16.6.3 Development of Appropriate Wastewater and Sludge Treatment Technology for Controlling CH_4 and N_2O Emission Applicable to China (Final Report)

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ABSTRACT

With the aim to improve nitrogen removal and to control the emissions of green-house gases as well, some comparative investigations were made between the processes of SND (simultaneous nitrification and denitrification) and SQND (sequential nitrification and denitrification) for treating domestic wastewater.

The results showed that both SND and SQND had good nitrogen removal abilities, with TN removal 41.42% for SND and 45.65% for SQND, and the later one is a little higher than former one by 4.23%, but SND obtained a less lower N_2O emission than SQND; that DO concentration played important role in aerobic phase of SND for TN removal, and it indicated the tendency that TN removal efficiency went higher as DO became lower.

It seems that intermittently aerated SBR system, a kind of modified SQND process, is a hopeful wastewater treatment process for nitrogen removal, for that it obtained a TN removal efficiency of 81.8%, much higher than those obtained from SND and SQND.

Key words Simultaneous nitrification and denitrification, Nitrogen removal, Green-house gases, N_2O emission

1.Introduction

Biological processes with both denitrification and nitrification for removal of nitrogen are becoming increasingly important because of the need for controlling eutrophication in many areas. N_2O, an important kind of green-house gas, is closely related to the process of nitrification and denitrification, and its emissions from sewage disposal with nitrification and denitrification have been known since the late 1970s. This source has been largely neglected in recent budgets of N_2O sources, but it can also contribute a significant part to the global N_2O emissions. The estimated global emission of N_2O from sewage disposal with nitrification and denitrification was considered to be in the range of 0.3 – 3 Tg/year[1]. To reduce this sum of N_2O emission, many efforts should be made in the field of sewage disposal because of the vast variety and high complexity of its processes.

Nitrification is generally considered to be autotrophic conversion of ammonia through nitrite to nitrate. Whereas, denitrification was initially considered to be strictly an anoxic process, since denitrifiers, as facultative aerobes, prefer to use the DO (even at DO concentration as low as 0.1 mg/l) for respiration. This prevents the use of nitrate or nitrite as terminal electron acceptors[2]. From traditional theory, nitrogen could only be removed by sequential nitrification and denitrification (SQND), and this theory has been guiding the sewage treatment practice for many years up to now. Therefor, almost all the sewage treatment facilities constructed before were and are operated in the system of SQND.

In recent years, some important progress about the theory of nitrification and denitrification has been made that many heterotrophic organisms have been found to be able to nitrify organic and inorganic nitrogen compounds, given the dominance of heterotrophs over autotrophs in most ecosystems[3, 4] ; that aerobic denitrifiers are also heterotrophic nitrifiers and as such, convert ammonia directly into gaseous end products[3]; and that, on the other hand, certain species of bacteria are able to denitrify aerobically [5].

So, some inferences could be obtained from the newly found theory about nitrification and denitrification that both actions of nitrification and denitrification can be accomplished in one process, and in some conditions, the process of simultaneous nitrification and denitrification (SND) may have some advantages over the process of sequential nitrification and denitrification [6, 7].

Although a great few investigations have been done into SND process, mostly in the way of pure culture studies, little has been done systematically about its comparison with SQND process and about its green-house gases emission problem

This investigation is mainly focused on some comparative studies about nitrogen removal and about N_2O emission between SND and SQND.

2. Materials and Methods

2.1 Experimental Apparatus

Four SBR reactors of same size were divided into two sets, with 2 reactors of each set used for SND and SQND, respectively. The diameter of each reactor is 150 mm and height of 500 mm. The apparatus was kept in a thermostat room with the working temperature changeable as required.

The system was operated by an automatically controlling system. The HRT of the cycle and the operation time of each stage, such as fill, anoxic (in case of SQND), oxic, settling and discharge, could be easily changed according to the requirements .

2.2 Substrate Composition

Artificial wastewater and sewage from a nearby residential quarter were used as influents, but for the purpose of scheduled research, artificial wastewater was mainly used. The artificial wastewater was made in the way to obtain a similar quality of the wastewater discharged from some China cities, the characteristic of which is that the concentration of NH_4-N is a bit high and the ratio of TOC / TN is relatively low. In order to investigate the effect of TOC / NH_4-N ratio on N_2O emission, concentration of NH_4-N was kept constant but TOC (or COD) was changed as required (Table 1).

Table 1 Constituents of Artificial Wastewater

Constituents	Concentration
NH_4Cl	20-40 mg · l^{-1}as N
Sugar (as TOC)	60-400 mg · l^{-1} as required
Na_2CO_3	100-280 mg · l^{-1}
Phosphate buffer solution*	0.5 ml · l^{-1}
Magnesium sulfate solution*	0.5 ml · l^{-1}
Calcium chloride solution*	0.5 ml · l^{-1}
Ferric chloride solution*	0.5 ml · l^{-1}

* Made in the same prescriptions as reagents of BOD_5 measurement (Standard Method, 1989).

2.3 Sludge Cultivation

10 liters of seeding sludge was taken from the recycling sludge line of a wastewater plant near Tsukuba city, and was cultivated in the same operation way as SBR systems of SND and SQND, respectively, fed with the artificial wastewater for about half a month. Then, the sludge was moved into the 4 reactors, and after that the experiment started.

2.4 Experimental Procedures

The operation cycle of SND system includes the following procedures: 10 minutes for fill, 11 hours for oxic, 40 minutes for settling, and 10 minutes for discharge. And for SQND system, the procedures are: 10 minutes for fill, 2 hours for anoxic, 9 hours for oxic, 40 minutes for settling, and 10 minutes for discharge. Wastewater treated by every cycle was 2.65 l which is 50 % of the total available volume.

2.5 Analysis

Influent and effluent samples were analyzed for NH_4-N, NO_2^--N, NO_3^--N, TN, TOC and COD. Among them, NH_4-N, NO_2^--N, NO_3^--N and TN were analyzed with TRAACS-800 instrument (Roy and Knowles, 1994), and TOC was analyzed with TOC-5000 instrument (Shimadzu Co.). N_2O concentration was analyzed with GC-8A instrument (Wu et al, 1996).

3.Results and Discussions

3.1 Nitrogen Removal

3.1.1 Comparative nitrogen removal between SND and SQND

Comparative investigations into nitrogen removal between the processes of SND and SQND were made, and the results were presented in Figures 1 and 2, respectively. As shown, both SND and SQND have good nitrogen removal efficiencies, with TN removal 41.42% for SND and 45.65% for SQND, and the later one is a little higher than former one by 4.23%.

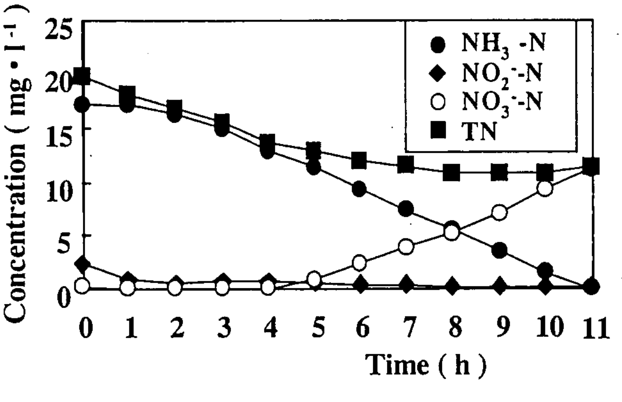

Fig. 1 Nitrogen removal by SND

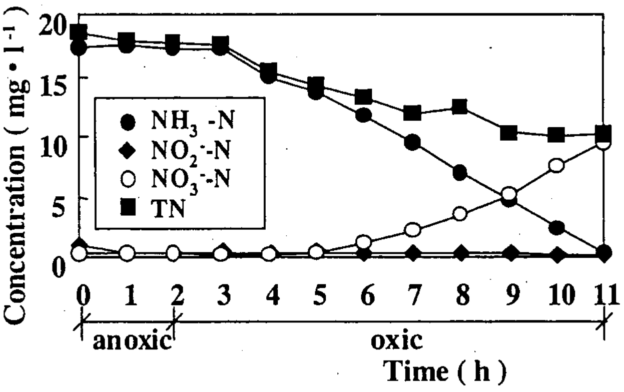

Fig. 2 Nitrogen removal by SQND

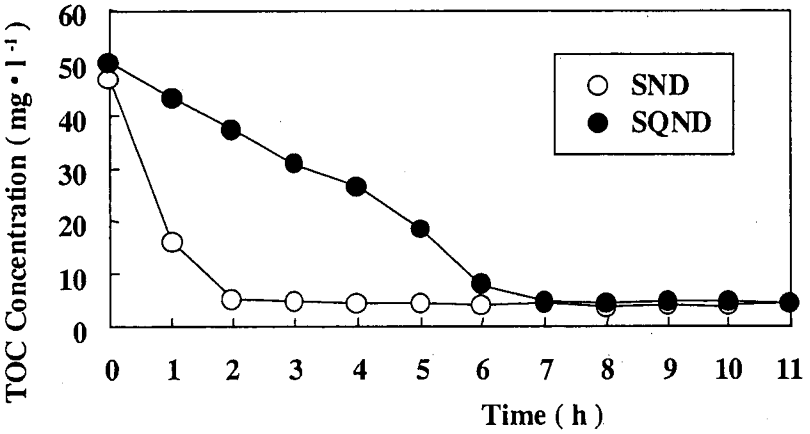

Fig. 3 TOC removal during one cycle of SND and SQND

It could be identified clearly from Figure 1 that aerobic denitrification did happen, mainly in the earlier stages of SND cycle. During the first 4 hours period from the start of oxic phase, NH_3-N decreased gradually but no increase of NO_3^--N and NO_2^--N could be found. Conclusions could be easily drawn that simultaneous nitrification and denitrification happened during this period, and as a result, TN removal was achieved. Similarly, Figure 2 also indicated this phenomenon during its earlier stage of oxic period (during 3 hours period after beginning time). During the later stage of the oxic period of both SND and SQND, ammonia nitrogen was reducing and nitrate increasing, and meanwhile, there was no evident TN removal. Therefore, it is suggested that nitrification became the dominant action during the later oxic period both in the processes of SND and SQND. And, the TOC analysis throughout the cycle, indicated that carbon source plays an important role in the process of denitrification for that TOC decreased quickly as the oxic time goes on, and only during the earlier stage of the oxic period denitrification happened when carbon source was still sufficient for its use (Figure 3).

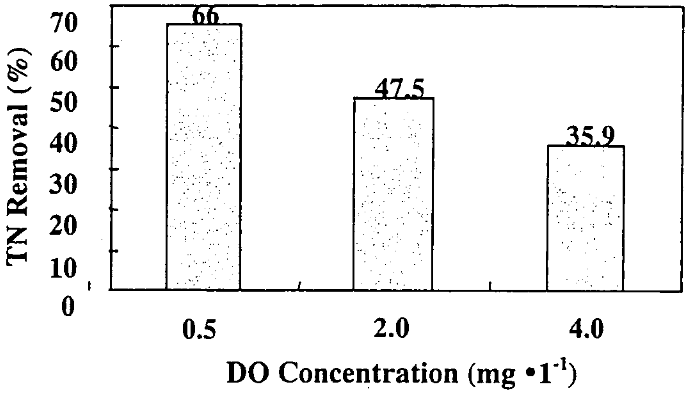

Fig. 4 Effect of DO on nitrogen removal

3.1.2 Effect of DO concentration on nitrogen removal

As shown in Figure 4, DO concentration could exert important effect on total nitrogen removal by SND process. With the same influent quality, COD 400 mg/l and NH_3-N 40 mg/l, different total nitrogen removal efficiencies of 35.9%, 47.5% and 66% were obtained under different DO concentration of 4, 2 and 0.5 mg/l. It could be naturally concluded that nitrogen removal efficiency goes higher as DO concentration becomes lower. The micro-zone theory and oxygen transport depth may be able to give some explanation to the experimental results here. Under lower DO condition, it is easier to create more and better anaerobic (or anoxic) micro-zones inside the bio-flocs, and thereby, two functions of denitrification, such as anoxic denitrification and aerobic one could contribute for nitrogen removal at the same time. As a result, better TN removal could be obtained under lower DO condition.

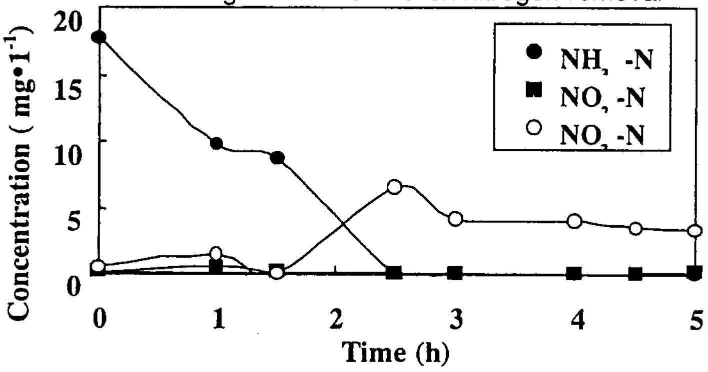

Fig. 5 Nitrogen removal by intermittent aeration system

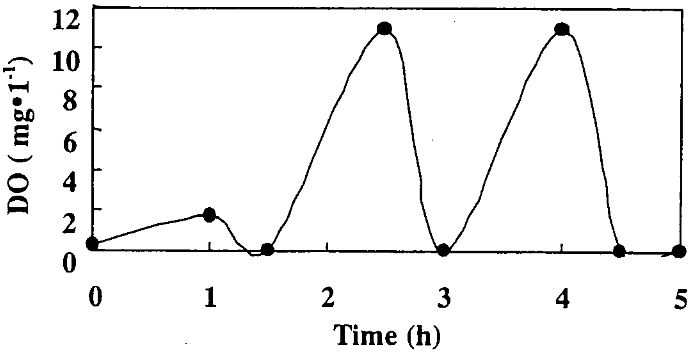

Fig. 6 DO concentration during one cycle of intermittent aeration system

3.1.3 Intermittent aeration SBR system

Inspired by the results from SND and SQND processes, intermittent aeration SBR system (It may be called intermittent sequential nitrification and denitrification, ISQND) was tested, which is actually a modified SQND process but may be also similar to SND in some way. The intermittent aeration process was operated in the mode of every 1 h for oxic and every other 0.5 h for anoxic with 3 hours for oxic and 1.5 hours for anoxic in total during one cycle, respectively.

With more than 30 cycles' experiments for intermittent aeration system lasting longer than one week, results indicated that intermittent aeration process could produce a higher efficiency for nitrogen removal, which can reach as high as 81.8% as shown in Figure 5.

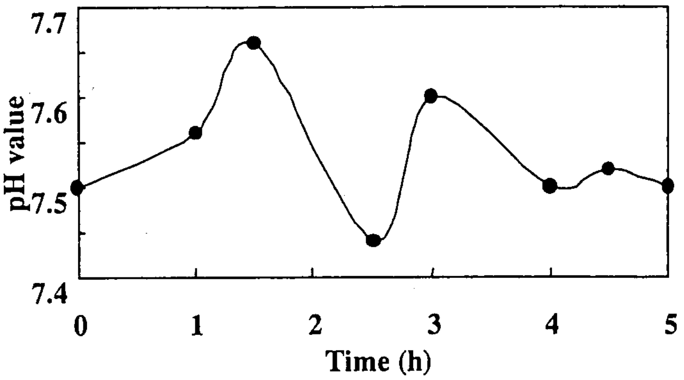

Fig. 7 pH value during one cycle of intermittent aeration system

The reason for improvement of nitrogen removal may be that repeated denitrification could produce alkalinity to maintain a suitable pH value for nitrification in the next step (Figure 6 and Figure 7), and as a result of this, cost of adding alkali into wastewater could be reduced; that nitrification rate in the oxic period could become faster because of removal of nitrate and nitrite in time during the anoxic period in advance of the next oxic step.

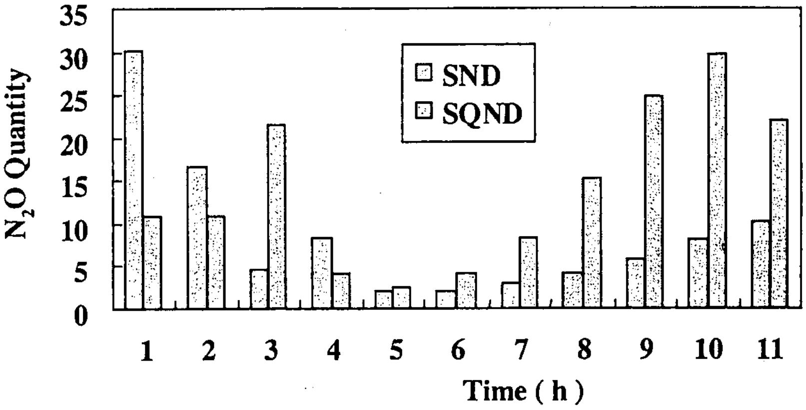

Fig.8 N_2O emission throughout One Cycle

It seems that SBR system operated in the way of intermittent aeration is a hopeful wastewater treatment for nitrogen removal. But some further investigations about the process is still needed to be done, such as investigation into its characteristic of N_2O emission.

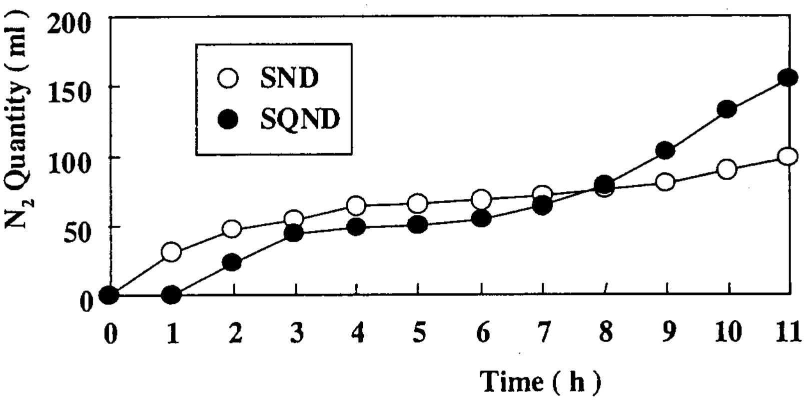

Fig.9 Accumulated N_2O emission in one cycle

3.2 Green-house gases emission

3.2.1 Comparison of N_2O emission between SND and SQND

During one cycle period, SND process was kept aerating for 11 hours and SND was operated 2 hours for anoxic first and then 9 hours for oxic. Sampling was made every hour for N_2O analysis and the amount of N_2O emission then calculated by multiplying its concentration and airflow.

Figure 8 illustrates the amounts of N_2O escaped from SND and SQND throughout one cycle, and Figure 9 indicates the accumulated amounts of N_2O emission within the cycle. The results showed that the total N_2O emission from SND process within one cycle is less than that from SQND, but N_2O emission of SND during the first 2 hours was higher than that of SQND during the same time when SQND was undergoing the anoxic condition.

Being different from the SND, the N_2O emission of SQND during the anoxic time was calculated from the N_2O in upper static air in balance with the wastewater below inside the reactor.

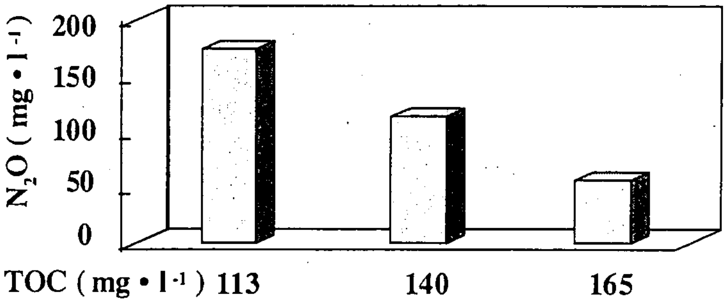

Fig.10 Influence of TOC in N_2O emission

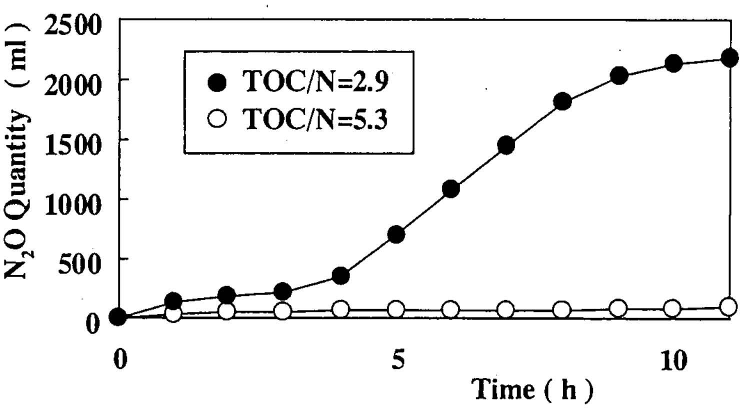

Fig.11 N_2O emission from SND at different TOC/N

Figure 8 also indicates that the N_2O emission during the period of the third hour from SQND is very high, and it seems that most of the N_2O was produced during the anoxic time before and still dissolved in wastewater in balance with above air, and it was stripped out from water by aeration as the oxic hour started, resulting in high N_2O concentration in airflow as the aeration just started.

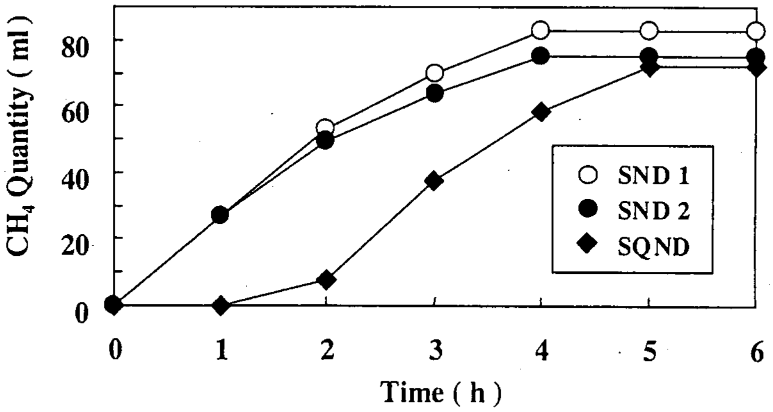

Fig.12 CH_4 emission from different process

3.2.2 *Effect of TOC/N*

Comparative experiments were made by using different influents with varied TOC but fixed NH_4-N concentration, and the results were shown in Figure 10 and 11. Figure 10 indicates the effect of TOC/N on N_2O emission during the anoxic process of SQND. And the

Figure 11 showed effect of TOC/N on the N_2O emission throughout one cycle of SND process. All the results illustrated that insufficient carbon source in influent resulted a higher N_2O emission, and the ratio of TOC to NH_4-N may have important effect on both nitrification and denitrification processes.

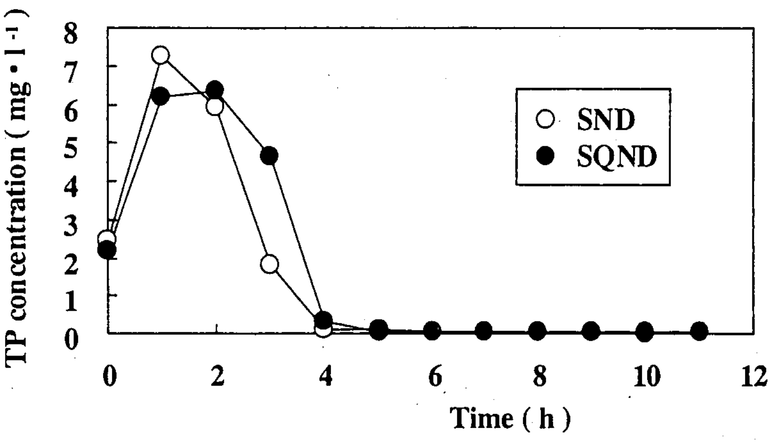

Fig.13 Phosphorus removal by DND and SQND

3.2.3 CH_4 Emission

Investigations into CH_4 emission from the processes of SND and SQND were also made, though CH_4 emission may not be so important to anoxic and oxic processes in this studies. In Figure 12, the runs of SND1 and SQND had the same TOC concentration of 130.18 mg/l for influent, and that of SND2 was 69.85 mg/l. Figure 12 indicated that there was no clear difference of CH_4 emissions between SND and SQND, but that lower TOC in influent could lead to a lower CH_4 production.

3.3 Phosphorus Removal

Both processes of SND and SQND have very satisfactory results for phosphorus removal, as shown in Figure 13. The rule is very evident that phosphorus was released from sludge at the anoxic stage of SQND and at the earlier stage of SND when DO was very low, and then absorbed by the sludge when DO became high at the later period of the cycle. This result indicated that phosphorus could be mainly absorbed by the activated sludge in aerobic process, and good separation of the sludge with effluent is required for phosphorus removal, and a careful disposal of the wasted sludge as well.

4.Conclusions

Both SND and SQND have good nitrogen removal efficiency, with TN removal 41.42% for SND and 45.65% for SQND, and the later one is a little higher than former one by 4.23%, but SND could produce a lower N_2O emission than SQND.

The ratio of TOC to NH_3-N has important effect on N_2O emission, and insufficient carbon source leads to higher emission of N_2O.

With the same influent quality, COD 400 mg/l and NH_3-N 40 mg/l, different total nitrogen removal efficiencies of 35.9%, 47.5% and 66% were obtained under different DO concentration of 4, 2 and 0.5 mg/l. It could be easily concluded that nitrogen removal efficiency goes higher as DO concentration becomes lower. The micro-zone theory and oxygen transport depth may be able to give some explanation to experimental results here.

It seems that SBR system operated in the way of intermittent aeration (ISQND) is a hopeful wastewater treatment process for nitrogen removal, for that it has the highest efficiency for nitrogen removal among the three processes tested in this study. But some further investigations about the process are still needed to be done, such as N_2O emission with the process.

5.References

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