INCREASING THE RATE OF AMMONIA NITROGEN REMOVAL FROM WASTEWATER BY BAFFLED ALGAL REACTORS WITH RECYCLE LINE UNDER LABORATORY CONDITIONS

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NÂNG CAO HIỆU QUẢ XỬ LÝ AMMONIA NITROGEN TRONG NƯỚC THẢI THÔNG QUA THÍ NGHIỆM TRONG PHÒNG VỚI DÒNG TÁI SỬ DỤNG

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1. Introduction

Domestic wastewater contains a number of nitrogen-containing compounds and ammonia nitrogen is the one of the most challenging compounds to remove from wastewater. Hence, it is a very important to eliminate the substance before this wastewater returned to bodies of water. Biological treatment to take away nitrogen concentrations from wastewater with the activated sludge system, by applying nitrification and denitrification is less expensive and more effective. Thus, it has been used as a standard method worldwide to achieve low nitrogen emissions [1], [2]. Traditional and novel biological nitrogen elimination technologies are being reviewed. Recent studies dealing with temperature, dissolved oxygen, nitrate concentration, salinity, pH or free ammonia concentration as factors affecting the ammonia nitrogen removal efficiency have also been incorporated with biological treatment processes [3], [4].

Techniques to eliminate the majority of pollutants from these effluents are essential in developed countries and are becoming increasingly important from an environmental and human health point of view in the developing countries [5]. In order to achieve high levels of ammonia nitrogen removal, the baffled algal reactors with recycle line setup had been implemented. The modifications setup for the study were based and followed on the design the process model of Wuhrmann, Ludzack-Ettinger, and Modified Ludzack-Ettinger process-MLE as cited by [6] for single sludge nitrogen removal, for the carbon oxidation and nitrification system.

The combined-modified system shows the interesting advantages of the system compare with the conventional process, such as air supplies, enhanced residual substance treatment by returning one more time to the main reactors, and others [7]. From the point of view, the combined system can achieve high removal efficiency of ammonia nitrogen, on order 86-90%. Moreover, the modified system is providing an impressive reduction of COD, $BOD₅$ from influent. It also shows an interesting advantage in comparing with another system, such as enhanced the residual substance treatment, etc. [8].

The research objective focus on the behavior of baffled algal treatment reactors intend to obtain the highest ammonia nitrogen removal efficiency as well as COD, BOD₅ removal from wastewater without settable particles.

2. Research materials and methods

2.1. Material and treatments construction

The research had been done since 2015 with municipal wastewater without settle able particles taken from the effluent of primary treatment unit of wastewater treatment plant in Berlin. The experiment was established in three algal treatment reactors: two setups for the baffle's contributions in different design (upward and downward flow: **T1**; sideward flow: **T2**), the last setup was a Reference treatment reactor (without baffle: **T3**). All designs were combined with continuing flow. The system was operated 180days.

In the laboratory's condition, artificial lights board with photon flux of 0.72 x 1020 photons/m².s ($120 \mu E/m^2$.s), yielding 8.000lux, the intermittent light simulations 12:12 hours and were set up to provide light to the algae growth in the three reactors.

The three baffled algal reactors had no aeration system provided. Therefore, the oxygen concentration was completely generated by the algal via photosynthesis mechanisms. A mixed fresh cultural algal-bacterium is used for each reactor (fresh-weight of 0.5 g /l). There was no CO₂ addition to the reactors. The schematic of algal baffled reactors is shown in the Figure 1.

With the purpose of increasing the efficiency of substances removal from wastewater, a recycling line was used in order to obtain an optimal efficiency of ammonia nitrogen, COD, $BOD₅$ removal, which won't be possible to obtain by using the traditional methods due to the conventional nitrogen removal process requiring a large-scale renovation of the existing facilities. The recycle flow was 0.7 l/h (activated by pumping back 12 hours per day).

Figure 1. *The sketch of experiment implementation*

2.1. Material and sampling location

Weekly grabbed samples were analyzed in order to observe the concentrations trend throughout the week of the algal reactors. $NO₃$ -N, BOD₅, COD and NH₄⁺-N were analyzed by cuvette-test (Hach Lange). pH and oxygen concentration are measured by Electronic handout equipment. Algae specific growth inhibition test were carried out in Laboratory SIWAWI-Technical University of Berlin, the entire test followed by the principles of guideline for the testing of chemicals [9] and [10]. The opening conditions were described in the Table 1.

Table 1. *Operating conditions*

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Flowing rate: - Influent	$22.50 \frac{\text{1}}{\text{d}}$	
- Effluent	$22.30 \frac{1}{d}$	
Water evaporation	0.201/d	
- Treatment reactors	3 separated reactors	
Dimension	H: 60cm, W: 65cm; L: 85	
Capacity (effective capacity)	235 litres (in volume)	
- Collectors (3 separated units)	40 litres	
Hydraulic retention time (approximately)	10 days	
Artificial lights condition (light intensity)	Photon flux of 0.72×1020 photons/m ² s ($120 \mu E/m^2 s$), yielding 8.000lux	
Intermittent light simulations (day/night)	12 hours light	
Wastewater discharge type	Raw wastewater without settle particles	
Temperature on surface of the reactors	22° C (on average)	
Plastic baffles areas (reactors T1 and T2)	2.4m ²	

2.2. Sample anlalysis methods

- Oxygen concentration, pH media and temperature in wastewater: the oxygen concentration, dissolved oxygen, pH and temperature in the reactors and samples were measured by electronic equipment HQ40D with equivalence sensors.

- COD: According to ISO 15705, COD is the volume of oxygen equivalent to the mass of potassium dichromate that reacts with the oxidizable substances in the water under the working conditions of the method. COD is measured by adding 2ml of fresh sample into the COD cuvette. The reaction time is about 15 minutes of cooking time at 170°C by Dr. LANGE HT200S (in reality, it needed two hours from heating up to cooling down the cuvette). The sample must be homogenized before the analysis is performed. The digestion solution of COD cuvette was prepared by adding $K_2Cr_2O_7$ and concentrated H₂SO₄. Colorimetric measurements of COD were made using Photometer DR5000 with different wavelengths (348, 448 and 605 nm) for the different ranges of COD from 15 to 1.000 mg/l. The filtrated COD with membrane filter with size 0.45 µm (Millipore) was used to calculate the fractions of COD.

- NH₄⁺-N: concentration was determined by using the Hach Lange DR5000 Spectrophotometer at wavelengths 550-694nm and adequate cuvette test kits for NH₄+-N: LCK 302, 302, 305, 304 to obtain the NH⁴ + -N concentration in the wastewater. According to DIN 38406 E5, the principle to determining NH₄⁺-N in wastewater by using Hach Lange Cuvette test is dependent on the rate of reaction of Ammonium ions at the pH value of 12.6 with hypochlorite ions (created in an alkaline medium by hydrolysis of dichloroisocyanuric acid ions) and salicylate ions in the presence of sodium prusside-sodium as a catalyst for the blue dye indophenol blue. A filtered sample about 0.2–5 ml in volume, filtered via a membrane filter (0.45 μ m), is added to the cuvette and allowed to react for 15 minutes

- NO₃ -N: the analysis principle is based on the reductant of nitrate to nitrite by added hydrazine. The nitrite then undergoes diazotization with sulphanilamide and azo coupling with N-naphtyl-ethylendiamindihydrochlorid (NED) which is measured photometrically at 370-546 nm with cuvette test kits for LCK 339 (Hach-Lange 1989; DIN 38 406 E5; DIN 38 405 D9).

3. Results and Discussion

Table 2 describes the characteristics of raw wastewater without settle-able particles in the influent to the reactors, as well as the water quality at the effluent of each reactor. The determination of load removal efficiency was done following the calculation methods described in heading 2- Research materials and methods. The results show that the reactor T1 and T2 obtained high efficiency of nitrogen removal as well as COD, BOD₅ removal efficiencies. It also indicates to higher oxygen concentrations being produced in the baffled algal reactors in comparison with the un-baffled reactor.

Basically, in water, nitrogen exists in the form of NH_4^+ , NH_3 , NO_3 and NO_2 . The term ammonia refers to two chemical species which are in equilibrium in water: $NH₃$ (un-ionized) and NH₄⁺ (ionized). Ammonia's toxicity is primarily attributable to the un-ionized form (NH₃), as opposed to the ionized form (NH_4^+) . In general, the higher the pH, the more the NH_3 and the greater the toxicity. When dissolved in water, normal ammonia (NH3) reacts to form an ionized species called ammonium (NH₄⁺).

$$
\text{org. N (e.g. H2N-CO-NH2)} + H2O \rightarrow NH4+ + OH-
$$

NH₃ + H₂O \leftrightarrow NH₄⁺ + OH⁻

A nitrite in water is mostly produced by bacteria of the genus *Nitrosomonas*. Nitrite is less toxic than ammonia but is still toxic [11]. High levels of nitrite can kill many aquatic organisms. Fortunately, a further nitrification reaction can occur:

Noted: - pH(): unitless*

-This results are obtained from owned study)

Using natural algal material to remove nutrients such as ammonia and ammonium nitrogen from wastewater is not a new idea and has been applied in warmer climates. Several projects have improved and developed the techniques for producing algae in terms of nutrient removal capacity. The basis of this method are the algae as photosynthetic, autotrophic organisms that can assimilate nutrients from the water to use it for their biomass growth and oxygen production (N/biomass average 8%, and 0.16-5% for phosphorus according to [12], [13].

Some reports proved that algal growth could remove up to 90% of the phosphorus or nitrogen from wastewater [14]. In biological treatment reactors, algae may provide heterotrophic organisms in secondary treatment with oxygen.

3.1. Ammonium nitrogen (NH4+-N) removal efficiency

3.1.1 Ammonium nitrogen (NH4+-N) removal trend

The ammonia nitrogen concentrations in the influent and the effluent of the three reactors were monitored and are shown in the Figure 2. The figure also shows the results obtained from three different reactors and under the different light saturation conditions.

The ammonia nitrogen concentration discharged into treatment reactors was 63 mgNH₄+-N/l, approximately. When 12 hours light conditions were applied, the ammonium nitrogen concentration in the effluent of the reactor T1 was approximately 6 mgNH $_4$ ⁺-N/l and for that of T2 was 7 mgNH₄⁺-N/l during the entire study period, except for the reactor T3, where NH₄⁺-N was 30mg/l.

Weekly sample analysis also pointed that about 90% of the influent ammonia nitrogen was removed by the reactor T1, and 88% by the reactor T2 (a maximum removal rate of 99% of ammonia was obtained for the reactors T1 and T2). The reactor T3 achieved a removal of only 53% of ammonia nitrogen from the influent. Based on the obtained result under 12 hours lighting conditions, it can be pre-concluded that both reactor T1 and T2 have performed better ammonia removal than reactor T3 (without baffles).

The concentration of ammonia in the reactor T3 was measured by quick-test from 02.07 to 17.09.2012 to roughly get the range of the ammonia concentration. These results were not considered for the evaluation. Under the same conditions (provided light, temperature), the ammonia nitrogen removal efficiencies in the baffled reactors T1, T2 were higher than T3.

Figure 2. *NH⁴ + -N mg/l measured at the influent and effluent of the algal treatment reactors*

3.1.2. Ammonia nitrogen (NH4+-N mg/l) efficiency

As the results shown in the Figure 3, $NH₄⁺-N$ removal efficiency of approximately 90% of NH_4 ⁺-N from the influent could be achieved with loading rates of around 5-7 gNH₄⁺-N/(m³·d). The relative correlation in both baffled algal reactors were $R²= 0.67$ -0.68. There was no observation number of removal efficiency at loading rates over 7 gNH_4^+ -N/(m³·d).

From the Figure 3, it could also be seen that the optimum loading rate is found to be 5-7 gNH_4^+ -N/(m³ \cdot d). In this case, the trend of log₁₀ was used to evaluate the results. The trend shows a better correlation between the evaluated results and the theoretical results than the linear or exponential trend.

In the research about removal of nutrients in various types of constructed wetlands, Udo Wiesmann *et al.,* found that the processes that affect removal and retention of nitrogen during wastewater treatment in constructed wetlands (CWs) include NH₃ volatilization, nitrification, denitrification, nitrogen fixation, plant and microbial uptake, ammonification, nitrate reduction to ammonium, etc. Removal of total nitrogen in this study varied between 40 and 55% with removed load ranging between 250 and 630 gN/ $(m^2 \text{yr})$ depending on CWs type and inflow loading [15].

Brazil described the performance and operation of a rotating biological contactor in a tilapia recirculating aquaculture system. The system obtained an average TAN-total ammonia nitrogen areal removal rate of about 0.42 g/m^2 day [7]. For three different applied filter medium types in commercial farms and for a range of hydraulic surface loading conditions, the highest observed TAN areal removal rate for a trickling filter was 1.1 gTAN/ $m²$ day, with an average TAN areal removal rate of 0.16 g/m^2d [16]. Lyssenko and Wheaton reported total ammonium nitrogen areal removal rates of 0.64 g/(m²day) [17]. Table 3 shows the comparison of removal efficiency NH₄⁺-N under 12 hours light conditions by treatment reactors with other results.

3.1.3. Nitrogen mass balance in the baffled algal reactor (T1):

Based on result obtained from the calculation, the nitrogen mass balance in this study shows a nitrification rate of 6.1 mg/(l.d), a denitrification rate of 5.5 mg/(l.d) and an ammonia nitrogen assimilation efficiency of 79%, the culculaed result of nitrogen mass balance shows in Figure 4.

Figure 4. *Nitrogen mass balance in the baffled reactor T1*

3.2. COD remonal

3.2.1. COD removal trend

The measurements of COD at the influent and the effluent of the three experiments at the different measurement times are shown in the Figure 5. The average COD measured at the influent was 594 mg/l. The average value of COD was 82mg/l (min: 44 mg/l, max: 143 mg/l) at the effluence of the algal and 57 mgCOD/l at the effluence of the duckweed experiment (min: 33 mg/l, max: 124 mg/l). The COD decreased about 86% in the algal and 91% in the duckweed treatment, whereas the COD load was approximately $70 \text{ gCOD/(m}^3 \cdot d)$.

Figure 5. *COD measured in the influent and at the effluent of the experiments.*

3.2.2. COD removal effficiency

The Figure 6 shows the variation of COD in the influent and at the outflow of the different reactors at the different measuring times under 12 hour lighting conditions. The trend of log_{10} was used to detect a better trend for the evaluated result with the theoretical results.

Figure 6. *The comparison of the COD removal and COD load into the reactors*

An average of 124-127 mg COD/l at the effluence (unfiltered samples) of the reactor T1 and T2 was measured during the entire period of the study. The high removal rates of COD when COD load rates were 60-78 $g/m³d$ were obtained from the reactor T1, with the relative correlation factors being $R^2=0.91$ and $R^2=92$ for the reactor T2. Meanwhile, the reactor T3 achieved only 78% COD from the influent. The relationship correlation factor of the reactor T3 was approximately $R^2=0.77$.

It obviously shows that for the reactors T1 and T2, the removal efficiency of COD is closer to the theoretical value than that for the reactor T3. Lower total COD removal in the reactor T3 was due to the absence of baffles. The mixed algal culture here does not have enough ability to remove COD. Moreover, the lack of the baffles (reactor T3) could make a shortcut flow from inlet to outlet. However, the results from the three reactors showed, there were slightly different of removal efficiencies of COD between them.

Ma *et al.,* as cited in Breisha found that approximately 82% of COD was removed by a benchscale continuous flow system with Terramycin crystallization solution [3]. Gálvez *et al.* indicated that heterotrophic denitrifying bacteria depend on the type of carbon sources while the C/N ratios were 2.5, 1.08 and 1.1 for sucrose, ethanol and methanol assays, respectively [18].

In comparison with other process, it could indicate that the COD removal efficiencies of the reactor obtained high efficiency. The comparison of the current study and other results obtained from different processes is describes in the Table 4 below.

3.3. BOD⁵ removal

3.3.1. BOD⁵ removal trend

The average BOD_5 mass loading of the system from feeding tanks was 284 mg $BOD_5/(1 \cdot d)$. In algal experiments, the effluent $BOD₅$ was 20 mg $BOD₅/l$ (min: 6 mg/l, max: 62 mg/l) and 21 mgBOD5/l was detected in duckweed experiment (minimum value of 9 mg/l, maximum: 70 mg/l). Meanwhile, the reference treatment achieved only 86.7 mg $BOD₅/l$ during the entire length of study (Figure 7). It is important to recognize that the performances of the reactor T1 and T2 in the systems are working well with respect to the BOD₅ removal with long photoperiods and are reduced to an efficiency of 40-45% for short photoperiods.

Figure 7. *BOD⁵ (mg/l) in the influent and at the effluent of the three reactors*

3.3*.2. BOD⁵ removal efficiency*

The average BOD₅ mass load rate into the reactors was 27 gBOD₅/(m³d). In the reactors T1 and T2 with light provided for 12 hours per day, the $BOD₅$ was detected to be about 30-37 mgBOD₅/l, respectively. Meanwhile, in the reactor T3 about 102 mgBOD_5 /l at the effluent was achieved. The results showed that the performances of treatment reactor T1 and T2 are working well with respect to the BOD_5 removal. The effluents of BOD_5 concentration in the baffled reactors showed that the highest organic matter removal occurred in the system where the baffles contributed to the reactors. The BOD_5 removal efficiency in the reactor T1 and T2 was up to 89% when 12-hour light condition was saturated with a loading rate of 30-38 $g/(m^3)d$). The efficiency correlation factor of the reactor T1 was $R^2=0.9$ and T2: $R^2=0.8$ (see in the Figure 8).

There is no observation for removal efficiency when the loading rate was more than 38 g $BOD₅/(m³d)$. The lowest performance of $BOD₅$ removal was noted in the reactor T3, where only 62% efficiency of BOD₅ removal from the influent was obtained. The BOD₅ concentration was generally corresponding to the aeration conditions and oxygen production of algae. The linear trend was used to provide a better comparison between evaluated results with theoretical result than the result obtained from the log_{10} or exponential trend.

High removal efficiencies of COD and $BOD₅$ in the reactors T1 and T2 could be attributed to faster algal development compared to the reactor without baffles. High $BOD₅$ and COD removal rates can be obtained under suitable temperature and for high biomass production by the algae [19]-[22].

The reason of the $BOD₅$ degradation in the treatment reactor due to more attaching surface and high oxygen concentration has been reviewed in the part of the experiment descriptions. The result of this research agrees with [23], [24]. They are prevailed that high $BOD₅$ removal efficiency (46-50%) can be achieved in facultative conditions. Constable *et al.*, obtained the same result when they studied a pond without baffles and concluded that more efficient nitrification could be obtained from ponds with an attachment surface than from the pond without baffles [25]. Harrison and Daigger pointed that the ammonia removal efficiency in trickling filters could reach high values when BOD loading of BOD_{bubble}3.4-3.6 kg/(m^3 d) was present [26].

Figure 8. *Comparison of BOD⁵ removal with BOD⁵ load into the reactors*

The $BOD₅$ removal efficiencies of this research were lower than the removal efficiency obtained from advanced activated sludge process 98% BOD₅ (DWA 2011), 85-93% trickling filter with low standard rate [27], [28], [4]. Table 5 shows the comparison the result of $BOD₅$ removal efficiencies in this study with several the results obtained from research using algae for nutrient removal from wastewater. Table 5 below shows the comparison of the BOD₅ removal efficiency under 12 hours light conditions by treatment reactors with another result*.*

Table 5. *Comparison of BOD⁵ removal efficiency under 12 hours light conditions by treatment reactors with another result*

Removal efficiency % BOD5		System
Alaerts et al. (1996) [23]	95-99	Full-scale duckweed treatment plant
Zirschky and Reed (1988) [24]	80	Algal and duckweed systems
Zimmo (2003) [25]	85	Algal based waste stabilization pond
Markou and Georgakakis (2011)	90	Blue-green algae reactors with agro-industrial wastes
$[26]$		and wastewaters
von Sperling et al. (2005) [4]	80-90	Constructed wetland
von Sperling et al. (2005) [4]	< 50	USAB
DWA (2011)	>98	Activated Sludge process
Own study	89	Vertical baffled algal reactor
	86	Horizontal baffled algal reactor

Lower values of produced oxygen could be observed in reactor T3 in comparison to the reactors T1 and T2. It may be the result of lower diffusion of oxygen into the water, low space for the attached algae and shortcut flow, leading to reduced photosynthetic activity, and thus limited oxygen production.

The biomass in reactor T3 is expected to play a minor role in $BOD₅$ removal due to lacking space provided by baffles. Apparently, the growth rate of algae in baffled reactors is faster than in reactor T3, and can increase their biomass in a shorter time. Zimmo (2003) suggested that an increased organic loading into the system can improve the algae's organic removal performance [29]. However, the surplus biomass may be lost through the outflow or may contribute to the sedimentation of the die-off algae [30]. Lai and Lam (1997) also found the relationship between organic nitrogen and the biomass production of algae. This is also reflected in higher Kjeldahl nitrogen values in the sludge [31]. In summary, BOD₅ removal efficiency in baffled algal reactors (86-89%) is higher than in the reactor without baffles (62%). Good correlation between $BOD₅$ removal rates and BOD₅ load was found in each reactor.

3.4. NO³ - -N

A comparison of nitrate concentration of the three reactors is shown in the Figure 9. The average nitrate concentration in the influent to the reactors was 0.4 mg NO_3 -N/l, while at the effluent of reactor T1 and T2, the average nitrate concentrations were 5.5-7.5 mg/l, respectively.

The reference reactor showed the lowest nitrate concentration followed by reactor T1 and T2. It is interesting to note that at the time when the nitrate variations were measured, ammonia was often effectively removed in the algal treatment reactors. This may indicate to occurrence of the nitrification and/or denitrification processes.

The low results of NO₃-N again highlight the influence of the reduced photosynthesis activity under the short lighting intervals. Hence, the reduction in the oxygen produced affects the nitrifying bacteria oxidizing ammonia to nitrates.

- Nitrate plays an important role for ammonia removal

Nitrate is the initial component involved in the denitrification process. The group of bacteria carrying out the denitrification process require absence of oxygen in water [32]. Under aerobic conditions, the dissolved oxygen favors the activity of the nitrifying bacteria. In contrast, at low levels of oxygen i.e. concentrations less than $0.5 \text{ mgO}_2/l$ (anoxic conditions) the biological denitrification process can be expected to occur. Both *Nitrosomonas* and *Nitrobacter* require ammonia nitrogen as the nutrient for growth.

The experiment results show that the concentrations of nitrate at the effluents of the reactors T1 and T2 were 5.5 and 7.5 mgNO₃-N/l respectively. Additionally, the concentrations of the produced oxygen in the reactors T1 and T2 were $5.9-6.2 \text{ mgO}_2/l$. Therefore, the reactors T1 and T2 have the positive impact on the nitrification performance, even there was the low nitrate concentration influenced to the treatment reactors.

From the results obtained from the reactors T1 and T2 (88-90% of ammonia removal efficiency), it is clear to see that the produced nitrates contributed as an important factor to the rise of the nitrification and denitrification rates. Breisha (2010) pointed out that nitrogen tolerant bacteria include nitrate respiring bacteria and true denitrifies [33].

Under the high nitrate concentration, the population of nitrate tolerant bacteria multiplies faster than nitrate intolerant bacteria. Thus, it increases the efficiency of ammonia removal.

Several researchers suggested that the nitrification-denitrification process is unlikely to be the principal mechanism of nitrogen removal from wastewater on using algal ponds [34], [35].

Moreover, the results obtained from this study indicate that assimilation as well as nitrification and denitrification processes were important mechanisms for the removal of nitrogen. If the detention time is long enough, there is a reduction in the concentration of oxygen in different parts of the reactors, the pH of the media is suitable and the algal population grows significantly.

4. Conclusion

It could be indicated that the baffled algal reactors have shown a high treatment performances in ammonia nitrogen removal in wastewater. The nitrification, denitrification and assimilation processes were the major mechanisms for nitrogen removal in baffled algae reactors. With the intermittent light regime of 12:12 hours (day/night) and temperatures around 22° C, the highest 86-89% of ammonium nitrogen removal efficiency was recorded.

In the baffled algal reactors approximately 81% of COD, 86-89% of BOD₅ removal efficiencies were observed without any aeration systems nor $CO₂$ addition, and 7.5 mgNO₃-N/l was created. A specific algal growth rate, μ_{spec} of 1.47/day was found at 20-22°C. Furthermore, the research revealed that, if the sludge is not removed from the system on time (every three weeks at least), the efficiency of ammonia nitrogen elimination will reduce could be explained by an increased decay rate of organisms and the recycling of organic matters into the water body.

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