



Enhanced removal of total nitrogen and total phosphorus by applying intermittent aeration to the Modified Ludzack-Ettinger (MLE) process



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ABSTRACT

In the conventional preanoxic Modified Ludzack-Ettinger (MLE) process, the nitrified mixed liquor cannot be completely returned to the preanoxic zone for denitrification. In this study, we applied intermittent aeration to the MLE process to enhance denitrification and total phosphorus (TP) removal. Pilot-scale test results indicated that, when treating municipal wastewater with a carbon (represented by five-day biological oxygen demand) to total nitrogen (TN) ratio (e.g. C/N ratio) of 2.4 or greater, the effluent ammonia, TN, and TP were generally less than 0.5 mg-N/L, 5 mg-N/L, and 0.5 mg-P/L, respectively. The removal efficiencies of both TN and TP were approximately 90%. Under extremely low temperature of 9.3 °C and low C/N ratio of 1.6 conditions, this process still removed 76% of TN and 56% of TP. Compared to the conventional MLE process, the implementation of intermittent aeration significantly improved TN and TP removal and also saved aeration energy by 10%. Existing MLE plants can be easily upgraded to this intermittent aeration mode to achieve enhanced nutrient removal while saving energy.

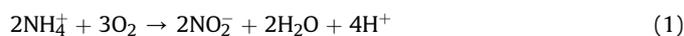
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1. Introduction

Eutrophication is one of the most significant concerns for surface water quality and drinking water supplies. This is especially true for sensitive water bodies such as Chesapeake Bay and the Gulf of Mexico in the United States, Dianchi Lake and Taihu Lake in China, and many others in the world (Oleszkiewicz and Barnard, 2006; Conley et al., 2009; Le et al., 2014). Accordingly, regulations on the discharge of total nitrogen (TN) and total phosphorus (TP) from wastewater treatment facilities have become more and more stringent, particularly in European countries and in North America (Oleszkiewicz and Barnard, 2006). Based on the directive 98/15/EC on urban wastewater treatment in Europe (European Commission, 1998), the effluent concentrations of TN and TP for large plants (>100,000 PE) must be less than 10 mg/L and 1 mg/L, respectively, or the removal efficiencies for TN and TP must be at least 80% and 70–80%, respectively (Oleszkiewicz and Barnard, 2006). In both the United States and Canada, the nutrient discharge permit is

determined based on the capacity of the receiving water body. Many wastewater treatment plants in the central region of North America do not need to remove nutrients. However, for sensitive water bodies, the discharge limits for TN and TP can be less than 3 mg/L and 0.07 mg/L, respectively (WERF, 2006; Oleszkiewicz and Barnard, 2006). In China, the most stringent national discharge limits for ammonia nitrogen, TN, and TP are 5, 15, and 0.5 mg/L, respectively (China EPA, 2002), and they were proposed to decrease to 1, 10, and 0.3 mg/L, respectively (China EPA, 2015). However, more than 50% of the treatment facilities in China do not even meet the current requirements (Jin et al., 2014). These new requirements have reached, or will exceed, the technology limits for biological nutrient removal (BNR) processes if chemical addition and filtration are not implemented (Oleszkiewicz and Barnard, 2006; WERF, 2006).

Conventional nitrogen removal pathways are employed by BNR plants. According to this pathway, ammonia is oxidized to nitrite by ammonia oxidizing bacteria, and then nitrite is oxidized to nitrate by nitrite oxidizing bacteria. Both reactions are accomplished under aerobic conditions, and this process is called nitrification. These reactions can be expressed as (Metcalf and Eddy, 2003):



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The nitrate is then reduced to nitrogen gas under anoxic conditions using organic matter as electron donors, and this process is called denitrification. When using the organic matter in the wastewater as the electron donor, the reaction can be expressed as (Metcalf and Eddy, 2003):



To utilize the influent organic matter as an electron donor for denitrification, a preanoxic zone is needed, and the nitrified mixed liquor in the aeration zone is returned to the preanoxic zone for denitrification (Metcalf and Eddy, 2003). This process is called Modified Ludzack-Ettinger (MLE) process. In this process, a fraction of the nitrified mixed liquor cannot be recycled back to the preanoxic zone (Metcalf and Eddy, 2003). As a result, complete denitrification cannot be achieved by the MLE process.

Phosphorus can also be removed biologically. In this process, phosphorus accumulating organisms (PAOs) need to be cultivated under alternating anaerobic and oxic conditions (Metcalf and Eddy, 2003). These PAOs uptake phosphorus under aerobic conditions. During sludge wasting, the excess sludge along with the sorbed phosphorus are removed simultaneously. Because the MLE process does not have a strict anaerobic zone, its TP removal is not ideal.

The anaerobic-anoxic-oxic (A²O) process and the University of Cape Town (UCT) process, aimed at removing both nitrogen and phosphorus, cannot achieve complete denitrification (Cosenza et al., 2014). They generally remove up to 70% of TN with effluent concentrations ranging between 8 and 15 mg/L (Metcalf and Eddy, 2003; Vaipoulou and Aivasidis, 2008; Lee et al., 2009; Wang et al., 2012). To further reduce the effluent TN concentration, a post-anoxic zone followed by a polishing aerobic zone is needed, and this process is called Bardenpho process. External carbon, including methyl alcohol, acetate, or other kinds of organic materials (i.e., food wastes), is added to the post-anoxic zone to improve denitrification. This will significantly increase the treatment cost. The step-feed anoxic-oxic process (Tang et al., 2007; Zhu et al., 2009; Mannina et al., 2016) could achieve excellent TN removal, while the TP removal was insignificant due to the lack of anaerobic condition within the process train. In the modified step-feed processes, such as UCT step-feed process (Ge et al., 2010, 2011; Majdi Nasab et al., 2016; Mannina et al., 2017), both TN and TP could be removed comprehensively with effluent concentrations less than 5 and 0.5 mg/L, respectively. However, these step feed processes are complex to operate (Chen et al., 2016).

In addition to the MLE process, alternating aerobic-anoxic (AAA) process, which employs intermittent aeration strategy, is also used for TN removal from municipal wastewater (Hao and Huang, 1996; Chachuata et al., 2005; Habermeyer and Sánchez, 2005; Fulazzaky et al., 2015). When the aeration is on, the ammonia is oxidized to nitrate. When the aeration is off, the nitrate is denitrified to nitrogen gas. The effluent ammonia and nitrate concentrations in the AAA process is highly dependent on the hydraulic retention time (HRT) and aerobic/anoxic durations. When the tank is large enough (i.e., long HRT) to dilute the ammonia during aeration off period and nitrate during aeration on period, this process could reduce both the ammonia and nitrate to less than 1 mg/L, respectively (Habermeyer and Sánchez, 2005). A long HRT, however, will increase the reactor size therefore the construction cost.

The nitrogen removal process based on novel pathways, such as the recently developed anaerobic ammonium oxidation (e.g. anammox) process, can theoretically remove nitrogen from wastewater with a very low 5-day biochemical oxygen demand (BOD₅) to TN ratio (C/N ratio) (Jetten et al., 2001; Hao et al., 2002;

Sun et al., 2010). However, the operational condition that achieves this nitrogen removal pathway is very strict. So far, they have not been used to treat raw municipal wastewater due to the large fluctuations in strength, flow rate, and temperature of the wastewater. Furthermore, the low nitrogen concentration in raw wastewater adds difficulty to the practical operation of the anammox process (Sun et al., 2010).

In this study, we integrated the internal mixed liquor return and the intermittent aeration into one process to extensively remove TN and TP without adding significant operational complexity. This process was evaluated using regular, limited C/N ratio, and very low C/N ratio municipal wastewaters under different temperature conditions in a pilot-scale, to provide an alternative to the existing enhanced nutrient removal (ENR) process.

2. Materials and methods

2.1. Process description

Fig. 1 shows the process schematic used for this research. It includes a mixing zone, an intermittent aeration zone, a post-aeration zone, and a clarifier. The mixed liquor in the intermittent aeration zone is continuously returned to the mixing zone (internal mixed liquor return). The process schematic is similar to the MLE process, except a mixing device is added to the intermittent aeration zone and a continuously aerated post-aeration zone is added after the intermittent aeration zone. To differentiate it from the MLE process, we name it intermittent MLE, or iMLE process. In the iMLE process, the aeration device in the intermittent aeration zone is operated in an on/off cycling pattern. When the aeration device is in operation, the intermittent aeration zone is under an aerobic condition, and the mixing zone is under an anoxic condition. The iMLE process operates exactly like the conventional MLE process to conduct nitrification within the intermittent aeration zone and denitrification within the mixing zone. The intermittent aspect is to turn off the aeration device while turning on a mixing device within the intermittent aeration zone, so that the intermittent aeration zone becomes anoxic to denitrify the residual nitrate. In the meantime, the return mixed liquor to the mixing zone does not contain any dissolved oxygen (DO) and contains a very low nitrate concentration, which turns the mixing zone to an anaerobic condition to cultivate PAOs. The on/off operation of the aeration device repeats, and their durations can be easily adjusted to achieve the best result for different influent qualities and environmental conditions. During the aeration off-period, a low concentration of ammonia will accumulate in the effluent of the intermittent aeration zone, and phosphate will also be released into the water. Therefore, a post-aeration zone is added to polish the effluent from the intermittent aeration zone. Within the post-aeration zone the residual ammonia is oxidized and the soluble phosphorus is re-uptaken by the sludge. As a result, by simply converting the aerobic zone of a MLE process into an intermittent aeration zone, and

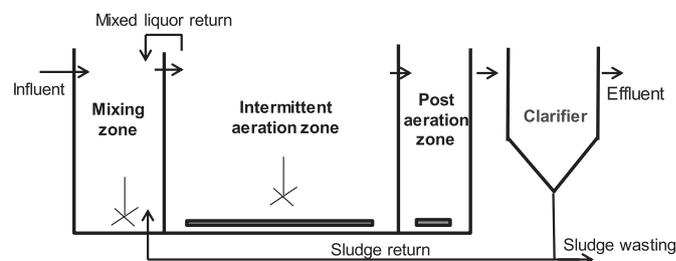


Fig. 1. Schematic of the intermittent Modified Ludzack-Ettinger (iMLE) process.

adding a post-aeration polishing zone, both nitrogen and phosphorus are extensively removed. Enhanced denitrification also allows the iMLE process to recover most of the oxygen credit in the nitrate form, thereby reducing oxygen demand.

A pilot-scale iMLE process was constructed using a standard 20-ft shipping container. The effective volumes of the mixing zone and the intermittent aeration zone were 6.3 and 14.3 m³, respectively, and the post-aeration zone was 1.25 m³. The aeration on/off operation in the intermittent aeration zone was controlled by a timer. The pilot-scale unit was installed at the Southeast Wastewater Treatment Plant in Rolla, Missouri, and fed with raw wastewater after the screening.

2.2. Experimental setup

The operational conditions for different phases are shown in Table 1. In Phase I, the process was started up naturally with raw wastewater, without adding any seeding sludge. It was operated using the intermittent aeration mode (iMLE mode). After approximately 5 weeks (Fig. 3(b)), complete nitrification was achieved (effluent ammonia <1 mg-N/L). Then, the performance of the iMLE process was evaluated using regular wastewater for 2 months (Phase II). In Phase III, the process was operated using the continuous aeration mode (MLE mode) for approximately one month, to compare the nutrient removal performance and energy consumption between the iMLE and MLE operations. During the MLE mode operation, the internal mixed liquor return rate was not changed.

In Phase IV, the TN removal performance of the iMLE process was tested at a reduced C/N ratio to evaluate the process performance under carbon limited conditions. Tap water and ammonium bicarbonate were added to the influent, to decrease the BOD concentration while maintaining a similar TN concentration as before. Because the temperature dropped significantly during Phase IV testing, the sludge wasting rate was reduced to increase the solids retention time (SRT) to compensate for the low temperature effect.

In Phases V and VI, the temperature of the reactor dropped to less than 10 °C. The C/N ratio was further reduced to 1.6, to test the reactor performance under extremely unfavorable conditions (both temperature and C/N ratio). Because the raw wastewater was very much diluted during these phases, only ammonium bicarbonate was added to the raw wastewater to decrease the C/N ratio. To quickly accumulate nitrifiers in the low temperature, the process was operated using the MLE mode first (Phase V). During Phase V, the added ammonia concentration was about 7 mg/L at the beginning, and finally reached 24 mg/L at the 170th day. When complete nitrification was achieved (effluent ammonia <1 mg-N/L), the operation changed into iMLE mode, marked as Phase VI. In Phases V and VI, the reactor temperature decreased to less than 10 °C and the SRT was increased to 35–45 days to compensate for the low temperature impact. To maintain a long SRT of 35–45 days, the inflow rate was reduced to decrease the hydraulic loading and increase the MLSS concentration in the reactor.

During all phases, the internal mixed liquor return ratio (mixed liquor return/inflow) was maintained at approximately 2, e.g. the

mixed liquor return flow is approximately 200% of the inflow rate. Different SRTs were maintained in accordance with the reactor temperatures shown in Table 1. The DO concentration in the intermittent aeration zone was controlled in the range of 0.2–3.0 mg/L during the aeration-on period using a DO controller. During the aeration-off period, a mixing device was turned on to provide necessary mixing within the intermittent aeration zone. The aeration time vs. the mixing time was 51 min vs. 45 min during the tests in Phases II and IV. In Phase VI, the aeration time vs. the mixing time was 60 min vs. 60 min.

During the performance test, the MLSS concentration and the sludge settling characteristics, as indicated by sludge volume index (SVI), were monitored. Composite influent and effluent samples were collected for water quality analysis. The concentrations of COD, SS, TN, and TP in the influent and effluent, and the effluent concentrations of ammonia and nitrate, were measured three times a week. The effluent nitrite was monitored during Phases IV and VI with external ammonium addition.

2.3. Cycling test

To better understand nitrogen and phosphorous removal during the iMLE mode operation, two cycling tests were conducted under stabilized conditions during Phases II and IV, respectively. In the cycling test, the concentrations of ammonia, nitrate, and phosphate in the intermittent aeration zone, the post-aeration zone, and effluent were monitored during one complete aeration on/off cycle. A composite influent sample was collected during the testing period for influent quality analysis. In the first cycling test, the concentrations of influent COD, SS, TN, ammonia nitrogen, and TP were 773, 281, 37.9, 22.5, and 16.9 mg/L, respectively. In the second cycling test, the concentrations of influent COD, SS, TN, ammonia nitrogen, and TP were 268, 46.0, 35.2, and 8.6 mg/L, respectively. The temperature in the reactor was approximately 20 °C and 9.7 °C during these tests, respectively, representing normal and low temperature scenarios.

2.4. Analytical methods

The analytical methods for different water quality parameters such as MLSS, COD, SS, TN, ammonia, nitrite, nitrate, and TP were described previously (Liu et al., 2012; Liu and Wang, 2012, 2013). The unit for all nitrogen species, including ammonium, nitrate, nitrite, and total nitrogen, are reported a mg-N/L.

3. Results and discussion

3.1. Sludge settling characteristics and COD removal

In the iMLE process, the anaerobic and anoxic conditions in the mixing zone, and the anoxic condition in the intermittent aeration zone, could inhibit the growth of filamentous bacteria (Metcalf and Eddy, 2003). During the entire experimental period, the average reactor temperature ranged from 8 to 25 °C (Table 1), and the

Table 1

Experimental setup (note: the hydraulic retention time (HRT) for the treatment zones, including the mixing zone, intermittent aeration zone, and post-aeration zone).

Phase	Duration (day)	Mode	Aeration: Mixing (min)	Flow (m ³ /d)	T (°C)	HRT (h)	SRT (d)	Wastewater
I	1–36	iMLE	51:45	57	23.6	9.2		Natural start-up with regular wastewater
II	37–98	iMLE	51:55	57	24.1	9.2	~10	Feeding with regular wastewater with C/N = 4.6
III	99–132	MLE	100% aeration	57	20.2	9.2	~10	
IV	133–152	iMLE	51:45	57–48–37	14	9.2–14.2	~18	Feeding with limited C/N = 2.4 wastewater
V	153–186	MLE	100% aeration	30	9.9	17.5	~35	Feeding with very low C/N = 1.6 wastewater
VI	187–223	iMLE	60:60	30	9.3	17.5	~45	

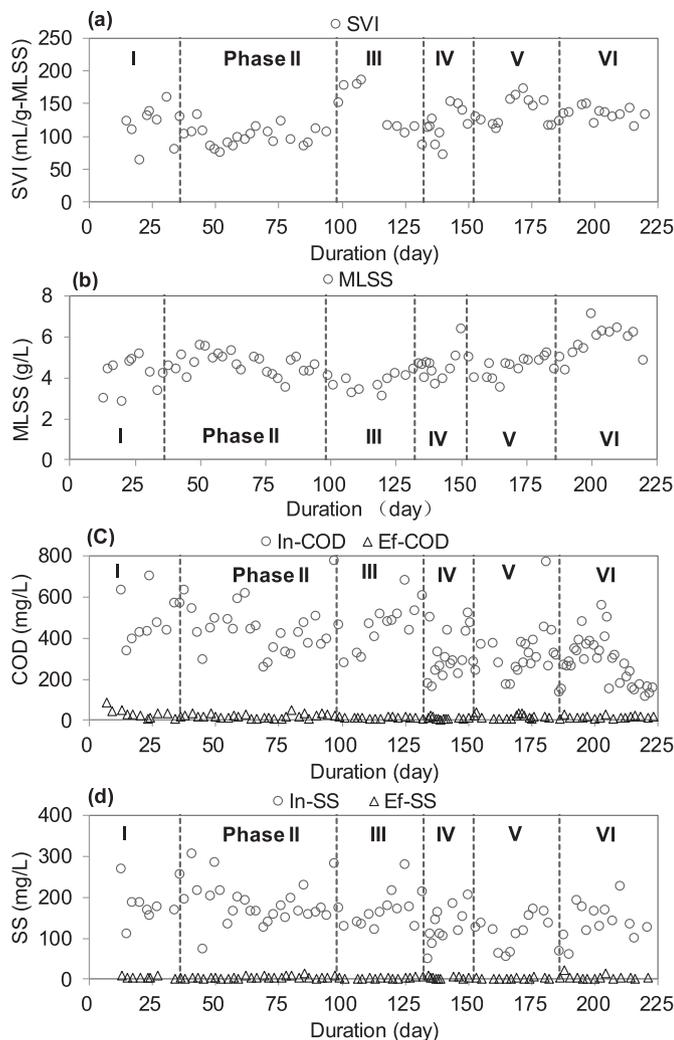


Fig. 2. (a) The SVI value and (b) the MLSS concentration for the mixed liquor in the intermittent aeration zone; the influent and effluent (c) chemical oxygen demand (COD) and (d) suspended solids (SS) concentrations.

average SVI ranged from 99 to 142 mL/g-MLSS (Fig. 2(a)). The average MLSS concentration in different phases ranged from 3.7 to 5.7 g/L (Fig. 2(b)).

As shown in Fig. 2(c) and Table 2, the average influent COD concentrations during Phases II, III, IV, V, and VI were 458 ± 140 , 451 ± 108 , 369 ± 107 , 349 ± 140 , and 279 ± 130 mg/L, respectively. Correspondingly, the average effluent COD concentrations were 20 ± 10 , 13 ± 5 , 11 ± 5 , 18 ± 10 , and 16 ± 7 mg/L, respectively. These low effluent COD concentrations indicated that intermittent aeration under the iMLE mode operation and low temperature did not impact organic matter removal. The average ratio of BOD₅/COD for the influent was determined to be 0.28 (data not shown).

From Phase II to Phase VI, the average effluent SS concentrations (unfiltered) were 4.7 ± 3.4 , 3.0 ± 1.2 , 4.3 ± 1.9 , 2.5 ± 1.1 , and 5.4 ± 4.6 mg/L (Fig. 2(d)). The SS removal efficiencies were 97–98% during the entire experimental period.

3.2. Nitrogen removal in iMLE and MLE modes

The influent and effluent nitrogen concentrations are shown in Fig. 3 and also summarized in Table 2. After the 31st day, the effluent ammonia concentration became less than 1 mg-N/L, indicating that one month was needed to achieve complete nitrification if the iMLE process started naturally (no seeding sludge addition) in the summer.

In Phase II, the influent TN ranged from 14.5 to 44.4 mg-N/L, with an average concentration of 28 mg-N/L (Table 2). Approximately 70% of influent TN was ammonia and 30% was organic nitrogen. As shown in Fig. 3(b), the effluent ammonia was consistently lower than 1 mg-N/L, with an average value of 0.3 ± 0.2 mg-N/L (Table 2). The effluent nitrate concentration ranged from 0.5 to 2.1 mg-N/L, with an average value of 1.2 ± 0.5 mg-N/L. As shown in Fig. 3(a), the effluent TN ranged from 2.2 to 4.4 mg-N/L, with an average of 3.2 ± 0.6 mg-N/L. This indicated that 89% of TN was removed. The sum of the average effluent ammonia + nitrate was 1.5 mg-N/L, indicating that both nitrification and denitrification were completed and approximately 1.7 mg-N/L of effluent TN was organic nitrogen + nitrite nitrogen. In the effluent, dissolved organic nitrogen generally ranged from 1 to 2 mg-N/L (Pehlivanoglu-Mantas and Sedlak, 2008; Parkin and McCarty, 1981), indicating that no significant nitrite accumulated in Phase II operation.

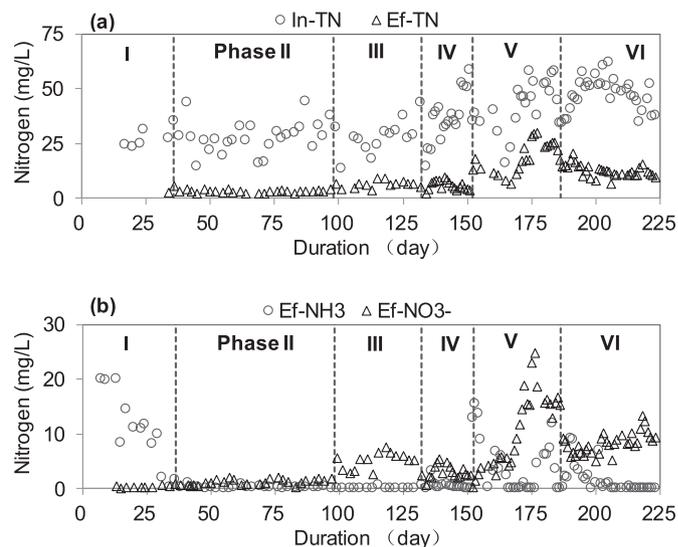


Fig. 3. (a) The influent and effluent TN concentrations and (b) the effluent ammonia and nitrate concentrations.

Table 2
Summary of the treatment performance at steady condition in different phases.

Phase	COD (mg/L)			TN (mg-N/L)			Ef-NH ₃ (mg-N/L)	Ef-NO ₃ (mg-N/L)	TP (mg-P/L)		
	In.	Ef.	% Rem.	In.	Ef.	% Rem.			In.	Ef.	% Rem.
II	458 ± 140	20 ± 10	96	28.0 ± 7.3	3.2 ± 0.6	89	0.3 ± 0.2	1.2 ± 0.5	4.3 ± 0.8	0.5 ± 0.2	90
III	451 ± 108	13 ± 5	97	27.0 ± 6.2	6.4 ± 1.5	76	0.1 ± 0.2	5.2 ± 1.5	4.2 ± 1.2	0.4 ± 0.3	91
IV	369 ± 107	11 ± 5	97	44.7 ± 9.8	4.8 ± 1.8	89	0.3 ± 0.1	2.7 ± 1.1	4.5 ± 1.2	0.3 ± 0.1	93
V	349 ± 140	18 ± 10	95	48.6 ± 7.1	22.6 ± 5.0	54	2.8 ± 3.7	16.7 ± 3.4	3.1 ± 0.9	1.4 ± 0.7	56
VI	279 ± 130	16 ± 7	94	47.9 ± 7.3	11.5 ± 1.9	76	0.3 ± 0.3	9.3 ± 1.7	2.7 ± 0.8	1.2 ± 0.4	56

In Phase III, the reactor was operated using the MLE mode, and the intermittent aeration zone was continuously aerated. The average influent COD of 451 mg/L was almost the same as that in Phase II with 458 mg/L. The influent TN ranged from 13.8 to 37.9 mg-N/L, with an average of 27 ± 6 mg-N/L, that was also similar to that in Phase II with an average value of 28 mg/L. As shown in Fig. 3(b), the effluent ammonia concentration decreased slightly, to approximately 0.1 mg-N/L, due to continuous aeration operation. However, the effluent nitrate concentration increased significantly in Phase III, ranging from 2.7 to 6.6 mg-N/L, with an average of 5.2 ± 1.5 mg-N/L. Compared to Phase II (iMLE mode), the effluent nitrate increased by more than 3 times. Due to the increase in the effluent nitrate, the effluent TN also increased to a range of 3.6–9.6 mg-N/L, with an average of 6.4 ± 1.5 mg-N/L. Although this effluent TN was still low, it was a 100% increase compared to the iMLE mode operation for this particular test. This result suggests that, compared to the MLE process, the iMLE process has similar nitrification performance, but much better denitrification performance and, therefore, much better TN removal efficiency. Better denitrification also leads to a greater oxygen recovery from nitrate, which can reduce the oxygen demand and save aeration energy (Metcalf and Eddy, 2003). Based on the recorded operational times of the blowers and mixers, we determined that the unit power use under the iMLE mode operation and the MLE mode operation was 2.65 and 3.01 kWh/kg-BOD removed, respectively. As a result, the iMLE process saved 10% of the aeration energy needed for biological reactions (aeration + mixing). The actual energy saving should be greater than this value because the MLE mode was operated at a temperature of 4 °C lower than that during the iMLE mode operation. This energy saving of the iMLE process was also contributed by a greater DO deficit at the initial stage when the aeration was turned on to switch the intermittent aeration zone from the anoxic condition to the aerobic condition.

3.3. Nitrogen removal with limited carbon source

A low C/N ratio can decrease TN removal efficiency because of the limitation of the electron donor needed for denitrification. We wanted to explore how the iMLE process performed when the C/N ratio was reduced, which may be the case for some wastewater. The system was tested with a diluted influent in Phase IV. Tap water was used to dilute the influent to decrease the BOD₅ concentration. Ammonium bicarbonate was also added to maintain a similar TN concentration in the influent as before. Therefore, the influent C/N ratio was reduced. The temperature was also significantly dropped during this Phase, making nitrification and denitrification more difficult. The results are shown in Fig. 3.

In Phase IV, the average concentration of influent BOD₅ (data not shown) and TN was 89 mg/L and 39 mg-N/L, respectively, with an average C/N ratio of 2.4. This C/N ratio was about one half of that for the regular wastewater tested in Phase II. As shown in Fig. 3(b), the effluent ammonia initially increased to above 3 mg-N/L, mainly due to the lower temperature. Then, the SRT was increased for approximately 18 days to compensate for the low temperature effect (Table 1). In the meantime, the inflow rate was reduced to 48 m³/d on the 137th day and to 37 m³/d on the 148th day. The inflow rate was lowered by reducing the tap water flow, so that the loads of BOD and TN were not reduced. After reducing the sludge wasting, the effluent ammonia concentration reduced to less than 1 mg-N/L on the 137th day. Between the 137th and 143rd days, the effluent nitrate concentration was approximately 5 mg-N/L, with a TN removal efficiency of 75%. From the 144th day, however, the effluent nitrate decreased, and the TN removal efficiency increased. From the 144th to the 151st day, the average effluent ammonia, nitrate, and TN were 0.3, 2.7, and 4.8 mg-N/L, respectively,

indicating that complete nitrification had been achieved and that 89% of the influent TN had been removed. The effluent nitrite was measured during Phases IV (with external ammonium addition), and its concentration after 147th day was always less than 0.5 mg-N/L (Table S2). Compared to the effluent quality in Phase II, the effluent ammonia concentration was at a similar level, while the effluent nitrate and TN increased by 1.5 and 1.6 mg-N/L, respectively. The slight increase in effluent TN concentration was due to the increased nitrate. These results strongly indicated that the iMLE process could achieve excellent TN removal from municipal wastewater with a limited organic source (C/N = 2.4), even when the temperature was around 14 °C.

3.4. Nitrogen removal under extremely unfavorable conditions

During Phases V and VI, the process was loaded with wastewater that had an even lower C/N ratio when the reactor temperature was 9–10 °C. Ammonium bicarbonate was added directly into the influent to increase the influent TN concentration, without tap water dilution (the BOD in the raw influent was low during Phases V and VI). With the addition of ammonium bicarbonate, the average C/N ratio was reduced to 1.6. To compensate for the adverse effect of low temperature, the influent flow was further reduced to 30 m³/day and the SRT increased to 35–45 days (Table 1). To quickly accumulate nitrifiers to handle the increased ammonia loading, the process was initially operated using the MLE mode (with continuous aeration), marked as Phase V. When complete nitrification was achieved, the operation mode was changed back to iMLE (Phase VI). The results from Phases V and VI are also shown in Fig. 3.

With the MLE mode from the 170th to 186th day in Phase V, the average influent and effluent TN concentrations were 48.6 ± 7.1 and 22.6 ± 5.0 mg-N/L, respectively, indicating that only 54% of the TN was removed. The effluent nitrate concentration was 16.7 mg-N/L. As shown in Fig. 3(b), when the iMLE mode was applied on the 186th day, the effluent nitrate concentration decreased significantly. The effluent ammonia increased after changing to the iMLE mode initially. However, it decreased to less than 1 mg-N/L after the operational DO was increased from the 1–2.5 mg/L range to the 1.5–2.5 mg/L range, starting from the 202nd day. This indicated that the iMLE process could still achieve complete nitrification when both the SRT and the DO were sufficient. Under a relatively stable condition from the 202nd day in Phase VI, the average influent and effluent TN concentrations were 47.9 ± 7.3 and 11.5 ± 1.9 mg-N/L, respectively, with a removal efficiency of 76%. Therefore, the iMLE process had improved TN removal by approximately 20% compared to the MLE process when treating the same low C/N ratio wastewater.

3.5. Phosphorus removal

As shown in Table 2, during Phases II, III, and IV, the average influent TP were 4.3, 4.2, and 4.5 mg-P/L, respectively. Considering the average influent BOD₅/COD ratio of 0.28, the average C/P ratio, as indicated by BOD₅/TP, was approximately 29 for the influent. As shown in Fig. 4, the effluent TP was consistently low during Phases II, III, and IV, with average concentrations of 0.45 ± 0.22 , 0.39 ± 0.29 , and 0.31 ± 0.08 mg-P/L, respectively. In Phases II and IV, the TP removal efficiency was 90% and 93%, respectively. This high TP removal efficiency reflects the technology limits of biological processes without chemical addition and filtration.

During the MLE mode operation (Phase III), surprisingly, the TP removal efficiency was similar to that in Phase II with the iMLE operation. Guerrero et al. (2011) also found that, after changing a pilot-scale plant from the A²O operation to MLE operation, no detrimental effect on phosphorus removal was observed. However,

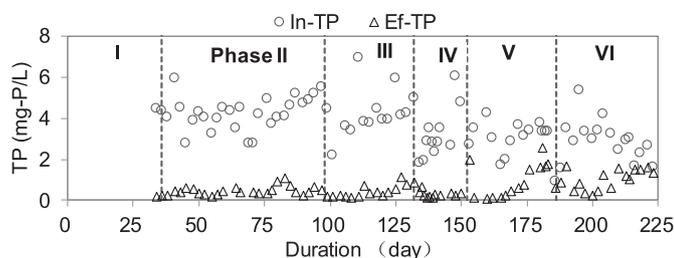


Fig. 4. The influent and effluent TP.

this may last for only a short time period. The effluent TP was approximately 0.17 mg-P/L from the 99th to the 111th day and ranged from 0.22 to 0.72 mg-P/L between the 113th to the 122nd day, and from 0.55 to 1.11 mg-P/L between the 125th to the 129th day. This indicated that, after a longer operation time using the MLE mode, the PAOs could be washed out through sludge wasting, gradually reducing the TP removal efficiency.

After changing to the MLE operation mode on 155th day while feeding with very low C/N ratio wastewater (Phase V), low effluent TP concentration (<0.5 mg-P/L) was initially detected. However, its effluent concentration increased gradually from 169th day. From day 170th to 186th day, the averaged effluent TP concentration was 1.4 mg-P/L with removal efficiency of 56%. In Phase VI, the influent and effluent TP under steady condition were 2.7 and 1.2 mg-P/L, respectively, with a removal efficiency of 56%. This suggests that the removal efficiency of TP was reduced significantly, possibly through the combined effects of low temperature, high nitrate concentration, and long SRT. Low temperature and high nitrate concentration made the mixing zone less ideal to cultivate PAOs, and long SRT reduced total biomass production.

4. Discussion

When treating a raw municipal wastewater with C/N = 4.6 using the iMLE process, the effluent ammonia, nitrate, and TN

concentrations were 0.3, 1.2, and 3.2 mg-N/L, respectively, and the TN removal efficiency was approximately 90%. Both nitrification and denitrification were completed. When treating carbon limited wastewater (C/N = 2.4), the iMLE process could still reduce the TN to approximately 5 mg-N/L. For both types of wastewater, the averaged TP concentrations in the unfiltered effluent were less than 0.5 mg-P/L with removal efficiencies of approximately 90%. These low effluent TN and TP concentrations reflect the limits that a biological process can achieve if no chemicals (such as an external carbon source and phosphorus precipitation agents) are used. The combination of intermittent aeration and internal mixed liquor return played a critical role in the success of the iMLE process to achieve enhanced nutrient removal.

Fig. 5 shows the cycling test results when treating regular wastewater for one complete aeration on/off cycle. As shown in Fig. 5(a), when the aeration device was turned on, the intermittent aeration zone became aerobic. As shown in Fig. 5(b), the initial ammonia concentration in the intermittent aeration zone was approximately 3.8 mg-N/L, which decreased to less than 1 mg-N/L in just 30 min. Based on the ammonia curve shown in Fig. 5(b), the apparent ammonia oxidation rate was 6.9 mg-N/L·h in the first 30 min. For the pilot scale reactor, the HRT in the intermittent aeration zone was only 6 h at an inflow rate of 57 m³/d (15,000 gpd). Because the influent TN during the test was 38 mg-N/L, it brought in 3.2 mg-N/L of additional ammonia into the intermittent aeration zone in 30 min. Therefore, the actual ammonia oxidation rate was about 10.1 mg-N/L·h, and the specific oxidation rate was 2.4 mg-N/L·h·g-MLSS. As a result of nitrification, the nitrate concentration increased at a rate of 4.2 mg-N/L·h in the first 30 min (Fig. 5(b)). During the aeration-on period, nitrate was still removed in the preceding mixing zone because some mixed liquor was continuously returned to the mixing zone. Consequently, the nitrate concentration increased at a rate slower than that of ammonia oxidation. Meanwhile, the soluble phosphate carried into the reactor by the influent and that was released from the PAOs during the aeration-off period was removed from the solution by PAOs. As shown in Fig. 5(b), the phosphate concentration decreased to 0.2 mg-P/L in just 17 min after the aeration device was turned on.

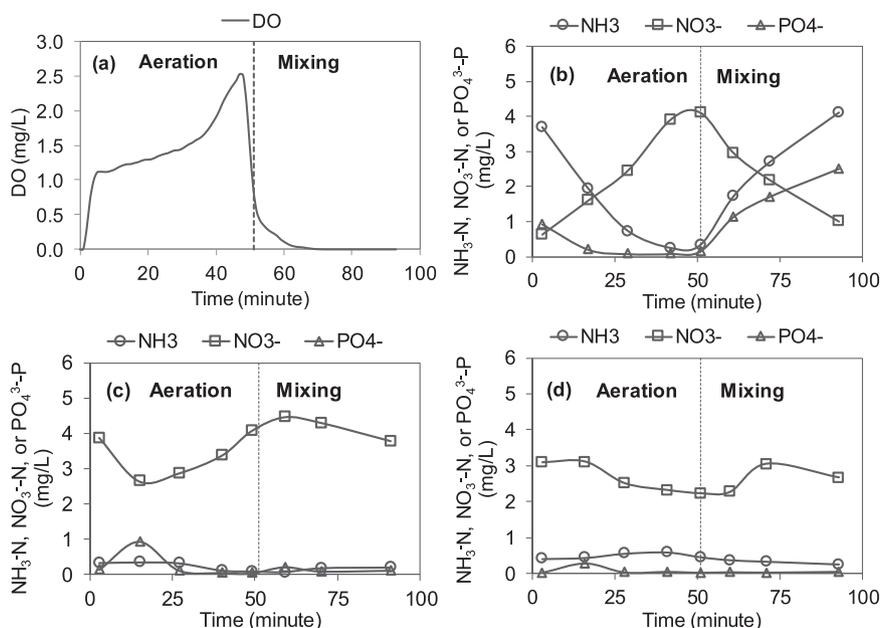


Fig. 5. Cycling test results in Phase II. (a) DO concentration in the intermittent aeration zone. The concentrations of ammonia, nitrate, and phosphate in the intermittent aeration zone (b), post-aeration zone (c), and effluent (d).

As shown in Fig. 5(c) and (d), during the aeration-on period, both the ammonia and phosphate in the post-aeration zone and the effluent were very low.

When the aeration was off, the DO in the intermittent aeration zone dropped to zero within 15 min (Fig. 5(a)). The intermittent aeration zone became anoxic due to the presence of nitrate. Because of the dilution, the accumulation of ammonia in the intermittent aeration zone during the aeration-off period was insignificant, with the maximum value of 4.75 mg/L, assuming effluent ammonia was negligible. As shown in Fig. 5(b), 4 mg-N/L of ammonia had accumulated at the end of the aeration-off period because some ammonia was carried out of the intermittent aeration zone by the continuous flow. The ammonia carried to the post-aeration zone was converted to nitrate due to the continuous aeration. This is validated by Fig. 5(c) and (d), which show that the ammonia concentrations in the post-aeration zone and in the effluent were consistently low. With aeration being off, the nitrate was reduced to nitrogen gas within the intermittent aeration zone. As shown in Fig. 5(b), the nitrate concentration in the intermittent aeration zone decreased at a rate of 4.2 mg-N/L·h. At the end of the aeration-off period, the nitrate concentration was less than 1 mg-N/L, indicating that 45 min was long enough to accomplish full denitrification. Note that, if the HRT in the intermittent aeration zone is extended, the accumulation of ammonia during the aeration-off period will decrease due to more dilution, which will reduce the TN concentration in the effluent of the intermittent aeration zone. The post-aeration zone and the clarifier worked as buffers. As a result, the effluent nitrate concentration during one complete aeration on/off cycle was relatively consistent, ranging from 2 to 3 mg-N/L, as shown in Fig. 5(d). As shown in Fig. 5(b), when the aeration was turned off, the phosphate in the intermittent aeration zone was quickly released, indicating that denitrification and phosphate release occurred at the same time. In the post-aeration zone, however, the released phosphate was removed again. Consequently, the phosphate in the effluent was consistently low, as shown in Fig. 5(d). The low nitrate in the return mixed liquor (<2 mg-N/L) from the intermittent aeration zone

could make the preceding mixing zone under an anaerobic condition. This will cultivate PAOs for phosphorus removal.

When treating wastewater at a very low temperature with a very low C/N ratio of 1.6 in Phase VI, the removal efficiency of the TN and TP decreased, from 90% to 75% for TN and from 90% to 56% for TP. These results indicated that the iMLE process was losing TN and TP removal capability. Another cycling test was carried out on the 219th day in Phase VI. Fig. 6 shows the major operating and water quality parameters in the intermittent aeration zone, the post aeration zone, and the effluent. The phosphate concentration remained constant in all zones during a complete aeration on/off cycle. No phosphate was released or removed during the aeration off or on period. The average TP concentration in the intermittent aeration zone, post-aeration zone, and the effluent were 1.70 ± 0.10 , 1.61 ± 0.11 , and 1.74 ± 0.18 mg-P/L, respectively, indicating that the biological phosphate removal process did not occur. In the intermittent aeration zone, nitrate concentration at the end of the aeration-off period was still 8.8 mg-N/L; therefore, the denitrification was not complete. Such a high nitrate concentration made it impossible to turn the preceding mixing zone to anaerobic during the aeration-off period. In addition, the low temperature and the extended SRT contributed to the low phosphorus uptake. Therefore, the TP removal efficiency decreased.

When treating regular strength municipal wastewater, the intermittent aeration exposed the activated sludge in the iMLE process to alternating anaerobic and anoxic conditions within the mixing zone, and alternating anoxic and oxic conditions within the intermittent aeration zone. This promotes the accumulation of PAOs for TP removal and denitrifiers for TN removal. When the aeration device was turned on, nitrate accumulation in the intermittent aeration zone was the concern, as shown in Fig. 6(b). If this aeration-on period lasted for too long, dilution would not be sufficient to yield a low nitrate concentration in the intermittent aeration zone, and the result would be a high effluent TN concentration. When the aeration was turned off, the concern was ammonia accumulation within the intermittent aeration zone. A small post-aeration zone would not be sufficient to remove the

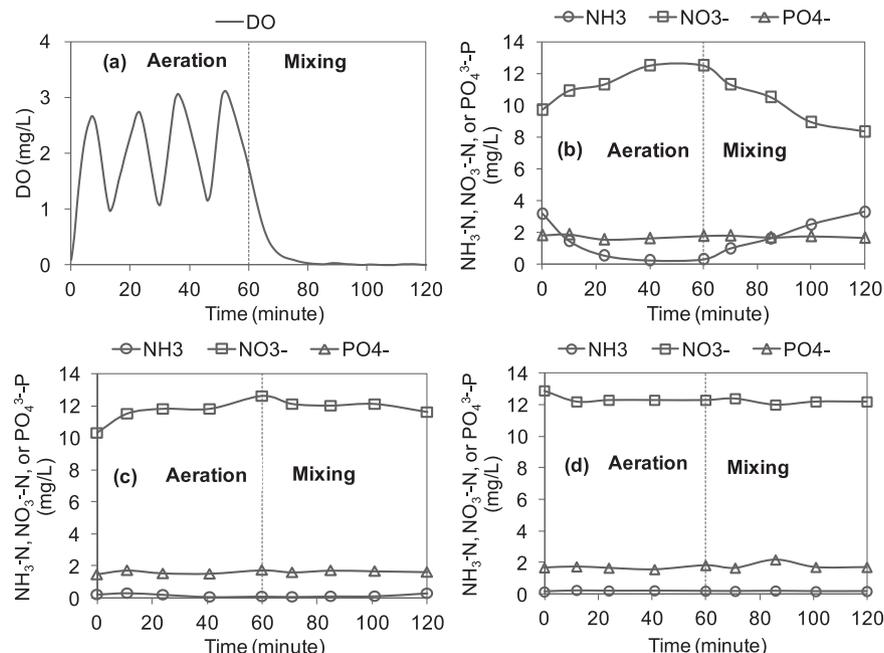


Fig. 6. (a) DO concentration change in the intermittent aeration zone during one cycle of aeration on/off in Phase VI. The changes in the ammonia, nitrate, and phosphate in the (b) intermittent aeration zone, (c) post-aeration zone, and (d) effluent, during one cycle of aeration on/off in Phase VI.

residual ammonia, which would show up in the final effluent. Additionally, if the aeration-off period was too long, the PAOs would be over exposed to an anaerobic condition and cause secondary phosphorus release, which would damage the PAOs. Therefore, the duration of the aeration on- and off-periods should be balanced to avoid high ammonia and nitrate concentrations in the final effluent, which also depend on operating conditions such as temperature, SRT, and wastewater strength. The cycling test data shown in Fig. 5 indicate that, under a temperature of approximately 20 °C, 51 min of aeration vs 45 min of mixing had achieved these goals. With a low temperature of approximately 9.5 °C, it would have taken longer to deplete the residual oxygen after aeration. In addition, the activities of nitrifiers and denitrifiers were also reduced. Therefore, the aeration on and off durations for each cycle should be extended.

Based on conventional pathways that are used in the A²O and UCT processes, the key to an advanced wastewater treatment process to remove both TN and TP is to create cycling anaerobic, anoxic, and oxic conditions through different unit processes to effectively utilize the limited organic matter in wastewater. However, for the A²O and UCT processes, part of the nitrate in the mixed liquor of the final aerobic tank does not go through the anoxic condition for denitrification. As a result, the TN removal efficiency would generally be about 70%, with an effluent concentration of higher than 8 mg-N/L (Metcalf and Eddy, 2003; Vaiopoulou and Aivasidis, 2008; Lee et al., 2009; Wang et al., 2012). The practical effluent TN concentration could easily be greater than 15 mg-N/L, exceeding the Class 1A standard set by China EPA (2002). If TN needs to be reduced further, a post-anoxic zone with external carbon addition must be added, such as the Bardenpho process. The addition of external carbon would increase operation cost significantly. The step-feed anoxic-oxic process (Tang et al., 2007; Zhu et al., 2009) could achieve excellent TN nitrogen with removal efficiency greater than 90%, while the removal of TP was insignificant due to the absence of alternating anaerobic-aerobic condition. The modified step-feed processes, such as UCT step-feed process (Ge et al., 2010, 2011; Majdi Nasab et al., 2016), could achieve comprehensive removal for both TN and TP with their effluent concentrations less than 5 and 0.5 mg/L, respectively. However, these step-feed processes are complicated to operate and the control of the flow distribution is challenging (Chen et al., 2016). The AAA process could complete nitrogen removal with a long operation HRT (Habermeyer and Sánchez, 2005). However, a longer HRT indicated greater capital cost. Kim et al. (2014) developed a modified A²O process by applying intermittent aeration and adding a post-aeration zone, termed ABA² process. Although this process provides some energy savings, it removes only 66–73% of TN with effluent concentrations of 7.5–12 mg-N/L, which is similar to the conventional MLE and A²O process. Compared to the iMLE process, this ABA² process has a lower TN removal efficiency and employs one more functional zone so it is more complex to operate.

Compared to the above BNR processes, the iMLE process, which integrates intermittent aeration and the internal mixed liquor return, could easily improve TN efficiency by 15% and also achieve advanced TP removal without enlarging the reactor footprint, adding additional unit processes, and using any chemicals. Meanwhile, the operation of the iMLE process is very similar to the proven MLE process, except that an alternating aeration control and a mixing system are added to the majority of the MLE's aeration zone, and leave the end portion of the aeration zone continuously aerated. With this change, the process not only significantly improves TN removal, but also improves TP removal and saves aeration energy. The aeration on and off durations can be easily adjusted (manually or automatically), to achieve the best results under various influent quality and environmental conditions.

5. Conclusions

The novel iMLE process, which implemented intermittent aeration to the regular MLE process, could achieve enhanced TN and TP removal without using any chemicals. When treating a regular and a carbon-limited municipal wastewater (C/N ratio ≥ 2.4), the TN and TP removal efficiencies were approximately 90%, and their concentrations in the unfiltered effluent were generally less than 5 and 0.5 mg/L, respectively. When treating a wastewater with a very low C/N ratio of 1.6 under a very low temperature of 9.5 °C, the iMLE process still removed 76% of the TN. The iMLE process also saved 10% of aeration energy compared to the conventional MLE process. The combination of intermittent aeration and internal mixed liquor return played a critical role in the success of the iMLE process to achieve enhanced nutrient removal and energy saving.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jclepro.2017.08.017>.

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