hours), one sludge thickener (HRT = 6–18 hours) and two final sedimentation tanks in parallel (HRT = 2–4 hours), as shown in Fig. 1. The sludge collected through sedimentation is treated in a sludge thickener, followed by dehydration and drying for disposal. Polymers and activated carbon are added prior to the aeration tanks to enhance the removal of compounds which are difficultly treated by the biological process. The designed daily and maximum treatment capacities are 3,800 and 4,000–5,000 cubic meter per day (CMD), respectively. The treatment throughput is typically reduced by half in summer because the industrial activities in these complexes produce less wastewater during the periods.

In-Situ Continuous Measurement

An *in-situ* measurement with a floating chamber was developed by modifying the design by Bao *et al.* (2015) and Sebastian *et al.* (2013) to continuously monitor the GHG concentrations in the WWTP (Fig. 2). The equipment consisted of a floating chamber, a dust removal unit, a rotameter, a sampling pump, a sampling bag, and a continuous GHG analytical instrument. The chamber was designed to create the best mixing and sampling conditions without altering the emission of gases at the surface. Air sample emitted from water surface were completely mixed with the make-up air in the floating chamber and filtered for removal of particles in the air. A Teflon tube was connected to the top of the floating chamber and air sample was transported and analyzed *in-situ* with an instrument (Teledyne Analytical Instruments Series 7600, USA) that directly measured the CO_2, CH_4, and N_2O concentrations.

The quantifiable concentration ranges of three GHGs

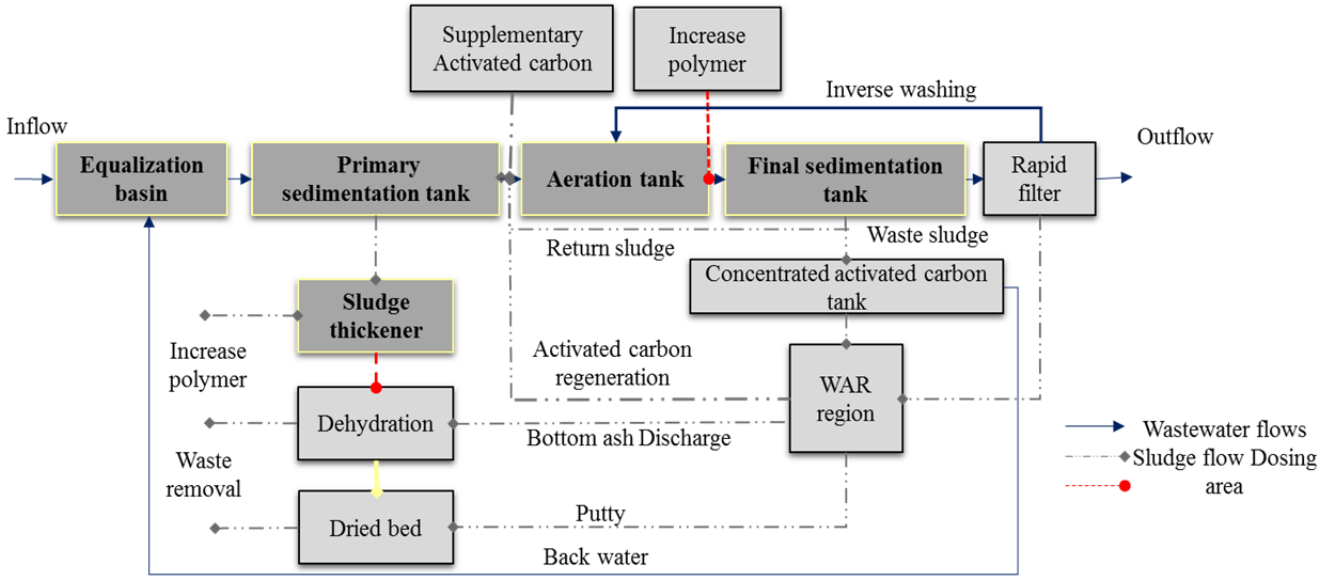

Fig. 1. Treatment processes of the WWTP selected in this study.

ranged from 0 to 2,500 part per million (ppm). At the beginning of every field analysis, standards with pre-determined concentrations were analyzed to ensure the quality of the data (< 20% relative percentage difference).

The continuous GHG monitoring was undertaken in winter and summer to represent the data in summer and winter, respectively. At each monitoring site, the GHG monitoring was conducted for 24 hours a day and lasted for 6 days. The GHG concentrations were measured and recorded every five minutes. The monitoring sites included the equalization tank, primary sedimentation tank, aeration tank, final sedimentation tank, and sludge thickener. The meteorological data including the air temperature and relative humidity were obtained through a nearby meteorological station operated by the Central Weather Bureau of Taiwan. The atmospheric temperatures were 26°C and 33°C in winter and summer January and Jun of 2014, respectively.

Grab Sampling Method

For comparison with the results by the *in-situ* continuous monitoring, air samples were collected by using tiffin sampling bags, followed by laboratory analysis to determine the GHG concentrations with the USEPA Standard Method TO-17 (USEPA, 1999). The CO_2 and CH_4 concentrations were analyzed by using a gas chromatography coupled with a flame ionization detector (GC-FID) and a methane converter (G1530A, Agilent, USA). The GC-FID was equipped with a 15 m × 0.32 mm I.D. PLOT-Q capillary column with 2.0 μm film thickness (Agilent, USA). One μL of sample was injected in the splitless mode. The column temperature was programmed as follows: an initial oven temperature of 150°C ramped at 10 °C min^{-1} to 300°C, then 30 °C min^{-1} to 220°C, and held for 6 min. The methane conversion temperature was 375°C. The FID was performed in the flame ionization mode and the ion source temperature was 300°C. Data acquisition was performed with a source temperature at 300°C. The method detection limits (MDLs) of CO_2 and CH_4 were 1 and 0.5 mg L^{-1}.

The N_2O concentrations were analyzed by using a GC coupled with an electron capture detector (GC-ECD) (G3440B, Agilent, USA). The GC-ECD was equipped with a 30 m × 0.53 mm I.D. PLOT/Q capillary column with 4.0 μm film thickness (Agilent, USA). One μL of sample was injected in the splitless mode. The column temperature was programmed as follows: an initial oven temperature of 40°C ramped at 10 °C min^{-1} to 120°C, then 30 °C min^{-1} to 250°C, and held for 5 min. The source temperature was 250°C. Data acquisition was performed with a source temperature at 250°C. The MDLs of N_2O was 0.5 mg L^{-1}. All of the samplings and analyses conducted in this study were at least duplicated.

Emission Flux and Emission Factor Calculations

The emission fluxes or emission factors were estimated to quantify the extents of three GHGs released into the atmosphere in wastewater treatment. The emission fluxes are expressed as the weight of a GHG divided by a unit area and time, while the emission factor considers the weight of the activity emitting the GHG (e.g., grams of a GHG emitted per gram of COD removed). These factors facilitate the discussion of GHG emissions between the WWTP selected in this study and those investigated in other studies. In this study, the emission fluxes of three GHGs were calculated by using Eq. (1) (Hobson, 2012; Taiwan EPA, 2013; Yan and Liu, 2014; Chen *et al.*, 2017).

$$E = \frac{Q_a \times C_{GHG}}{A_C} \times 10^{-3} \quad (1)$$

where E denotes the GHG emission flux [g m^{-2} min^{-1}]; Q_a denotes the volumetric flowrate of sampling air [L min^{-1}] (2 L min^{-1} ± 5%) in the floating chamber; C_{GHG} denotes the GHG concentration in the floating chamber [g m^{-3}]; A_C denotes the interfacial areas between the air and water

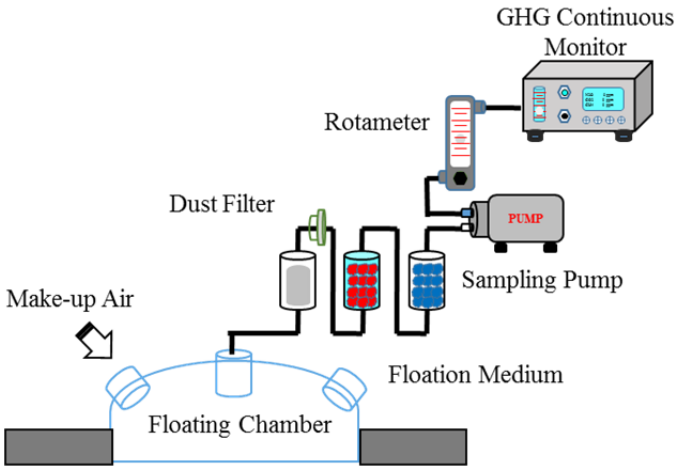

Fig. 2. Scheme of the *in-situ* continuous measurement to determine the GHG concentrations in the wastewater treatment processes.

phases in the floating chamber [m^2] (0.76 m^2). The total GHG emissions and emission factors of each wastewater treatment unit were estimated by using Eqs. (2) and (3), respectively.

$$TE = E \times A_T \times 1440 \quad (2)$$

$$EF = \frac{TE}{Q_w \times (C_{wi} - C_{we})} \quad (3)$$

where TE denotes the total GHG emission in each wastewater treatment unit within a day [g day^{-1}]; A_t denotes the total surface area of wastewater in each treatment unit; EF denotes the GHG emission factor in each wastewater treatment unit [g kg^{-1}]; Q_w denotes the volumetric flowrate of wastewater [m^3 day^{-1}]; C_{wi} and C_{we} denote the chemical oxygen demand (COD) or total Kjeldahi nitrogen [mg m^{-3}] in the influent and effluent of each wastewater treatment unit.

Water Quality Sampling and Analysis

The methods used to sample and analyze the water quality parameters of concern followed the standard methods developed by the National Institute of Environmental Analysis in Taiwan Environmental Protection Administration (Taiwan EPA). Wastewater samples taken at the both inflow and outflow of each unit of WWTPs were preserved in brown glass bottles that were stored in an ice box and then transferred back to the laboratory for water quality analysis. The water quality parameters analyzed included MLSS, SS, pH, COD, total Kjeldahl nitrogen, NH_4^+-N, NO_3^--N, and NO_2^--N following the Standard Methods NIEA W210.58A, NIEA W424.52A, NIEA W515.54A, NIEA W420.50B, NIEA W437.52C, NIEA W458.50, and NIEA W459.50B, respectively.

RESULTS AND DISCUSSION

GHG Analysis by Two Approaches

Table 1 summarizes the influent wastewater quality information in the sampling and analysis periods. The effluent water quality met the regulations determined by the Taiwan EPA, as the concentrations of COD, SS, total nitrogen were required to be below 100, 30, and 20 mg L^{-1}, respedively. The average removal efficiencies of MLSS, SS, COD, NH_4^+-N, NO_2^--N, NO_3^--N, and TKN in two seasons were in the range of 96–97%, 79–86%, 76–79%, 84–86%, 90–93%, 83–88%, and 92–95%, respectively.

The standard gas concentrations of CO_2, CH_4, and N_2O were 929.0, 104.7, and 52.4 ppm, respectively. Every five-minute measurement was recorded to quantify the differences between the three GHG concentrations for a total of 10 times. The mean and standard deviation ($\bar{X}$ ± SD) of the measured CO_2, CH_4, and N_2O concentrations were 926.0 ± 5.0, 103.4 ± 0.6, and 52.1 ± 0.5 ppm, respectively. The accuracies of CO_2, CH_4, and N_2O measurements using the continuous GHG monitoring method (Teledyne Analytical Instruments, Series 7600) were 99.68, 98.76, and 99.43%, respectively, with the relative errors of 0.32, 1.24, and 0.57%, respectively. Results obtained from the precision analysis showed that the relative standard deviation (RSD) of CO_2, CH_4, and N_2O were 0.56, 0.59, and 1.02%, respectively. Accordingly, the continuous GHG monitoring method demonstrated high accuracy and precision for in-situ on-line GHG measurement.

The GHG concentrations analyzed by the in-situ continuous monitoring and the grab sampling followed by GC analysis were compared in Figs. 3 and 4 that illustrate the GHG concentrations analyzed in the primary sedimentation tank of the WWTP in two seasons. The solid and dash lines in the figures represent the averages of the daily GHG concentrations analyzed by two analytical methods, respectively. The primary sedimentation tank was chosen because it is the 1st treatment step that receive the wastewater with a concentration expectedly higher than those in the subsequent treatment processes. It was initially assumed that the GHG concentrations could be relatively high in this step, making the location proper for comparison between the two analytical approaches. In the results, the *in-situ* continuous monitoring was more effective to capture the

Table 1. Influent wastewater quality of the WWTP during the monitoring periods.

Psarameter	Summer			Winter		
	Range	Average ± SD	Removal (%)	Range	Average ± SD	Removal (%)
Flow rate (CMD)	2867–3473	3170 ± 172		2984–3681	3332 ± 2157	
MLSS (mg L^{-1})	1853–2742	2272 ± 223	97	1581–2692	2082 ± 174	86
SS (mg L^{-1})	41–267	152 ± 6	96	35–263	114 ± 19	90
pH	7.5–7.9	7.7 ± 0.2	–	7.2–7.8	7.7 ± 0.4	-
COD (mg L^{-1})	115–344	217 ± 19	86	105–312	209 ± 23	93
HRT (hr)	4–8	–	–	4–8	–	–
NH_4^+-N (mg N L^{-1})	32–96	63 ± 21	79	33–114	72 ± 24	83
NO_2^--N (mg N L^{-1})	3.2–4.6	3.9 ± 0.5	76	3.9–5.1	4.6 ± 0.5	88
NO_3^--N (mg N L^{-1})	3.5–4.9	4.1 ± 0.5	79	4.1–5.6	4.8 ± 0.5	95
TKN (mg N L^{-1})	51–113	78 ± 24	84	66–129	86 ± 24	92

temporal variations of three GHG concentrations in the sampling periods. The average daily concentrations by the grab sampling method with different sampling frequencies were compared with those determined by the *in-situ* continuous measurement.

The relative percentage difference (RPD) was used to quantify the differences between the results measured by two approaches (the *in-situ* continuous monitoring and the grab sampling with different sampling frequencies) as shown in Table 2. Figs. 3 and 4 compare the averaged data measured by two approaches. These two approaches were conducted to simultaneously sample and measure GHGs over a sampling period of 10 min. The air flowrate of the grab sampling was 1.0 L min^{-1} and the volume of a Tedlar bag used for sampling GHGs is 10 liters. Thus, the sampling time was set as 10 min in order to fill the entire bag for further GC analysis. In summer, when the grab sampling was conducted once a day (i.e., every 24 hours), the RPDs of three GHG concentrations ranged from 2–52%. The RPDs dropped to 1–40% and 2–41% when the sampling frequency increased to twice (i.e., every 12 hours) and thrice a day (i.e., every 8 hours), respectively. When the grab sampling was undertaken four times a day (i.e., every 6 hours), the RPDs ranged from 0.1 to 17%. It is worth noting that the CO_2 emissions were different from those of CH_4 and N_2O. Given the high CO_2 background concentration (402 ppm) in the atmosphere, the CO_2 concentration increase due to the emission from the primary sedimentation tank could be limited, causing relatively lower RPDs for CO2 concentration comparison. Similar findings were observed in winter. The ranges of the RPDs between the average daily concentrations of three GHGs were 0.6–33%, 0.2–19%, 0–11%, and 0–8%, when the grab sampling was conducted once, twice, thrice, and four times a day. Monitoring the GHG concentrations continuously or with a sufficiently intensive sampling frequency is important given the strong temporal variations of GHG emissions in wastewater treatment. Additionally, by using these two approaches, the CO_2 concentrations ranged from 445 to 458 ppm (3% RPD between two approaches) and from 485 to 490 ppm (1% RPD between two approaches) in two monitoring seasons. The CH_4 concentrations by two approaches were within 35–38 ppm (7% RPD between two approaches) and 54–50 ppm (7% RPD between two approaches), while the N_2O concentrations ranged from 22 to 24 ppm (9% RPD between two approaches) and from 27 to 28 ppm (4% RPD between two approaches) in two seasons. The concentration variations between two seasons appeared to be limited, attributable to its stable influent wastewater quality through the operation.

In this study, the amounts of CO_2, CH_4, and N_2O productions in WWTPs in summer were 48027, 342, and 223 g day^{-1}, respectively, while those in winter were 57272, 283, and 487 g day^{-1}, respectively. Our results showed that the GHG productions obtained from this study were lower than Japan (CH_4: 263 g day^{-1}; N_2O: 196 g day^{-1}) but higher than USA (CH_4: 472 g day^{-1}; N_2O: 533 g day^{-1}) and Australia (CH_4: 467 g day^{-1}) (IPCC, 2006a, b; Ministry of the Environment, 2012).

Diurnal Variation of CO_2 Concentration

Figs. 5(a) and 6(a) provide the temporal variations of CO_2 concentrations in different treatment technologies of the WWTP in summer and winter, respectively. The equalization tank was the process with relatively higher emissions in both seasons. In summer, the CO_2 concentrations ranged from 1,923 to 2,340 ppm with an obvious diurnal variation during equalization (the average was 2,221 ppm). The concentration dropped to the range of 450 to 600 ppm (the average was 528 ppm) and became more stable in the primary sedimentation tank, possibly due to the characteristic of its close system. Since the aeration tank is open to the atmosphere, the CO_2 concentration was potentially enhanced and the concentration stably ranged from 2650 and 2750 ppm (the average was 2675 ppm) through 24 hours. In the sludge thickener, besides its fully sealed condition, sewage organic matter could be decomposed by microorganisms consuming oxygen, resulting in anaerobic state due to shortage of dissolved oxygen and forming excess CO_2 and CH_4 (Wei *et al.*, 2008). The CO_2 concentration ranged from 2,500 to 3,600 ppm with the average of 2893 ppm. The primary sedimentation tank is an open system in which appreciable levels of floating flocs could be overflowed across the weir. In this process, tiny flocs that contained microorganisms were partially suspended on the water surface, as the microorganisms could continuously decompose organic matters in wastewater, increasing the CO_2 concentration. The CO_2 concentration in the final sedimentation tank was within the range of 450 to 550 ppm

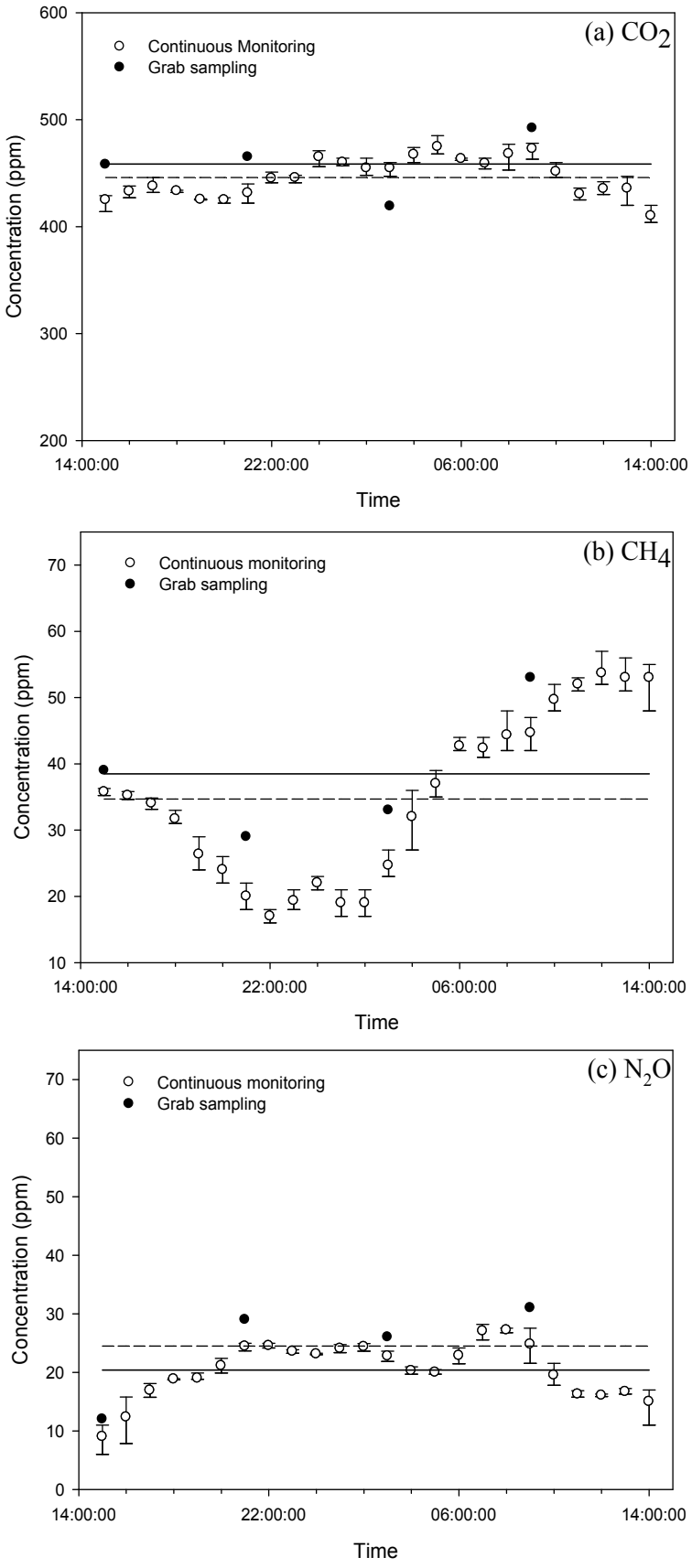

Fig. 3. GHG concentrations observed by the *in-situ* continuous measurement and the grab sampling method in summer. The solid and dash lines represent the average daily concentrations by the *in-situ* continuous measurement and the grab sampling method, respectively.

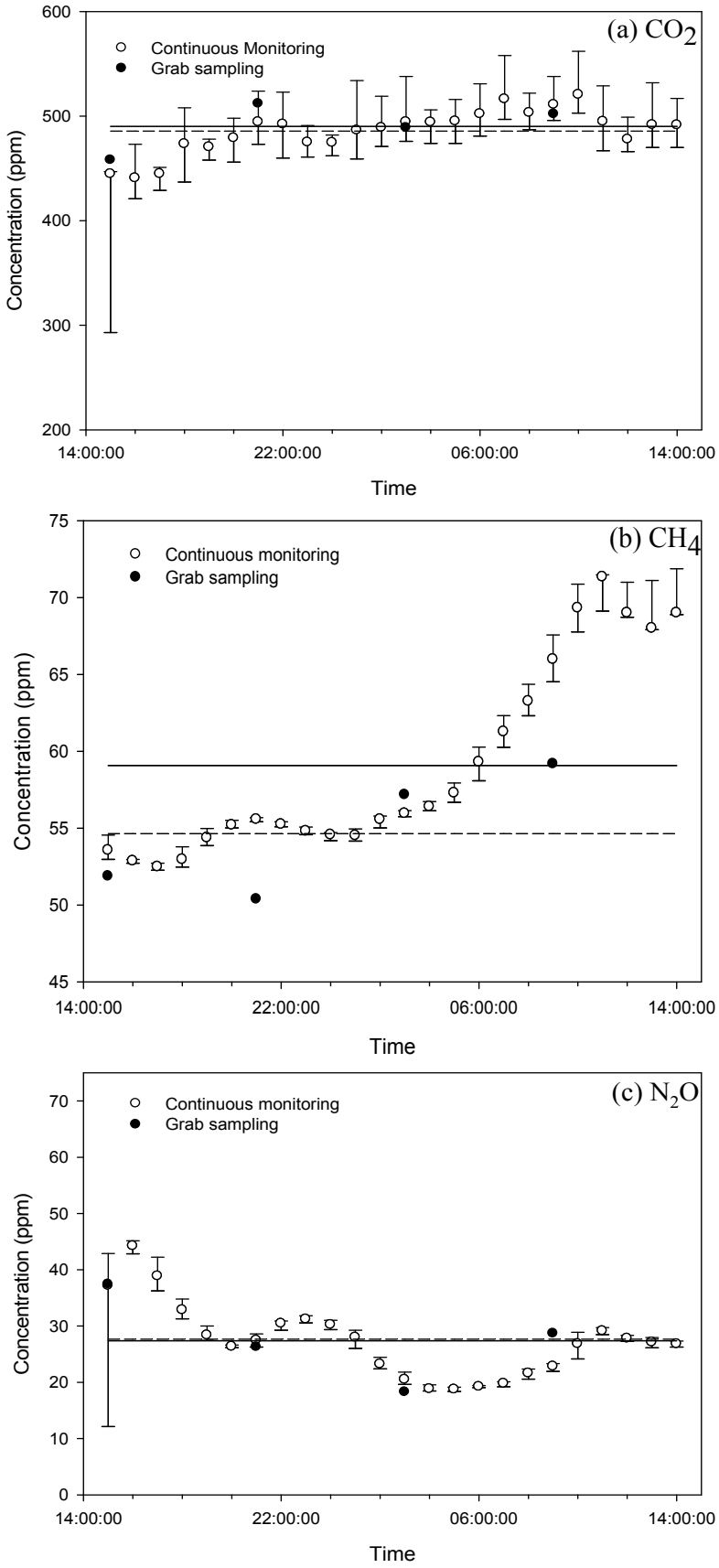

Fig. 4. GHG concentrations observed by the *in-situ* continuous measurement and the grab sampling approach in winter. The solid and dash lines represent the average daily concentrations by the *in-situ* continuous measurement and the grab sampling method, respectively.

Table 2. Relative percent differences (RPDs) between the GHG concentrations measured by the *in-situ* continuous monitoring and the grab sampling methods.

Season	Species	Once a day	Twice a day	Thrice a day	Four times a day
Summer	CO_2	1.5–8.5	0.6–3.5	0.2–2.3	0.1
	CH_4	4.6–53.2	1.7–24.3	2.7–10.8	11.3
	N_2O	27.0–52.0	24.8–39.7	9.5–40.5	16.7
Winter	CO_2	0.6–5.8	0.2–2.9	0.1–3.0	0.9
	CH_4	3.4–15.3	1.7–14.4	6.2–10.7	8.1
	N_2O	3.7–33.3	14.3–18.5	0–1.4	0

The numbers are shown as percentage (%).

(the average was 488 ppm). The results showed that the CO_2 emission in the final sedimentation tank was generally lower than that from the primary sedimentation tank. It is possible that the temporal variations of CO_2 concentrations in the wastewater treatment processes were mostly relevant to the occurrence of excess microbial decomposition of organic matters (Gaur *et al.*, 1971).

Slightly different CO_2 concentration patterns were found in winter (Fig. 6(a)). The average concentration in the WWTP in winter was 5,643 ppm, significantly higher than that in summer. The temporal variations of the CO_2 concentration were less significant due to the lower water temperature in winter. The CO_2 concentration during equalization ranged from 4,000 to 5,800 ppm. The close system characteristic of the primary sedimentation tank reduced the concentration to 40–500 ppm with an average of 485 ppm. Similar to the result in summer, the CO_2 emission was enhanced in the open aeration tank (the concentration ranged between 2,000 and 2,387 and the average was 2,222 ppm). In the sludge thickener and final sedimentation tank, the CO_2 concentrations ranged from 1,650 to 2,100 ppm and from 500 to 700 ppm, and the average concentrations were 1898 and 587 ppm, respectively.

Diurnal Variation of CH_4 Concentration

Figs. 5(b) and 6(b) show the temporal variations of CH_4 concentrations in summer and winter, respectively. The sludge thickener was the process with relatively strong emissions in both seasons. In summer, a moderate diurnal variation was observed in the equalization tank, with an average concentration of 95 ppm. The concentration in the primary sedimentation tank was low, ranging from 15 to 35 ppm with an average of 24 ppm. The lowest CH_4 concentration was found in the aeration tank, attributed to limited anaerobic reactions occurred in this process (Monteith *et al.*, 2005). In the sludge thickener, the concentration ranged from 55 to 95 ppm with an average concentration of 74 ppm. The close system and decomposition of sewage organic matter resulted in an anaerobic state due to hypoxia, accelerating the CH_4 formation. The CH_4 concentration in the final sedimentation tank was moderate by sludge settlement causing anaerobic condition, ranging from 18 to 52 ppm with an average of 35 ppm.

In winter, the sludge thickener and final sedimentation tank still dominated the CH_4 emissions in the WWTP. The concentration during equalization tank was lower possibly attributable to wastewater quality variation. Similar to the CO_2 concentrations in winter, the diurnal variations of CH_4 concentrations were not obvious. The CH_4 concentrations ranged from 10 to 15 ppm, from 20 to 42 ppm, from 37 to 55 ppm, from 56 to 69 ppm in the primary sedimentation tank, aeration tank, sludge thickener, and final sedimentation tank, respectively. The average concentrations in these processes were 11, 30, 45, and 59 ppm, respectively. Overall, the CH_4 concentrations mainly occurred in the equalization tank, sludge thickener, and final sedimentation tank in both seasons. CH_4 is known to be formed thorough anaerobic reactions and affected by water quality including the temperature, DO, and COD concentrations (El-Fadel and Massoud, 2001), explaining the increasing CH_4 concentrations in the sludge thickener and final sedimentation tank. One possible explanation for high CH_4 concentration during equalization was that dissolved CH_4 was formed when wastewater was distributed from the source to WWTP in pipelines with limited oxygen concentrations and was emitted after the water initially flowed into the WWTP (El-Fadel and Massoud, 2001).

Carbon mass balance in the WWTP was also investigated. The total organic carbon (TOC) concentrations in the source and treated water of the WWTP were 96 and 11 mg L^{-1} in summer, respectively. Given the negligible pH difference between the source and treated water, it is expected that the inorganic carbon removal though the treatment processes was limited. The sum of the CO_2 and CH_4 emissions was divided by the TOC removal in the WWTP to estimate the fraction of carbon loss due to CO_2 and CH_4 emission (15–18%). The rest fraction of carbon was considered to be assimilated into sludge.

Diurnal Variation of N_2O Concentration

Figs. 5(c) and 6(c) illustrate the temporal variations of N_2O concentrations in different treatment processes in summer and winter, respectively. Comparing with CO_2 and CH_4, N_2O was the species with the most significant diurnal variations. In summer, the N_2O concentrations in the processes were mostly less than 12 ppm. However, given its high global warming potential (GWP) (IPCC, 2014b), these appreciable levels of N_2O were still important contributors for GHG emission from the WWTP. In winter, the N_2O concentrations were relatively higher. Obvious diurnal concentration variations were noticed, particularly in the equalization and aeration tanks. With the complex formation as an intermediate of either nitrification or denitrification (Kampschreur *et al.*, 2008; Kampschreur *et*

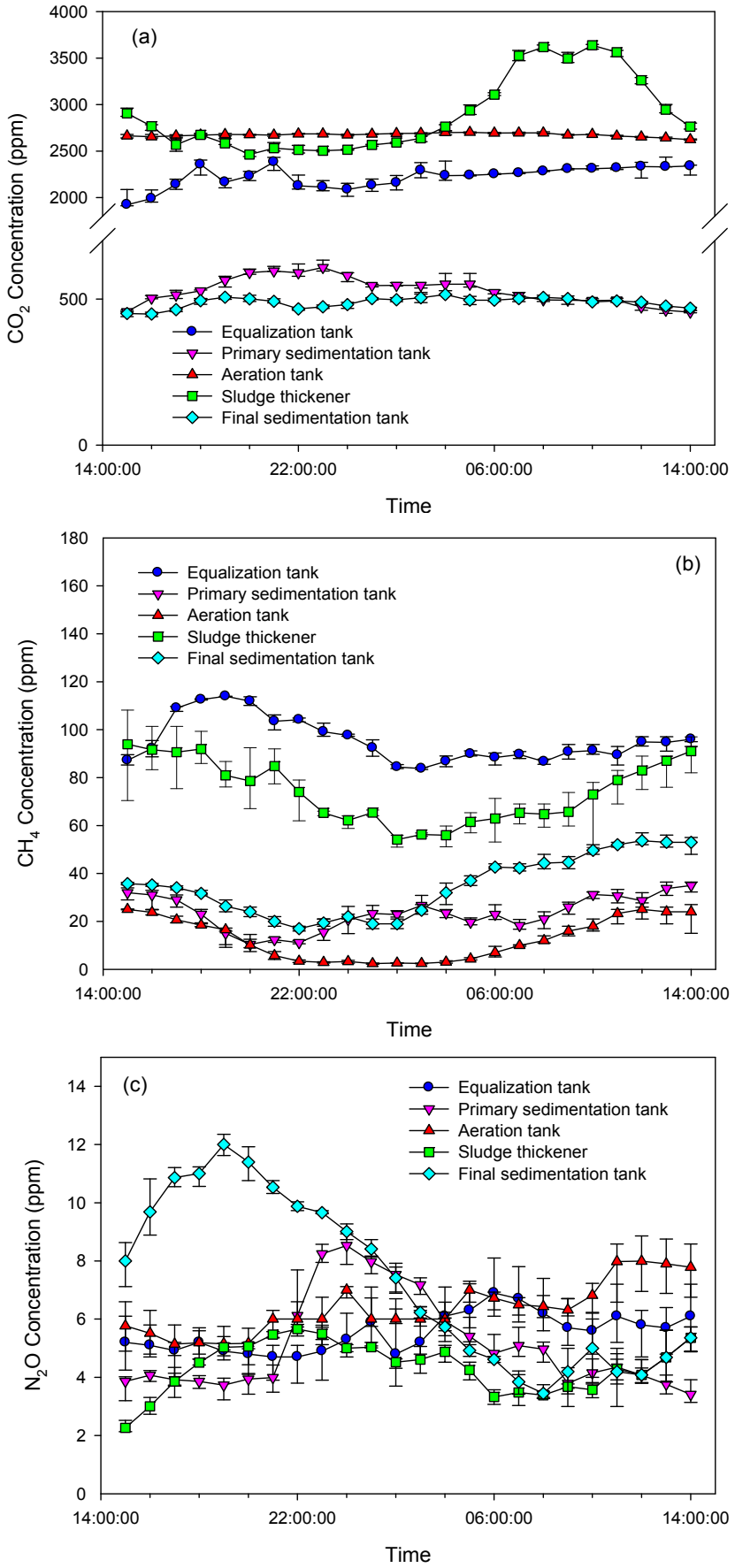

Fig. 5. Temporal variations of the GHG concentrations observed in different treatment units in summer.

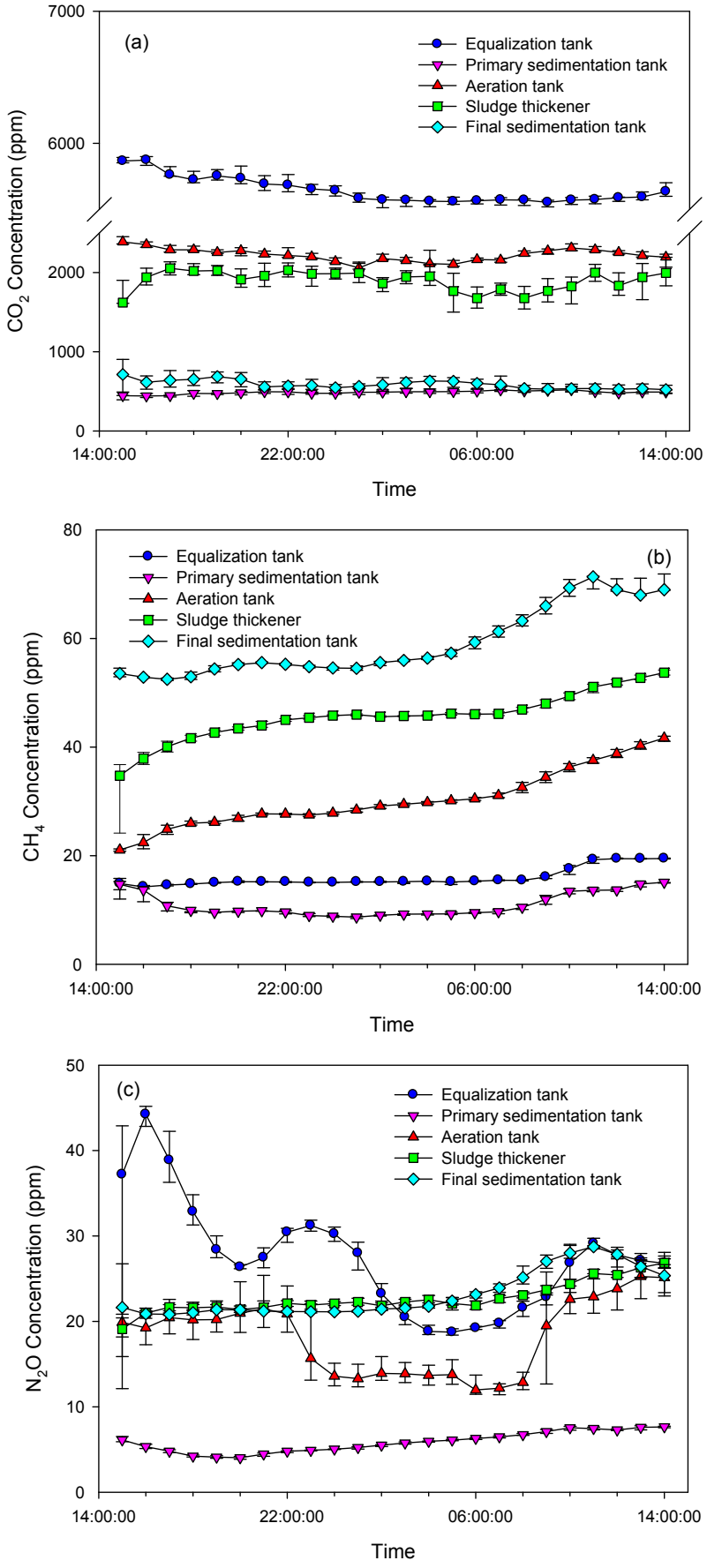

Fig. 6. Temporal variations of the GHG concentrations observed in different treatment technologies in winter.

al., 2009), the *in-situ* continuous monitoring seemed to be more important and necessary to investigate the formation and emission of N_2O in wastewater treatment.

In addition to biological reactions, chemical pathways such as reactions between nitrite and hydroxylamine producing NO and N_2O as well as nitrite reduction lead to N_2O formation in WWTPs (Van Cleemput, 1998). Although aerobic treatment is the main technology in the WWTP investigated in this study, anaerobic conditions could still occur in some locations if the oxygen-transfer was limited or inhibited possibly due to insufficiently mixing or thick microbial flocs. In addition, similar to CH_4 observed during equalization, N_2O could be formed when the wastewater was transported to the WWTP in pipelines with limited oxygen concentrations and was emitted after the wastewater initially flowed into the WWTP. These mechanisms might be relevant to N_2O formation with limited presence of microorganisms during equalization. However, as the biological and chemical N_2O formations are influenced by dissolved oxygen (DO), nitrogen concentration, and COD to nitrogen ratio (Kampschreur *et al.*, 2008; Tallec *et al.*, 2008; Foley *et al.*, 2010), it is difficult to distinguish the N_2O formation between chemical and biological processes in WWTPs (Kampschreur, 2009).

Emission Flux and Emission Factor Estimations between Two Approaches

Tables 3 and 4 summarize the estimated GHG emission fluxes and factors by using the concentrations from the *in-situ* continuous measurement and grab sampling method, respectively. While the sampling frequency of the grab sampling method was four times a day, the differences between the emission fluxes and factors by two methods were negligible. The equalization tank and aeration tank represent two dominant technologies that possessed high CO_2 emission fluxes (up to 56,948 and 11,749 g CO_2 day^{-1}) and emission factors (up to 1.64×10^{-1} and 1.75×10^{-1} kg CO_2/kg COD), respectively. The equalization and final sedimentation tanks were the main sources of CH_4 emissions (the emission fluxes were up to 193 and 171 g CH_4 day^{-1}, as the emission factors were up to 7.97×10^{-4} and 2.38×10^{-3} kg CH_4/kg COD, respectively). The N_2O emission fluxes in the equalization and final sedimentation tanks were up to 193 and 180 g N_2O day^{-1}, as the emission factors were up to 5.32×10^{-4} and 2.47×10^{-3} kg N_2O/kg COD, respectively.

Although the literature reviewed in this study had reported no uncertainty analysis for GHG emission factor, this study described the GHG emission factors with uncertainty analysis. All emission factors have showed 50% and 95% confidence intervals. Input data (CH_4 and N_2O) are assumed to have log-normal probability distributions, represented by median values and 95% confidence interval uncertainties. The total uncertainty levels for CH_4 and N_2O emission factors, expressed as 95% confidence intervals, were 6.3 and 5.3%, respectively. Uncertainties in the trend were 6.9 and 4.9%, respectively. The confidence interval width was expected to be narrow when estimating the sample size. Enough sample size is required to maintain the confidence interval narrow with high accuracy. This article provides

Table 3. Emission fluxes of three GHGs estimated by using the *in-situ* continuous monitoring and grab sampling methods.

Emission flux		*In-situ* continuous measurement				Gab sampling method				
			CH_4		N_2O		CH_4		N_2O	
Treatment units	Season	CO_2 (g day^{-1})	CH_4 (g day^{-1})	CO_2e* (g day^{-1})	N_2O (g day^{-1})	CO_2e* (g day^{-1})	CO_2 (g day^{-1})	CH_4 (g day^{-1})	CO_2e* (g day^{-1})	N_2O (g day^{-1})	CO_2e* (g day^{-1})
Equalization	Summer	38867	189	4728	110	32631	38843	193	4815	112	33495
Primary sedimentation		1212	23	568	16	4857	1230	25	620	18	5245
Aeration		3625	25	635	32	9596	3544	27	673	32	9506
Sludge thickener		450	4	105	1	179	423	6	143	1	387
Final sedimentation		3873	101	2525	64	18923	3924	98	2458	62	18357
Total		48027	342	8561	223	66186	47964	349	8709	225	66990
Equalization	Winter	39354	40	1010	191	56948	39322	37	917	193	57508
Primary sedimentation		1365	11	280	16	4857	1378	11	284	16	4750
Aeration		11528	58	1455	96	28519	11749	51	1272	94	27973
Sludge thickener		292	3	65	4	1073	288	2	60	3	1004
Final sedimentation		4733	171	4263	180	53610	4855	158	3943	173	51629
Total		57272	283	7073	487	145007	57592	259	6476	479	142864

* CO2e denote the CO_2 equivalent that describes the amount of CO_2 that would have the same global warming potential (GWP) when measured over 100 years.

Table 4. Emission factors of three GHGs estimated by using the *in-situ* continuous monitoring and the grab sampling method.

Emission factors		In-situ Continuous monitoring			Gab sampling and analysis		
Treatment units	Season	CO_2 (kg CO_2/kg COD)	CH_4 (kg CH_4/kg COD)	N_2O (kg N_2O/kg N)	CO_2 (kg CO_2/kg COD)	CH_4 (kg CH_4/kg COD)	N_2O (kg N_2O/kg N)
Equalization tank	Summer	1.64E-01	7.97E-04	4.61E-04	1.58E-01	7.82E-04	4.51E-04
Primary sedimentation tank		1.04E-02	1.96E-04	1.40E-04	1.12E-02	1.86E-04	1.48E-04
Aeration tank		1.72E-01	1.20E-03	1.53E-03	1.75E-01	1.24E-03	1.51E-03
Sludge thickener		1.61E-04	1.50E-06	2.22E-07	1.63E-04	1.53E-06	2.31E-07
Final sedimentation tank		6.38E-02	1.66E-03	1.05E-03	6.42E-02	1.68E-03	1.12E-03
Total		4.10E-01	3.85E-03	3.18E-03	4.09E-01	3.89E-03	3.23E-03
Equalization tank	Winter	1.09E-01	1.12E-04	5.32E-04	1.13E-01	1.19E-04	5.37E-04
Primary sedimentation tank		9.56E-03	7.86E-05	1.14E-04	9.42E-03	7.92E-05	1.16E-04
Aeration tank		7.28E-02	3.67E-04	6.04E-04	7.33E-02	3.62E-04	6.12E-04
Sludge thickener		6.95E-05	6.26E-07	8.52E-07	6.96E-05	6.28E-07	8.43E-07
Final sedimentation tank		6.49E-02	2.34E-03	2.47E-03	6.45E-02	2.38E-03	2.42E-03
Total		2.56E-01	2.90E-03	3.72E-03	2.60E-01	2.94E-03	3.69E-03

the sample size calculation for precise confidence interval of standardized effect in one-way ANOVA (Fig. 7). Finally, using the confidence interval of effect sizes to calculate an appropriate sample size with different combination of population parameters under required confidence level. At 95% confidence interval, the CH_4 and N_2O levels were significant ($p = 0.03$). The emission factors were 3.4×10^{-3} kg CH_4/kg COD and 3.5×10^{-3} kg N_2O/kg N in WWTPs. The emission factor levels of GHGs emitted from the WWTPs were statistically significant at $p < 0.05$ ($p = 0.03$). This information strengthens the reliability of comparison among the GHG emissions in this and those early studies.

In addition to their high emissions during sedimentation due to anaerobic reactions, the emission fluxes and factors of CH_4 and N_2O were relatively higher in the early stages of wastewater treatment, notably during equalization. High organic matter concentrations in wastewater and the treatment technology open to atmosphere were two possible explanations. The emission factors estimated in this study were compared with those in the literatures and Intergovernmental Panel on Climate Change (IPCC) (see Table 5). It is worth noting that the methane correction factors (MCFs) by the IPCC have been considered in Table 5 (e.g., the MCF values are 0.25 and 0.2–0.8 kg CH_4/kg COD for a concentrated aerobic treatment unit and an anaerobic sludge digester, respectively). The comparison showed that the emission factors of GHGs obtained from this study were less than those in Netherlands, Australia, and IPCC, but higher than those in Japan.

CONCLUSIONS

Two different approaches including the *in-situ* continuous monitoring and typical grab sampling approaches were applied to analyze the concentrations of CO_2, CH_4, and N_2O in different treatment processes of a petrochemical WWTP. One important finding was that both analytical approaches could provide reliable information regarding the GHG emissions in the wastewater treatment units, although a number of considerations such as wastewater quality and treatment technologies could potentially affect the GHG emissions in the WWTP. However, the assumption for this conclusion was that a sufficient sampling frequency (e.g., once every 6 hours) was required for the typical grab sampling method in order to effectively resolve the diurnal variations of GHG levels emitted from the wastewater treatment units. With this assumption, consistent emission factors of three GHGs between two approaches were obtained, as the emission factors of CH_4 and N_2O were higher in the equalization tank (1.12×10^{-4} to 7.97×10^{-4} kg CH_4/kg COD and 4.51×10^{-4} to 5.37×10^{-4} kg N_2O/kg N) and the final sedimentation tank (1.66×10^{-3} to 2.38×10^{-3} kg CH_4/kg COD and 1.05×10^{-3} to 2.47×10^{-3} kg N_2O/kg N), respectively. Only with proper and reliable sampling and analytical methods to determine the GHG emissions from wastewater treatment processes, it becomes possible to correctly identify the characteristics of GHG emissions and to develop strategies to curtail the GHG emissions from such an important source in response to regulatory measures

and international treaties. Compared with previous literatures and IPCC, the GHG emission factors of CH_4 and N_2O obtained from this study were lower than those measured in Netherlands, Australia, and IPCC, but similar to those measured in Japan. Furthermore, the emission factor levels of GHGs emitted from the WWTPs were statistically significant at $p < 0.05$ ($p = 0.03$), which strengthens the reliability of comparison among the GHG emissions in this and those early studies.

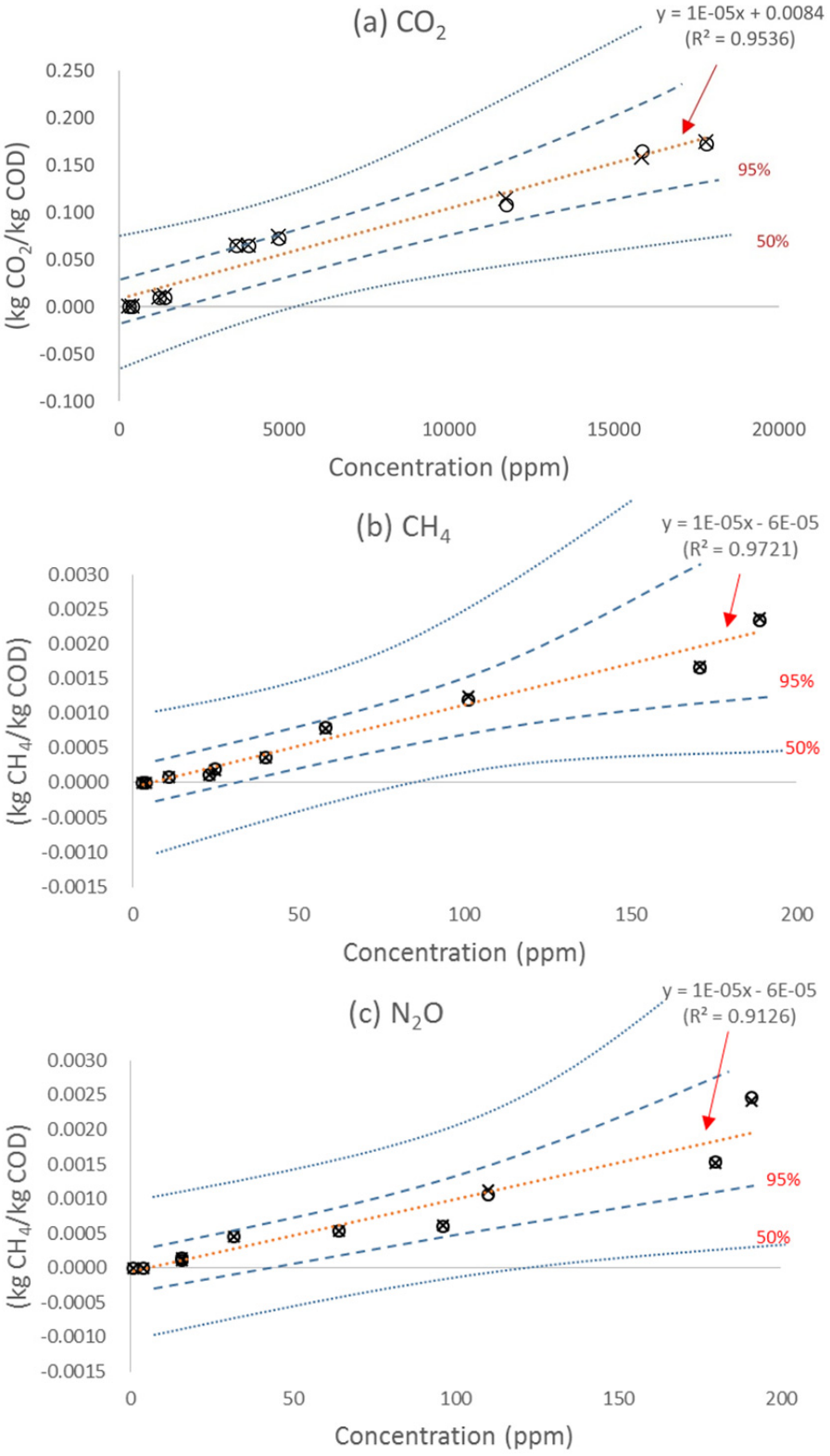

Fig. 7. Uncertainty analysis of GHG emission factors given the concentrations emitted in the WWTP.

Table 5. GHG emission factors estimated in this and early studies including the IPCC.

Countries	GHGs	Emission factors	Units	Literatures/Sources
United Kingdom (UK)[a]	CH_4	0.075–0.2[f]	kg CH_4/kg COD	IPCC, 2006a
California, USA[b]	CH_4	0.25–0.6[f]	kg CH_4/kg COD	IPCC, 2006a
Japan[c]	CH_4	4.9E-03[f]	kg CH_4/kg COD	Ministry of the Environment, 2012
	N_2O	4.3E-03	kg N_2O/kg N	Ministry of the Environment, 2012
Netherlands[d]	CH_4	5.6E-02[f]	kg CH_4/kg COD	RIVM, 2013
Australia[e]	CH_4	0.075–0.2[f]	kg CH_4/kg COD	IPCC, 1997; Australian Government, 2013
Taiwan	CH_4	3.4E-03[f]	kg CH_4/kg COD	This study
	N_2O	3.5E-03	kg N_2O/kg N	

[a] IPCC, 2006a IPCC Guidelines for National Greenhouse Gas Inventories, 2006.
[b] IPCC, "Revised 1996 IPCC Guidelines for National Greenhouse Gas Emission Inventories," Three volumes: Reference manual, Reporting Guidelines and Workbook. IPCC/OECD/IEA, 1997.
[c] Ministry of the Environment, "National Greenhouse Gas Inventory Report of Japan", Japan, 2012.
[d] IVM, "Greenhouse gas emissions in the Netherlands 1990–2011," National Inventory Report, 2013.
[e] Australian Government, "Australian National Greenhouse Accounts - National Inventory Report the Australian Government Submission to the United Nations Framework Convention on Climate Change," 2013.
[f] Emission factors of GHGs herein were determined by multiplying MCF of 0.25 or 0.2–0.8.

ACKNOWLEDGEMENTS

The authors would like to express their sincere appreciation to Ministry of Science and Technology (MOST) and Environmental Protection Administration (EPA) of R.O.C (Taiwan) under the auspicious of NSC102-EPA-F-009-002 for their financial supports to accomplish this research. We also thank the staff of the Ren-Da Wastewater Treatment Plant for their kind cooperation and assistance for the sampling and analytical works.

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Received for review, August 19, 2017
Revised, September 14, 2017
Accepted, September 15, 2017