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A tannin–based agent for coagulation and flocculation of municipal wastewater as a pretreatment for biofilm process

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ABSTRACT

The effects of a commercially produced Tannin-based coagulant and flocculant (Tanfloc) in a biofilm process pilot plant treating municipal wastewater were investigated. The investigated flow rates were 10, 14 and 18 L/min for the entire pilot plant, with two additional flows of 22 and 24 L/min were used for flocculation and sedimentation processes only. There was no clear deterioration in flocculation efficiency; even at 24 L/min, where the flocculation time was only 7.5 min. In terms of the clarification process, the enhancement was remarkably good; especially at high flows. Without Tanfloc, the removal efficiencies in the clarifier were less than 20%, 40%, 22% and 8% for turbidity, total suspended solids, biochemical oxygen demand and total phosphate, respectively. Meanwhile, when Tanfloc was used, they achieved 75%, 61%, 60% and 16% for the same respective pollutants. A significant rise in dissolved oxygen level in the aeration tank was observed when Tanfloc was applied (promising saving of energy during aeration). For instance, a dissolved oxygen level of 3 mg/L measured in experiments without Tanfloc, witnessed a climb to 6 mg/L when Tanfloc was used. In addition, volatile suspended solids concentration in the aeration tank decreased when Tanfloc was used (promising less production of sludge). Other measurements of total suspended solids (mg/L), chemical oxygen demand (mg/L) and biochemical oxygen demand (mg/L) in the experiments without Tanfloc were in the range (12–36), (60–104) and (24–50), respectively. Remarkably Tanfloc was able to reduce these measurements to low levels of (9–26), (28–68) and (7–24). In conclusion, the results suggest Tanfloc as promising agent to enhance the performance of clarification in a biological treatment unit. In light of this enhancement, Tanfloc could be used to upgrade existing treatment plants or design compact treatment units.

1. Introduction

Conventional sewage treatment plants construction and operation are highly expensive and require spacious area. In order to limit this problem, several attempts were conducted to develop and modify the conventional treatment units or provide other alternative methods with less requirement of construction, operation or land cost.

Reducing of organic load on the biological unit in the treatment plant is one of the attempts that may contribute to reduce the requirement of volume and oxygen for this unit. By reducing the organic load, faster treatment process is expected and consequently, exploitation of the same tank for the treatment of higher flow is anticipated. Moreover, since aeration of wastewater is the main consumer of energy in treatment plant (Dotro et al., 2011; Zhou et al., 2013), it will be a great benefit to reduce the oxygen requirement. United States Environmental Protection Agency (2010) stated that Oxygen requirement is a reflection of diurnal pattern of organic load, based on that, less requirement of oxygen coincides with having less organic load.

Practically, enhancement of sedimentation process is one of the alternatives to reduce the organic load on the biological unit (Tchobanoglous et al., 2003). Enhancement of sedimentation

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process could be achieved by preceding coagulation and flocculation into the process. An argument about the feasibility of this strategy is anticipated, however feasibility depends on the characteristics of the targeted wastewater (portion of suspended organic matters to total organics), availability of land, power cost, flocculant cost, primary sludge production expected increment due to capturing of more solids and anticipated decrement as a consequence of compaction when more solids are captured (Beltrán-Heredia and Sánchez-Martín, 2009), secondary sludge production (expected decrement due to slower biofilm growth rate as a consequence of lower influent organic load (Andreoli et al., 2007) and anticipated increment due to less compaction effect when the separated biofilm solids are less).

Flocculation has been used in combination with other treatment processes to treat a variety of wastewater. It has been used in full scale treatment plants with filtration as a post treatment process for municipal wastewater (Wang et al., 2017). Moreover, it has been used in pilot plants as a pretreatment (Sánchez-Martín et al., 2010) with sedimentation and slow sand filtration, and as a post treatment (Bongiovani et al., 2015) with filtration to remove the trace of organic matters. Finally, it has been used in batch-wise experiments in combination with oxidation and filtration (Hashem et al., 2016) and in coupling with membrane filtration and Fenton reactions (Gonçalves et al., 2017) and with photo-Fenton oxidation and sequential batch reactor (Rodrigues et al., 2017).

While coagulation is neutralization of negatively charged coloidals, flocculation is the accumulating of these neutralized colloids together to form bigger particles which can be easily settled (Tran et al., 2012). Coagulants and flocculants are those materials which possess the ability to achieve coagulation and flocculation. Conventional coagulants and flocculants are aluminum and ferrous salts. Several environmental (Nair and Ahammed, 2015) and public health (de Paula et al., 2014; Kakoi et al., 2017) problems aroused due to extended use of these conventional chemicals. Because of that, great efforts were paid to provide organic environment friendly alternatives to be substituted for the conventional inorganic chemicals. For example, Chitosan (Renault et al., 2009), Moringa oleifera seeds (Amagloh and Benang, 2009), Tannin (Beltrán-Heredia et al., 2010), Jatropha curcas seeds (Abidin et al., 2013) in addition to biopolymers exerted by certain species of microorganisms (Aljuboori et al., 2014) were exploited as environmental friendly coagulants. While some alternatives are produced in commercial quantity, others are still in the limits of lab scale production.

Tanfloc is a natural organic coagulant and flocculant, mainly consists of tannin which is extracted from *Acacia mearnsii De Wild* tree, this tannin is cationized by a certain chemical procedure under the name of Mannish reaction. In this reaction, a mixture of formaldehyde, quaternary Nitrogen (NH_4Cl) and hydrochloric acid (HCl) is stirred and heated, and then tannin extract is added, this process takes several hours until a viscous mixture contains 40% solids is produced, evaporation process is the last step to produce Tanfloc in its powder form. . The modified tannin possesses additional characteristics (cationic charge and higher molecular weight which is around 1.7 KD) that improve its ability to coagulate and flocculate colloids (Sánchez-Martín et al., 2009). Unlike conventional inorganic coagulants, Tanfloc is a biodegradable material. The value of biochemical oxygen demand (BOD_5)/chemical oxygen demand (COD) ratio for the aqueous solution of Tanfloc is 0.62 which indicates high biodegradability (Hameed et al., 2016).

Tanfloc has been used as a pretreatment agent in pilot plants comprise only physical/chemical treatment units such as sedimentation/slow sand filtration (Sánchez-Martín et al., 2010) and multi-layer filter/chlorination (Bongiovani et al., 2015). The aim of this study is to evaluate the effect of Tanfloc as a pretreatment agent on the performance of a pilot plant comprises biological treatment unit.

2. Materials and methods

2.1. Materials

2.1.1. Tanfloc

Tanfloc was purchased in powder form and used in the experiments as an aqueous solution with pre-determined concentrations depending on wastewater flows in the experiments.

2.1.2. Raw municipal wastewater

The municipal wastewater which is used in the experiment was produced from the hostel of Faculty of Engineering at Universiti Putra Malaysia. This hostel accommodates for 336 students with one central canteen. The main characteristics of the municipal wastewater are listed in Table 1.

2.1.3. Biofilm carrier

Cosmoballs (trademark of Pakar Management Technology/Malaysia) were used as biofilm carrier, they are hollow spherical polyethylene media with eight holes, each hole is 1 cm diameter, specific gravity of 0.9, average diameter of 8 cm, specific surface area of 160 m^3/m^2 and 2000 pieces occupy 1 m^3. The cosmoballs were packed in mesh bags used for fishing (100 pieces/bag) to maintain packed bed condition and immersed in the aeration tank with a filling ratio of 50%. This filling ratio is recommended by the manufacturer and it falls in the range that used by the researchers (Gu et al., 2014; Pal et al., 2012).

2.1.4. Pilot plant

The pilot plant was installed in the hostel mentioned earlier. It is made of transparent PVC and consists of the following units as illustrated in Fig. 1.

2.1.4.1. Raw water tank. Raw water tank (1250 L) is the first unit in the pilot plant. It receives wastewater from the sump of the real treatment plant in the hostel which is preceded by a screen unit. A submersible pump in this tank is used to convey the wastewater to the successive units of the pilot plant. A gate valve and flow meter were used to control the flow of this pump.

2.1.4.2. Dosing pump. Two dosing pumps (Seko) were able to eject a total amount of 10 L Tanfloc solution per hour. They were taking the solution from the buckets and dosing it to the coagulation tank.

2.1.4.3. Coagulation tank. This tank has the dimension of length,

Table 1
Raw wastewater characteristics.

Parameter	Unit	Value
Turbidity	NTU	38–79
Total suspended solids (TSS)	mg/L	52–120
Total dissolved solids (TDS)	mg/L	205
BOD_5	mg/L	50–146
COD	mg/L	134–352
Conductivity	μ s/cm	413
Nitrate (NO_3)	mg/L as N	0.4–0.9
Nitrite (NO_2)	mg/L as N	0
Ammonia Nitrogen (NH_3- N)	mg/L as N	15–29
Total Phosphate (PO_4)	mg/L as p	3.5–7.9
Temperature	C	27–29
pH		6.6–7.9

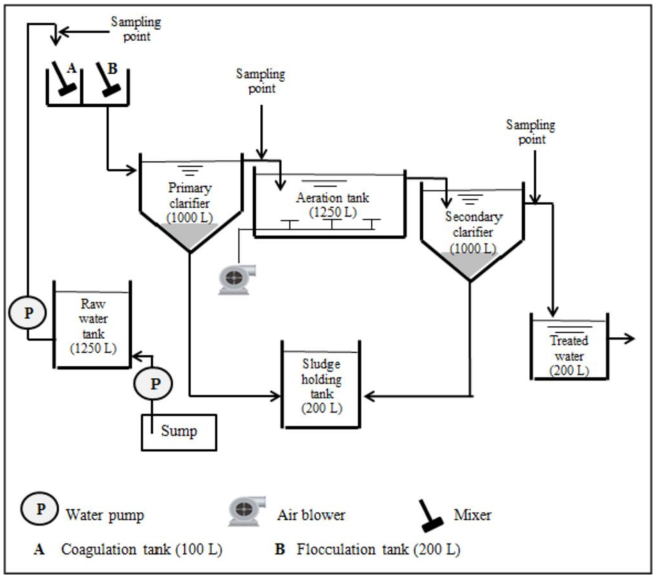

Fig. 1. Layout of the pilot plant.

width, and depth of 50, 40, 56 cm (100 L net volume). It is equipped with a pH meter and mixer with power of 0.37 KW driving a disc – type radial flow impeller with diameter of 20 cm at 180 rpm.

2.1.4.4. Flocculation tank. This tank is designed with length, width and depth of 85, 50 and 52 cm (200 L net volume). It is supplied with a mixer with power of 0.18 KW which is driving at 30 rpm a two layer axial flow pitched blade impeller, each layer contain 4 blades, the total length of every two opposite blades is 34 cm.

2.1.4.5. Primary clarifier. This unit has a square surface area of 100 cm dimension and a total depth of 135 cm including 35 cm depth inverse pyramid bottom for the sludge zone (1000 L volume excluding sludge zone). The wastewater is flowing from the flocculation tank through a 10 cm PVC pipe to a 30 cm diameter and 40 cm depth PVC cylinder located at the center of the clarifier acts as a baffle to uniformly distribute the wastewater. The effluent weirs is V notch type, each V is 15 cm width and 7 cm depth.

2.1.4.6. Aeration tank. A high density polyethylene tank with a net volume of 1250 L is equipped with two aquarium air pumps (HAILEA), 125 W power. According to the performance curve of this pump, one pump is able to blow around 115 L/min of air under the pressure of 0.012 MPa which is approximately the same of the water head pressure in the aeration tank, each pump has a plastic distributor of air with 26 nozzles connected to flexible hose ending with aquarium porous stone.

2.1.4.7. Secondary clarifier. This unit is identical to the primary clarifier.

2.2. Methods

2.2.1. Start up the pilot plant

The cosmoballs were put in the aeration tank and the pilot plant was running at a flow of 10 L/min. After consistent operation, COD was monitored for the influent and effluent of the pilot plant. Since a real municipal wastewater was used, fluctuation in wastewater quality was expected; consequently, fixed removal efficiency was not anticipated. Due to that, when 80% removal efficiency of COD was achieved (Approximately, after 20 days) and no significant increment was observed, the experimental performance data were taken.

2.2.2. Sampling strategy

All the experiments started at 8 a.m. as shown in Table 2, at that time the wastewater inside the pilot plant was produced at night and early morning and it was expected to be in low pollution level, consequently, we tried to get rid of the collected wastewater by doing two runs for every single experiment. The first one was passed without sampling, while samples (1.5 L for each sample) were taken for influent and effluent in the second run (duration of one run is the summation of the hydraulic retention time (HRT) of all the units in the pilot plant). Three to four samples were taken from the influent to cover the run duration and only one sample was taken from each of primary clarifier, aeration tank and secondary clarifier at the end of the second run.

2.2.3. Experiments plan

After stabilization of the pilot plant, from 2 to 3 experiments were completed every week. The sequence of the experiments is as shown in Table 2. Wastewater flow was varied from 10 L/min up to 18 L/min.

Suja and Donnelly (2008) collected data after 2 weeks started from the day that the flow has been changed to ensure stabilization and acclimation of biofilm to the new condition, Pedros et al. (2007) waited for 3 weeks for the same reason. However, both of them used constant characteristics wastewater. In the current case, fluctuating municipal wastewater was used in the experiment, consequently, maintaining of fixed organic load was not feasible, because of that steady state of biofilm after each variation of flow was not anticipated, in spite of that, 10 days waiting period was

Table 2
Sequence of experiments.

Experiment no.	Flow (L/min)	Tanfloc	HRT (one run (hr))	Starting time 1st run	Finishing time 2nd run	Tanfloc solution conc. (g/L)	Dosing rate of dosing pump (L/hr)
1	10	Without	5.7	8 a.m.	7.5 p.m.	–	–
Duplicate 1	10	without	5.7	8 a.m.	7.5 p.m.	–	–
2	10	with	5.7	8 a.m.	7.5 p.m.	2.1	10
Duplicate 2	10	with	5.7	8 a.m.	7.5 p.m.	2.1	10
3	14	Without	4.2	8 a.m.	4.5 p.m.	–	–
Duplicate3	14	Without	4.2	8 a.m.	4.5 p.m.	–	–
4	14	with	4.2	8 a.m.	4.5 p.m.	2.94	10
Duplicate 4	14	with	4.2	8 a.m.	4.5 p.m.	2.94	10
5	18	Without	3.3	8 a.m.	2.5 p.m.	–	–
Duplicate5	18	Without	3.3	8 a.m.	2.5 p.m.	–	–
6	18	with	3.3	8 a.m.	2.5 p.m.	3.78	10
Duplicate6	18	with	3.3	8 a.m.	2.5 p.m.	3.78	10

spent to tolerate any changes that might take place in biofilm behavior.

When Tanfloc was used in the experiment, it was added at a dose of 35 mg/L which has been already determined as the optimum dose in lab experiments (Hameed et al., 2016). When Tanfloc was used, submersible pump in the raw water tank and the mixer in coagulation tank were off temporary and Tanfloc was added manually at the required dose to the flocculation tank, slow mixing was allowed for a certain retention time based on the flow of that experiment to enable the flocs to be formed. After retention time was finished, the following four actions were taken: adding the required amount of Tanfloc to the coagulation tank, running the mixer of coagulation tank, running the submersible pump in the raw water tank and finally, running dosing pumps.

2.2.4. Analytical methods

All samples were taken in 1.5 L plastic bottles and analyzed for pH, turbidity, and BOD_5 at the same day then they were kept at 4 °C to be analyzed for COD, ammonia nitrogen, phosphate, and TSS on the next day. All analytical methods were done based on the American Public Health Association Standard Methods (Eaton et al., 2005). Method (5210) was followed to determine BOD_5, (5220 C) for COD, (2540 D) for TSS, (4500-NH¬3 C) for ammonia nitrogen, (4500-P C) for total phosphate determination after digestion with persulfate digestion method (4500-P B). Sludge volumetric index (SVI) according to (2710 D), flocculated wastewater samples were taken from the flocculation tank and transferred to 1 L imhoff cone in order to be settled for 1 h to determine the volume of settled sludge according to (2540 F), TSS was determined for the same flocculated wastewater. SVI was calculated based on Eq. (1) (Eaton et al., 2005):

$$SVI = \frac{settled\ \ sludge\ \ volume\left(\frac{mL}{L}\right) \times 1000}{Suspended\ \ \ solid\left(\frac{mg}{L}\right)} \tag{1}$$

Distribution of flocs size in the flocculated water was determined using Malvern instruments (MASTERSIZER/HYDRO 2000 MU). For more explanation, it should be known that MASTERSIZER instrument is basically a measurement of the volume distribution, this means that it measures the total volume of particles and gives the percentage of particles volume for the particles that lie below d (0.1), d (0.5) and d (0.9).

3. Results and discussion

3.1. Evaluation of flocculation process

To evaluate the impact of mixing time on the efficiency of flocculation process, five different experiments were conducted with different flows of wastewater, the first experiment with the flow of 10 L/min and the last one with the maximum flow which was limited by the wastewater pump capacity to 26 L/min. Three factors, flocs size distribution, residual turbidity and SVI, were investigated as indicators for the efficiency of flocculation process as shown in Tables 3–5.

Table 3 shows the values of d (0.1), d (0.5) and d (0.9), raw wastewater sample was tested to have an approximate idea about particle size distribution, since flocculation works mainly on non-settelable particles (the smaller in size); another sample was taken from settled raw wastewater and tested for particle size distribution. However, diurnal flocculation in size distribution is acceptable based on varied diurnal activities. No clear deterioration in floc size was noticed when the flow was increased up to 26 L/min, indicating that short flocculation time (7.5 min) was enough to achieve efficient flocculation process and longer time will not produce bigger flocs. The floc size in this experiment is smaller than floc size noticed in jar test experiment for the same wastewater (Hameed et al., 2016), this could be justified by the effect of continuous flow in the pilot plant that affects the real retention time compared to the batch mixing in the jar test. Furthermore, the best geometry of flocculation tank is the circular not the rectangular, our rectangular tank negatively affects the concept of velocity gradient (Tchobanoglous et al., 2003).

Table 3
Flocs size distribution (μm).

	Flocculation time (min)	d (0.1)	d (0.5)	d (0.9)
Raw wastewater	–	2	18	97
Settled raw	–	5	26	74
10 L/min	20	16	38	93
14 L/min	14	19	45	111
18 L/min	11	12	31	81
22 L/min	9	16	41	115
26 L/min	7.5	18	42	96

Table 4
Residual turbidity in the beaker.

	Flow (L/min)				
	10	14	18	22	26
HRT (minutes)	20	14.3	11	9	7.5
Initial turbidity (NTU)	47	53	59	55	53
After 10 min turbidity (NTU)	11	14	16	15.5	16
Removal efficiency %	76	73	73	72	70

For more investigation about the effect of increasing flow on the flocculation performance, samples were taken from flocculation tank at each experiment and kept in a 500 mL beaker for 10 min; a syringe was used to suck water 2 cm below water surface in the beaker to test the residual turbidity as shown in Table 4. Practically, there was no clear deterioration noticed in the removal efficiency confirming that as short retention time as 7.5 min is enough to achieve efficient flocculation process. Barrado-Moreno et al. (2016) investigated the influence of mixing time in the range of 10–60 min on the percentage of algal removal, and he found that the effect was negligible. Beltrán-Heredia and Sánchez-Martín (2009) investigated the effect of mixing time on the removal efficiency of turbidity in municipal wastewater using jar test, the results showed that no clear deterioration even at a mixing time of 5 min.

Sludge characteristics were investigated by determination of SVI as shown in Table 5, these values are comparable to the values of SVI which are gotten by Beltrán-Heredia and Sánchez-Martín (2009) (170 mL/g at Tanfloc dose of 40 mg/L), taking in consideration that the SVI is a function of TSS concentration which is variable according to water characteristics. It could be noticed that the values of SVI for the flows 18, 22, 26 L/min are better than that of 10, 14 L/min, this could be understood by observing that the TSS concentration is higher in the last three flows compared to the flow of 10, 14 L/min, which increases the opportunity of sludge compression that leads to smaller sludge volume.

3.2. Evaluation of clarification

The characteristics of wastewater before and after primary clarifier are shown in Table 6, and removal efficiencies were calculated and depicted in Table 7. It is worthy to mention that the samples taken for influent and effluent of primary clarifier were grab samples with time interval between influent and effluent equal to the HRT. Consequently influent characteristics may have little change within this period of time; furthermore, theoretical

Table 5
Sludge volume index.

Flow														
10 L/min (HRT = 20 min)			14 L/min (HRT = 14.3 min)			18 L/min (HRT = 11 min)			22 L/min (HRT = 9 min)			26 L/min (HRT = 7.5 min)		
TSS (mg/L)	Sludge volume (mL/L)	SVI (mL/g)	TSS (mg/L)	Sludge volume (mL/L)	SVI (mL/g)	TSS (mg/L)	Sludge volume (mL/L)	SVI (mL/g)	TSS (mg/L)	Sludge volume (mL/L)	SVI (mL/g)	TSS (mg/L)	Sludge volume (mL/L)	SVI (mL/g)
115	14	122	110	14	127	130	13	100	150	12	80	125	12	96

Table 6
Wastewater characteristics before and after primary clarifier.

	Flow																			
	10 L/min (HRT = 100 min)				14 L/min (HRT = 71 min)				18 L/min (HRT = 55.5 min)				22 L/min (HRT = 45 min)				26 L/min (HRT = 39 min)			
	Without Tanfloc		With Tanfloc		Without Tanfloc		With Tanfloc		Without Tanfloc		With Tanfloc		Without Tanfloc		With Tanfloc		Without Tanfloc		With Tanfloc	
	In	out	in	out	in	out	In	out	in	out	in	out	in	out	In	out	in	out	in	out
Turbidity (NTU)	52	44	49	16	47	39	43	15	42	45	53	13	–	–	52	20	–	–	52	23
TSS (mg/L)	90	55	70	30	90	70	65	25	70	67	75	30	–	–	70	40	–	–	80	55
COD (mg/L)	352	150	206	102	160	144	165	70	144	122	166	76	–	–	172	100	–	–	188	104
BOD_5 (mg/L)	78	64	73	33	62	56	56	28	73	57	93	37	–	–	76	33	–	–	67	45
Total phosphate (mg/L)	7.1	7.8	4.3	3.8	4	3.7	3.5	3.1	3.4	3.6	5	4.1	–	–	3.5	3.3	–	–	5	5.2

Table 7
Removal efficiencies of pollutants in primary clarifier.

Flow										
% removal	10 L/min		14 L/min		18 L/min		22 L/min		26 L/min	
	Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc
	%	%	%	%	%	%	%	%	%	%
Turbidity	15	67	17	65	0	75	–	61	–	56
TSS	39	57	22	61	4	60	–	43	–	31
COD	57	51	10	58	15	54	–	42	–	45
BOD_5	18	55	10	50	22	60	–	56	–	33
Total phosphate	0	12	8	11	0	16	–	6	–	–

HRT may differ from the real one due to the expected dead zones.

Turbidity removal efficiencies are shown in Table 7, it is clear that the efficiency for experiment without Tafloc were low at flows 10 and 14 L/min and it deteriorated to zero at the flow of 18 L/min due to short HRT (55 min) as depicted in Table 6. The enhancement of clarifier performance was obvious when Tanfloc was introduced; while high removal efficiency (more than 60%) was observed up to the flow of 18 L/min, a slight declination was noticed at flow of 22 L/min and a significant deterioration was shown at 26 L/min as a consequence of extremely short HRT (39 min). Beltrán-Heredia and Sánchez-Martín (2009) and Singh et al. (2016) stated higher removal efficiency of turbidity. However, their studies were conducted in jar test.

TSS behavior is similar to turbidity behavior. By increasing the flow from 10 to 18 L/min, removal efficiency deteriorated dramatically from 39% to 4%. After introducing Tanfloc, removal efficiency enhanced significantly to 60% and stabled to this level even when the flow increased to 18 L/min indicating the formation of big flocs that could be settled even if the HRT was as low as 55 min. The case was different at 22 and 26 L/min where the HRT was extremely short led to a serious deterioration in removal efficiency.

COD removal efficiency for 10 L/min was high for the experiment without Tanfloc because high initial concentration incident while they were low for 14 and 18 L/min. The effect of Tanfloc was very clear and the removal efficiencies increased to more than 50% for 14 and 18 L/min, while they were less at 22, 26 L/min due to short HRT. Furthermore, COD removal resulted from organic suspended solids removal. Singh et al. (2016) achieved around 63% and Beltrán-Heredia and Sánchez-Martín (2009) 60% removal efficiency of COD in their jar test study.

BOD_5 removal efficiency for experiments without Tanfloc was very low, while significant improvement was shown when Tanfloc was introduced. Clear declination was noticed at flow of 26 L/min due to short HRT. BOD_5 removal is the consequence of organic suspended solids removal. Singh et al. (2016) achieved around 58% and Beltrán-Heredia and Sánchez-Martín (2009) 60% removal efficiency of BOD_5 in their jar test study.

Phosphate removal efficiencies were very low without Tanfloc, while there was a slight increment when Tanfloc was introduced in the experiments with flows 10, 14, 18 L/min. At 26 L/min there was no removal efficiency due to short HRT. Some suspended solids in wastewater contains phosphate such as food residue and body waste (Eaton et al., 2005), it is expected that phosphate removal was achieved incidentally by removing of those solids from wastewater.

It is obvious from the previous discussion that removal efficiency decreased as HRT decreased, however this trend is not very clear in some cases due to the fluctuation in wastewater characteristics since the tested water is municipal wastewater not synthetic one. Moreover, in spite of extremely short HRT in case of 26 L/min flow, significant removal efficiency was observed when Tanfloc was introduced.

3.3. Evaluation of aeration tank (the biological unit)

Three pivotal points were studied in the evaluation of aeration tank: treatment efficiency, oxygen saving and estimation of volatile suspended solids (VSS). It is worthy to mention that regardless of the influent raw water temperature, it was almost stable at 30 °C in the aeration tank; indeed large amount of air bubble causes this stability. Moreover, pH values for raw and treated water were in the range of (6.6–7.5).

3.3.1. Treatment efficiency

Data for monitoring of raw, primary clarifier and secondary clarifier effluents are depicted in Tables 8–10 (values are mean ± standard deviation for duplicated experiments). Removal efficiencies of primary and secondary clarifier for a certain experiment were calculated based on the average values of the raw wastewater for that experiment.

TSS data that is mentioned in Table 8 showed some difference when Tanfloc was applied. Generally, introducing of Tanfloc improves solids removal in primary clarifier, consequently in the process overall.

Improvement in COD and BOD_5 due to application of Tanfloc was significant in this study as shown in Tables 9 and 10. It is expected that the main contributor to BOD_5 and COD are the organic solids that require more time to be decomposed compared to soluble organic materials (Tchobanoglous et al., 2003). When Tanfloc was used, removal efficiencies were better because most of the organic solids were removed in primary clarifier.

Regarding Ammonia nitrogen, no removal efficiency was observed (data are not shown). It is known that bacteria require a certain time to be adapted to a new environment. It is speculated that the inconsistent operation especially that related to flow variation due to clogging problem and the need for frequent adjustment might disturb Ammonia oxidizing bacteria, keeping in mind that they are known by their low growth rate and substrate consumption rate, and that might prolong the lag time. Another possibility is that the short retention time in aeration tank that might be insufficient for this kind of bacteria.

Generally, due to anticipated fluctuation of municipal wastewater characteristics, some confusion in results occurred. Indeed, even if samples of raw wastewater covered the whole period of experiment, still it is anticipated that some fluctuation might take a place between each two successive samples. However, this concept is more critical for primary clarifier than completely mixed aeration tank.

3.3.2. Dissolved oxygen study

Dissolved oxygen (DO) level decreased significantly as flow was increased as depicted in Table 11. . According to United States Environmental Protection Agency (2010), the oxygen demand in aeration tank is a reflection of influent organic load. It seems that the more organic materials the more bacterial activity and the more oxygen consumption, that is why oxygen demand follows the same trend of influent organic load. The increment of flow leads to organic load increment, consequently, more oxygen demand and less DO concentration. Similarly, at each flow rate DO level was higher in case of using Tanfloc because a high percentage of organics were already removed in the primary clarifier. It is worth mentioning that DO level was calculated based on the average of three readings during the experiment period and organic load was estimated based on the BOD_5 for the effluent of primary clarifier which was only one sample in the experiment, giving the potential of deviation from the reality.

One experiment was achieved using Tanfloc at flow of 18 L/min. The results are shown in Table 12. Although one of the two identical air pumps was off, the DO level was 2.4–2.6 mg/L at organic load of 0.046 (kg/hr), promising the potential to save 50% of the required

Table 8
Removal efficiencies of TSS (mg/L) in primary clarifier and secondary treatment (standard deviation shown in parentheses).

Flow (L/min)	Without Tanfloc				With Tanfloc			
	Raw	P. clarifier	% removal	S. clarifier	Raw	P. clarifier	% removal	S. clarifier
10	73.5 (13.4)	67 (2.8)	8.5 (12)	23.5 (2.1)	75.5 (13.4)	35.5 (9.2)	53 (4.2)	20.5 (7.8)
14	85.5 (10)	61.5 (2.1)	27.5 (12)	22.5 (6.3)	71 (1.4)	41 (2.8)	42 (2.8)	9.5 (0.7)
18	77.5 (10)	68 (11.3)	12.5 (2.1)	24 (17)	76.5 (2.1)	37.5 (0.7)	50.5 (0.7)	22 (1.4)

Table 9
Removal efficiencies of COD (mg/L) in primary clarifier and secondary treatment (standard deviation shown in parentheses).

Flow (L/min)	Without Tanfloc					With Tanfloc				
	Raw	P. clarifier	% removal	S. clarifier	% removal	Raw	P. clarifier	% removal	S. clarifier	% removal
10	191.5 (5)	169.5 (13.4)	11.5 (9.2)	64 (5.7)	66.5 (2.1)	162 (25.5)	89.5 (3.5)	44.5 (6.4)	38.5 (14.8)	76.5 (6.3)
14	205.5 (13.4)	174 (1.4)	15.5 (5)	84 (17)	59 (11.3)	170.5 (13.4)	92.5 (16.3)	45 (14.1)	47 (5.7)	72.5 (0.7)
18	207 (7.1)	200 (5.7)	3.5 (0.7)	102 (2.8)	51 (2.8)	207 (7.1)	98 (2.8)	52.5 (0.7)	64.5 (5)	69 (1.4)

Table 10
Removal efficiencies of BOD_5 (mg/L) in primary clarifier and secondary treatment (standard deviation shown in parentheses).

Flow (L/min)	Without Tanfloc					With Tanfloc				
	Raw	P. clarifier	% removal	S. clarifier	% removal	Raw	P. clarifier	% removal	S. clarifier	% removal
10	94.5 (9.2)	83 (8.5)	12 (17)	39.5 (0.7)	58.5 (5)	72 (0)	33 (9.9)	54 (14.1)	12.5 (10.6)	82.5 (10.6)
14	90.5 (23)	79.5 (17.7)	11.5 (3.5)	37 (18.4)	60.5 (10.6)	87.5 (12)	46 (5.7)	46.5 (13.4)	23 (1.4)	73.5 (5)
18	102.5 (0.7)	79 (1.4)	23 (1.4)	42 (8.5)	59 (8.5)	80.5 (0.7)	28.5 (2.1)	64.5 (2.1)	13.5 (0.7)	83.5 (0.7)

Table 11
DO variation in Aeration Tank.

	Flow					
	10 L/min		14 L/min		18 L/min	
	Organic load (kg/hr)	DO (mg/L)	Organic load (kg/hr)	DO (mg/L)	Organic load (kg/hr)	DO (mg/L)
Without Tanfloc	0.046	3.2	0.056	2.1	0.084	2.0
With Tanfloc	0.024	6	0.042	5.8	0.032	4.5
increasing		87%		176%		125%

Table 12
Performance of the pilot plant with one of the two identical air pumps was off.

	Turbidity (NTU)	TSS (mg/L)	BOD_5 (mg/L)	COD (mg/L)	NH_3-N (mg/L)
Influent (average)	66	69	98	218	26
Effluent	11	9	23	58	24
% removal	83	87	77	73	8

energy.

3.3.3. Estimation of VSS

In biofilm system, biomass is attached to the carrier, as bacteria grow, the biofilm increases in thickness, and bacteria flocs start to separate from the biofilm mostly due to hydrodynamics. The separated flocs (VSS) are settled in the secondary clarifier producing the secondary sludge.

The concentrations of VSS are depicted in Table 13. Generally, biofilm growth rate (consequently separation of biofilm) is a reflection of influent organic load. Accordingly, it is expected to observe lower concentration of VSS in the aeration tank as the influent organic load is less (in the experiments with Tanfloc). This concept is shown in some of the experiments in Table 13 and it is not clear in others, may be due to the low concentrations of VSS; indeed, moderate and low polluted wastewater does not stimulate biofilm to grow rapidly. Furthermore, as mentioned earlier, organic load was estimated based on the BOD_5 for the effluent of primary clarifier which was only one measurement in the experiment, giving the potential of deviation from the reality since it is not an average. Moreover, there is a possibility that some of the separated biofilm flocs were settled in the aeration tank in spite of the fact that it is a completely mixed tank. Finally, depending on its position in aeration tank, cosmoball might act as a shelter for the separated biofilm flocs because of its hollow design. However, turbidity measurements show clearly that the low turbidity coincides with low organic load (experiments with Tanfloc).

3.4. Statistical analysis

An Independent-Samples *t*-test was conducted to compare the difference between experiments without Tanfloc and with Tanfloc removal efficiencies at flow of 10, 14 and 18 L/min for COD and BOD_5. For removal efficiencies of COD, the results in Table 14 show that, there is no significant difference between cases without Tanfloc and with Tanfloc removal efficiencies at flow of 10 and 14 L/min ($p > 0.05$). But, the results indicate a significant difference between cases without Tanfloc and with Tanfloc removal efficiencies at flow of 18 L/min ($p < 0.05$). For removal efficiencies of BOD_5 similarly, the results reveal no significant difference between experiments without Tanfloc and with Tanfloc removal efficiencies at flow of 10 and 14 L/min ($p > 0.05$). However, the results show a significant difference between those without Tanfloc and with Tanfloc removal efficiencies at flow of 18 L/min ($p < 0.05$).

4. Conclusions

The Conclusions that could be drawn from this study are:

Even at short retention time in flocculation tank (7.5 min), Tanfloc showed a high potential to form big flocs with size distribution d (0.1), d (0.5) and d (0.9) of 18, 42 and 96 μm respectively

Table 14
Independent sample t − test of removal efficiencies of COD and BOD_5.

Variable	without Tanfloc		with Tanfloc				
	M	SD	M	SD	*df*	*t*	*p*
Removal efficiencies of COD							
Flow of 10 L/min	66.50	2.12	76.50	6.36	2	−2.108	0.170
Flow of 14 L/min	59.00	11.31	72.50	0.71	2	−1.684	0.234
Flow of 18 L/min	51.00	2.83	69.00	1.41	2	−8.050	0.015
Removal efficiencies of BOD_5							
Flow of 10 L/min	58.50	4.95	82.50	10.61	2	−2.900	0.101
Flow of 14 L/min	60.50	10.61	73.50	4.95	2	−1.571	0.257
Flow of 18 L/min	59.83	8.49	83.50	0.71	2	−4.069	0.039

Table 13
VSS concentration and turbidity for the effluent of aeration tank.

		Flow					
		10 L/min		14 L/min		18 L/min	
		Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc	Without Tanfloc	With Tanfloc
1st experiment	Organic load (kg/hr)	0.053	0.024	0.056	0.0353	0.0842	0.032
	VSS (mg/L)	30	32	53	28	54	44
	Turbidity (NTU)	42	12.4	44	15	51	20
Duplicate	Organic load (kg/hr)	0.046	0.0156	0.077	0.042	0.0864	0.029
	VSS (mg/L)	46	51	53	19	34	39
	Turbidity (NTU)	38	11	45	16	40	28

promising a better tendency to be settled. The SVI for the sludge produced from these flocs was 96 mL/g, indicating high ability to be compacted.

Enhancement of clarification process due to Tanfloc application was very clear. While efficiency of TSS removal in the clarifier was only 4% at a flow of 18 L/min (HRT = 55.5 min), 60% was achieved when Tanfloc was used. Even at high flow of 26 L/min (HRT = 39 min), removal efficiency of 31% was achieved when Tanfloc was applied promising the potential to shorten HRT and exploit smaller clarifier.

Enhancement in aeration tank performance was noticed due to Tanfloc effect on reducing organic load, BOD_5 range dropped from (24–50) to (7–24) mg/L when Tanfloc was introduced. Similarly, COD dropped from (60–104) to (28–68) mg/L. Furthermore, dissolved oxygen level in aeration tank jumped to almost double its value when Tanfloc was introduced to the biological process; it touched the threshold of 6 mg/L promising a saving in power and exploit smaller air blower. In addition, a noticeable decrease in VSS concentrations in the aeration tank when Tanfloc was used could promise future lower sludge production.

Finally, the aforementioned results suggest Tanfloc as promising agent to enhance the performance of clarification that benefits biological treatment units. In light of this enhancement, Tanfloc could be used to upgrade poor performing treatment plants or to get compact treatment units.

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