

Acetate injection into anaerobic settled sludge for biological P-removal in an intermittently aerated reactor

K.H. Ahn¹, H. Yoo², J.W. Lee¹, S.K. Maeng¹, K.Y. Park¹, and K.G. Song¹

¹Environment and Process Technology Division, Korea Institute of Science and Technology (KIST)

P.O. Box 131 Cheongryang, Seoul, Korea, 136-650

²Civil and Environmental Engineering Dept. Stanford University, Stanford, CA, USA

(* Author to whom all correspondence should be addressed.)

Tel: 82-2-958-5842 Fax: 82-2-958-5839 E-mail: khahn@kistmail.kist.re.kr

Abstract Injecting acetate into the sludge layer during the settling and decanting periods was adopted to enhance phosphorus release inside the sludge layer during those periods and phosphorus uptake during the subsequent aeration period in a KIST Intermittently Decanted Extended Aeration (KIDEA) process. The relationship among nitrification, denitrification and phosphorus removal was investigated in detail and analyzed with a qualitative floc model. Dependencies of nitrification on the maximum DO level during the aerobic phase and phosphorus release on residual nitrate concentration during the settling phase were significant. High degree of nitrification resulted that phosphorus release inside the sludge layer was significantly interfered with nitrate due to the limitation of available acetate and the carbon sources from influent. Such limitation was related to the primary utilization of organic substance for denitrification in the outer layer of the floc and the retarded mass transfer into the inner layer of the floc. Nevertheless, effects of acetate injection on both denitrification and phosphorus release during the settling phase were significant. Denitrification rate after acetate injection was two times as high as that before acetate injection, and phosphorus release reached about 14 mg PO₄³⁻-P/g MLVSS/hr during the decanting phase after the termination of denitrification inside the sludge layer. Extremely low level of maximum DO (around 0.5 mg/L) during the aerobic phase may inhibited nitrification, considerably, and thus nearly no nitrate was present. However, the absence of nitrate increased when the phosphorus release rate was reached up to 33 mg PO₄³⁻-P/g MLVSS/hr during the settling and decanting phase, and nearly all phosphorus was taken up during subsequent aerobic phase. Since the sludge layer could function as a blocking layer, phosphorus concentrations in the supernatant was not influenced by the released phosphorus inside the sludge layer during the settling and decanting period. Phosphorus removal was directly (for uptake) and indirectly (for release) dependent on the median and maximum DO concentration during the aerobic phase, and those optimal values may exist within the range from 0.2 to 0.6 mg/L and 0.4 to 1.2 mg/L, respectively.

Keywords Acetate injection; enhanced biological phosphorus removal; intermittent aeration; optimal DO concentration; sludge layer

Introduction

Background

It is generally known that in order to achieve the enhanced biological phosphorus removal (EBPR) system has to be subjected to alternating anaerobic and aerobic condition. Under the anaerobic condition, phosphorus-accumulating organisms (PAOs) take up easily degradable organic matter from wastewater. The organic matter is then stored as poly-β-hydroxybutyrate (PHB) or poly-β-hydroxyvalerate (PHV). The energy required for the storage of PHB/PHV is produced by the PAOs by decomposing polyphosphate from intracellular store. As a result, the PAOs will release phosphate in connection with the storage of organic matter under the anaerobic condition. Under the aerobic condition, the PAOs consume PHB/PHV. The energy produced is used by the PAOs for growth and storage of phosphate in polyphosphate store. Such storage of phosphate is called the “excess uptake of

phosphorus” and has been adopted for phosphorus removal from wastewater in various treatment plant configurations.

When simultaneously removing a large amount of nitrogen along with phosphorus from the wastewater, EBPR would lower the denitrification capacity because of the competition between the denitrifiers and PAOs for organic substrate. This would be the case only if PAOs are completely different from the denitrifiers. If the PAOs (or part of them) are able to denitrify under anoxic condition, the extent of competition for organic substrate would be lowered. It has been demonstrated by Comeau *et al.* (1987) and Gerber *et al.* (1987) that the PAOs can take up phosphate under the anoxic condition utilizing nitrate as an electron acceptor. Especially, such denitrifying dephosphatation would be beneficial for removing phosphorus from wastewater with low COD:P ratio, as was the case for this experiment.

Description of process used for experiment

The process adopted in this study was an upgraded form of intermittently decanted extended aeration (IDEA) process (first developed in 1965 in Australia), named KIST Intermittently Decanted Extended Aeration (KIDEA) process. One of the chronically separated processes, IDEA, consisting of three phases (aeration, settling and decanting) was developed to overcome the seasonal fluctuation of influent without equalization basin by continuous feeding. The IDEA process was modified to introduce raw sewage to the bottom of the single bioreactor continuously and uniformly throughout the operation via specially designed inlet system, which eliminated the necessity of sludge mixing and saved the mixing energy by means of the uniform supply of organic loadings. Thus, the proposed process does not require a separate mixing period because not only the settling and decanting periods but also the initial portion of the aeration period can provide anoxic/anaerobic condition.

Figure 1 (a) and (b) shows the KIDEA process description and operation mode, respectively. The operation mode was discriminated by aeration, settling and decanting period. Based on the difference of the DO level, such periods can be defined as aerobic, anoxic and anaerobic period, respectively.

Since “Anoxic” is defined in this process as the state or condition where denitrifying bacteria (including the denitrifying P-removing bacteria) carry out denitrification independent of the DO concentration in the surrounding aquatic environment, anoxic condition may take place in the entire reactor during the initial aeration period, and in the settled sludge layer during the settling and decanting periods. “Anaerobic” condition takes place inside the settled sludge layer after all the nitrite and nitrate as well as oxygen are used up as electron acceptors. Since the influent wastewater and acetate are fed into the sludge layer even at the settling and decanting phase, phosphorus release can be effectively accomplished inside the sludge layer under the anaerobic condition without adversely affecting the effluent quality. “Aerobic” condition appears after a certain retardation time in an aeration period has elapsed. Then, nitrification and aerobic excess phosphorus uptake takes place as much as molecular oxygen sufficiently is supplied.

In this study, acetate as an additional carbon was injected into the sludge layer during the settling and decanting periods in order to induce phosphorus release and PHB/PHV synthesis only inside the sludge layer. The released phosphorus would not migrate rapidly into the supernatant resulting the deterioration of the effluent water quality. Also, the released phosphorus would be excessively taken up by the PAOs during the aerobic phase and so the population of the PAOs would be increased. Compared to the generally known bio-P removal process with chemical supplementation such as PhoStrip, acetate injection into the KIDEA does not need to have a separate reactor to precipitate the stripped phosphorus and also produces less sludge production.

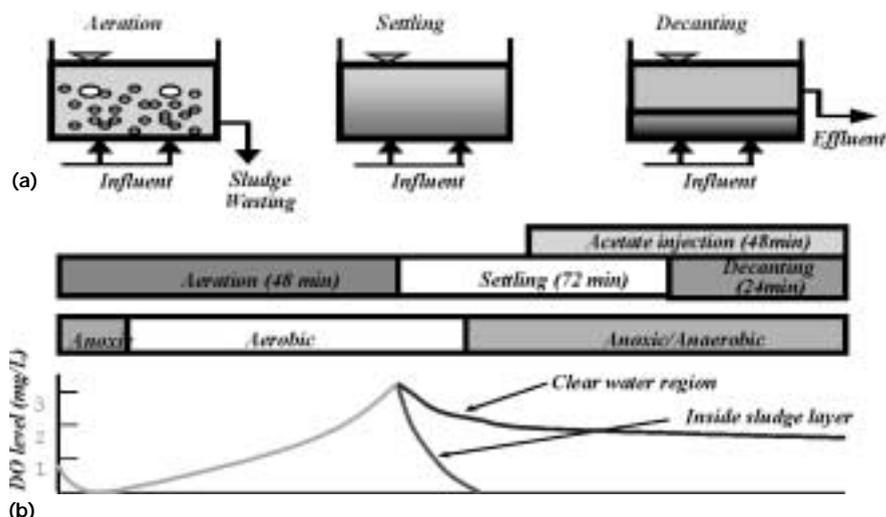


Figure 1 KIDEA process description and operation strategy: three periods of the process (a), and lengths of the three periods, corresponding conditions and the ideal DO pattern (b)

Qualitative microbial floc model for N and P removal

Acetate was frequently adopted as an external carbon source into the settled sludge layer to overcome the shortage of organic carbon content in the influent wastewater for effective N and P removal. The hypothesized concepts for nitrogen and phosphorus removal when the acetate is injected into sludge layer during settling and decanting periods are shown in Figure 2, which describes the condition (shades) and reactions (slashed lines) of a microbial floc during the one cycle of KIDEA process. When acetate injection begins 24 minutes into the settling period, the floc is rapidly under anaerobic condition (P-3 and N-3) and eventually saturated with acetate (P-4 and N-4) because the concentration of acetate solution is sufficiently high. First, any electron acceptor (nitrate or oxygen) available in the floc will be used up by the heterotrophs. Then, active phosphorus release along with PHB/PHV production takes place, utilizing acetate. After aeration begins, acetate present is aerobically degraded, first in the outmost region of the floc. Once all acetate appeared (P-5), aerobic phosphorus uptake takes place. From nitrification in this outmost region, nitrate is produced in the floc (N-5). In the middle region of the floc, heterotrophs carry out reduction of the nitrate using any available acetate (N-5). In the middle region (P-5), another heterotrophs, DPB can simultaneously carry out phosphorus uptake along with denitrification if they conducted phosphorus release in advance using acetate. In the inmost region phosphorus release continues until all acetate is consumed. Once acetate is completely utilized in the floc (P-1), aerobic phosphorus uptake and denitrifying phosphorus uptake take place in the outer and inner regions, respectively. As the oxygen supply continues during the nitrification, the aerobic and anoxic fractions inside floc are increased resulting in the phosphorus uptake inside the DPB and PAOs, Phosphorus uptake continues to take place. At the same time, aerobic and denitrifying consumption of the influent organic substrate takes place by the heterotrophs (N-1). When the aeration period ends and the settling period starts, aerobic region disappears because of quick oxygen consumption by heterotrophs. Then, the outer and the inner region of the floc become anoxic and anaerobic (P-2 and N-2), respectively.

The heterotrophs in the outer region carry out denitrification (N-2) utilizing organic substrate in the influent. If the stored PHB/PHV is still available for DPB, denitrifying phosphorus uptake continues in the outer region (P-2). In the inner anaerobic region, phosphorus release commences once acetate is available.

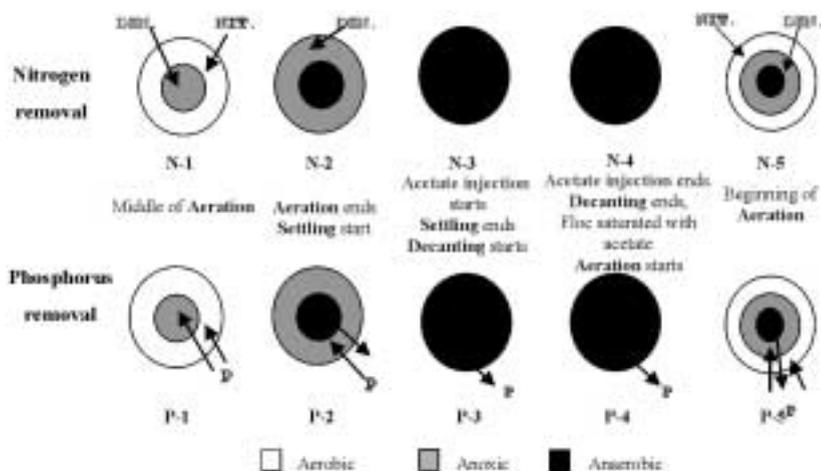


Figure 2 Reactions and condition in the microbial floc during one cycle for the KIDEA process

The objectives of this study are to examine the effects of acetate injection into sludge layer on the phosphorus behavior inside the sludge as well as denitrification efficiency, and furthermore dependencies of phosphorus removal on nitrate during the settling and decanting period along with DO level during the aeration period. It was another purposes of this study to determine the optimal DO levels for nitrogen and phosphorus removal.

Materials and methods

Experimental set-up and reactor operation

Activated sludge taken from Kwangdongli Sewage Treatment Plant (Kyungki-Do, Korea) served as the seeds. Mixed liquor suspended solids (MLSS) concentration in a reactor was initially set at approximately 8,000 mg/L, and the reactors were operated for the next 115 days. Sample analysis commenced after 23 days of acclimation period and continued for the next 92 days. The reactor had circular bottom with diameter of 20 centimeters, and the maximum working volume of 15 L. Influent wastewater port was located at the center of the bottom of the reactors. The reactor was operated with identical hydraulic retention time (HRT) of 36 h, and solid retention time (SRT) of 25 days. The influent wastewater was domestic wastewater obtained from Korea Institute of Science and Technology (KIST)'s residential apartments, and kept at 4°C. Nitrogen gas was intermittently pumped (10 minute on-off cycle) into the wastewater storage tanks to provide mixing. The reactor was operated at 17–20°C.

Cyclic operation was controlled by a programmable logic controller (PLC). The required time for the aeration, settling and decanting in a cycle was 48, 72 and 24 minutes, respectively; the duration of each cycle was 144 minutes (2.4 h) i.e. 10 cycles per day. Excess activated sludge was wasted for 1 minute before the end of the aeration period every cycle. Acetate was injected into reactor 2 during the last one-third (24 minutes) of the settling period, and the entire decanting period (24 minutes). 144 mL of acetate solution was injected into the reactor every cycle. The concentration of acetate solution was 4,430 mg/L, which was equivalent to providing additional 600 mg/L theoretical COD to the influent wastewater. 10 L of wastewater was fed to each reactor everyday, and all influent, effluent, acetate injection, and wastage streams for excess sludge were controlled using variable-speed Masterflex pumps. Air was diffused through aquarium-type diffuser stones and the supply rate was controlled using flowmeter. Supplied air was generally in the range of 0.3–1.0 L/min. A DO level control system was purposely not used to observe nitrogen and

phosphorus removal rates at various DO levels, and to obtain the optimal DO levels. DO concentration was measured by using DO meter (YSI Inc. Ohio, USA).

Track studies

Samples were collected 13 times during one cycle at different intervals. During the aeration period, the mixed liquor was collected and filtered through 0.45 mm glass fiber paper for analysis. After the settling period started, two samples were collected at each time of sampling – supernatant sample was collected by immersing 50 ml beaker into the water surface and sludge sample was collected by immersing a pipette carefully into the sludge layer. The tip of the pipette hit the bottom centre of the reactor, and then about 30 mL of the sample with settling/settled sludge was collected, which was immediately filtered. Soluble NO_2^- , NO_3^- and PO_4^{3-} were analyzed in all samples by using Ion Chromatography (DKK LIC-10). Analysis of others was performed according to the Standard Methods (1995).

Results and discussion

Nitrification, nitrate and phosphorus removal

Figure 3 describes the concentration of ammonia ($\text{NH}_3/\text{NH}_4^+$), nitrate (NO_3^-) and orthophosphate (PO_4^{3-}) in the effluent and the mixed liquor samples collected at the end of aeration. In both types of the samples, the concentration of orthophosphate appeared to be high when the nitrate concentrations were high, due to the limitation of electron donor for denitrification and phosphorus release. The nitrite (NO_2^-) concentration of the samples was lower than the detection limit.

Relationship between nitrate and orthophosphate concentration is shown in Figures 3(a) and 3(b). In spite of the poor correlation, the data may suggest that the orthophosphate concentration generally increased with the increase of nitrate concentration. Most of the orthophosphate was removed when the nitrate concentration was low. Few data points show high concentration of orthophosphate even when nitrate concentration was low. These data points were probably related to the extremely low DO condition during the aeration period which was too low to remove phosphorus as well as nitrification. Under extremely low DO conditions, limitation in electron acceptors either oxygen or nitrate resulted in the insufficient phosphorus uptake during the aeration period.

When acetate was introduced into the sludge layer during the settling and decanting period, nitrate interfered with phosphorus release and the production of PHB/PHV in the PAOs. One of the main reasons for the interference in phosphorus release by nitrate might be that heterotrophic denitrifiers have competitive advantage in utilizing acetate over the PAOs. If this were the case, phosphorus release would not take place until nitrate concen-

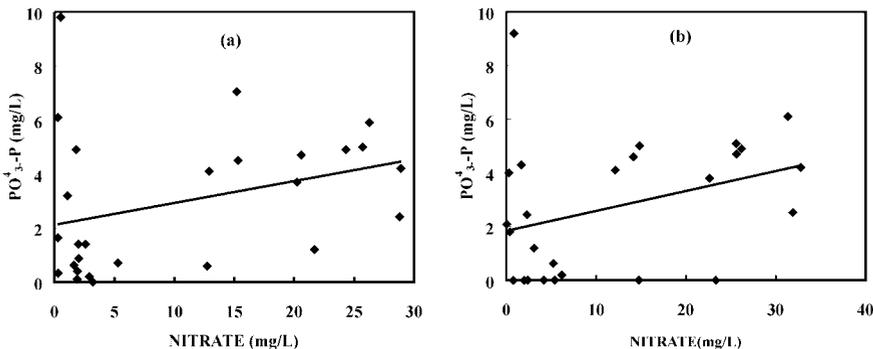


Figure 3 Relationship between nitrate and orthophosphate in the effluent (a), and the samples at the end of aeration (b)

tration fell below a certain threshold value. Wachtmeister *et al.* (1987) have reported that the phosphorus release to consumed-acetate ratio decreased under the presence of nitrate compared to that under complete anaerobic condition.

Track studies

In order to investigate the relationship between nitrate and orthophosphate more closely inside the sludge layer, track studies were carried out through out cycle. From the track studies, the rates for nitrate reduction (i.e. denitrification) and phosphorus release were determined. Strong dependency of the phosphorus behavior on the nitrate is shown in Figure 4. Also, nitrate concentration appeared to be desperately affected by DO concentration during the aerobic phase. Figure 4 (a) and (b) was discriminated by the maximum DO level during the aerobic phase, which remarkably influenced on the behaviors of both nitrogen and phosphorus. When the maximum DO concentration during the aeration period was as high as 1.4 mg/L, nitrification properly took place. On the contrary, when DO concentration was kept lower than 0.5 mg/L, nitrification appeared to be significantly interfered with, and therefore nitrification was ignorable as shown in Figure 4 (b).

In Figure 4 (a), nitrate concentration reached 14.9 mg/L at the end of aeration, and the effluent contained 12.2 mg/L. Simultaneous nitrification and denitrification (SND) was evident because the sum of ammonia and nitrate concentration at any time during the cycle did not add up to the influent ammonia concentration. In the moderately nitrified reactor, nitrate concentration decreases to nearly zero inside the sludge layer throughout the settling and decanting period. At this time, corresponding the specific denitrification rate was 3.2 mg NO_3^- -N/g MLVSS/hr. Once the acetate injection started, the specific denitrification rate remarkably increased up to 6.3 mg NO_3^- -N/g MLVSS/hr inside the sludge layer. However, nitrate concentration in the supernatant was slightly diminished throughout the settling and decanting period. As a part of the blocking layer, sludge layer retarded the migration of nitrate between the supernatant and sludge layer, and therefore a large fraction of nitrate in the supernatant remained without entering the sludge layer.

The added acetate could be utilized for phosphorus release inside the sludge layer after the primary utilization for denitrification. In this experiment, termination of the denitrification appeared when the decanting period was almost over. However, in spite of nitrate existence, phosphorus began to release in the sludge layer after 24 minutes of retardation time

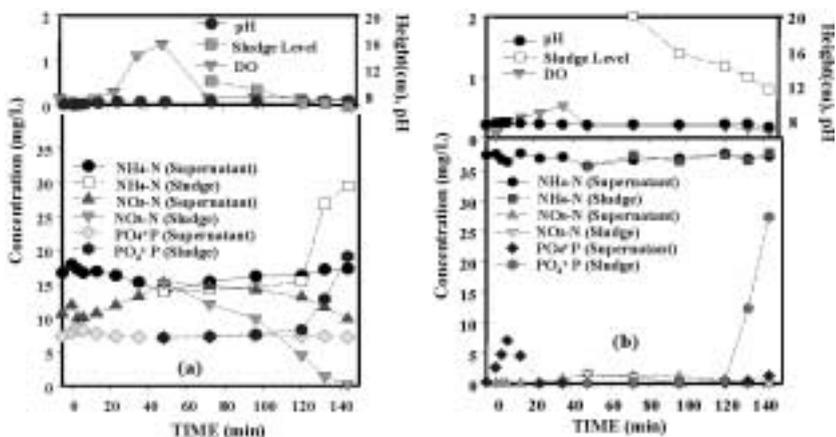


Figure 4 Behaviors of nitrogen and phosphorus both in the supernatant and the sludge layer depending on DO condition in cyclic studies. Results obtained on day 40 (a) and day 79 (b) in reactor 2. (Minutes 0–48 aeration, 48–120 settling, 120–144 decanting, 96–144 acetate injection; “(S)” indicates values observed near the influent inlet in the sludge layer)

elapsed since acetate injection started. Such result means the termination of the denitrification was locally accomplished near the influent inlet region earlier as shown in Figure 2. During the latter 24 minutes of the decanting period, the phosphorus concentration in the sludge layer increased up to 20 mg/L from 8.7 mg/L along with reduction of the nitrate concentration from 4.8 to 0.2 mg/L. During the decanting period, the specific phosphorus release rate was 14 mg PO₄³⁻-P/g MLVSS/hr. However, the phosphorus release was remarkably increased when the nitrification was hardly carried out as shown in Figure (b). Unlike the moderately nitrified reactor, the phosphorus concentration in the hardly nitrified reactor was increased up to 26.3 mg/L with the rate of 33 mg PO₄³⁻-P/g MLVSS/hr during the same period. The phosphorus release rate in the hardly nitrified reactor was two times higher than that of the moderately nitrified reactor.

Interestingly, phosphorus release continued until the initial part of aeration normally when phosphorus uptake was carried out. The phosphorus release during the aeration period could be understood in Figure 4(b), where the released phosphorus concentration reached as high as 7.3 mg/L in about 10 minutes. Many researchers reported that PAOs could release phosphorus under anaerobic, anoxic and aerobic condition whenever acetate or propionate was available, and the degree of release became greater under the anaerobic condition (Hascoet and Florentz, 1985; Gerber *et al.*, 1987; Kuba *et al.*, 1993, 1997). The phosphorus release during the initial part of aeration indicates that the acetate was not fully used up during the settling and decanting periods. Also, this result indicates much of acetate probably remains near the influent inlet at the bottom center of the reactor, even if acetate is intended to be evenly and quickly distributed into the sludge layer. The surplus acetate was locally concentrated near the influent inlet area, and then the substantial amount of acetate could be consumed for phosphorus release by the PAOs under the completely mixed condition during the aeration period.

Phosphorus uptake commenced only after acetate was fully consumed during the aerobic phase. In the moderately nitrified reactor shown in Figure 4 (a), phosphorus uptake was fairly limited, whereas complete phosphorus uptake took place in the hardly nitrified reactor shown in Figure 4 (b). Such difference was strongly associated with the amount of the previously released phosphorus. Phosphorus concentration in the moderately nitrified reactor was slightly decreased resulting in the relatively high concentration of phosphorus as high as 7.1 mg /L even after the aerobic phase finished. While, in the hardly nitrified reactor, phosphorus concentration was rapidly decreased to zero only for 10 minutes.

The measured denitrification and phosphorus release rates are somewhat overestimated, since the samples were collected near the influent inlet where acetate concentration was the greatest. At regions farther away from the inlet, lower rates are expected. Inside the settled sludge layer, the nitrate reduction rate is a critical factor for phosphorus release and is simultaneously dependent on the intensity of migration of the acetate which is a function of the acetate solution concentration and flux including both diffusion and convection inside the layer.

It may be concluded that the higher the spread efficiency of acetate solution, the higher denitrification and phosphorus release rates for the whole settled sludge layer, and the higher phosphorus removal.

Optimal DO concentration for phosphorus removal

The DO concentration was very low at the beginning of aeration, usually below 0.2 mg/L, and then kept increasing up to its maximum value at the end of aeration. The mid-value between the minimum and maximum DO concentration can be termed the median DO concentration. It can be surmised that optimal median as well as maximum DO concentration values exist for phosphorus removal in this process, since direct or indirect dependencies of

phosphorus on DO concentration were evident. For phosphorus uptake, either the median or the maximum DO concentration plays a major role. Also, maximum DO concentration has an indirect influence on the phosphorus release, which may be directly affected by nitrate concentration depending on DO concentration. If DO concentration exceeds the optimal value, excessive nitrification would take place, resulting in high concentration of nitrate in the sludge layer. Such nitrate would interfere with phosphorus release of the PAOs. On the contrary, the limitation of DO would result in the insufficient phosphorus uptake, in spite of satisfactory phosphorus release inside the sludge layer. Furthermore, nitrification would be nearly absent due to insufficient oxygen supply. Thus, under this condition, excessive amount of ammonia and phosphorus may be discharged without suitable treatment.

Figure 5 shows the existence of optimal median and maximum DO concentration for phosphorus removal during the aeration period in the KIDEA process with acetate injection. The minimum DO concentration was usually very low, and it is very difficult to reveal the relationship between the minimum DO concentration and the phosphorus removal from this study. From Figure 5 (b) and 5 (c), the optimal maximum and median DO concentration for phosphorus removal could be found out, respectively. In this study, the optimal DO concentration for phosphorus removal was determined as the DO concentration when the phosphorus removal efficiency was higher than 60 percent. As shown in Figure 5 (b) and 5 (c), the optimal maximum and median DO concentration for phosphorus removal existed within the range from 0.4 to 1.2 mg/L and from 0.24 to 0.65 mg/L, respectively. 11 out of 18 points were included within the wanted level, whereas 7 points were excluded from that level.

Only around 60 percent of the measured data within optimal DO range resulted in the satisfactory phosphorus removal efficiency (60%). The exclusion of these 7 data may result from the other factors such as the variations in the influent (based on COD, N and P), the biomass, temperature, pH and so on. Schön *et al.* (1993) reported that optimal DO concentration for phosphorus release and uptake was between 0.1 and 0.5 mg/L. However, it is too difficult to find out the optimal DO value for phosphorus removal because “phosphorus removal” is dependent on both the phosphorus release and uptake at the same time. From this study, it can be derived that phosphorus removal would be considerably dependent on the optimal DO concentration, even if it also tends to be relied upon such various factors.

Conclusion

A qualitative microbial floc model for nitrogen and phosphorus removal was developed and tested under conditions where highly concentrated acetate solution is injected into the sludge layer in this process. By examining the effluent and end-of-aeration samples, it was

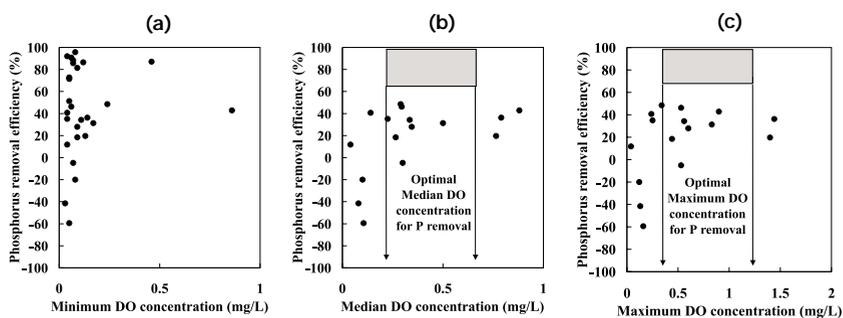


Figure 5 Influence of various minimum (a), maximum (b) and median (c) DO concentration values on Phosphorus removal in reactor 2

found that high degree of nitrification interfered with phosphorus release inside the sludge layer in the presence of acetate. Nitrate needed to be previously denitrified utilizing acetate before effective phosphorus release could occur. In the sludge layer before acetate injection, the average (maximum) specific denitrification rate was 3.2 mg NO_3^- -N/g MLVSS/hr. After acetate was introduced, the average (maximum) rate increased nearly two-fold to 6.3 mg NO_3^- -N/g MLVSS/hr. When approximately 15 mg/L was present at the end of aeration, (maximum) phosphorus release rate in the sludge layer reached about 14 mg PO_4^{3-} -P/g MLVSS/hr during the decanting period and the significant amount of phosphorus was discharged in the effluent. When nitrate was nearly absent at the end of aeration, specific phosphorus release rate reached about 33 mg PO_4^{3-} -P/g MLVSS/hr, and nearly all phosphorus was removed. Phosphorus release took place during the initial part of aeration period because the acetate was not fully used up during the settling and decanting period.

Optimal maximum, median and average DO concentration values during aeration for phosphorus removal seems to exist. These optimal DO concentrations may only apply to system like KIDEA or SBR because the DO is keep changing through out the one cycle in a single reactor. For phosphorus removal, the optimal median DO concentration was within the range from 0.2 to 0.6 mg/L and the maximum concentration was within the range from 0.4 to 1.2 mg/L. From the phosphorus release during the initial part of aeration in all track studies, it was concluded that the injected acetate solution was not distributed evenly and quickly in the sludge layer. Some portion of the injected acetate failed to contribute to PHB/PHV production. If the dispersion efficiency of the acetate solution is increased by increasing the flux of acetate injection and/or installing a special inlet mechanism, denitrification rate and the phosphorus release rate in the sludge layer should increase considerably. Furthermore, if a system for optimizing the DO concentration is installed, stable and high phosphorus removal should be achieved. Acetate injection and phosphorus release inside the settled sludge layer did not affect the quality of the effluent, indicating that the layer acted as an effective blocking layer between sludge layer and supernatant.

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