



Effect of particle size distribution in wastewater on the performance of nutrient removal process

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ABSTRACT

The present investigation aims to explore the effect of particle size distribution in wastewater on the performance of sorption denitrification phosphorus removal process (S-DN-P process). The wastewater was obtained from the Wassmansdorf sewage plant in Berlin, which was denoted as the wastewater (WW). Further, the filtrates of wastewater fractions, obtained by sequential filtration using different pore size filters i.e. 3, 0.45, and 0.1 μm , were denoted by WW(3), WW(0.45), and WW(0.1). The P-removal was obtained to be 16.6, 9.0, 6.2, and 8.0 mg/L, respectively, for the wastewater samples WW, WW(3), WW(0.45), and WW(0.1). P-removal was decreased with decreasing pore size, except for the fraction WW(0.1). It was further observed that the ratios, COD:NO₃-N:Acetate:P, were found to be 8.04:1.93:3.55:1, 19.94:2.82:6.88:1, 16.29:3.26:10.23:1 and 13.50:2.54:7.41:1, respectively, for the fractions WW, WW(3), WW(0.45), and WW(0.1). Moreover, approximately 3.5 mg acetate/mg P removed for WW, 7 mg acetate/mg P removed for WW(3) and WW(0.1), and 10 mg acetate/mg P removed for the WW(0.45).

Keywords: Biodegradation; Particle size distribution; Sorption denitrification phosphorus removal process; Sequencing batch biofilm reactor; Wastewater treatment

1. Introduction

The sorption denitrification phosphorus removal (S-DN-P) process was biofilm-based large-scale denitrification and enhanced the biological phosphorus removal (EBPR) process. The S-DN-P process was performed in sequencing batch biofilm reactor (SBBR), in which wastewater was initially subjected to sorption (sorption phase), subsequently to denitrification (i.e. denitrification phase). The S-DN-P process was based on nitrate rather than oxygen respiration i.e. the aerobic phase of the biological phosphorus removal (bio-P) process was replaced by an anoxic phase [1,2].

The easily degradable substrates that were present in the wastewater at the beginning of anaerobic phase were used simultaneously for both processes (i.e. denitrification and bio-P removal) [3].

Contaminants present in the wastewater are complex mixtures of particulate and soluble constituents and the treatment of wastewater samples strongly depends on the size distribution of the contaminants [4,5]. Therefore, characterization and the effect of size distribution of the contaminants in the wastewater are important parameters to be studied in order to make a better understanding of the complex interactions that occur in the unit's operations and treatment processes. Particulate contaminants in wastewater usually occurred in the size range from <0.001 to over 100 μm

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[6]. In several studies, contaminants in wastewater were separated into four sizes by successive sedimentations, centrifugation, and filtration [7–9]. The fractions were classified by the size as settleable, supracolloidal, colloidal, or soluble fractions. An important conclusion drawn from these studies was that particles smaller than $1.0\ \mu\text{m}$ can be degraded biochemically with a significantly higher rate than the particles larger than $1.0\ \mu\text{m}$. In a line, De Haas et al. [10] suggested to increase the efficiency of the biological treatment of wastewater by grinding the larger particles of wastewater to a size of about $1.0\ \mu\text{m}$. Klopp and Koppe [11] reported that fractions of particles bigger than $2\ \mu\text{m}$ were accompanied with maximum organic portion. Similarly, wastewater fractions with particles less than $0.002\ \mu\text{m}$ were mainly composed of dissolved pollutants with low molecular size.

This paper intends to observe the effect of different wastewater fractions, obtained by filters having the pore sizes 3, 0.45, and $0.1\ \mu\text{m}$ in the treatment of wastewaters in particular to obtain the relationship between the substrate consumption and bio-P removal with denitrification in the S-DN-P process.

2. Materials and methods

2.1. Experimental design

Four different types of wastewater samples were obtained to evaluate the effects of particles in wastewater on P-removal and denitrification: wastewater (WW), wastewater filtered with a $3\ \mu\text{m}$ filter (WW(3)), wastewater filtered with a $0.45\ \mu\text{m}$ filter (WW(0.45)), and wastewater filtered with a $0.1\ \mu\text{m}$ (WW(0.1)) filter. Chemical composition of the wastewater from the Wassmanskorf sewage plant in Berlin (Germany) was determined to be 185–298 mg/L of biological oxygen demand (BOD), 300–490 mg/L of total chemical oxygen demand (TCOD), 20–48 mg/L of total nitrogen, and 8–15 mg/L of total phosphorus (TP). Moreover, the residual oxygen was measured and it was found to be within the range of 0.2–0.3 mg/L in the anaerobic phase.

The batch tests of the S-DN-P-cycle were performed in a lab-scale reactor, as shown in Fig. 1. The anaerobic sorption phase (S-phase) was performed first for two hours followed by the reactor that was drained completely. The anoxic denitrification phase (DN-phase) was performed by filling the lab-scale reactor with nitrifying water. The composition of nitrifying water is represented in Table 1. Finally, tap water was added until the total volume of this solution reached 500 mL. Removal of the residual chlorine

in the tap water was done by aerating the tap for 15 min with constant stirring followed by keeping this tap water in the open environment for one day. Anoxic phase was then performed for 4 h. Each test was performed with a 500 mL solution, which is about the same portion of thickened Bio-flow media. The final batch volume was varied between 750 and 800 mL. All the four fractions were tested in parallel. The mixing of wastewater samples was done with a magnetic agitator.

For microbial media, approximately 9-mm Bio-flow granules made from polyethylene and polypropylene were used. The Bio-flow media may help to increase the total quantity of biomass per ring in the SBBR. The total volume of SBBR and the volume of poured water were 1.5 and $0.6\ \text{m}^3$, respectively.

2.2. Sampling and measurements

The test was initiated with an injection of nitrogen gas. The first sample was drawn immediately after the nitrogen injection, whereas subsequent samplings were done with a definite time interval (S-phase: 0, 10, 20, 30, 60, and 120 min; DN-phase: 0, 10, 20, 30, 60, 120, 180, and 240 min). All the samples, except for TCOD and TP analyses, were immediately filtered with a $0.45\ \mu\text{m}$ membrane filter and placed in a refrigerator. The tests of all samples were performed under controlled and identical environmental conditions, i.e. pH (7.0 ± 3), time, and temperature ($25^\circ\text{C} \pm 2$). The $\text{PO}_4\text{-P}$, $\text{NO}_3\text{-N}$, $\text{NO}_2\text{-N}$, and acetate (AC) concentrations were analyzed by using an ion chromatograph, according to DIN standards (German Institute for Standardization). The TP, TCOD, and filtered chemical oxygen demand (FCOD) were obtained photometrically by using a spectrophotometer (UV-Vis 1,240, Tecator Co., Germany).

3. Results and discussion

3.1. Consumption of substrates in the sorption phase

The amount of substrates present in the wastewater fractions was optimized by determining TCOD. The TCOD consumed ($\text{TCOD}_{0\ \text{min}} - \text{TCOD}_{120\ \text{min}}$) during the sorption phase was found to be 133.50, 179.50, 101.00, and 108.00 mg/L for the WW, WW(3), WW(0.45), and WW(0.1), respectively. Further, the ratios of the TCOD-consumption for these four wastewater fractions after 2 h of operation were found to be 26% for the WW, 34% for the WW(3), 19% for the WW(0.45), and 21% for the WW(0.1). (cf. Fig. 2(a)). The degradation rate (i.e. TCOD-consumption) of the WW (3) fraction was found to be higher than those of the

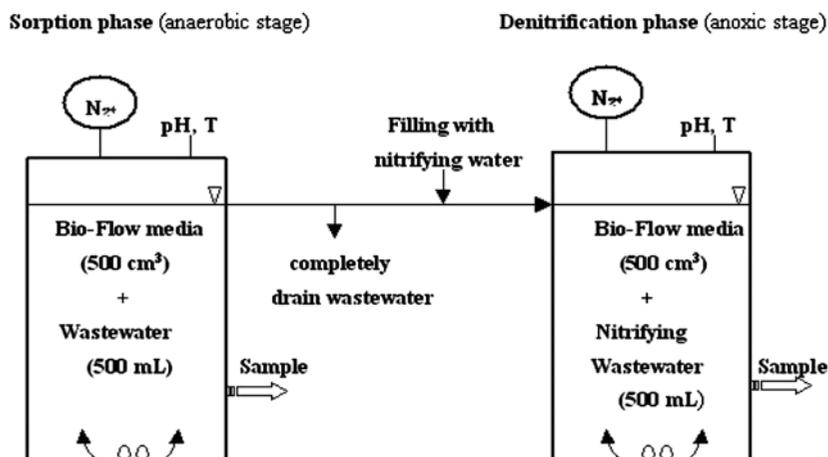


Fig. 1. A schematic of the batch plant.

Table 1
Composition of wastewater fractions

Fractions	Sorption phase (anaerobic phase)	Denitrification phase (anoxic phase)
WW	500 mL filling materials + wastewater (500 mL)	50 mg/L NO ₃ -N from 152 mg/L NaNO ₃ + 0.5 mL buffer (20 mg/L PO ₄ -P from KH ₂ PO ₄ and K ₂ HPO ₄) + 1 mL trace elements in 500 mL tap water
WW(3)	500 mL filling materials + 3 μm filtered wastewater (500 mL)	50 mg/L NO ₃ -N from 152 mg/L NaNO ₃ + 0.5 mL buffer (20 mg/L PO ₄ -P from KH ₂ PO ₄ and K ₂ HPO ₄) + 1 mL trace elements in 500 mL tap water
WW (0.45)	500 mL filling materials + 0.45 μm filtered wastewater (500 mL)	50 mg/L NO ₃ -N from 152 mg/L NaNO ₃ + 0.5 mL buffer (20 mg/L PO ₄ -P from KH ₂ PO ₄ and K ₂ HPO ₄) + 1 mL trace elements in 500 mL tap water
WW(0.1)	500 mL filling materials + 0.1 μm filtered wastewater (500 mL)	50 mg/L NO ₃ -N from 152 mg/L NaNO ₃ + 0.5 mL buffer (20 mg/L PO ₄ -P from KH ₂ PO ₄ and K ₂ HPO ₄) + 1 mL trace elements in 500 mL tap water

other three fractions. Similarly, the FCOD-consumption ($FCOD_{0\text{ min}} - FCOD_{120\text{ min}}$) was obtained for these four fractions and was found to be 106.40, 111.50, 98.50, and 95.00 mg/L, respectively, for the WW, WW (3), WW(0.45), and WW(0.1). This clearly indicated: the lower the TCOD-consumption, correspondingly lower the FCOD-consumptions. Moreover, the WW(3) possessed the highest FCOD-consumption efficiency i. e. approximately 27% in comparison to 26, 24, and 23% obtained, respectively, for the WW, WW(3), WW (0.45), and WW(0.1) fractions (Fig. 2(b)). The FCOD-consumption/TCOD-consumption ratio during the sorption phase was the highest for the WW(0.45) (0.98), followed by the WW(0.1) (0.88), WW (0.80), and WW(3) (0.62). Higher FCOD-consumption/TCOD-consumption ratios indicated that enhanced levels of

degradable contents were present in the wastewater [12,13].

The composition of wastewater greatly influences the elimination of biological phosphorus. The content of easily degradable substances, which contribute to the intensity of the anaerobic phosphate release, is an important parameter. Particularly, acetate plays a significant role as one of the C sources and facilitates the degradation process, which finally caused for biological P-removal from wastewater [4,14]. Hence, the consumption of acetate (AC-consumption: $AC_{0\text{ min}} - AC_{120\text{ min}}$) was analyzed for all these fractions and was found to be 58.9, 61.9, 63.4, and 59.3 mg/L for the WW, WW(3), WW(0.45), and WW(0.1), respectively. This showed that the WW and WW(0.1) possessed almost identical values. Although the WW(0.45)

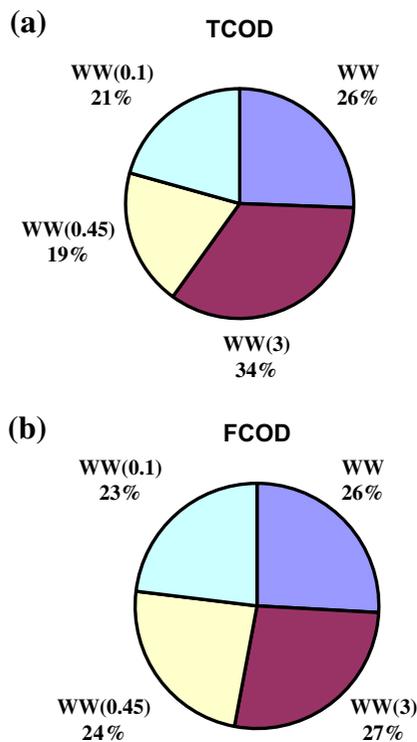


Fig. 2. TCOD (a) and FCOD (b) consumption in the sorption phase.

showed comparably the highest acetate content among these studied fractions however, the other fractions showed less change with the acetate content values. Acetate, which is smaller than $0.1\ \mu\text{m}$ pore size, was supposed to pass through these filters. Thus, the acetate consumption cannot depend on the filter size as it was present evenly in soluble form in the wastewater.

Furthermore, the ratios of AC-consumption/TCOD-consumption were calculated and found to be 0.44, 0.34, 0.63, and 0.55, respectively, for the WW, WW(3), WW(0.45), and WW(0.1). Although the WW(3) showed a lower value for the AC-consumption/TCOD-consumption in comparison to the other studied

fractions however, the overall values for all of these fractions were very much comparable to one another.

3.2. $\text{NO}_3\text{-N}$ -decomposition and substrate uptake in the denitrification (DN) phase

Decomposed amounts of the nitrate ($\text{NO}_3\text{-N}_{0\ \text{min}} - \text{NO}_3\text{-N}_{240\ \text{min}}$) in the DN-phase were found to be 32.10 mg/L for the WW, 25.40 mg/L for the WW(3), 20.20 mg/L for the WW(0.45), and 20.00 mg/L for the WW(0.1). These results indicated that the $\text{NO}_3\text{-N}$ -decomposition in the anoxic phase was decreased with decreasing filter size. However, WW(0.45) and WW(0.1) showed almost identical values, which were less than the values obtained for other two fractions i. e. WW and WW(3). During the initial period, the DN-phase showed a higher oxygen concentration. However, this higher oxygen concentration was quickly utilized by these samples. Nitrate, an electron acceptor present in the wastewater, is useful for denitrification and bio-P removal [15] in this DN-phase.

One of the most critical parameters in the denitrification process is the ratio of influent chemical oxygen demand to the nitrogen (COD/N), since it directly correlates the growth competition between the autotrophic and heterotrophic micro-organism populations [16,17]. The specific substrate consumption describes the stoichiometry between the organic substrate and the electron acceptor, which serves as an oxidizing agent in the respiration chain. The relationship between the organic matter consumed and the nitrogen reduced was determined by the denitrification process stoichiometry and the organic matter composition [18]. Table 2 shows the substrate-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios for these four fractions. The measured values of the TCOD-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios lie within the range of 5–6 g, which was regarded as a normal range for most microbial metabolic processes [19]. Table 2 clearly shows that, except for the WW(3), the measured

Table 2
Ratio of substrate consumption and $\text{NO}_3\text{-N}$ -decomposition

Fractions	TCOD-consumption/ $\text{NO}_3\text{-N}$ -decomposition (g TCOD/g $\text{NO}_3\text{-N}$)	FCOD -consumption/ $\text{NO}_3\text{-N}$ -decomposition (g FCOD/g $\text{NO}_3\text{-N}$)	Acetate(AC)-consumption/ $\text{NO}_3\text{-N}$ -decomposition (g AC/g $\text{NO}_3\text{-N}$)
WW	4.20	3.30	1.80
WW(3)	7.10	4.40	2.40
WW (0.45)	5.00	4.90	3.10
WW(0.1)	5.30	4.70	2.90

TCOD-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios were in a normal range for all these wastewater fractions. The TCOD-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios were 4.20, 7.10, 5.00, and 5.30 for the WW, WW(3), WW(0.45), and WW(0.1), respectively.

The EBPR process is widely employed to remove the nutrients from wastewater. However, the phosphate accumulating organism (PAO) and the heterotrophic denitrifiers (such as *Paracoccus denitrificans* and various *Pseudomonas*) were able to uptake P and reduce nitrate ($\text{NO}_3\text{-N}$), respectively. Therefore, the wastewater must contain sufficient carbon so as to enable their metabolism. It has been reported that 3–4 mg/L of acetate per 1 mg/L of $\text{NO}_3\text{-N}$ is required for complete denitrification [20]. In this study, the AC-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios were 1.80, 2.40, 3.10, and 2.90 for the WW, WW(3), WW(0.45), and WW(0.1), respectively. Furthermore, both values i.e. the FCOD-consumption/ $\text{NO}_3\text{-N}$ -decomposition and AC-consumption/ $\text{NO}_3\text{-N}$ -decomposition ratios were found to be lower for the WW in comparison to the other fractions studied.

3.3. P-release and substrate uptake in the sorption phase

The P-release in the reactor seemed to depend on the substrate consumption. Important characteristics of the influent wastewater for the bio-P removal process include the phosphate/COD-consumption ratios along with the nutritional components. Previous studies revealed that appropriate values for the P-release/TCOD-consumption ratios could lie between 0.01 and 0.1 g P/g TCOD [13,21]. In this study, the P-release/TCOD-consumption ratios for the WW, WW(3), WW(0.45), and WW(0.1) were found to be 0.03, 0.04, 0.10, and 0.06, respectively (Table 3). The WW(0.45) fraction showed a relatively higher P-release/TCOD-consumption ratio. In addition, this fraction showed the highest P-release, which could be explicable on the basis of substrate consumption observed previously. The P-release and AC-consumption ratios for the WW, WW(3), WW(0.45), and WW(0.1) were

found to be 0.08, 0.13, 0.17, and 0.11, respectively. The value for WW(0.45) was obtained higher those of other fractions studied, which inferred that the level of cell growth increased at a higher acetate concentration. The P-release/FCOD-consumption, P-release/TCOD-consumption, and P-release/AC-consumption ratios were found to be highest for the WW(0.45) fraction in comparison to the other three fractions. These results showed that, in the WW, approximately 10 mg/L of acetate was consumed per 1 mg/L of P-release, whereas in the WW(0.45), 5 mg/L of acetate was consumed per 1 mg/L of P-release.

3.4. Relationship to the P-uptake and $\text{NO}_3\text{-N}$ -decomposition in the denitrification phase

The majority of microbiological conversion was considered a major mechanism of increased P-removal. However, the electron acceptor, i.e. nitrate, was mainly used for other purposes i.e. the growth of micro-organisms. Comparing the level of P-uptake with the $\text{NO}_3\text{-N}$ -decomposition in the DN phase, it was noted that the complete reduction of nitrate to molecular nitrogen could not occur, as part of the nitrate was converted to nitrite [22]. Therefore, it was observed that $\text{NO}_3\text{-N}$ -decomposition affects the P-uptake in the DN-phase. The WW showed a maximum P-uptake of 21.2 mg/L, followed by the WW(3) and WW(0.45) with 16.8 mg/L, and lastly the WW(0.1) with 14.8 mg/L (Table 4).

The dissolved fractions in the WW(0.45) and WW(0.1) possessed identical $\text{NO}_3\text{-N}$ -decomposition values. Increasing the $\text{NO}_3\text{-N}$ concentration, the residual P concentration was decreased accordingly, which indicated that P release was decreased at the higher concentration of $\text{NO}_3\text{-N}$. The $\text{NO}_3\text{-N}$ -decomposition (32.10 mg/L) in the WW was observed to be higher compared to other fractions, but the P-release (4.60 mg/L) was lower than the other fractions. Furthermore, it was reported that optimum P-uptake and $\text{NO}_3\text{-N}$ -decomposition could be around 0.5 [14,23]. In this study, the P-uptake/ $\text{NO}_3\text{-N}$ -decomposition ratios

Table 3
P-release and substrate uptake in the sorption phase

Fractions	P-release (mg/L)	P-release/substrate consumption		
		(g P/g TCOD)	(g P/g FCOD)	(g P/g AC)
WW	4.60	0.03	0.04	0.08
WW(3)	7.80	0.04	0.07	0.13
WW(0.45)	10.60	0.10	0.10	0.17
WW(0.1)	6.80	0.06	0.07	0.11

Table 4
P-uptake and NO₃-N-decomposition in the DN-phase

Fractions	P-uptake (mg/L)	P-removal (mg/L)	P-uptake/NO ₃ -N-decomposition (g P/g NO ₃ -N)
WW	21.20	16.6	0.66
WW(3)	16.80	9	0.66
WW(0.45)	16.80	6.2	0.83
WW(0.1)	14.80	8	0.73

Table 5
The ratio of COD:NO₃-N:AC:P for the wastewater fractions

	COD:NO ₃ -N:AC:P
WW	8.04:1.93:3.55:1
WW(3)	19.94:2.82:6.88:1
WW(0.45)	16.29:3.26:10.23:1
WW(0.1)	13.5:2.54:7.41:1

were 0.66 for the WW and WW(3), 0.88 for the WW (0.45), and 0.73 for the WW(0.1).

Further, the ratio of COD: NO₃-N: AC: P for these wastewater fractions could enable us to predict the dependency of wastewater fractions on denitrification and P-removal in the S-DN-P process. Hence, the ratio COD: NO₃-N: AC: P was calculated and is represented in Table 5. It was noted that for 1 mg/L of P-removal, around 7 mg/L of acetate was used with the WW(0.45) and WW(0.1), whereas around 10 mg/L of acetate was used with the WW(0.45). Generally, 7–10 mg/L of acetate caused around 1 mg/L of P-removal [24]; thus, a greater acetate consumption leads to more cell growth and hence higher P-removal. The WW fraction showed a lower AC/P ratio (3.55) than the other three fractions. Nevertheless, the WW showed fairly good P-removal. The P-removal values were 16.60 (86.18%), 9.00 (60.43%), 6.20 (54.91%), and 8.00 (55.22%) mg/L for the WW, WW(3), WW(0.45), and WW(0.1), respectively. The WW(0.45) possessed a higher P-release than the other three fractions, but the P-uptake of the WW was determined to be around 5–7 mg/L, which was greater than the other fractions.

4. Conclusions

The effects of size distribution particles in wastewater on denitrification and biological P-removal in the S-DN-P process were investigated. From the reactor

operation, the P-release/TCOD-consumption and the P-uptake/NO₃-N-decomposition ratios were found to be higher (0.10 and 0.83) for the WW(0.45) fraction than the other fractions. The P-release/TCOD-consumption, P-uptake/NO₃-N-decomposition, and AC/P ratios increased as the pore size decreased, except for the WW(0.1). AC/P ratio indicated that in the WW(3) and WW(0.1), approximately 7 mg/L of acetate was consumed for 1 mg/L of P-removal, whereas in the WW(0.45), 10 mg/L of acetate was consumed for 1 mg/L of P-removal. The WW showed a lower AC/P ratio (3.55) compared to the other three fractions studied. Nevertheless, the WW showed a fairly good P-removal (16.60 mg/L). The WW(0.45) showed a greater P-release (10.6 mg/L) than the other three fractions, but the P-uptake in the WW (21.20 mg/L) was determined to be approximately 5–7 mg/L, which was greater than the other fractions. It was observed that decreasing the particle size fraction from WW to WW0.45 caused an apparent decrease with substrate uptake, P-removal along with the NO₃-N-decomposition. However, the fractions WW(0.45) and WW(0.1) showed almost similar values or a very slight increase.

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