

Operation performance of an A/O process combined sewage sludge treatment and phosphorus recovery using human urine

Fanzhe Zeng, Wenbiao Jin and Qingliang Zhao

ABSTRACT

A novel sewage sludge treatment process is developed in which sludge anaerobically phosphorus (P) released with the temperature control/ultrasonic treatment and recovery with human urine are incorporated to a conventional anaerobic/aerobic (A/O) process. The results showed that temperature affected the anaerobic P release and the maximum orthophosphate (PO_4^{3-}P) release rate was 21.68 mg $\text{PO}_4^{3-}\text{P}/(\text{g MLVSS}\cdot\text{h})$ at 20 °C. The optimal specific energy of ultrasonic treatment was 15,000 kJ/kg TS, at which the solubilization degree of soluble chemical oxygen demand (SCOD) was 37.93%, which verified that the anaerobic sludge flocs were broken and the organic matter was obviously released. Human urine and P-rich sludge stream could be verified as a feasible way of P recovery in the form of struvite. The output of P in the combined A/O treatment process consisted of three pathways (i.e., effluent wastewater, sewage sludge, and P recovery). The influent P could be recovered by 22.84% and about 1.48 g/d potential struvite could be recovered from the anaerobic sludge flow using 0.27 L/d-human urine. The mass balances of COD and nitrogen (N) indicated that the combined A/O process also improved the organic mineralization and the removal of N.

Key words | anaerobic/aerobic process, human urine, phosphorus recovery, temperature control, ultrasonic treatment

Fanzhe Zeng

Qingliang Zhao (corresponding author)
State Key Laboratory of Urban Water Resource and Environment (SKLUWRE), School of Environment, Harbin Institute of Technology, Harbin 150090, China
E-mail: qlzhao@hit.edu.cn

Wenbiao Jin

School of Municipal and Environmental Engineering, Harbin Institute of Technology, Shenzhen 518055, China

INTRODUCTION

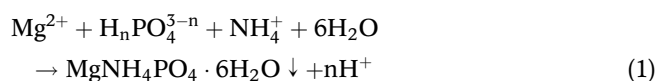
With the acceleration of urbanization and the dramatic increase of urban population, the amount of water to be treated in wastewater treatment plants (WWTPs) increases significantly. The wastewater in WWTPs are characterized by their high loads of carbon (C), nitrogen (N) and phosphorus (P), which need to be treated before discharging into natural water bodies to avoid environmental pollution (oxygen depletion, toxicity issues, eutrophication and algal bloom) (Singh *et al.* 2015; García *et al.* 2017). The anaerobic/aerobic (A/O) process is the most conventional and operational technology of biological mainstream wastewater treatment, which is favored for the high efficiency and low energy consumption (Steen 1998). Most novel treatment processes of WWTPs are based on the A/O foundation, thus, it is important to emphasize the plant-wide modeling as an integrated process and analyze the interactions among the A/O treatment systems (Jeppsson *et al.* 2013). But the restriction of this process was mainly because the P removal

efficiency was relatively low. In the anaerobic tank, a high concentration of phosphorus accumulating organisms (PAOs) in the activated sludge, which could hydrolyze polyphosphates and make phosphorus release. Then the wastewater and sludge enter into an oxygenated chamber, where the bacteria begin to absorb and enrich phosphorus (Saunders *et al.* 2003). In the settling tank, settled sewage sludge is returned to be treated by the A/O process and the supernatant could be recovered as a crystalline product. As sewage sludge enriches in P and is also considered as a perfect resource to produce soluble carbon source (Morgan-Sagastume *et al.* 2014). Thus, to realize the environmental sewage sludge recovery was the fundamental step to develop the sustainable A/O treatment process.

Anaerobic P release is verified as an effective and competitive technology, which is necessary to control the competition between PAOs and another group of microorganisms known as glycogen accumulating organisms

(GAOs). Different operating and environmental conditions have been identified as determinant factors to understand the competition between PAOs and GAOs. Among them, temperature appears to play an important role in sewage sludge activity and phosphorus release (Mulkerriens *et al.* 2004; Huang *et al.* 2011). Because PAOs could store volatile fatty acids (VFAs) as poly- β -hydroxyalkanoates (PHA), while intracellularly stored poly-phosphate (poly-P) and glycogen produce the energy for the P release process. But the anaerobic treatment in WWTPs always existed the low-performance rate of the organic matter hydrolysis, thus ultrasonic treatment, an emerging technology, is recognized as an efficient and promising method for improving the sludge reduction and organic matter solubilization. The ultrasonic process leads to the formation of bubble cavitation, which could implode and yield mechanical shear forces with free radicals (e.g. $\cdot\text{H}$, $\cdot\text{OH}$, HO_2) (Cai *et al.* 2018). The sludge with ultrasonic treatment could be obviously destroyed and organic matter could be released from the sludge, mainly depended on the supplied energy and ultrasonic frequency (Bougrier *et al.* 2005). However, a limited study reported the implementation of sequential anaerobic treatment with temperature control and ultrasonic treatment for the full-scale wastewater treatment.

Meanwhile, phosphorus, a non-renewable source, is considered as the key ingredient in fertilizers for plant growth (Cordell *et al.* 2009; Zhang *et al.* 2013). Therefore, P recovery from WWTPs, especially from phosphorus-rich sewage sludge, is a promising technology for chemical precipitation process in the wastewater treatment process. Struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) precipitates could be used as agricultural fertilizer for crop productivity as presented in Equation (1), where $n = 0, 1$ or 2 (Battistoni *et al.* 2005). Thus, how to enhance phosphorus release for the further crystallization is meaningful.



Mixed human urine and anaerobic sludge supernatant have been verified to be used for phosphorus recovery as the form of struvite (Tilley *et al.* 2008; Zeng *et al.* 2018). Both of them are environmental stream enriched with phosphorus and human urine is also an easily accessible ammonia nitrogen resource ($\text{NH}_4^+\text{-N}$). When human urine is stored, urease-driven hydrolysis leads to a high $\text{NH}_4^+\text{-N}$ concentration in the collection tank. Moreover, the urine matrix is preferable for struvite formation

because of a high pH (>8.0). The initial pH of human urine is near to the optimal pH condition and hence much less alkaline chemicals is required. Therefore, the potential benefit for P recovery with urine needs to be investigated in the actual water treatment process to solve the problem of high-cost sludge disposal. In this study, a sequential A/O wastewater treatment process is developed with the combination of three-sub-processes, i.e., conventional anaerobic/aerobic tank, anaerobic phosphorus release tank with temperature control and ultrasonic treatment and human urine/sludge crystallization tank to reduce wastewater treatment costs and realize environmental phosphorus recovery. The schematic diagram is shown in Figure 1.

The aims of this study are: (1) to understand the effect of temperature and ultrasonic treatment on the anaerobic sludge; (2) to study an approach for mechanistic description of phosphorus mass balance to ensure the accuracy of measurements and analyses; (3) to predict the novel A/O system performance in practical use (conventional A/O treatment process, anaerobic sludge process with temperature control and ultrasonic treatment, crystallization process with human urine for phosphorus recovery).

METHODS AND MATERIALS

Experimental equipment

The pilot-scale combined treatment system was composed of a conventional A/O reactor, an anaerobic phosphorus release tank with sewage sludge treatments (temperature controlling and ultrasonic treatment), and a crystallization tank. The A/O module consisted of a bioreactor with a specific volume ratio of anaerobic and aerobic tanks (i.e., anaerobic tank 12.8 L, aerobic tank 38.4 L) and a settling tank (10 L). Dissolved oxygen (DO) concentration in the aerobic tank was controlled between 1.5 and 2.5 mg O_2/L . The wastewater flow and sewage sludge sample were obtained from a wastewater treatment plant (WWTP) in Shenzhen, China. The plant adopted a biological treatment by an anaerobic/aerobic (A/O) process and the plant influent was mainly from the urban domestic and industrial wastewater.

The sewage sludge was settled in order to obtain the sewage sludge highly enriched in phosphorus (sludge retention time = 25 d) and the effluent in the stable operating system that was discharged conformed to Grade 1-A of the *Discharge Standard of Pollutants for*

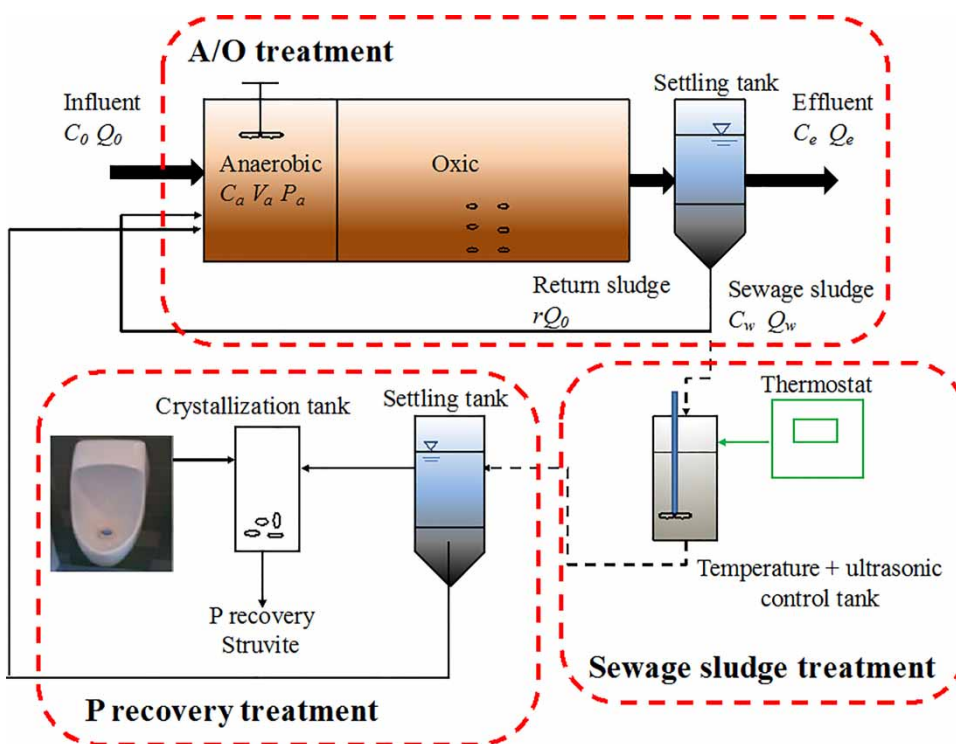


Figure 1 | Schematic of the A/O treatment plant.

Municipal Wastewater Treatment (GB 1898–2002) in China. After the mechanical dehydration on the sewage sludge, the water content of sewage sludge was nearly 98% and the sludge reflux ratio (r) was 70% to the anaerobic tank. The collected sewage treatment amount was about 5 L. The A/O reactor was operated under the following conditions: influent flow rate = $0.1 \text{ m}^3 \text{ d}^{-1}$, hydraulic retention time = 9 h (anaerobic tank 2 h, aerobic tank 2 h, settling tanks 1.5 h, anaerobic tank with temperature controlling and ultrasonic treatment 2 h).

The steady system was operated in continuous flow mode for 120 d after the startup period, which could be divided into four phases. In phase I (1–15 d), conventional operation strategy was adopted for the A/O treatment. In phase II (16–30 d), different temperatures were controlled for anaerobic phosphorus release to determine the optimal temperature, and in phase III (31–60 d) optimal temperature controlling with ultrasonic treatment was added into the excess sludge for sludge disintegration and then the sludge was recycled to the bioreactor. In phase IV (61–120 d), controlling crystallization tanks were added for phosphorus recovery in the form of struvite after the sewage sludge treatments. After P recovery, the sludge was recycled to the bioreactor. The A/O reactor was run continuously, while the anaerobic sewage sludge treatments and P recovery

were run intermittently to determine the operating performance of this combined A/O treatment process.

Batch experiments of phosphorus release with temperature control and ultrasonic treatment

The anaerobic phosphorus release experiments were carried out under 5, 10, 20 and $30 \text{ }^\circ\text{C}$ ($\pm 0.5 \text{ }^\circ\text{C}$) controlled by several thermostats (XT-SM8, Dongguan, China) to optimize the temperature as well as clarify the effect of temperature on the release of P from the sludge degradation. Sewage sludge (5 L) with the initial total solid concentration (8.5 g/L TS) was collected from the settling tank of the A/O treatment process and then subjected to the anaerobic P-release treatment in the capped 20 L-phosphorus release tanks with temperature control. The initial HAc concentration for PAOs anaerobic experiments was kept constant at 6.25 C-mmol/L and the pH was controlled to be constant at 7.0 by adding HCl or NaOH. Active biomass concentration was used for the determination of the specific rates by considering the PAO biomass composition ($\text{CH}_{2.09}\text{O}_{0.54}\text{N}_{0.20}\text{P}_{0.015}$) (Smolders et al. 1994).

During the ultrasonic treatment, the anaerobic sludge with optimal temperature controlling was conducted using a high amplitude power probe with intermittent stirring and the

probe was dipped inside the sludge to a depth of 2 cm above the bottom of the tank. The samples were ultrasonically treated to obtain certain specific energy (SE) of 0, 5,000, 10,000 and 15,000 kJ/kg TS (the ultrasonic treatment conditions are shown in Table S1 (available with the online version of this paper), which were calculated according to Equation (2)).

$$SE = \frac{P \times t}{V \times TS} \quad (2)$$

where P is the ultrasonic power (W), t is the ultrasonic time (s), V is the sludge volume (L). The operating frequency was 20 kHz and the instrument had a supply power of 750 W. Ultrasonic power was fixed at the maximum and the power efficiency was recorded during the ultrasonic treatment. The solubilization degree (SD) of organic components was calculated based on Equation (3).

$$SD = \frac{SCOD_T - SCOD_0}{TCOD_0} \times 100\% \quad (3)$$

where SD is the solubilization degree (%), $SCOD_T$ and $SCOD_0$ are the soluble chemical oxygen demand of ultrasonic sludge and raw sludge in the anaerobic phosphorus release tank (mg/L), respectively. $TCOD_0$ is the total chemical oxygen demand of raw sludge (mg/L).

Phosphorus recovery experiment

The human urine samples were collected from female student restrooms on campus with 1 M sodium hypochlorite at room temperature and the human urine sample was directly injected into the sealed container. Stale urine contained sufficient NH_4^+ -N concentration, which could be the optimal choice of ammonia for struvite precipitation. Table S2 (available online) shows the typical properties of the actual stored urine sample in the public toilet, which was collected to achieve a stable ammonia concentration for 15 days. The percentage of Mg^{2+} concentration in the crystallization tank was about 2.0 ± 0.2 mg/L, and a small number of precipitates was observed because of the occurrence of 'spontaneous precipitation' (i.e. $Mg_3(PO_4)_2$) in the urine samples (precipitate formed without addition of any artificial reagents). This phenomenon was correlated with the results by Udert et al. (2003). The supernatant of the anaerobic sludge after sewage sludge treatments and human urine were mixed by additional magnesium (1M- $MgCl_2$) at the certain molar ratio. Each experiment was carried out three times in parallel and the average values were recorded.

Identification of microbial populations

In order to understand the quantification of PAOs among the microbial communities, fluorescence *in situ* hybridization (FISH) analysis was widely performed (Mielczarek et al. 2013). Sludge samples (1 mL) were firstly fixed in 4% paraformaldehyde-phosphate-buffered saline (PBS) solution for 2 h at 4 °C. Secondly, the fixed sludge samples were washed twice in PBS, re-suspended in PBS-ethanol solution (1:1, vol/vol) and then stored at -20 °C. The FISH probes used in this study were Cy5-labeled EUBmix (comprising equal amounts of probe EUB338, EUB338-II, EUB338-III) for all bacteria, 6-FAM-labelled PAOmix (comprising equal amounts of probe PAO462, PAO651, PAO846 (Carvalho et al. 2007) to target *Accumulibacter*. Then, during the hybridization process, hybridization buffer solution (8 μ L) and 1 μ L fluorescent probe titrated on slides at 46 °C for 25 h by a confocal scanning laser microscope (HITACHI, Tokyo, Japan). Population distributions were quantified by the MATLAB image processing toolbox according to the method described by López-Vázquez et al. (2008).

Analytical methods

During the study period, the wastewater characterization in terms of major conventional parameters was performed on a daily basis. All analyses, including ammonia nitrogen (NH_4^+ -N), total phosphorus (TP), orthophosphate (PO_4^{3-} -P), soluble chemical oxygen demand (SCOD), TS, total chemical oxygen demand (TCOD), and mixed volatile suspended solids (MLVSS) concentrations were performed in accordance with *Standard Methods* (APHA 2012). The concentrations of TP and PO_4^{3-} -P in the liquid phase were measured by colorimetry using spectrophotometer (T6, Beijing Purkinje General Instrument, Beijing, China). Then, the PO_4^{3-} -P contents in liquid samples were filtered through a 0.45 μ m membrane. VFAs were measured by the method proposed by Lahav et al. (2002) by gas chromatography (6890N, Agilent, USA) and PHA was analyzed by the method proposed by Acevedo et al. (2012) using a Bruker 430-GC gas chromatograph equipped with an FID detector and a BR-SWax column. The suspension pH was measured using PHS-3C type pH meter (Shanghai Hongyi Co. Ltd, Shanghai, China). Glycogen was determined by a modification of the method of Lanham et al. (2012), using the following conditions: 20 mg sludge, 0.6 M HCl and 5–6 h of digestion time at 105 °C. The digested supernatant was discarded and diluted 25–50 times. The fluorenone reagent (5 mL) was added in the diluted sample with a water bath and

determined the absorbance at 625 nm. Struvite crystals were analyzed by X-ray diffraction (XRD, D/max-Rb, Rigaku Japan) and scanning electron microscopy (SEM, S-3400N, HITACHI, Tokyo, Japan), coupled with an energy dispersive X-ray micro-analysis detector. A one-way analysis of variance (ANOVA) was performed to assess the sole index for anaerobic P release using Excel (Microsoft, USA), and the data displayed corresponding to the mean \pm standard deviation of the target parameters.

RESULTS AND DISCUSSION

Phosphorus release kinetics of sewage sludge at different temperatures

In order to enhance the ability of phosphorus release in the full-scale treatment process, the sewage sludge anaerobically treated at different temperatures. Organic substrates transformation and biomass composition were determined in the anaerobic temperature controlling tank (Table 1), which showed that the effect of temperature on the anaerobic

Table 1 | Organic substrate transformation and biomass composition at different temperature values

Parameter	5 °C	10 °C	20 °C	30 °C
MLSS (mg/L)	2,500	2,320	2,420	2,030
MLVSS (mg/L)	2,240	2,130	1,870	1,680
Polysaccharide (mg/g)	24.78	15.02	16.78	14.02
Ash (mg/L)	260	190	550	350
Poly-P (mg/L)	142.11	77.89	451.58	261.58
Gly (mg/L)	164.38	182.42	157.92	155.23
PHA (mg/L)	178.91	179.53	202.54	109.46
Active biomass (mg/L)	1,896.71	1,768.05	1,509.54	1,415.31
Pr (mg $PO_4^{3-}P/$ (g MLVSS·h))	12.12	15.33	21.68	19.02
Activity decay rate (d^{-1})	0.1030	0.069	0.1331	0.1927
HAc uptake rate (C-mol/C-mol biomass·h)	0.041	0.045	0.17	0.18
P/HAc (P-mol/C-mol)	0.95	0.72	0.53	0.22
Gly/HAc (C-mol/C-mol)	0.83	0.70	0.79	0.81
PHA/HAc (mg-C/mg-C)	1.13	1.10	1.08	0.71

Ash = MLSS-MLVSS; poly-P = Ash-MLVSS*5/95; Active biomass = MLVSS-PHA-GLY; Pr = $\Delta PO_4^{3-}P/(MLVSS \cdot \Delta t)$.

phosphorus release kinetics. The observed phosphorus content of the biomass increased from 0.0397 mg-P/mg-MLVSS to 0.0463 mg-P/mg-MLVSS as the temperature increased from 5 °C to 20 °C and decreased to 0.0454 mg-P/mg-MLVSS at 30 °C, which verified that the activity of PAOs significantly decreased and the capacity of PHA synthesis inhibited with the decreased ratio of PHA/HAc.

The contents of both PHA accumulation and glycogen consumption increased significantly during 5–20 °C. Meanwhile, while the storage of PHA was extremely limited and the phosphorus concentration significantly decreased when temperature increased to 30 °C. During the anaerobic phosphorus release, COD consumed was stored in the form of PHA while glycogen was decomposed to provide energy for releasing orthophosphate. When a large amount of PHA accumulated inside the microbial cells, the cells became extremely fragile and was easy to be disrupted (Kuroda *et al.* 2002). In addition, PHA could be oxidized for glycogen replenishment, biomass growth, and aerobic maintenance purposes. Thus, it influenced not only the solubility of certain substances (i.e. oxygen, ammonia) but also the reaction rates of biochemical processes.

Especially, the anaerobic acetate uptake rate at 30 °C (0.18 C-mol/C-mol biomass·h) was comparable to that at 20 °C. The similar result was also demonstrated by Law *et al.* (2016), which verified that GAOs could co-exist with PAOs community. But the lower ratio of P/HAc indicated the inactivity of the TCA cycle and the orthophosphate concentration decreased at 30 °C, which suggested that either there was a reduced level of GAOs activity or that PAOs relied more on polyphosphate hydrolysis and less on the glycolysis of glycogen as the energy source at high temperature.

At 30 °C, GAOs might become the principal group and glycolysis was the only energy source and reducing power, while PAOs could use intracellular poly-P to provide energy. And 6.24% less active biomass at 30 °C was presented compared with that at 20 °C, which verified that higher temperature needed greater energy requirements for maintenance. Moreover, compared to the low-temperature operation, COD could be taken up by biomass corresponding to less P release in the bulk, resulting in less MLVSS concentration presented. Therefore, the anaerobic phosphorus release ability of sludge depended on temperature strongly and the optimal temperature for GAOs was higher than that of PAOs. The similar results were demonstrated by Panswad *et al.* (2003).

The maximum orthophosphate release rate (Pr) was 21.68 mg $PO_4^{3-}P/$ (g MLVSS·h) under the temperature of 20 °C. Meanwhile, the biomass compositions of poly-P

content (451.58 mg/L at 20 °C) were contrasted to the phosphorus percentage of anaerobic sludge. The FISH results showed that the quantities of microbial populations (*Accumulibacter*) in the anaerobic phosphorus tank (Figure S1, available with the online version of this paper). PAOs contained a high fraction of an EBPR community in full-scale plants (61.4% of the total population). In the long-term experiments (phase II), phosphorus was largely released at the optimal temperature controlling (20 °C), which was beneficial for further phosphorus recovery.

Anaerobic sludge disintegration under ultrasonic treatment

According to the Erden & Filibeli (2010), the ultrasonic treatment led to an obvious change of temperature, which might deteriorate the disintegration of sludge because of the decrease of cavitation intensity. Thus, the ultrasonic treatment proceeded with the optimal temperature control for the sludge disintegration (phase III). The different ultrasonic specific energy treatments in the range of 0–15,000 kJ/kg TS were found to improve the solubilization of organic components (Figure 2). The sludge solubilization efficiency relied on the specific energy, which could improve the settleability and disintegration of anaerobic sludge. The SCOD of the anaerobic sludge at the optimal temperature increased from 262 mg/L to 1,290, 2,435 and 3,423 mg/L at 5,000, 10,000 and 15,000 kJ/kg TS, respectively. The SD of SCOD was 37.93%, which verified that the anaerobic sludge flocs were broken and the organic matter was obviously released. In addition, MLSS and MLVSS concentrations decreased rapidly, and the decrease of MLVSS/MLSS ratio (from 77.27% in raw sludge to 52.70% for ultrasonically treated sludge at 15,000 kJ/kg TS specific energy input) indicated that the ultrasonic treatment could promote the sludge stabilization and a certain percentage of organic organisms was mineralized. This result was comparable with the results of Bougrier et al. (2005). Thus, due to the better biodegradation of anaerobic sludge, phosphorus and ammonium concentrations in the sludge supernatant were higher than that in phase II, which has a direct effect on the following phosphorus recovery process. The ultrasonic treatment used for the disintegration process was quite effective and contributed to the increase of phosphorus in the supernatant.

According to Figure 2(c), the ultrasonic treatment did not improve computer simulation technology (CST) reduction and reduced the filterability of sludge. When the SD was low (12.35%), the effect on filterability was not obvious, while the SD increased to 26.08%/37.93%, the tiny particles

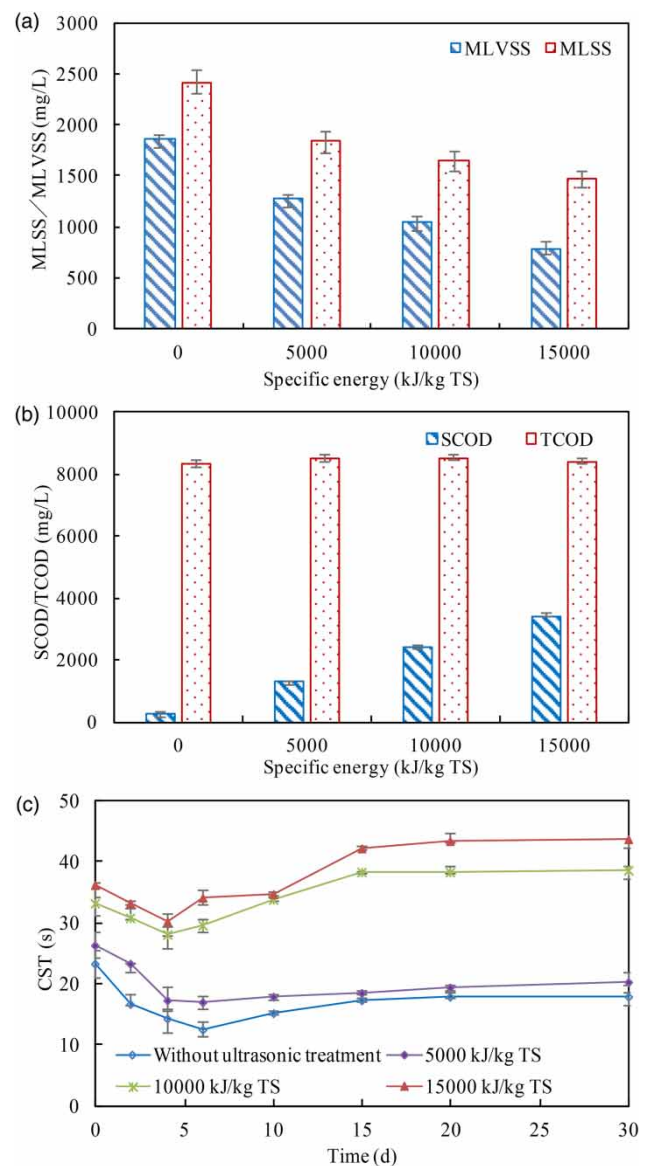


Figure 2 | Changes of organic compounds with different specific energy values.

were resisted to the sludge dewaterability. The increase of CST was also confirmed that the ultrasonic treatment enhanced the sludge disintegration causing the destruction of sludge structure and the release of phosphorus into the liquid phase. The similar results were indicated by Xie et al. (2008). Thus, the sequential ultrasonic and temperature controlling anaerobic treatment was beneficial for the anaerobic sludge disintegration and phosphorus release.

Phosphorus recovery with human urine in the crystallization tank

The volumetric ratio of human urine and sludge flow in the crystallization tank (phase IV) was determined to meet the

demand for struvite recovery. Human urine was a qualified ammonia nitrogen source, which could satisfy the molar ratio of Mg, N, and P (1:1:1) for struvite precipitation. As a certain amount of calcium was present, hydroxyapatite (HAP) could be also precipitated. But struvite content was much higher than HAP in the cyclization tank because the NH_4^+ content and the high alkalinity in the human urine affected the HAP formation. According to our previous study (Zeng *et al.* 2018), the optimal molar ratio of $[\text{Mg}^{2+}]:[\text{NH}_4^+-\text{N}]:[\text{PO}_4^{3-}\text{P}] = 1.2:1.05:1$ was used to meet the demand of the phosphorus recovery in both actual human urine and sludge flow, thus, the urine/sludge volume ratio was determined as 0.9%.

In addition, the ammonia nitrogen content in human urine samples gradually increased because of the hydrolysis process, thus, more phosphorus could be recovered with longer hydraulic retention time. The operation performance was first unstable, which might be because the treatment system did not adjust to the abrupt change of environment. The mean orthophosphate content in the effluent of the crystallization tank was 0.85 mg/L when the treatment system became stable, and the phosphorus removal efficiency maintained as much as 98.9% (Figure S2, available online).

However, only 95% phosphorus removal as struvite was achieved when $[\text{Mg}^{2+}]:[\text{PO}_4^{3-}\text{P}]$ molar ratio was 1.5:1 (Jaffer *et al.* 2002). Struvite precipitated as a white solid when MgCl_2 was added. The main elements of the precipitate were Mg, N, P and O and the SEM image confirmed that the precipitate was almost pure struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$). The XRD pattern also showed the typical peaks of struvite (Figure 3), which verified that struvite, the dominating compounds in the crystallization tank, could be recovered.

In conclusion, phosphorus recovery by human urine and anaerobic sludge flow was verified as a technical feasibility method. Human urine was variable, which depended on gender, hydrolysis time, person and many other factors, but the urine matrix was favorable for phosphorus recovery as the form of struvite because most phosphorus could be harvested and the ammonia concentration was quite sufficient for struvite precipitation. In this combined process, about 1.48 g/d potential struvite could be recovered from the anaerobic sludge flow using 0.27 L/d-human urine. Compared with other methods for the production of struvite (Martí *et al.* 2017), phosphorus recovery with anaerobic sludge supernatant and nitrogen/phosphorus in human

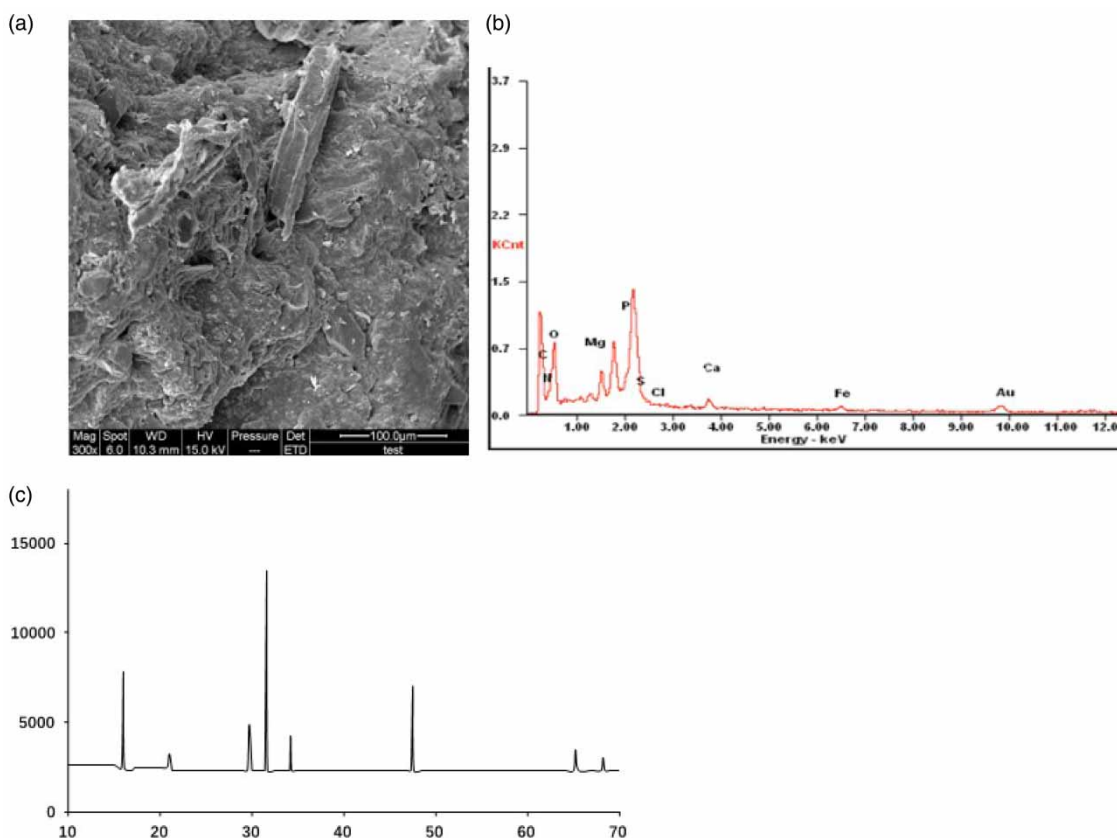


Figure 3 | SEM picture of the precipitates (a), elements analysis via EDS (b) and XRD pattern (c) of the precipitates with anaerobic sludge supernatant and human urine.

urine could gain much more struvite and fit the merits for environmental impacts. The recovery of P-based products alternatives for agriculture and industry, thus, struvite-based products, could be directly used as the effective and environmental fertilizer. The newest trends in WWTPs assumed to separate collecting of urine and fecal waste, which gave the opportunity for phosphorus recovery system with both urine and anaerobic sludge supernatant in the nearest future. The concept of ‘no waste generation’ was achieved in this study, which was described as a powerful and inspecting tool for phosphorus recovery and the whole sewage sludge management process.

Utilization in the combined A/O treatment system

The main parameters of influent wastewater, effluent wastewater in the combined A/O system at the optimal sewage sludge treatment conditions (20 °C controlling and 15,000 kJ/kg TS) during the entire operation periods were presented in Figure 4. In phase I, the average influent and effluent TP concentrations were 5.84 mg/L and 0.64 mg/L, respectively, while the effluent concentrations of COD and $\text{NH}_4^+\text{-N}$ were 77.5 and 5.9 mg/L, respectively. The concentration of COD and its removal efficiency obviously decreased in phases II and III, which verified that temperature controlling and ultrasonic treatments could promote the degradation of organic compounds. However, the phosphorus was continuously accumulated in the anaerobic tank with a rapid increase of the TP concentrations in phase II and III, which clarified that phosphorus was largely accumulated after the sewage sludge treatments. In phase IV, most of the supernatant P was recovered in the form of struvite and removed into the crystallization tank. However, P recovery process had an insignificant influence on the effluent concentrations of COD, while the $\text{NH}_4^+\text{-N}$ concentration rapidly decreased (30–50 d) until a stable level was reached (0.5 mg/L).

The differences between the conventional and combined A/O processes of the mass balance of COD, N, and P were illustrated in Figure 5. The influent phosphorus distributed into two pathways in the conventional A/O treatment: effluent wastewater and sewage sludge. 88.91% of P in the influent finally belonged to excess sludge, while 11.09% existed in the effluent flow. The inputs of the combined A/O process were the influent wastewater and the human urine, while the outputs of the combined A/O process included three pathways (i.e. effluent wastewater, sewage sludge and struvite derived from the crystallization tank). The mass balance was disturbed in the in phases II and III because of the anaerobic phosphorus significant

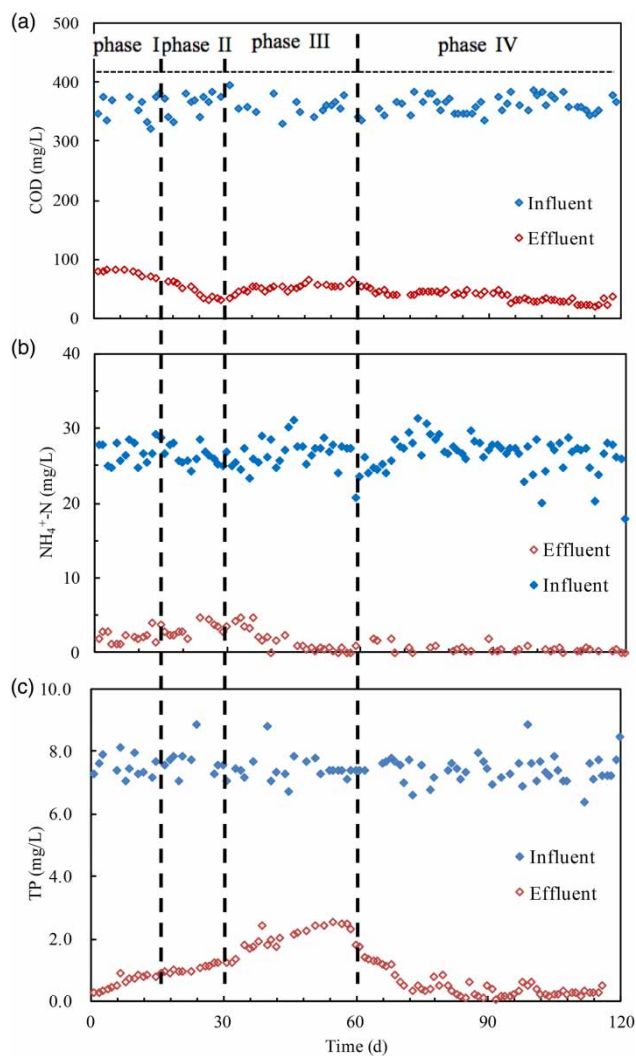


Figure 4 | Operation performance of the combined treatment system in terms of effluent pH (a), $\text{NH}_4^+\text{-N}$ (b), TP (c).

release. However, the mass balance of P regained balanced in phase IV. The effluent and sewage sludge of P decreased to 4.18% and 72.98%, respectively, because of the sewage sludge reduction by ultrasonic treatment and P recovery by the inflow of human urine. About 22.84% of the influent P was removed and recovered in the form of struvite, which was verified that the combined process was sustainable for the phosphorus removal in WWTPs and the novel phosphorus recovery method could be used in the full-scale application.

The proportions of COD and N discharged with sewage sludge also decreased obviously because the sewage sludge was reduced by the ultrasonic treatment. Especially, the removal efficiency of N increased significantly owing to the outputs (gas emission, effluent discharge, excess sludge

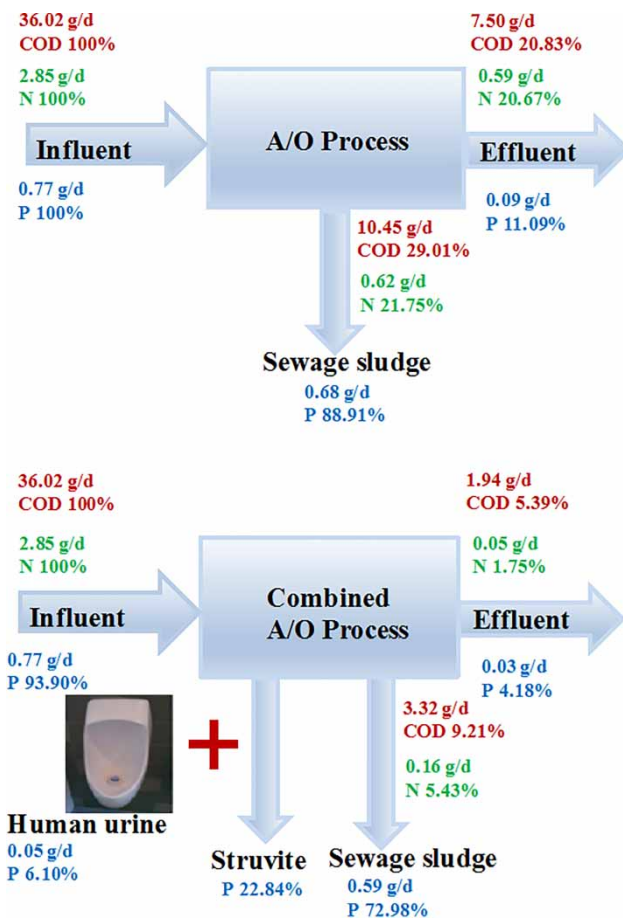


Figure 5 | Mass balances of COD, N, and P in conventional A/O and combined A/O processes of the pilot-scale system.

discharge, and struvite crystallization). The combined A/O process also had a significant influence on the removal of N (the proportion of effluent N decreased from 20.67% to 1.75%), owing to the high efficiency of the struvite crystallization process. A steady-state model of wastewater treatment process was developed based on the mass balance to predict the system performance and understand the distribution of nutrients (C,N,P) in the practical use (Saktaywin *et al.* 2005; Le *et al.* 2018). This study described the removal model of different nutrients in the combined A/O process and showed the destination of P in WWTPs, which could be straightforward and applied to other WWTPs without data collection.

CONCLUSIONS

This study investigated the long-term operating performance of a combined A/O process wastewater treatment plant with temperature control and ultrasonic treatment for anaerobic

P release and further phosphorus recovery by human urine. Based on the experimental results obtained, the conclusions were presented as follows:

- (1) The A/O process combined with sewage sludge treatments (temperature control and ultrasonic treatment) and P recovery using human urine was feasible.
- (2) The sequential ultrasonic and temperature-controlled anaerobic treatment were beneficial for the anaerobic sludge disintegration and phosphorus release. The phosphorus removal efficiency maintained as much as 98.9%. The maximum orthophosphate release rate was 21.68 mg $\text{PO}_4^{3-}\text{P}/(\text{g MLVSS}\cdot\text{h})$ under the temperature of 20 °C. In addition, the anaerobic sludge flocs were broken and the organic matter was obviously released with the solubilization degree was 37.93% at 15,000 kJ/kg TS specific energy input. Organic organisms of anaerobic sludge were also mineralized by ultrasonic treatment.
- (3) In this combined process, the influent P could be recovered by 22.84% from the supernatant. About 1.48 g/d potential struvite could be recovered from the anaerobic sludge flow using 0.27 L/d-human urine, and the P recovery was also beneficial for the removal of N.
- (4) The further studies could focus on P recovery from the anaerobic sludge supernatant with sewage sludge and human urine in the full-scale combined treatment process.

Supplementary data of this work can be found in online version of the paper.

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