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# Embedding constructed wetland in sequencing batch reactor for enhancing nutrients removal

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## **Abstract**

For conventional biological wastewater treatment processes, achieving stable and satisfactory nutrients removal is still a big challenge due to the probable lack of sufficient organic. A novel concept was firstly proposed and preliminarily investigated in a lab-scale sequencing batch reactor (SBR) by integrating with alum-sludge based constructed wetland (CW). This integrated novel system owns the striking features of adding carriers of wetland substrate (i.e. the dewatered alum sludge in this case) in SBR system for robust phosphorus adsorption while enriching the aesthetic value of CW in the SBR system. The preliminary 3-month trial with municipal wastewater has demonstrated average removal of 96%, 99% and 90% for BOD, TP and TN, respectively. The decoupling of phosphorus removal from organic allows more organic to cater to nitrogen removal. The introduction of biofilm and its interaction with suspended sludge in SBR supported high simultaneous nitrification and denitrification (SND) efficiency, ranging between 55%-88%, which dramatically contributes to total nitrogen depletion.

## **Keywords**

Alum sludge; Constructed wetland; Activated sludge; Phosphorus removal; Simultaneous nitrification and denitrification (SND)

## **INTRODUCTION**

Discharges from municipal and industrial wastewater treatment plants (WWTPs) have been identified as one of the major sources of aquatic pollution in industrialized countries. Among these pollutants, the release of nitrogen and phosphorus into river and lake could induce eutrophication and expose detrimental effect on aquatic organisms. In addition, most current WWTPs are also confronting growing connected population and rising quantity of wastewater, thereby urgency to be upgraded. All these factors address significant pressure on biological wastewater treatment processes, especially treating wastewater with low organic.

Many techniques have been proposed and operated in full-scale WWTPs to achieve these purposes, such as fluidized bed reactor (Islam et al., 2014), moving bed biofilm reactor (Barwal and Chaudhary, 2014; Javid et al., 2013), membrane bioreactor (Hazrati and Shayegan, 2011), integrated fixed-film activated sludge system (IFAS, Veuillet et al., 2014; Malovanyy et al., 2015). However, these approaches rooted in

improving the intrinsic problems of treatment technology rather than a defect of wastewater, such as low C/N ratio. For most municipal treatment plants, in order to achieve phosphorus and nitrogen removal simultaneously, pre-anoxic (i.e. UCT) is always adopted. However, higher internal recycle ratio should also be accompanied which undoubtedly increases the energy consumption. By the contrast, although post-anoxic setup abandons internal recycle, external carbon is a must to realize denitrification.

Actually, several operation-intensified methods have been proposed and taken into practice in full-scale. As summarized in Table 1, step feeding, multiple aerobic-anoxic stages, achieving denitrifying phosphate removal and simultaneous nitrification-denitrification by granular sludge or biofilm are among the widely employed methods. Both these methods achieved satisfactory effluent. However, all these methods need to be carefully monitored as influent fluctuates all the time. The distribution quantity in each point of step feeding, controlling oxygen concentration and nitrate supply for DPB is the key step in each strategy. In another word, these methods are a little bit sensitive and vulnerable.

**Table 1.** Typical enhancement of nitrogen removal by various methods

Influent (C/N/P, mg/L)	Removal efficiency (C/N/P, %)	Reactor	Remark	Reference
300/30/10	92/88/100	SBR	DPB cultivated for nitrogen removal	Lee et al., 2001
502/61/9	85/95/98		Step feeding strategy	Li et al., 2007
450/45/6.5	94/84/82		Pre-anoxic+anoxic-aerobic (3 pairs)	Ghehi et al., 2014
1500/150/75	96/88/80		Biofilm formed on the fixed-bed	Rahimi et al., 2011

In the context of these facts, a novel idea and process, named green biofilm-suspended sludge system (GBS), was firstly proposed by integrating alum sludge based CW into AS. In the present study, a lab-scale AS was retrofitted in order to preliminarily demonstrate the performance of this novel GBS, especially nitrogen removal. Moreover, attention was also addressed on the interaction of suspended sludge with biofilm in nitrogen removal, especially achieving simultaneous nitrification and denitrification (SND).

### Concept of GBS

GBS is derived from the integration of alum sludge based CW into conventional AS while its superiority is more than simply combining those two processes. The name of this novel process, Green Biofilm-Suspended sludge system, refers to most of its characteristics.

The dramatically aesthetic value because of vegetation firstly makes GBS a natural “green” ecosystem. The inherited pleasing appearance from CW turns wastewater

treatment plant, as depicted in Figure 1, to wastewater treatment park and habitat of more creatures<sup>15</sup> which is different from conventional ugly and odorous of WWTPs (Choi et al., 2012).



**Figure 1.** Conceptual scheme of GBS in a full-scale (left – conventional biological reactor; right – GBS)

For enhancement of nutrients removal, the matrix of alum sludge in GBS could act as carriers for supplying biofilm where much more nitrifiers would be attached to intensify nitrogen oxidation (Malovanyy et al., 2015). Moreover, alum sludge could also robustly remove phosphorus from wastewater through chemical adsorption, which has been confirmed by vast previous studies (Babatunde et al., 2008). Meanwhile, introducing phosphorus adsorption could allow more organics flowing to anoxic tank for denitrification in conventional AS. It means this novel process targets on remedying problems of not only AS but also the intrinsic defect of wastewater. Three functions could be achieved by only one step of introducing alum sludge based CW into the tank, which is the essence of GBS. What should be highlighted is that using alum sludge is in line with the policy of “reduce, reuse and recycle”.

The conceptual intention of GBS is to build a more sustainable process for wastewater treatment. Introducing alum sludge into AS has the potential to probably save a large quantity of chemical dosages, such as external organic carbon and ferrous/aluminum salts. It means volume of GHG could be offset indirectly. Actually, another appealing property of GBS compared with conventional AS is the potential carbon sink due to the existence of vegetation. This concept demonstrated the trial toward achieving sustainable wastewater treatment.

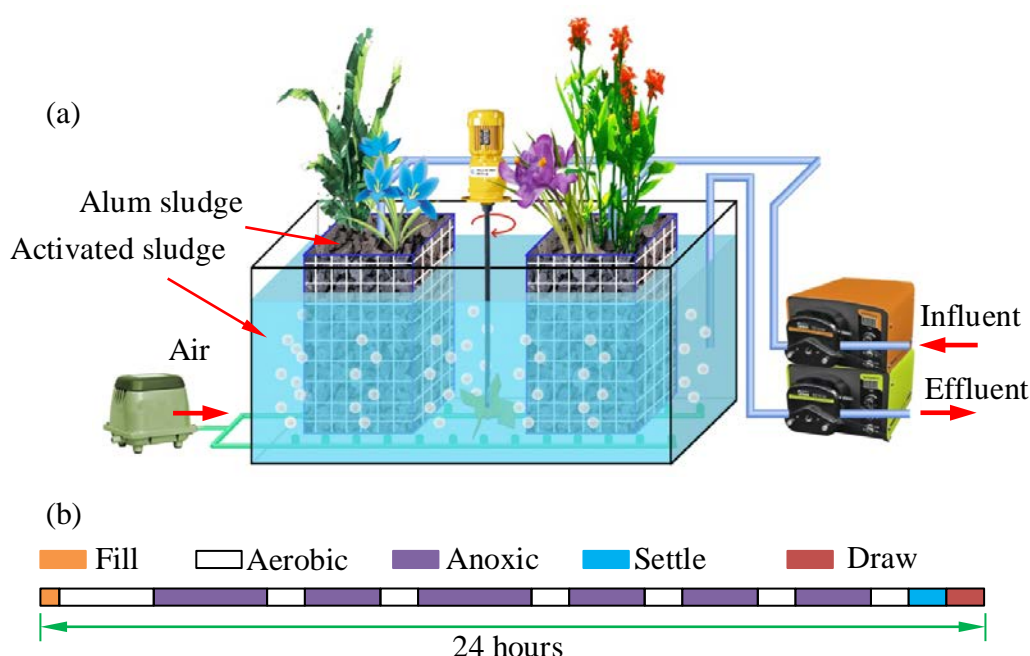
## **MATERIAL AND METHODS**

### **Lab-scale reactor and operation**

As depicted in Figure 2a, a plastic bucket (L×W×H: 54.5×35.0×42.0 cm) was refitted into a GBS system with a total volume of 65 L. Alum sludge<sup>18</sup> (10 kg in total, particle size around 2×2×2 cm) was filled into two mesh bags (20×16×30 cm) with mesh size of 0.2×0.2 cm. Then the two alum sludge bags with vegetation were hung in the bucket as floating CW. The actual working volume is 54 L. Air diffusers were placed

in a line at the bottom of the reactor and connected to an air compressor. A mixer was also installed in order to homogenize the suspended sludge.

At the very beginning, 20 L activated sludge collected from one WWTP in Dublin was seeded into the reactor to form initial suspended sludge (SS) concentration of  $2,000 \text{ mg}\cdot\text{L}^{-1}$ . 3 L mixed liquor was discharged every day at the end of last aerobic stage to keep the SS around  $2,000 \text{ mg}\cdot\text{L}^{-1}$  (SRT=18 d). The reactor was operated in SBR mode with four stages of fill, alternating aerobic and anoxic conditions, settle and draw of which the time distribution was depicted in Figure 2b. On and off of each stage was controlled automatically by timers and other parameters were summarized in Table 2. At the beginning of every cycle, the same volume of drained wastewater in the last cycle (33 L) was filled into the reactor by a peristaltic pump with an exchange ratio of 0.6 and an HLR of  $0.61 \text{ m}^3\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ .



**Figure 2.** Schematic description of alum sludge based GBS in lab-scale (a) and the time distribution of one cycle (b)

**Table 2.** Operation parameters of alum sludge based GBS

Parameter	Value	Parameter	Value
Cycle time (h)	24	DO (aerobic, $\text{mg}\cdot\text{L}^{-1}$ )	2.5-4
HLR ( $\text{m}^3\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ )	0.6	SS ( $\text{mg}\cdot\text{L}^{-1}$ )	1,500-2,000
OLR ( $\text{g}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ )	$242\pm 178$	Exchange ratio	0.6

HLR – Hydraulic loading rate, OLR – Organics loading rate

The performance of GBS over the whole operation period was monitored twice to three times per week including the influent/effluent quality of COD, BOD<sub>5</sub>, TN, NH<sub>4</sub><sup>+</sup>-N, NO<sub>x</sub><sup>-</sup>-N (NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup>), and TP. In order to understand the pollutants evolution in each stage, the pollutants profiles of one cycle were monitored and conducted respectively in two different days (20<sup>th</sup> and 50<sup>th</sup>). The cycle monitors were started after introducing the wastewater into reactor and samples were collected at the

end of each stage and filtered through 0.45  $\mu\text{m}$  membrane filter and then analyzed for SCOD, TN,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_x^-\text{-N}$ , and TP.

## Wastewater

Piggery wastewater was chosen as the influent of the present study. It was collected from an animal farm at Newcastle, Co. Dublin, Ireland and then diluted to municipal wastewater level with COD of  $400 \pm 80 \text{ mg}\cdot\text{L}^{-1}$ , TN of  $30 \pm 9 \text{ mg}\cdot\text{L}^{-1}$  and TP of  $15 \pm 4 \text{ mg}\cdot\text{L}^{-1}$  as real influent of the reactor. At the last month of operation, influent was changed to synthetic wastewater with COD, TN and TP of 400, 40, 10  $\text{mg}\cdot\text{L}^{-1}$  respectively and prepared according to Nguyen et al (2010).

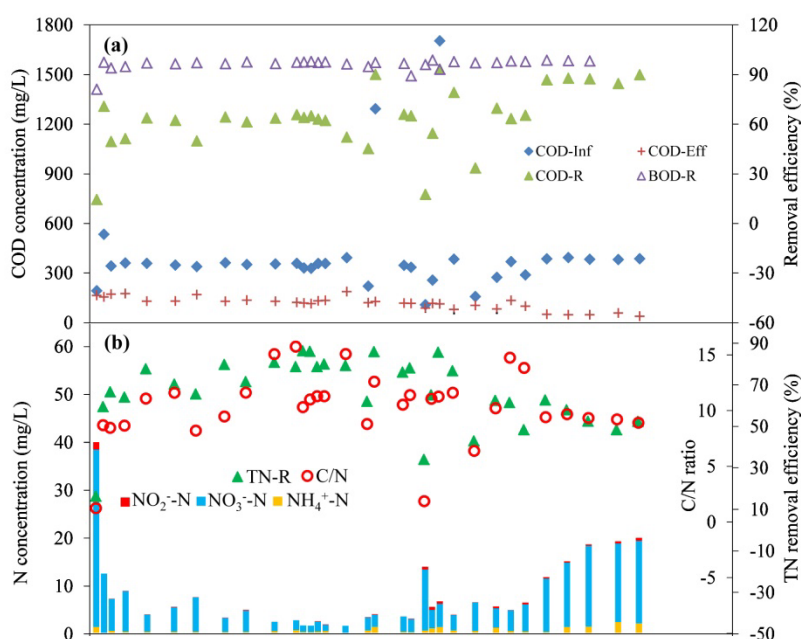
## Analytical methods

A HACH DR/2400 spectrophotometer was used to analyze COD, TN,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_x^-\text{-N}$  and TP according to its standard manual book.  $\text{BOD}_5$  was analyzed with a Hach BODTrak instrument (Hu et al., 2012). pH and DO were monitored with a pH meter (Orion 920 A+, Thermo) and a microprocessor oximeter (Oxi 325, WTW). Nitrification rate ( $N_R$ ) and denitrification rate ( $D_R$ ) were tested according to Hu et al (2012).

## RESULTS AND DISCUSSION

### Overall performance

The lab-scale GBS was operated for almost 3 months to preliminarily investigate its performance over pollutants removal. As depicted in Figure 3a, the COD content in influent was almost constant around 400  $\text{mg}\cdot\text{L}^{-1}$  except few singular points. For the whole stage, the removal efficiency of COD and BOD kept around 62% and 96% in average with SS concentration around 2,000  $\text{mg}\cdot\text{L}^{-1}$  in the reactor.



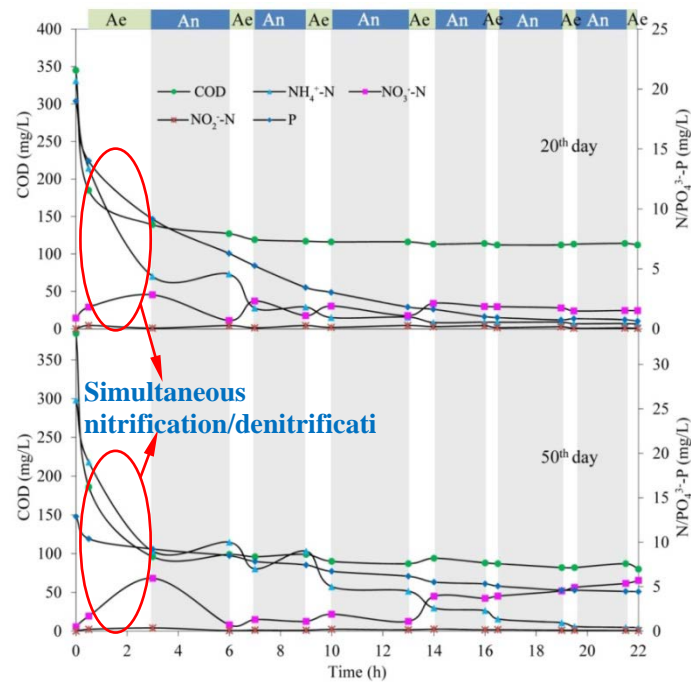
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**Figure 3.** Overall performance of alum sludge based GBS in removing organic and N (Effluent) (Inf – Influent, Eff – Effluent, R – Removal efficiency)

As can be seen in the chart (Figure 3b), 95% of  $\text{NH}_4^+\text{-N}$  could be converted to  $\text{NO}_x^-\text{-N}$  averagely at any time. The total nitrogen (TN) removal efficiency increased steadily from 70% at the beginning to around 90% at the 30<sup>th</sup> day. Given the unchanged C/N ratio, the improved performance should be induced by successfully formed biofilm on alum sludge. The low TN removal efficiency at the beginning or end was due to  $\text{NO}_3^-\text{-N}$  accumulation. The  $\text{NO}_3^-\text{-N}$  concentration in the effluent decreased obviously as biofilm was being cultivated from the 1<sup>st</sup> to 25<sup>th</sup> day. The existence of biofilm obviously did benefit to denitrification. As stated in previous IFAS, introducing biofilm could increase nitrification rate (Kim et al., 2010; Regmi et al., 2011) and also accelerate the consumption of BOD. However, the nitrate accumulation was mitigated with the biofilm under constant COD quantity. It is supposed that some specific schemes changing the distribution of organics were conceived, such as SND process (Virdis et al., 2011).

### **Cyclic pollutants removal profiles**

The cyclic COD, N and P evolutions were depicted in Figure 4. The removal of COD almost finished in the first aerobic stage. TN removal could be completed in the former 10 hours in both two cycles while the residual TN in the 50<sup>th</sup> day was obviously higher.  $\text{NH}_4^+\text{-N}$  was oxidized quickly in the first aerobic stage as well despite the competitive effect of heterotrophic bacteria. It is noteworthy that no corresponding amounts of  $\text{NO}_x^-\text{-N}$  accumulated in bulk liquid (Red cycle in Figure 4). Given the fact that minor  $\text{NO}_2^-\text{-N}$  was detected in the cycle as well as the high DO kept, possible ANAMMOX process could be excluded. Thus, it should be SND (Robertson and Kuenen, 1990; Holman and Wareham, 2005) that accounted for the depletion of  $\text{NO}_3^-\text{-N}$  in aerobic stage. Based on the definition of Hu et al. (2012) the highest gross SND efficiencies ( $E_{\text{SND}}$ ) in two cycles were calculated to be 88% and 55% respectively in the first aerobic stage.



**Figure 4.** Evolution of pollutants in a whole cycle during 20<sup>th</sup> and 50<sup>th</sup> (Ae – aerobic condition, An – anaerobic condition)

### Interaction between suspended sludge and biofilm in achieving robust SND

Similar to IFAS, alum sludge in GBS really induced benefits to N removal as expected. It is also noteworthy that robust SND was indeed observed in current study which utilizes organic more efficiently (Ju et al., 2006; Zhu et al., 2007). Actually, information about SND in IFAS system with co-existence of suspended sludge and biofilm is still scarce. Only Rutt et al. (2006) observed and mentioned SND in a full-scale IFAS system. That is because previous IFAS studies were usually conducted in a continuous reactor in which every tank exclusively remains one unique condition. The SBR in the present study may have advantage in achieving high SND efficiency as in line with the fact that SND in anoxic-aerobic mode is much higher than fully aerobic mode (Zhang et al., 2015).

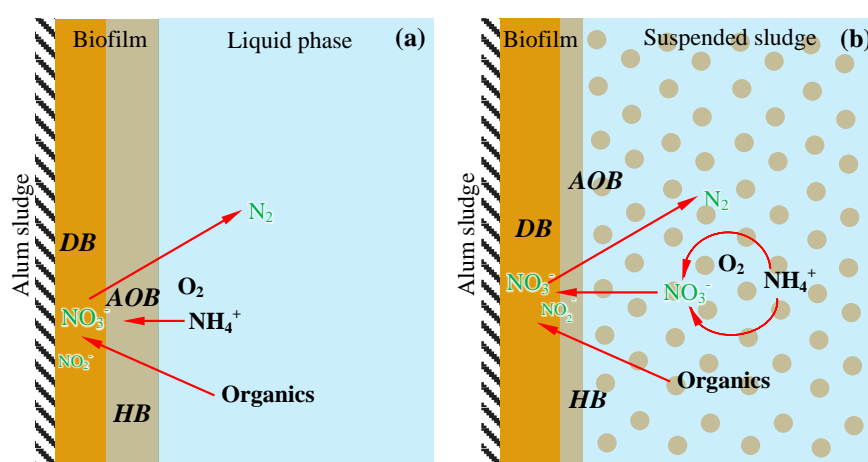
In order to investigate the mechanism of SND process in current study and differences from previous studies, the nitrification and denitrification rates of suspended sludge and biofilm were measured in jar test respectively.

The results (SI, Table S2) showed that suspended sludge undertook 80% ( $1.75 \text{ mg N} \cdot \text{L}^{-1} \cdot \text{h}^{-1}$ ) of nitrification along with 20% ( $0.32 \text{ mg N} \cdot \text{L}^{-1} \cdot \text{h}^{-1}$ ) on biofilm. The carriers did not contribute more as similar as that in IFAS (Regmi et al., 2011). Veuillet et al. (2014) ever observed in their reactor with the carriers circulating between all the tanks that 93% of nitrification took place in suspended sludge. In SBR reactor, nitrifiers seem to prefer growing in suspended sludge in order to compete with heterotrophic microorganisms for the substrate. On the other hand, the  $D_R$  in suspended sludge and biofilm were almost the same and the total  $D_R$  ( $3.85 \text{ mg N} \cdot \text{L}^{-1} \cdot \text{h}^{-1}$ ) was almost double of  $N_R$  ( $2.20 \text{ mg N} \cdot \text{L}^{-1} \cdot \text{h}^{-1}$ ). The accumulated nitrate in the first aerobic stage was just



because only denitrifying bacteria on the biofilm ( $1.77 \text{ mg N} \cdot \text{L}^{-1} \cdot \text{h}^{-1}$ ) could exert function in an aerobic condition which was lower than total  $N_R$ .

According to  $N_R$  and  $D_R$ , a preliminary demonstration of SND scheme in GBS was proposed and depicted in Figure 5 compared with that with biofilm only. The differences between two schemes were the existence of suspended sludge and the thickness of the top layer on biofilm. In the present scenario, the anaerobic condition in the deep layer should be protected by suspended sludge with the top layer to resist the  $O_2$  erosion. On the other hand, the existence of suspended sludge could also reduce the transportation kinetics of organics getting into the deep layer. Exactly, the organics stored by alum sludge could supply as a carbon source for denitrification as alum sludge enable to adsorb part of organics.



**Figure 5.** Proposed schematic diagrams of SND process in Biofilm only (a) and Biofilm combined with suspended sludge (b) (HB – Heterotrophic bacteria, AOB – Ammonium oxidizing bacteria, NOB – Nitrite oxidizing bacteria, DB – Denitrifying bacteria)

Overall, compared with SND in biofilm only (Puzanva et al., 2001), the SND achieved in present study with suspended sludge and biofilm together could probably have several advantages:

- High nitrification rate by getting rid of oxygen, alkalinity diffusion limitation;
- Improved mass transfer (Nitrate and organics) for denitrification as top layer was getting thin;
- Sufficient organics supply for denitrification in the deep layer with the adsorption effect of alum sludge to organics.

Therefore, the SND process happened in suspended sludge with biofilm together should be given further study. Overall, by introducing alum-sludge CW into activated sludge, nitrogen removal enhancement was achieved with the saved organic originally for PAOs. At the same time, with the biofilm on alum-sludge robust SND process came into function to contribute to nitrogen removal process.



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## Potential carbon-sink property of GBS

Generally, municipal WWTPs are carbon dioxide (CO<sub>2</sub>) source due to infrastructure construction, electricity consumption and chemical dose to purify wastewater. Specifically, greenhouse gas (GHG) emitted from WWTPs includes CO<sub>2</sub> induced by energy consumption (fossil fuel), nitrous oxide and methane (N<sub>2</sub>O and CH<sub>4</sub>) released from wastewater in biological processes. CO<sub>2</sub> from wastewater is not considered provided its biogenic fact. Although the contribution of GHG emission from wastewater sector is not significant, in the context of global warming and climate change, many studies and practical trials have been conducted to achieve net zero GHG emissions over the life time of WWTP – also known as the concept of “carbon neutrality”. Actually, carbon neutrality has partly or fully emerged in limited case studies of WWTPs by applying novel technologies, reducing energy consumption and/or enlarging energy reclamation.

GBS system obviously provides us a novel approach to contribute the carbon neutrality of WWTPs through the CO<sub>2</sub> sequestration function of vegetation. Additionally, reduction of chemical dose for phosphorus removal and possible phosphorus recovery could offset GHG emission indirectly. Based on the calculation of de Klein and van der Werf (2014), annual carbon sequestration by vegetation in total biomass varied between 617 and 977 g C·m<sup>-2</sup>·year<sup>-1</sup>. Undoubtedly, the vegetation in GBS will do benefit to “carbon neutrality” of WWTPs due to its net carbon-sink characteristic. It means that GBS system could let WWTPs step closer to “carbon neutrality”.

## CONCLUSION

The three-month preliminary study of GBS demonstrated its satisfactory performance in nutrients removal, especially the P removal without an anaerobic stage. The robust phosphorus removal by alum sludge and nitrogen removal by SND process in GBS make it a potential way to upgrade current WWTPs. Particularly, this GBS system provided us a novel scenario to remove pollutants economically as alum sludge is a kind of easily, locally accessible matrix compared with other commercial carriers. Moreover, the carbon-sink property of GBS could enjoy more advantage over other technology in the context of climate change.

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