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Assessment of tetracycline antibiotic removal from hospital wastewater by extended aeration activated sludge

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ABSTRACT

Antibiotics are very effective in the treatment of bacterial infections that can be very dangerous for environment, human beings and animals. The purpose of this study was to evaluate and analyze the efficiency of the extended aeration activated sludge (EAAS) system in the removal of tetracycline (TC) from hospital wastewater. This study employed a cross-sectional design. Data were collected through laboratory scale and sampling was done in three hospitals of Ahvaz city in Iran. During sampling, 44 samples were collected from untreated hospital wastewaters and post-treatment outlet wastewater treatment plants (WWTPs) in 2016. Samples were extracted for TC by ethanol and nitrogen gas. Finally, with 2 mL injected methanol, TC was analyzed by high-performance liquid chromatography (HPLC). During this study in different seasons, the highest TC concentration was observed in hospital WWTPs in summer (0.368 mg L^{-1}). According to the results of the study, the maximum removal percentage was observed in summer (98%). Also, the lowest TC concentration was observed in WWTPs in winter. The results showed that percentage of minimum removal of TC was in autumn. In addition, EAAS was the best process for removal of TC. Furthermore, it should be mentioned that in the hot season, the rate of TC removal was increased and conversely, it was decreased in the cold season.

Keywords: Tetracycline removal; Wastewater treatment; Antibiotic; Extended aeration; Activated sludge; Hospital

1. Introduction

In recent years, in Ahvaz, Iran, the sources of drinking water, agricultural and industrial water are threatened by a great number of pollutants [1–7]. Antibiotics have harmful effects on the environment and humans because of produce organic chemical matters [6,8–16]. The antibiotic type is the most important factor in the uptake and excretion of antibiotics [17,18]. Antibiotics are persistent and lipophilic and can maintain their chemical

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structures in the body, which have been used for a long time for therapeutic purposes [19]. Bacterial resistance is an important resistance which is attributed to the excessive use of antibiotics [12,20]. Nowadays, the increasing consumption of antibiotics for preventing and treating infections has health benefits for improving plants, animals and human that lead to public concerns about antibiotic resistance [9,21,22]. Accumulation in the environment, especially in wastewater is one of the most important characteristics of antibiotics [1,23–26]. Based on the available reports, the annual consumption of antibiotics in Europe is about 19.5 packs per 1,000 people [27]. According to different studies, after a poor absorption, about 70% of antibiotics, especially tetracycline (TC) are excreted from the body through urine and feces [12,13,21,28]. Excessive antibiotics in the environment can have a marked effect on human health. It can cause disruptions in ntestinal microflora balance, headache, insomnia, diarrhea, vomiting, dizziness, phototoxicity, breast cancer and in fact can disrupt the digestive system in humans [11,29]. Previous studies have shown that pharmaceutical residues, radioactive elements, pathogenic microorganisms and metabolites of pharmaceuticals are mostly contained in hospital wastewaters [13,30–37]. In the aquatic environment, TC is one of the most important pollutants that can be very dangerous [13,38,39]. The most important sources of TC are Surface water, groundwater, wastewater and seawater [13,28,40,41]. The presence of TC can choose microorganisms variation in natural environments, developing resistance in bacteria or pathogens [13,28,42,43]. Physical (reverse osmosis and nanofiltration) and chemical processes (advanced oxidation and biological processes) are important methods in treating antibiotics from wastewater. Biological treatment is a cost-effective process. Based on the results of several studies, activated sludge process, among other processes, is widely used throughout the world [12,13,21,28,44]. In the process of biological treatment, bioadsorption causes antibiotics removal especially TC [45–47]. Biological process of aerobic wastewater treatment such as activated sludge process has a good effect on antibiotic removal [48–50]. The present study tried to assess the efficiency of extended aeration activated sludge system for TC antibiotics removal in influent and effluent wastewater from three hospitals.

2. Materials and methods

2.1. Physicochemical parameters

The present study used an experimental design which examined TC antibiotics removal from hospital wastewater by extended aeration activated sludge. This study was done in order to evaluate the potential effects of pH, temperature, total suspended solids (TSS), biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD) on removal of TC in three hospitals of Ahvaz city (located in south-western Iran) by wastewater treatment plants (WWTPs) system in 2016. This study has two main stages, namely sampling and analysis. The method used in this study for determination of BOD, COD and TSS was based on the standard methods [51,52]. The initial concentration of samples had been tested for determination of BOD, COD and TSS. The influent component of the wastewater treatment plant is presented in Table 1. BOD_5, COD, TSS, pH, temperature, MLSS, *F*/*M*, Qr/*Q*, Q_c, SVI and HRT were factors affecting the influent component in treatment plant (Table 1).

Table 2 shows the biosynthetic coefficient common for activated sludge process in WWTPs.

2.2. Chromatogram from analysis of TC

In order to determine the wastewater samples, low levels of antibiotics chromatography are usually applied. Mass spectrometry and HPLC systems are methods for measurement of drug candidates or metabolites [52,53]. Fig. 1 presents a chromatogram for a mixture of TC. According to the

Table 1
The average influent component of the wastewater treatment plant

Parameter	Wastewater (average)
BOD_5, mg L^{-1}	250
COD, mg L^{-1}	510
TSS, mg L^{-1}	500
pH	7.95
Temperature, °C	23.8
Mixed liquor-suspended solids (MLSS), mg L^{-1}	2,819.36
Food-microorganism ratio (F/M ratio), d^{-1}	0.35
Return sludge flow rate/ wastewater flow rate (Qr/*Q*)	64.28
Contact angle (θc)	10.93
Sludge volume index (SVI), mg L^{-1}	144.89
Hydraulic retention time (HRT), h	6

Table 2
Biosynthetic coefficient common for activated sludge process in WWTP

Coefficient	Range
Yield coefficient (*Y*), mg biomass/mg BOD_5	0.6
Maximum rate of substrate utilization (*K*), g COD/g VSS d	5
Saturation constant (K_s), mg/L BOD	60
Decay coefficient (K_d), g VSS/g VSS d	0.1
Maximum specific growth rate (μ_{max}), d^{-1}	1

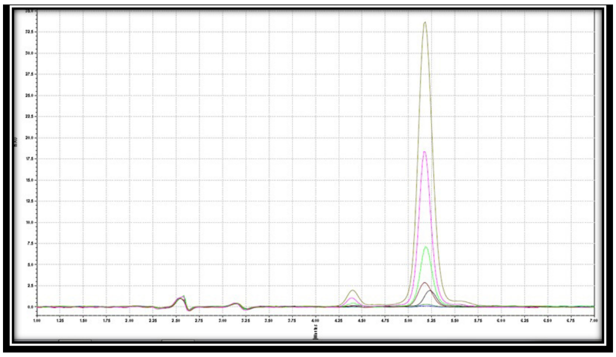

Fig. 1. Chromatograms of quantitation of ions for standard TC.

results presented in Fig. 1, peaks of TC were observed at the retention times of 2.4 min.

2.3. Sampling

Samples were collected from the influent and effluent of the wastewater treatment plants in Taleghani, Golestan and Abuzar teaching hospitals of Ahvaz city in Iran. In this study, for TC pollutants, 44 samples of three educational hospitals were taken and collected in 2016. Nine detection places were selected for sampling in three hospitals that had the same biological treatment (extended aeration activated sludge) during different seasons. 21 samples were collected using two glass bottles with a 1,000 mL volume. Then, McIlvain solution (5 mL) was added to the samples for dissolving TC. In the next stage, paper filter was placed in order to remove suspended solids by a Buchner funnel. Column or bed solid phase extraction (SPE) was activated with the addition of 2 mL of ethanol and 2 mL of deionized water to samples. Finally, the samples were analyzed by HPLC.

3. Results

Fig. 2 shows the trend of the TC treatment efficiencies, which is related to the WWTPs in the three hospitals. During the study, TC concentrations were generally reduced (80%–100%) by wastewater treatment process at WWTPs. The calculated percentages of removal related to TC in three hospitals were Golestan (64%), Taleghani (87%) and Abuzar (80%), respectively. As has been shown in Fig. 2, the average elimination of TC in Taleghani hospital (87%) was higher in comparison with those in other hospitals.

Fig. 3 indicates the BOD_5 removal efficiency in Golestan (85%), Taleghani (83%) and Abuzar (89%) hospitals in 2016, respectively. Furthermore, based on the results which are presented in Fig. 3, the COD treatment efficiency in three hospitals were in Golestan (85%), Taleghani (79%) and Abuzar (89%) hospitals, respectively, in 2016. Overall, Fig. 3 shows that the TSS elimination efficiency in Golestan, Taleghani and Abuzar hospitals was 85%, 82% and 89%, respectively.

Fig. 4 shows the relationships between removal efficiencies of physicochemical parameters (TSS, BOD_5 and COD)

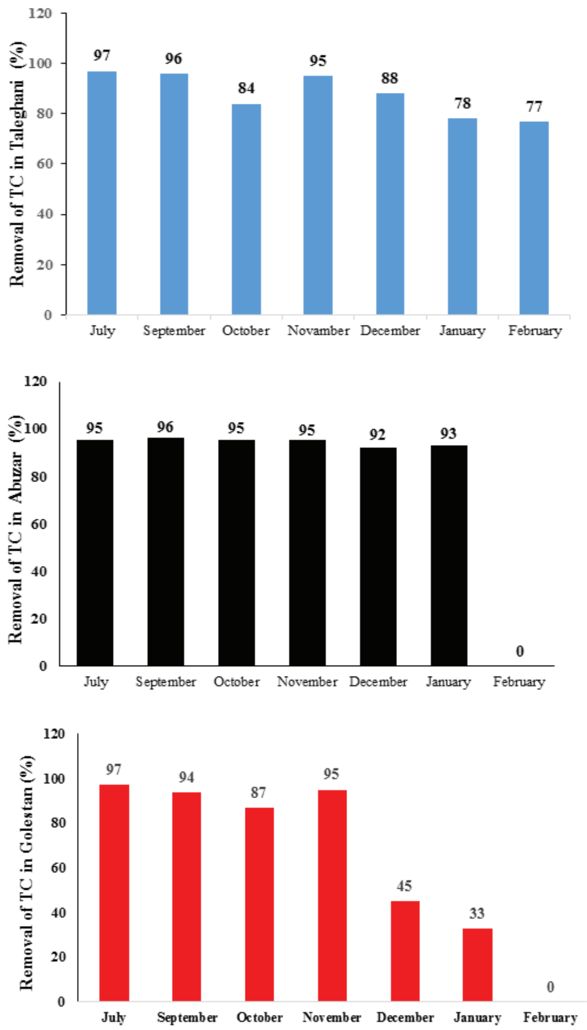

Fig. 2. TC removal efficiencies of the WWTP.

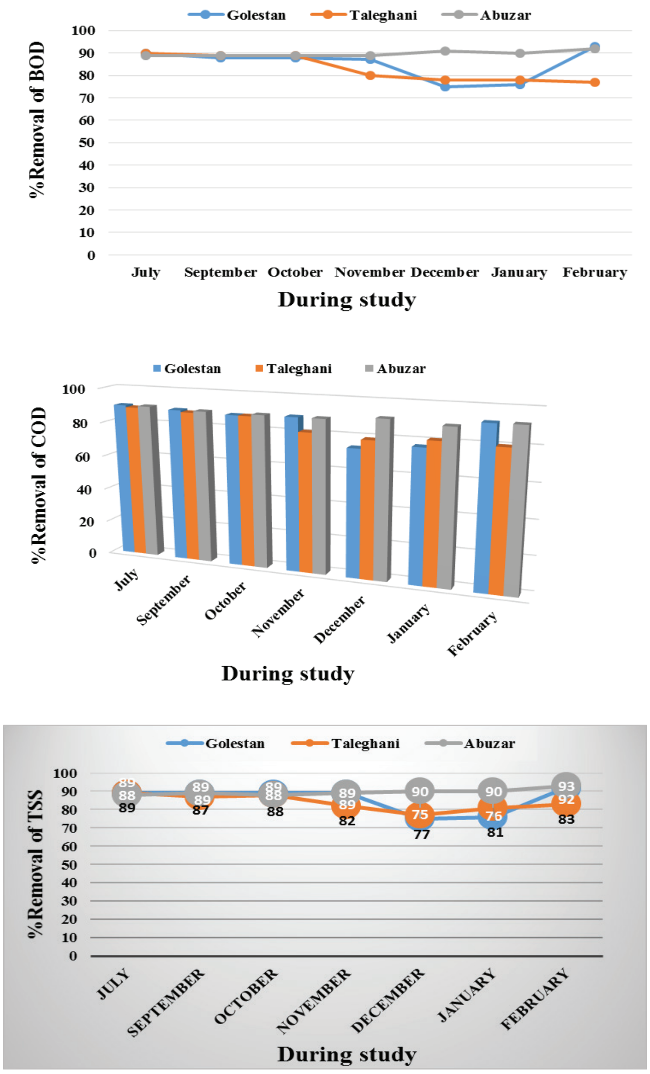

Fig. 3. Physicochemical parameters determined in the WWTP.

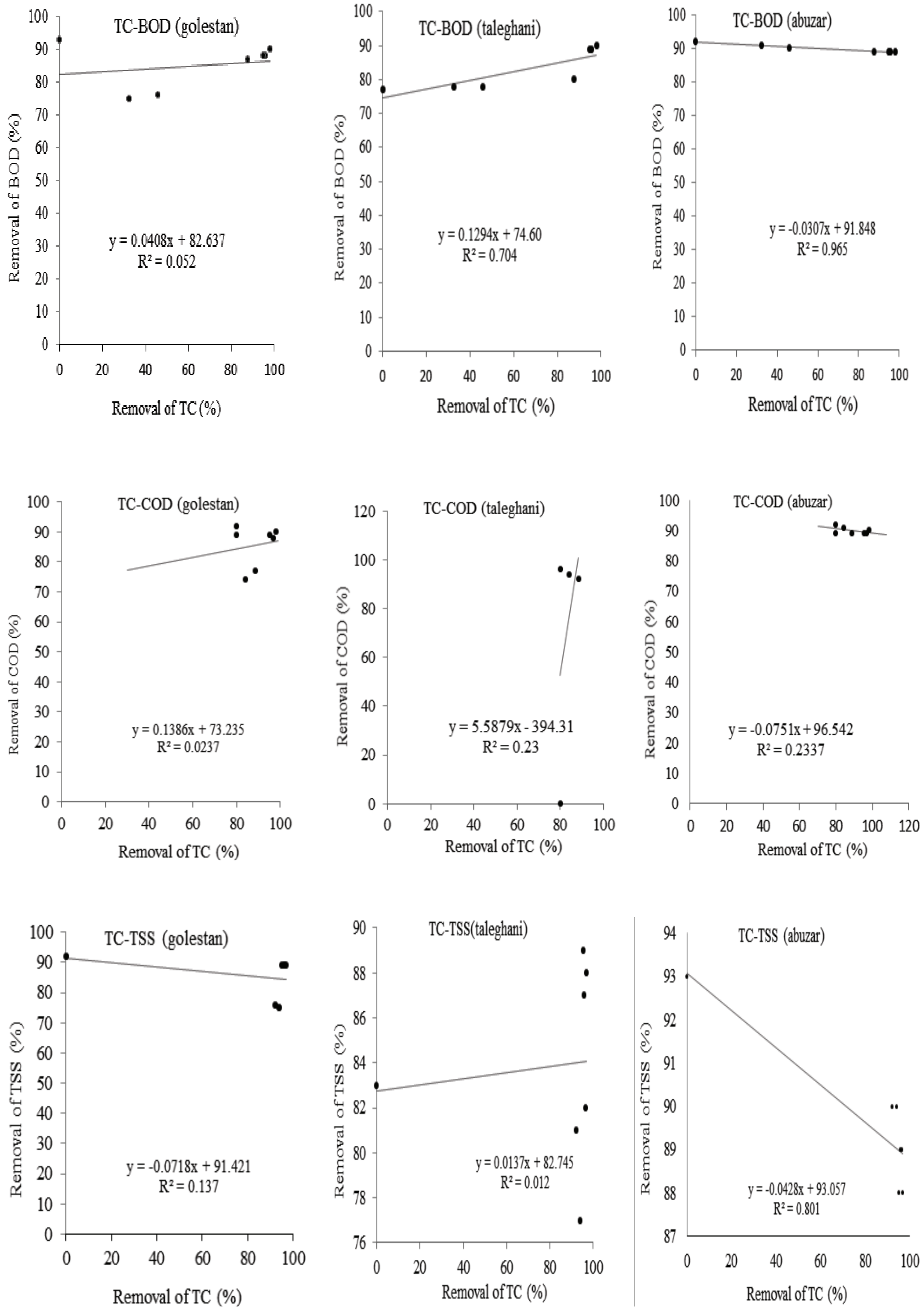

Fig. 4. Relationships between removal efficiencies of physicochemical parameters (TSS, BOD_5, COD) and TC.

and TC in Golestan, Taleghani and Abuzar hospitals in 2016. Based on the results presented in this figure, the correlation coefficients (R^2) between treatments TC and TSS, BOD_5 and COD were found to be 0.1, 0.01 and 0.8 in Golestan, Taleghani and Abuzar hospitals, respectively.

4. Discussion

The current study was an endeavor to assess TC antibiotic removal from the wastewater in Golestan, Taleghani and Abuzar hospitals in Ahvaz, Iran. In recent years, increased antibiotics consumption has been considered to be one of the serious threats to the public health and the environment of this metropolitan city. According to the results of this study, an average reduced TC by the wastewater treatment process at WWTPs was 80%–100%. Accordingly, a study conducted by Karthikeyan and Meyer [54] in Wisconsin in the United States demonstrated a relationship between Meyer, occurrence of antibiotics and treatment facilities. This study indicated that TC concentrations were generally reduced (70%–100%) by wastewater treatment process at WWTPs [54]. Based on the results of the study by Batt et al. [55], Watkinson et al. [39] and Lindberg et al. [56], the percentage of TC reductions was 68%–100%. Properties of the antibiotics, chemical structures, specific treatment processes and the sewage residence time are the major factors which are affected in antibiotic removal efficiency [53,29]. According to the results of this study, the highest rate of TC removal has occurred during treatment processes. Based on the previous study, the high removal rates of TC are attributed to sewage sludge, soils and sediments [57,58]. In the current study, the removal rate in the three hospitals was such that the average TC removal in Golestan hospital was 64%, while 97% was the highest removal rate in this hospital. TC removal rate in Taleghani hospital was 87%, while the highest removal rate in this hospital was 98%. The elimination of TC in the Abuzar hospital was 80%, while the highest removal rate was reported to be 95%. Furthermore, Abuzar hospital had the highest average treatment of TC. In another investigation which was done by Yu et al. [59] in 2009, the highest removal of antibiotics by extended sludge age biological process was determined to be 94%. The comparison of the result of this study and that of current study demonstrates that the final result is very similar. It should be noted that biological treatment process is one of the best processes for hospital wastewater treatment. Moreover, another study in Hanoi, Vietnam, by Duong et al. [33] evaluated the antibiotic resistance in hospital wastewaters. The results showed that the antibiotic elimination rate from wastewater was between 80% and 85% [33]. According to the results of this study, maximum removal efficiency of BOD_5 in the three hospitals was 90% for Golestan, 83% for Taleghani and 92% for Abuzar, respectively (Fig. 3). Also, the results of our study showed that the maximum treatment efficiency of COD in the hospitals was 92% for Golestan, 89% for Taleghani and 93% for Abuzar (Fig. 3). The calculated maximum removal efficiencies of TSS related to the WWTPs were 92%, 89%, and 93% in Golestan, Taleghani and Abuzar hospitals, respectively. In a study conducted by Michael et al. [41] on the effect of treatment of wastewater treatment plants (WTPs), COD removal was 93% and the removal of total organic carbon (TOC) was 78%. Relationships between the removal efficiencies of physicochemical parameters and TC are presented in Fig. 4. According to the results of our study, the higher relationships between removal efficiencies of TSS and TC was $TC_T>TC_G>TC_A$ (Fig. 4). Also, the findings of this study showed that the correlation coefficients (R^2) between treatment of COD and TC in Golestan, Taleghani and Abuzar hospitals were 0.02, 0.23 and 0.23. The order of importance of relationships between removal efficiencies of COD and TC (Golestan), TC (Abuzar) and TC (Taleghani) is as follows: $TC_G>TC_T>TC_A$ (Fig. 4). In terms of the relationships between removals of BOD and TC, the correlation coefficient ($R^2 = 0.05$) was obtained for TC in Golestan hospital. The correlation coefficient ($R^2 = 0.7$) was obtained for TC in Taleghani hospital. The correlation coefficient ($R^2 = 0.96$) was obtained for TC in Abuzar hospital. The order of importance of relationships between removal efficiencies of BOD and TC (Golestan), TC (Abuzar), TC (Taleghani) is as follows: $TC_G>TC_T>TC_A$. In a study conducted by Karthikeyan and Meyer [54] on the occurrence of antibiotics in wastewater treatment facilities in Wisconsin, USA, in 2006, it was found that the physicochemical parameters were weakly correlated with the removal of TC [54]. Hospital wastewaters consist of numerous pollutants, including antibiotics, heavy metals, Mercury, surfactants and organic matter that are very dangerous for humans and the environment. Extended aeration activated sludge (biological treatment), among many treatment processes, is a cost-effective method for the removal of antibiotics, especially TC. Biological treatment is a process that is capable of being an effective treatment process as the conventional methods.

4. Conclusion

Due to the limited dilution capacity of Karun River in Ahvaz, TC in wastewater especially hospital wastewaters can be very threatening for this river and its creatures. In this study, a detailed data analysis was carried out to find the removal of TC at Golestan, Abuzar and Taleghani educational hospitals of Ahvaz (located in the south-west of Iran), in 2016. In the present study, TC consumption is low in three hospitals of Golestan, Taleghani and Abuzar. Moreover, TC consumption rate is variable in warm and cold weather conditions. It should be mentioned that TC consumption is increased during infectious diseases, burns and surgical applications. Wastewater treatment with extended aeration can better remove TC (86%). Range of TC removal is from 80% to 97% with extended aeration. TC consumption was lowest during the weekdays of Friday and Saturday. However, it was high on Sundays.

Conflict of Interests

The authors declare no conflict of interests.

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