Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.940389

54 (2015) 1070–1079
April

Taylor & Francis
Taylor & Francis Group

Characteristics of direct CO_2 emissions in four full-scale wastewater treatment plants

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Received 25 December 2013; Accepted 23 June 2014

ABSTRACT

Direct CO_2 emissions from wastewater treatment plants (WWTPs), which are commonly excluded from the greenhouse gas emission inventory as proposed by the International Panel on Climate Change, should be partly taken into consideration, because some of the fossil organic carbons (i.e. detergents, cosmetics, and pharmaceuticals) are also biodegradable, leading to the emission of CO_2 during wastewater treatment. For the purpose of understanding the characteristics of direct CO_2 emissions during biological nutrient removal from urban WWTPs, we investigated four different processes. Full-scale tests were carried out continuously during a two-year period, consisting of the anoxic/anaerobic/oxic (A^2O) process, the anoxic/oxic (AO) process, the oxidation ditch, and the sequencing batch reactor (SBR) processes. Our experimental results show large differences in CO_2 fluxes in the various treatment tanks (or periods), e.g. the aerated units/periods in four different WWTPs all contributed more than 96.0% of the entire direct CO_2 emission caused by aerobic respiration and aeration stripping. On the other hand, CO_2 emissions from non-aerated areas were quite low giving rise to small amounts of CO_2 emissions during anaerobic metabolism. The direct emission of CO_2 when treating per m^3 wastewater (g CO_2/m^3 wastewater) corresponded with the following descending order among the four processes: SBR (347.34) > oxidation ditch (343.78) > A^2O (175.68) > AO (173.37). However, this order changed into SBR (0.97) > oxidation ditch (0.76) > AO (0.68) > A^2O (0.58) when treating per kilogram COD (kg CO_2/kg COD).

Keyword: Direct CO_2 emissions; Characteristics; GHG; Wastewater treatment plants

1. Introduction

Greenhouse gases (GHG), such as CO_2, CH_4, and N_2O, are causing adverse climate change [1]. CO_2, one of the most important GHG, responsible for most of the effect of global warming, accounting for about 45–61% of global GHG emissions [2]. CO_2, with its lifespan of 50 to 200 years in the atmosphere, has stable properties and cannot be easily decomposed [3].

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Presented at the 6th International Conference on the "Challenges in Environmental Science and Engineering" (CESE-2013), 29 October–2 November 2013, Daegu, Korea

Hence, it is important to reduce CO_2 emissions from all probable sources.

Human activities such as the production and consumption of fossil fuels, as well as agricultural and industrial activities, have caused an increase in CO_2 concentrations in the atmosphere. A CO_2 concentration of 314.42 ppm in the atmosphere was monitored in 1958, which has increased gradually and consistently to an average of 396.72 ppm in 2013 [4,5].

Studies have shown that wastewater treatment plants (WWTPs) are among the important sources of CO_2 emission, originating from energy consumption (mainly consuming electricity), the usage of chemicals, and microbial activities [6]. Of these, the first two are classified as indirect CO_2 emissions [7,8] while the last one is defined as direct CO_2 emission.

Microbial activities have been confirmed as causing large amounts of CO_2 emission in WWTPs [9,10]. The degradation of organic pollutants in urban sewage plays an important role in CO_2 emissions from WWTPs, mostly via biological processes [11–15]. Another major process is biomass respiration, which makes for lesser contributions to CO_2 emission [15]. In general, 90–95% of organic matter in sewage is removed during secondary treatments [16], of which a small part of biochemical oxygen demand is transformed into biogas and the other part turned into biomass which is then further converted into CO_2 via endogenous respiration [11]. In addition, anaerobic respiration can also emit CO_2 during wastewater treatment [17,18].

According to the protocols published by the International Panel on Climate Change, direct CO_2 emission from WWTPs should be excluded from GHG emission inventories, given that the impact factor of CO_2 production from biogenic sources is 0 kg CO_2 eq/kg CO_2, while CO_2 emission from fossil origins has an impact factor of 1 kg CO_2 eq/kg CO_2 [19]. However, in addition to organic matter resulting from non-fossil carbon sources consumed by human activities, fossil carbon products such as detergents, cosmetics, and pharmaceuticals in urban sewage can be mineralized and then emitted as CO_2 during biological treatment. These CO_2 emissions should be included into the total amount of GHG emission from WWTPs. For example, Griffith et al. [20] and Law et al. [21] pointed out that part of the carbon presented in wastewater could be of fossil origin and they are of the opinion that estimates of direct CO_2 are clearly of importance. According to Griffith et al. [20], 25% of wastewater dissolved organic carbon originates from fossil carbon, as estimated from mass balance calculations in a 12-WWTP-survey. Law et al. [21] carried out a further study in four municipal WWTPs, suggested that 39–65% of organic fossil carbon was incorporated into the activated sludge during secondary treatment and subsequently transformed to CO_2. Direct CO_2 emissions from biodegradable carbon sources during wastewater treatment, still require further study [11].

Therefore, in our investigation we studied the direct CO_2 emission in four different processes, based on the monitoring results of CO_2 emission characteristics per m^3 of treated wastewater, consisting of the variations in CO_2 flux from each treatment unit and the differences in total CO_2 emission. Simultaneously we summarized the tendency of CO_2 fluxes to change in the most critical treatment unit, i.e. the oxic tank in anoxic/anaerobic/oxic (A^2O) and anoxic/oxic (AO) processes, as well as the oxic area in oxidation ditches and the feeding and aeration period in the sequencing batch reactor (SBR) process, in order to arrive at a more precise calculation of CO_2 emission from WWTPs using these four processes. In the end we calculated the amount of direct CO_2 emission from each treatment process and suggest the optimal process for CO_2 reduction.

2. Materials and methods

In four WWTPs, each using a various treatment process (A^2O, AO, oxidation ditch, and SBR), located in Beijing, China, full-scale investigations were carried out during a two year period. From these urban sewage WWTPs, in which domestic wastewater was subjected to primary and secondary treatments with an average influent chemical oxygen demand (COD) of 397 mg/L (227–597 mg/L), 384 mg/L (239–534 mg/L), 488 mg/L (272–893 mg/L), and 414 mg/L (234–886 mg/L) in A^2O, AO, oxidation ditch, and SBR process, respectively. 90% of total COD was removed in these four WWTPs.

2.1. Monitoring points

CO_2 emission from the unit with an apparent gas-liquid interface was monitored in each of the four WWTPs. According to previous reports [22] and the tentative experiments performed by us, a larger flux of CO_2 was commonly found occurring in the aerated units (periods) than in the non-aerated units (periods). Hence, during the monitoring of CO_2 emissions in these four WWTPs, more monitoring points were established in the aerated units (periods) in order to obtain a better understanding of the characteristics of CO_2 emissions. The major treatment units are shown in Table 1.

Table 1
Surface areas, monitoring points/periods, and monitoring frequencies in various processes

WWTP	Unit	Surface area (m^2)	Numbers of monitoring points/periods	Monitoring Frequencies (/time)
A^{2}O	AGT	504	2	2
	PST	25,200	2	2
	AN	3,564	2	2
	ANA	3,564	2	2
	OT	25,011	6	12
	FC	23,562	2	2
AO	AGT	504	2	2
	PST	25,200	2	2
	AN	5,356	2	2
	OT	26,782	6	12
	FC	23,562	2	2
OD	AGT	320	2	2
	BST	762	1	1
	ANA	2,570	2	2
	AN-OD	36,540	3	6
	OX-OD	3,780	4	8
	FC	14,255	2	2
	SDT	57	2	2
	STT	760	2	2
SBR	SGT	28	2	2
	WDT	32	2	2
	FAP	11,872	2	8
	SP	11,872	2	2
	DP	11,872	2	2

Notes: "AGT", Aerated grit tank; "SGT", Swirl grit tank; "BST", Bio-selecting tank; "PST", Primary sedimentation tank; "WDT", Wastewater distribution tank; "ANA", Anaerobic tank (areas); "AN", Anoxic tank (areas); "OT", Oxic tank; "FC", Final clarifier; "OD", Oxidation ditch; "OX", oxic areas; "SDT", Sludge distribution tank; "STT", Sludge thickening tank; "FAP", Feeding and aeration period; "SP", Settling period; "DP", Declining period.

2.1.1. A^{2}O and AO processes

Along with wastewater flows, the major monitoring units in the A^{2}O WWTP were as follows: an aerated grit tank, a primary sedimentation tank, a biological treatment unit (consisting of an anoxic tank, an anaerobic tank, and an oxic tank) and a final clarifier. The 288 m long biological treatment unit was divided into three corridors, of which the first 64 m was equally divided into two parts, i.e. the anoxic tank and the anaerobic tank, while the remaining 224 m functioned as the oxic tank. The main monitoring points and water surface areas in each unit in the A^{2}O process are shown in Fig. 1(a).

As shown in Fig. 1(b), the major monitoring units for CO_2 emission in the AO process are similar to those in the A^{2}O process, although no anaerobic process was included during the biological treatment.

2.1.2. Oxidation ditch process

As seen in Fig. 1(c) the main monitoring units in the oxidation ditch process are as follows: an aerated grit tank, a bio-selecting tank, an anaerobic tank, an oxidation ditch tank (consisting of the anoxic areas and the oxic areas, equipped with 12 brush aerators to provide aeration and move the mixed liquor), a final clarifier, a sludge distribution tank, and a sludge thickening tank.

2.1.3. SBR process

As shown in Fig. 1(d), a swirl grit tank, a sewage distribution tank, and a SBR reactor were selected as the main monitoring units in the SBR process. A four-hour cycle was established for the operation of the SBR, i.e. two hours for the feeding and aeration period (both feeding and aerating were carried out during

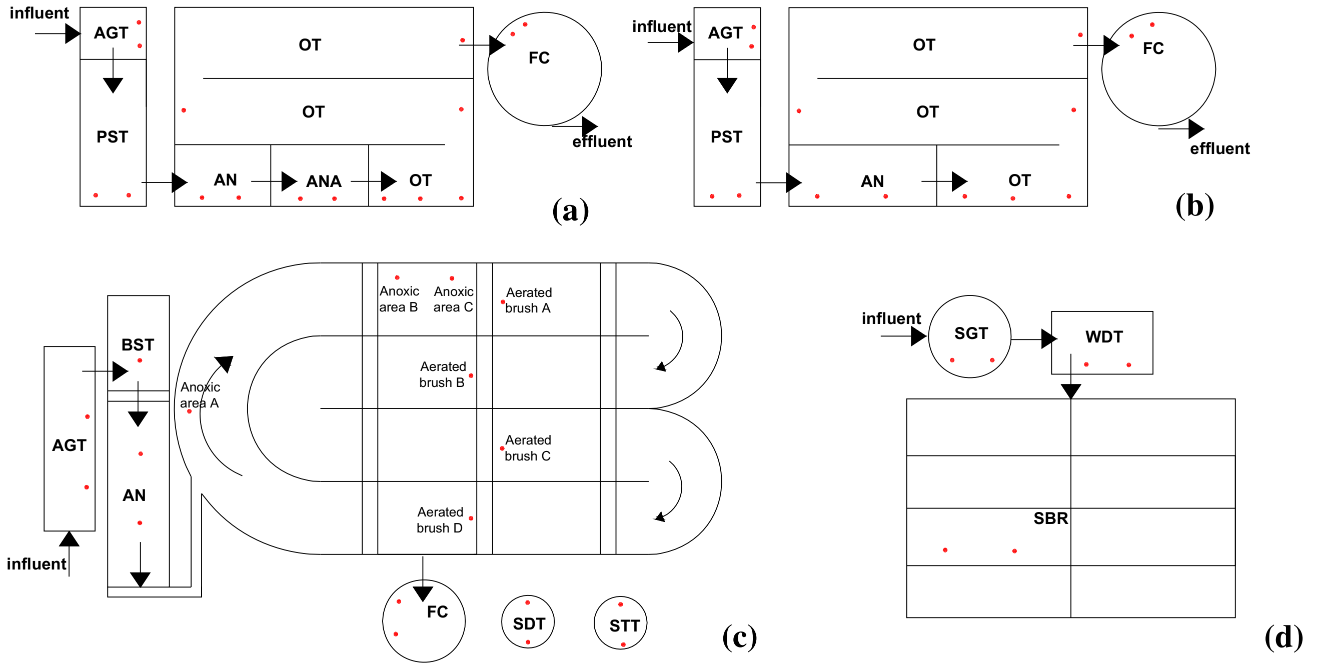

Fig. 1. Monitoring points in each process: (a) A^2O process, (b) AO process, (c) oxidation ditch, and (d) SBR process.

the first hour while only aerating occurred during the second hour), one hour for the settling period and one hour for decanting.

2.2. Monitoring frequencies

Surface areas, monitoring points and monitoring frequencies in each treatment unit of the various processes are shown in Table 1.

2.3. CO_2 monitoring in the biological units of various processes

Escalas et al. [23] pointed out that biodegradation of organic matters in the secondary biological treatment units in the WWTPs are largely responsible for the emission of CO_2. Meanwhile, our tests showed apparent differences in CO_2 flux in various areas/periods in the biological treatment units.

In order to study the characteristics of CO_2 emission from the biological units, two evenly distributed monitoring points were established at the water surface area in the anaerobic tank in the A^2O process and in the anoxic tank in both A^2O and AO processes. It appeared that more organic matter was degraded by aerobic respiration at the beginning of the oxic tank, leading to a higher flux in CO_2 under the same rate of aeration, than in other areas of the oxic tank and therefore, we established more monitoring points at the beginning, as shown in Fig. 1(a) and (b).

Similar to the set of monitoring points in the A^2O and AO processes, three points were set in the anoxic area and four in the oxic area in the oxidation ditch tank, as shown in Fig. 1(c). 2-year experiments showed that the dissolved oxygen concentration dropped below 1.5 mg/L, 5 m behind the aeration brush. So the first 5 m area behind the aeration brush was considered as the oxic area in the oxidation ditch tank.

In the SBR process, monitoring frequencies were based on the duration of each treatment. During the feeding and aeration period, four monitoring frequencies were established at each monitoring point at the same spacing interval.

2.4. CO_2 monitoring and analysis

2.4.1. Non-aerated areas

A static chamber gas collection technique [24] based on a closed chamber technique [22]. A sealed steel-made static chamber technique was used to measure CO_2 flux from non-aerated water surfaces. The static chamber with a thermo-detector and a pressure meter inside was fastened by the floating body and fixed well to minimize chamber movement due to surface turbulence, which could maintain a stable volume and gaseous pressure. Gas samples (in triple, RSD_g

(relative standard deviation) < 5.0%) were collected by 10-min intervals. In these above procedures, the pressure in the chamber was carefully supervised. The major advantage of this method was that it was able to maintain gas pressure stability in the chamber during the monitoring process.

2.4.2. Aerated areas

Polyethylene plastic bags were used to collect diffused gases from the aerated areas [24]. The pressure meter which is installed in the system can suddenly indicate the increase when the gases fulfilled the plastic bag (0.09 m^3). At the same time of sampling (in triple, $RSD_g < 5.0\%$), the pressure meter was monitored carefully. Gaseous samples of CO_2 were properly collected when the gas pressure increased sharply.

An Agilent 7890A gas chromatograph (GC, detection limits: 1.0 ppm) equipped with a flame ionization detector (GC-FID) was used for the analysis of CO_2 concentrations in the gas samples (RSD_{GC} (relative standard deviation of GC) < 2.6%, 5 ml gas sample for each test). COD concentrations (in triple, $RSD_{COD} \leq 5.0\%$) were obtained with a colorimetric method, using a Lian-Hua 5B-3B (China) in the range of 20–10,000 mg COD/L (RSD_C of Lian-Hua 5B-3B < 3.0%).

The gas flux, E (g $CO_2/m^2\,d^{-1}$), from the liquid surface in the static chamber was calculated with (Eq. (1)).

$$E = 1{,}440(\mathrm{d}c/\mathrm{d}t)\rho V/A_e \tag{1}$$

where dc/dt is the linear increase of the CO_2 concentration, ρ (g/m^3) is the CO_2 density at the temperature recorded, V (m^3) is the volume of the chamber above the liquid surface, and A_e (m^2) is the enclosed surface area. (Eq. (1)) assumes that CO_2 emissions are emitted at a steady rate at each monitoring period, without severe changes in gas pressure within the static chamber.

The gas flux, E (g $CO_2/m^2\,d^{-1}$), from the liquid surface in the plastic bags, was calculated with (Eq. (2)).

$$E = Q(C - C_a)p/A \tag{2}$$

where Q (m^3/d) is the air flow rate, C (ppm) is the concentration of gaseous CO_2 collected, C_a (ppm) is the concentration of ambient air, p (g/m^3) is the CO_2 density at the temperature recorded, and A (m^2) the gas monitoring area. As was the case in (Eq. (1)), we assumed that CO_2 emissions would be emitted at a steady rate in each monitoring period.

The CO_2 emission, E_v (g/m^3 wastewater) was calculated with (Eq. (3)).

$$E_v = EA_t/L \tag{3}$$

where E_v is the CO_2 emission (g/m^3 wastewater), E the CO_2 flux in the corresponding tank, A_t the total area in a corresponding tank and L (m^3) the capacity of wastewater treatment. Total CO_2 emission in each process was obtained by adding the CO_2 emission from each treatment unit.

3. Results and discussion

In the following sections, the CO_2 emission characteristics, consisting of CO_2 flux (g $CO_2/m^2\,d$) and the direct emission of CO_2 (g CO_2/m^3 wastewater) from each treatment unit in the four processes (A^2O, AO, oxidation ditch, and SBR), are illustrated. We discuss the characteristics of CO_2 emission from each of the biological treatment units. In the end, we compare the direct emission of CO_2 among the various processes and present the optimal process for the reduction of direct CO_2 emission.

3.1. CO_2 flux from each treatment unit in four processes

3.1.1. CO_2 flux in the A^2O process

CO_2 flux from each treatment unit in the A^2O process is shown in Fig. 2(a), ranked in descending order as follows: aerated grit tank, oxic tank, anaerobic tank, anoxic tank, final clarifier, and primary sedimentation tank. In the aerated grit tank, the average CO_2 flux was 3,728.44 g $CO_2/m^2\,d$, ranging from a minimum of 969.16 g $CO_2/m^2\,d$ to a maximum of 7,650.7 g $CO_2/m^2\,d$. This range is attributed to the various concentrations of dissolved excess CO_2 in the influent pipe, which can lead to CO_2 emission by aeration. Thus, the CO_2 flux from the aerated grit tank was at a high level and responsible for the large gap, given the vastly different conditions of the influent. Because of the limited biological activities and the stable water surface, the CO_2 flux from the primary sedimentation tank was much smaller than that from the aerated grit tank. It is seen that the CO_2 flux, at 5.04 g $CO_2/m^2\,d$ and with a range of 3.35 g CO_2/m^2–9.65 g $CO_2/m^2\,d$, was quite small in the primary sedimentation tank. In the anoxic tank, the CO_2 flux increased to 60.51 g $CO_2/m^2\,d$ and in the anaerobic tank to 67.90 g $CO_2/m^2\,d$, owing to biological denitrification, anaerobic respiration, and small fluctuations in the CO_2 flux found in these tanks in the A^2O process. The CO_2 flux from the oxic tank was very high with an average 2,904.77 g $CO_2/m^2\,d$, varying from 1,829.97 to 4,489.12 g $CO_2/m^2\,d$, due to aerobic respiration of microorganisms and the aeration stripping process. (CO_2

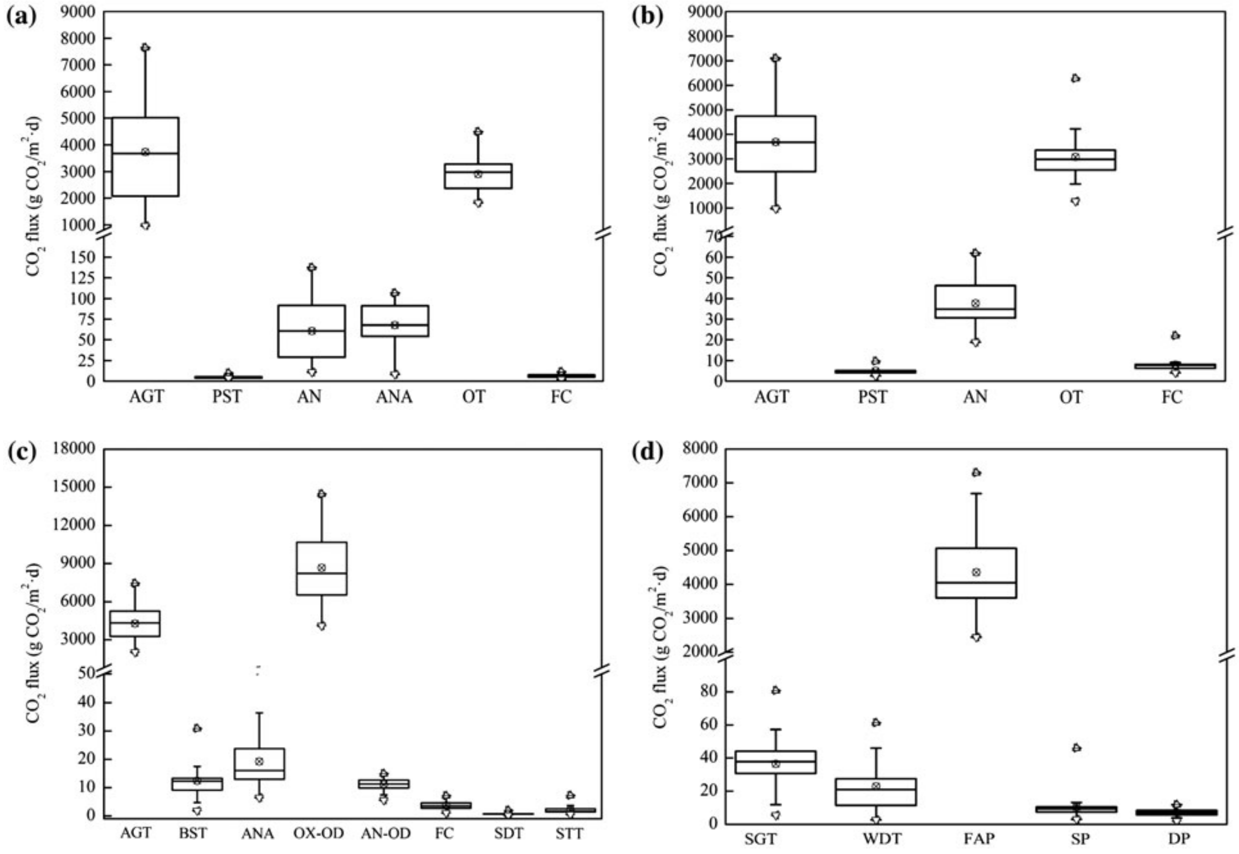

Fig. 2. CO_2 flux from different units over four processes: (a) A^2O process, (b) AO process, (c) oxidation ditch, and (d) SBR process.

produced in both aerobic and anoxic environment due to the decomposition of substrates, [10] and may exists as dissolved CO_2 in the wastewater in non-aerated reactors, those dissolved CO_2 may finally emit to the atmosphere by the effect of aeration in the aerated reactor. Analogously, this phenomenon can be found in other treatment processes.) The CO_2 flux decreased and gradually stabilized around 6.58 g CO_2/m^2 d in the final clarifier due to the stable state of wastewater and little microbial respiration.

3.1.2. CO_2 flux in the AO process

The CO_2 flux in each unit in the AO process is illustrated in Fig. 2(b), showing the same tendency of change as in the A^2O process. The two largest fluxes of CO_2 emission occurred in the aerated grit tank, ranging from a minimum of 910.47 g CO_2/m^2 d to a maximum of 7,050.18 g CO_2/m^2 d with an average of flux of 3,529.09 g CO_2/m^2 d and in the oxic tank with a range from 1,334.15 to 6,405.07 g CO_2/m^2 d), and an average CO_2 flux of 2,977.74 g CO_2/m^2 d. The two minimum CO_2 fluxes were found in the final clarifier (8.64 g CO_2/m^2 d) and the primary sedimentation tank (4.73 g CO_2/m^2 d). Given that there was no anaerobic tank in the AO process, relatively poor anaerobic respiration and denitrification occurred in the anoxic tank, leading to a lower CO_2 flux in the anoxic tank, with an average 39.93 g CO_2/m^2 d, than that in the A^2O process.

3.1.3. CO_2 flux in the oxidation ditch process

As shown in Fig. 2(c), the CO_2 flux in the aerated grit tank with an average of 4,272.00 g CO_2/m^2 d, ranged from a minimum of 2,033.16 g CO_2/m^2 d to a maximum of 7,433.38 g CO_2/m^2 d) in the oxidation ditch, similar to that in the A^2O and AO processes. The CO_2 flux was low (12.36 g CO_2/m^2 d) in the bio-selecting tank because of a low level of respiration and biodegradation (only to create a unique system of micro-organisms). The average CO_2 flux was 17.20 g CO_2/m^2 d in the anaerobic tank and 10.92 g CO_2/m^2 d in the non-aeration areas. The CO_2 flux in the aeration areas of the oxidation ditch tank was very high, with an average of 8,668.28 g CO_2/m^2 d, ranging from 4,116.43 to 14,466.92 g CO_2/m^2 d) due to the strong effect of stripping promoted by the aeration brushes. The average CO_2 flux in the sludge distribution tank was 19.17 and 11.07 g CO_2/m^2 d in the sludge thickening tank, showing the lowest level.

3.1.4. CO_2 *flux in the SBR process*

Fig. 2(d) shows the CO_2 flux in different units/periods in the SBR process. It was only 34.32 g CO_2/m^2 d in the swirl grit tank because this treatment unit used a swirling mixer instead of an aeration device, hence only a small amount of dissolved CO2 was emitted. The highest flux of CO_2 was found in the feeding and aeration period (4,117 g CO_2/m^2 d, ranging from 2,454.77 to 7,539.50 g CO_2/m^2 d), due to the aerobic respiration by microbes and the aeration stripping process. No apparent differences in the CO_2 flux (7.95 g CO_2/m^2 d for the settling period and 7.56 g CO_2/m^2 d for the decanting period) were found in the settling and decanting periods, because smaller amounts of CO_2 were generated and emitted in the non-aeration period for the anaerobic respiration and because of the stable flow of wastewater.

3.2. CO_2 *flux from biological treatment units in various processes*

Fig. 3(a) and (b) showed CO_2 fluxes in the biological tanks in the A^2O and AO processes. High fluxes of CO_2 were found at the first three monitoring sites in the oxic tank, while the CO_2 flux gradually declined at later monitoring sites. The reasons for the decrease in the CO_2 flux at the later monitoring sites (it had fallen by one-thirds to two-thirds, where the reduction of the CO_2 flux in the AO process was more marked than that in A^2O process) can be found in the following two issues: in the first instance, more CO_2 might be generated during higher load conditions; given the decrease in available organic matter, the microbial phase was likely to transform to the endogenous respiration period, leading to a low contribution of CO_2 emission. Secondly, CO_2 accumulated in the former treatment units was stripped out from the air at the front of the oxic tank by aeration. However, the CO_2 flux did not always decrease in the oxic tank. The highest of CO_2 flux (5–10% larger than at the first sampling point) was found up to tens of meters away from the entrance of the oxic tank, suggesting that biological adaptation was needed to achieve a maximum CO_2 flux.

In the oxidation ditch tank (Fig. 3(c)), the CO_2 flux was stronger than that in the A^2O and AO processes owing to the large effect of air stripping provided by the aeration brushes. The average CO_2 flux was high at each monitoring point, with a decrease from the first monitoring point to the second. A continuous decline can be seen, but the decrease was not very large.

Fig. 3(d) shows the changes in CO_2 flux in the SBR tank. As long as residual organic matter was available during the first hour during the feeding and aeration period, increasing amounts of CO_2 were generated and emitted during biological organic degradation. Although no influent was injected into the SBR tank after one hour in the SBR cycle, the CO_2 flux still remained stable around 4,000 g CO_2/m^2 d. After feeding and aeration period (2 h into the SBR cycle), the CO_2 flux showed a sharp decline from the very beginning and retained this low level, owing to non-aerobic respiration.

3.3. CO_2 *emission from each treatment unit in various processes*

CO_2 emissions (g CO_2/m^3 wastewater) from each treatment unit in the four processes are shown in Table 2. Average CO_2 emissions (Ave) with their maxima (Max) and minima (Min) are presented in order to illustrate the level of these emissions.

It can be seen that the total CO_2 emission from the A^2O process ranged from 105.07 g CO_2/m^3 wastewater to 287.64 g CO_2/m^3. At the same time, CO_2 emission from the oxic tank ranged from 103.75 g CO_2/m^3 wastewater to 275.71 g CO_2/m^3, accounting for about 96.0% of the total emission in the A^2O process, for not only did it have a high flux but also a large water surface area. In the AO process, CO_2 emission from the oxic tank (ranging from 85.98 g CO_2/m^3 wastewater to 346.55 g CO_2/m^3) accounted for a high percentage, i.e. about 96.6% of total emission, ranging from 87.11 g CO_2/m^3 wastewater to 356.96 g CO_2/m^3. In the oxidation ditch process, the total CO_2 emission ranged from 209.40 to 658.62 g CO_2/m^3 wastewater. CO_2 emission from the oxic areas (202.12–644.03 g CO_2/m^3 wastewater) was higher than that from the AO and A^2O processes, accounting for about 97.1% of the total emission from the oxidation ditch process. In the SBR process, CO_2 emission ranged from 128.58 to 654.73 g CO_2/m^3 wastewater, while the CO_2 emission from the feeding and aeration period ranged between 126.63 and 648.56 g CO_2/m^3 wastewater, this accounted for about 99.0% of total emission.

Although the surface areas (shown in Table 1) in the aerated grit tanks in the A^2O, AO and oxidation

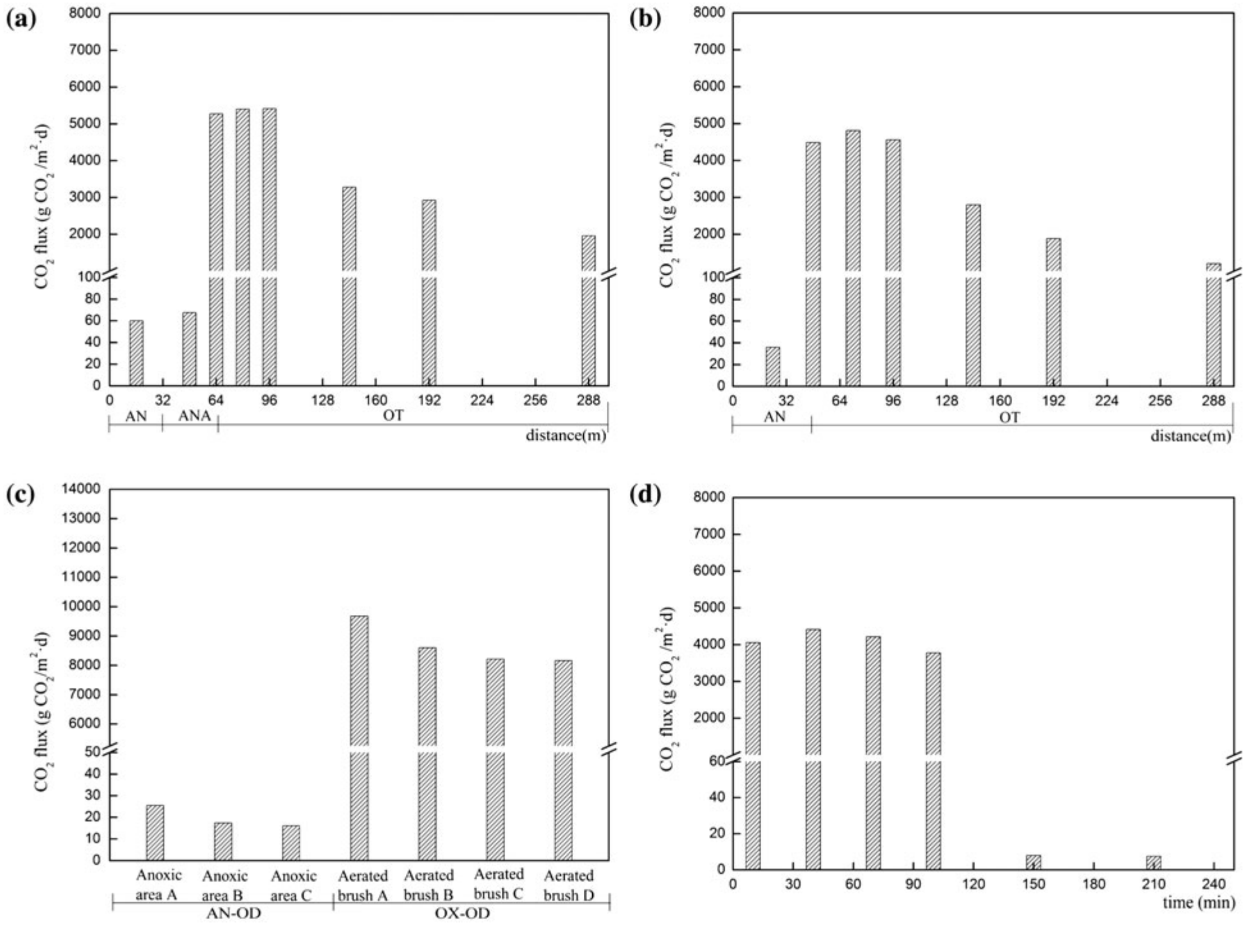

Fig. 3. CO_2 flux from secondary treatment over four processes: (a) A^2O process (b) AO process (c) oxidation ditch, and (d) SBR process.

Table 2
CO_2 emissions (g CO_2/m^3 wastewater) from each treatment unit in four different processes

WWTP		AGT	SGT	BST	PST	WDT	ANA	AN/AN-OD	OT/OX-OD/FAP	FC	SP	DP	SDT	STT
A^2O	Ave	5.25			0.28		0.47	0.51	168.65	0.52				
	Max	8.51			0.54		1.32	0.97	275.71	0.59				
	Min	0.96			0.12		0.16	0.05	103.75	0.03				
AO	Ave	4.38			0.25			0.34	167.99	0.41				
	Max	8.20			0.50			0.66	346.55	1.05				
	Min	1.07			0.02			0.02	85.98	0.02				
OD	Ave	5.39		0.06			0.27	2.20	335.87	2.08			0.04	0.07
	Max	7.59		0.20			0.71	3.13	647.03	2.80			0.05	0.11
	Min	3.50		0.03			0.16	1.50	202.12	2.05			0.02	0.02
SBR	Ave		0.14			0.15			343.86		1.83	1.36		
	Max		0.31			0.54			648.56		3.49	1.83		
	Min		0.03			0.02			126.63		1.06	0.84		

ditch processes were small, their CO_2 emissions still showed the second highest levels among the three treatment processes for the highest CO_2 flux. Instead, despite the large water surface area of the primary sedimentation tank and final clarifier, CO_2 emissions from these tanks accounted for a very low percentage

Table 3
Comparison of CO_2 emissions from A^2O, AO, oxidation ditch and SBR processes

WWTP	CO_2 emission (g CO_2/m^3 wastewater)	CO_2 emission (kg CO_2/kg COD)
A^2O	175.68	0.58
AO	173.37	0.68
Oxidation ditch	343.78	0.76
SBR	347.34	0.97

of total emission from the three WWTPs for their low CO_2 flux in these two units, ranging from 0.02 to 2.80 g CO_2/m^3 wastewater.

The CO_2 emissions of 0.02–3.49 g CO_2/m^3 wastewater in the anaerobic and anoxic tanks in the A^2O process, the anoxic tank in the AO process, the sludge distribution, and sludge thickening tanks in the oxidation process, as well as the swirl grit tank, settling period, and declining period in the SBR process, were entirely omitted because of these quite low CO_2 fluxes and their small surface areas.

3.4. Discussion

3.4.1. Comparison of CO_2 emissions from aerated and non-aerated areas

A significant gap in CO_2 fluxes was found between the aerated and non-aerated areas. The CO_2 emissions took account of more than 96.0% of the total emission from our four processes, similar to the CO_2 emissions from the biological treatment unit (92.0% of the total emission from a WWTP) found and reported by Czepiel et al. [22]. This phenomenon was attributed to the two following issues:

(1) Biological aerobic respiration. In the aerated areas, a small fraction of influent COD was degraded into CO_2 by the aerobic biomass and the rest was incorporated into the activated sludge, while some of the newly produced activated sludge was further transformed into CO_2 via endogenous respiration [11], and the CO_2 emission from aerobic reactor (period) consists of both biogenic and non-biogenic carbon though it could be the major fraction for non-CO_2 eq due to the aerobic decomposition.

(2) Air stripping caused by physical aeration. CO_2 can be easily stripped out from the wastewater to atmosphere by the effect of aeration, i.e. the CO_2 generated in both the former biological treatment unit and the oxic tank was stripped out altogether and almost completely from the aerated zone.

3.4.2. Comparison of CO_2 emission from the grit tanks

It is seen that the discrepancies between the average CO_2 fluxes in the aerated grit tank among the A^2O, AO, and oxidation ditch processes were relatively small. The CO_2 fluxes in the aerated grit tanks in our investigation were larger than those in a previous study [22], ranging from a minimum of 751 g CO_2/m^2 d to a maximum of 1,577 g CO_2/m^2 d, because the properties of influent wastewater and the conditions for pipe laying varied widely. Compared with the aerated grit tank, the swirl grit tank, contributed much less to CO_2 emission, given its physical stirring instead of aeration stripping. We conclude that, to a great extent, the CO_2 flux is a function of water turbulence.

3.4.3. Comparison of total CO_2 emissions

The total CO_2 emissions from the various processes are shown in Table 3. The table shows that the direct CO_2 emission was 175.68 g CO_2/m^3 wastewater in the A^2O process, 173.37 g CO_2/m^3 in the AO process, 343.78 g CO_2/m^3 in the oxidation ditch and 347.34 g CO_2/m^3 in the SBR process, indicating that the AO process contributed the lowest amount of direct CO_2 emission and may be considered as the optimal process in direct CO_2 reduction. However, if taking COD removal into consideration, the A^2O process shows the lowest direct CO_2 emission (0.58 kg CO_2/kg COD), which is clearly lower than that of the AO, oxidation ditch, and SBR processes. All the same, whatever the presence of fossil carbon in the influent water, the direct CO_2 emission caused by COD degradation shows a considerable amount of CO_2 which should not be ignored in the inventory of GHG emissions.

4. Conclusions

Large amounts of direct CO_2 emissions were found in four different wastewater treatment processes. The total direct CO_2 emission when treating wastewater using the A^2O, AO, oxidation ditch, and SBR processes was, respectively, 175.68, 173.37, 343.78 and 347.34 g CO_2/m^3 wastewater, of which the aerobic units/areas accounted for 96.0% (in the oxic tank in A^2O), 96.9% (in the oxic tank in AO), 97.1% (in the oxic areas of the oxidation ditch), and 99.2% (during the feeding and aeration period in SBR) of total emission. AO was the optimal process for CO_2 reduction when treating wastewater. However, the A^2O process contributes the lowest amount of CO_2 emission (0.58 kg CO_2/kg COD) when treating COD compared with the other three processes (0.68 kg for AO, 0.76 kg for the oxidation ditch, and 0.97 for SBR). We conclude therefore that, on the basis of our investigation the A^2O process is the optimal process for direct CO_2 reduction when taking COD removal into consideration. And it is worthy to make a further study to explore the proportion of CO_2 that comes from the fossil origin in different WWTPs.

Acknowledgment

This work was supported by the Environmental Public Welfare Fund (No. 201009053). As well, we thank the Beijing Urban Drainage Cooperation for its assistance.

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