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Performance evaluation of a full-scale ABS resin manufacturing wastewater treatment plant: a case study in Tabriz Petrochemical Complex

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Abstract

Background: The measurement data regarding the influent and effluent of wastewater treatment plant (WWTP) provides a general overview, demonstrating an overall performance of WWTP. Nevertheless, these data do not provide the suitable operational information for the optimization of individual units involved in a WWTP. A full-scale evolution of WWTP was carried out in this study via a reconciled data.

Methods: A full-scale evolution of acrylonitrile, butadiene and styrene (ABS) resin manufacturing WWTP was carried out. Data reconciliation technique was employed to fulfil the mass conservation law and also enhance the accuracy of the flow measurements. Daily average values from long-term measurements by the WWTP library along with the results of four sampling runs, were utilized for data reconciliation with further performance evaluation and characterization of WWTP.

Results: The full-scale evaluation, based on balanced data showed that removal efficiency based on chemical oxygen demand (COD) and biochemical oxygen demand (BOD_5) through the WWTP were 80% and 90%, respectively, from which only 28% of COD and 20% of BOD_5 removal had occurred in biological reactor. In addition, the removal efficiency of styrene and acrylonitrile, throughout the plant, was approximately 90%. Estimation results employing Toxchem model showed that 43% of acrylonitrile and 85% of styrene were emitted into the atmosphere above water surfaces.

Conclusion: It can be concluded that the volatilization of styrene and acrylonitrile is the main mechanism for their removal along with corresponded COD elimination from the WWTP.

Keywords: Performance evaluation, Acrylonitrile butadiene styrene (ABS), Petrochemical, Toxchem model

Citation: Shakerkhatibi M, Mosaferi M, Zorufchi Benis K, Akbari Z. Performance evaluation of a full-scale ABS resin manufacturing wastewater treatment plant: a case study in Tabriz Petrochemical Complex. Environmental Health Engineering and Management Journal 2016; 3(3): 151–158. doi: 10.15171/EHEM.2016.14.

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Introduction

Acrylonitrile butadiene styrene (ABS) is an engineering thermoplastic resin, containing styrene, butadiene and acrylonitrile (ACN) monomers. ABS is the most commonly utilized copolymer in industries including appliance, electrical and electronics, consumer goods, construction, and automotive (1-3). In recent years, the demand for ABS resin has increased globally (2). The production capacity of ABS in Iran is 70 000 tons per annum. Among this, 35 000 tons of ABS is produced in Tabriz Petrochemical Complex (TPC) each year. The emulsion grafting-blend production technology was employed to produce various grades of ABS resin in TPC. In this production technology, α-methyl styrene (1100 tons per annum), ACN (8300 tons per annum), butadiene and other auxiliary agents were utilized as feeding materials, resulting to a toxic, refractory and complicated liquid effluent. The wastewater from ABS production unit is one of the typical high strength petrochemical wastewaters (4,5). Typical characteristics of effluents from ABS industries are shown in Table 1.

Conventional treatment of high strength ABS wastewater is normally based on biological processes. According to the literature, among the available biological treatment technologies, activated sludge process (ASP) is the most

Table 1. Typical characteristics of effluent from ABS industries

Parameter	Unit	References					
		(34)	(40)	(8)	(2)	(41)	(4)
pH	-	6.8-7.4	6.2-7.5	6.6-7.3	2.6-7.9	2-3.5	2.5 ± 0.4
T	˚C	-	25-35	25-35	-	-	-
COD	mg/L	4282-6985	2950-4410	2200-4700	1550-8050	1100-1300	1200 ± 100
BOD	mg/ L	75-3700	1200-1600	800-2400	272-400	400-500	450 ± 50
TKN	mg/ L	626-949	-	340-670	-	80-100	100 ± 10
ACN	mg/ L	185-292	-	-	-	-	-

common method employed for treating ABS resin manufacturing effluents with high organic loads (6-8). Nevertheless, it is not efficient for the removal of chemical oxygen demand (COD) and nitrogenous compounds such as ACN and cyanides from such wastewater (8). On the contrary, the ASP has two major disadvantages in petrochemical wastewater treatment, including the possibility of emitting volatile organic compounds (VOCs) into the atmosphere, due to the turbulence and aeration and the relatively high suspended solids which remained in the effluent (9).

Generally, the performance of wastewater treatment plants (WWTPs) is evaluated according to the effluent recommendations of the design for disposal. The data resulting from the measurements on the influent and effluent of WWTP provides a general overview, demonstrating an overall performance of the WWTP. Nevertheless, these data do not provide the appropriate operational information for optimizing individual units involved in a WWTP. To solve this problem, full performance evaluation is needed on individual units of a WWTP. This evaluation is required to assess the effluent quality, in order to meet higher treatment requirement and to also know the feasibility of handling higher hydraulic and/or organic loadings. Over the past two decades, several studies have been carried out to evaluate the performance of both municipal and various industrial WWTPs, worldwide (10-17).

In TPC, the wastewater from ABS resin manufacturing unit is pre-treated utilizing ASP as in-plant control system before discharging into the central WWTP, operated as the end-of-pipe treatment system for all TPC wastewater streams. This study aims to evaluate the performance of each operation and process unit involved in a full scale WWTP treating ABS effluents. In this study, long term information collected from the petrochemical central laboratory, accompanied by the data obtained from experimental measurements was utilized as the data base for performance evaluation. To reduce the (random) measurement error, data reconciliation was employed to enhance the accuracy of the measured data.

Methods

Description of the WWTP

The case evaluated in this study is a WWTP located in TPC northwest of Iran. It was designed based on an average flow-rate of 800 m³/d, which consists of three screens (P1), a grit chamber (P2), an equalization basin (P3), a fine screen (P4), a dissolved air floatation (DAF) system (P5), a

biological extended aeration activated sludge reactor (S1) and a clarifier (S2) (Figure 1). The design and operational parameters of the full-scale WWTP are shown in Table 2.

Analytical methods

Four sampling runs were conducted on July, August and October, 2014. Twelve-hour composite samples were collected at the inlet and outlet of treatment units. Sludge samples were also collected from returned and waste streams of primary and secondary clarifiers. The grab samples were collected from returned streams. All measurements were carried out during dry weather flow conditions. Dissolved oxygen and temperature were measured *in situ* utilizing a DO-meter (AQUALYTIC-AL20OXi). Wastewater samples were analysed for TCOD (5220 B), BOD_5 (5210 B), total dissolved solids (TDS) (1030 E), TSS (2540 E), Alkalinity (2320 B), CN^- (4500-CN^- N), total phosphorus (TP), PO_4^{3-} (4500-P-A), NH_4^+ (4500-NH_3 D), NO_3^- (4500-NO_3^- B), NO_2^- (4500-NO_2^- B), total kjeldahl nitrogen (TKN) (4500 - N_{org} B). Sludge samples were analysed on COD, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) based on

Table 2. Design parameters of WWTP

WWTP unit	Number	Volume (m³)	Depth (m)	HRT (h)
Grit chamber (P2)	1	12	-	0.37
Equalization basin (P3)	1	508	3	14.9
DAF (P5)	1	24	3	0.5
Aeration basin (S1)	2	766	2	22.5
Clarifier (S2)	2	105	2.6	3.1
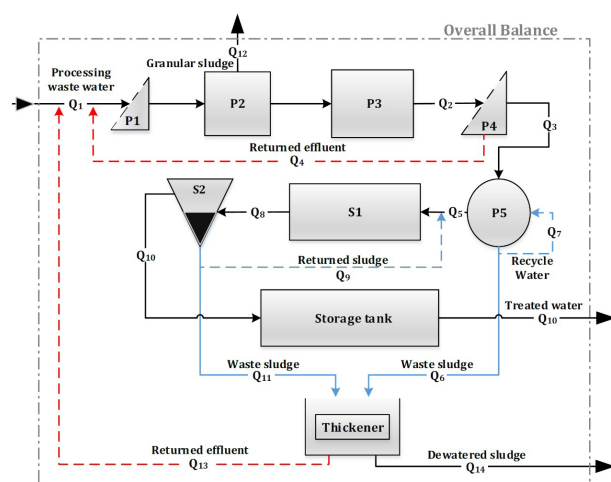
Figure 1. Schematic flow diagram for WWTP.

the standard methods for the examination of water and wastewater (18).

GC analysis

The collected samples (200 mL) from the influent and effluent of treatment units were extracted twice by 50 mL CH_2Cl_2 at the ambient temperature. Thereafter, 50 µL of each sample was injected into the GC (Varian Mod CP 3800) equipped with FID detector and a capillary column (CP-WAX 52 CB: 25 m × 0.32 mm × 1.2 µm) in order to determine the concentrations of ACN, acrylic acid, acrylamide and styrene. For ACN, acrylic acid and acrylamide analysis, the injector temperature was maintained at 170°C, while the column was programmed from 40 to 230°C at 40°C/min increments (3 minutes hold), and the detector was set at 230°C. For styrene analysis, the temperature was programmed from 40°C (1 minute) to 100°C at 20°C/min, maintaining this temperature for 1 minute. The detector and injector temperature were set to 300 and 230°C, respectively.

Data reconciliation

Measured data from WWTPs often contain gross errors, due to breakdown of the measuring devices, the process dynamics and variability of the influent loading. These errors led to mass imbalance of the WWTP and can also lead to incorrect process design, evaluation and modelling (3). Therefore, operational data should be verified before consideration (19,20). Data reconciliation (DR) is a technique proposed in the early 1950s for reducing the effect of random errors and improving (adjust) the measurement accuracies. This method, based on mass balance, is a well-known and generally used technique for detecting errors within data. The relationship between measurement value, true value and random measurement error can be mathematically written as (3,21,22):

$$y = x + \varepsilon \tag{1}$$

Where y is the vector of measurement values (noise free), x is the vector of model values and ε is the vector of random measurement errors. The aim of DR is to minimize the sum of squares of errors between measurements and model values, subject to a number of constraints (mean balance equations):

$$MIN\ (y-x)^T \psi^{-1} (y-x) \tag{2}$$

Subject to: $Ax = 0$

Where, ψ is the weight matrix and A is the process matrix (which is the balance equation).

A constrained optimization problem can be converted into unconstrained optimization problem by means of Lagrange multiplier method. Therefore, the problem can be solved analytically by introducing λ as the Lagrange multiplier (3,23):

$$L(y,\lambda) = (y-x)^T \psi^{-1}(y-x) - 2\lambda^T Ax \tag{3}$$

Substituting Equation (1) in Equation (3) gives:

$$L(y,\lambda) = \varepsilon^T \psi^{-1}\varepsilon - 2\lambda^T (Ay - A\varepsilon) \tag{4}$$

Considering ψ as the positive definite and the constraints are linear, the necessary and sufficient conditions for minimization are obtained after differentiating Equation (4) with respect to ε and λ and equating them to zero:

$$\frac{\partial L}{\partial \varepsilon} = 2\psi^{-1}\varepsilon + 2A^T \lambda = 0 \tag{5}$$

$$\frac{\partial L}{\partial \lambda} = A(y - \varepsilon) = 0 \tag{6}$$

Which yield Equations (7) and (8):

$$\varepsilon = -\psi A^T \lambda \tag{7}$$

$$\lambda = -(A\psi A^T)^{-1} Ay \tag{8}$$

The estimate of the process variable can be obtained as shown in Equation (9):

$$y_{estimated} = y - \varepsilon = y - \psi A^T (A\psi A^T)^{-1} Ay \tag{9}$$

This method was implemented in MATLAB and applied on measured flow rates.

Results

Characterization of the influent wastewater

Table 3 presents the influent wastewater characteristics as mean and standard deviation (mean ± SD). The data shown in Table 3 were calculated utilizing the results of 4 sampling runs and analyses throughout this study as well as comparison to the results of long-term measurements at the WWTP.

According to the data shown in Table 3, flow rate, TCOD, BOD_5, TSS, pH and cyanide are routine parameters which were measured periodically in recent years. Other parameters such as temperature, TKN, ACN and styrene were measured in this study to improve the evaluation of WWTP.

Data reconciliation

Table 4 shows the mass balance equations in a matrix format. The data shown in Table 4 show the interaction between the different flows and their effect on each com-

Table 3. Characteristics of the raw wastewater

Measurements	Units	Values		
		Measured (this study)	Long-term data	Design
Q_{ave}	m³/d	469 ± 57	538 ± 89	800
TCOD	mg/L	1345 ± 414	1377 ± 653	1200
BOD_5	mg/L	783 ± 473	647 ± 248	675
TSS	mg/L	649 ± 574	410 ± 354	300
pH	–	6.37 ± 0.16	6.16 ± 1.14	3
Cyanide	ppb	105 ± 31	126 ± 76	-
Temperature	˚C	34.5 ± 2.9	-	67
TKN	mg/L	61 ± 2	-	-
Acrylonitrile	mg/L	314 ± 100	-	-
Styrene	mg/L	123 ± 14	-	-

Table 4. Error diagnosis and data reconciliation of the flow measurements

Streams (m³/d)	Subsystem compartments					
	(P₁, P₂, P₃)	P₄	P₅	S₁	S₂	T
Q_1	469					
Q_2	-422	422				
Q_3		-529	529			
Q_4	45	-45				
Q_5			-472	472		
Q_6		-1				1
Q_8				-949	949	
Q_9				480	-480	
Q_{10}					-486	
Q_{11}					-3	3
Q_{12}	-0.5					
Q_{13}	1.7					-1.7
Q_{14}						-2.2
Errors	93.2	-152	56	3	-20	0.1

partment. Using the mass balance matrix as the input of the MATLAB program gives a unique solution and at the same time, increases accuracy of measured data.

Table 5 shows the results of the overall flow balance before and after reconciliation. As shown in Table 5, the measured data contained errors 19.7 m³/d. Calculation of operational conditions using these unbalanced data, leads to significant error which makes it practically impossible to employ such data for evaluation purposes (14). The standard deviation of balanced data obtained from reconciliation was decreased; and the overall mass balance of WWTP was satisfied.

Performance of individual units of the WWTP

Balanced flow rates were utilized in evaluating the performance of each treatment unit. Figure 2 shows the efficien-

cy of individual units of WWTP as the COD and BOD_5 removal. In addition, Table 6 illustrates the operating data of the biological reactor.

Figure 3 shows the daily variation of COD in the influent and effluent of WWTP during the study. As shown in Figure 3; in most cases, the effluent concentrations of COD were lower than design criteria (600 mg/l) across the study period. The effluent characteristics of WWTP are presented in Table 7. Figure 4 shows the fate of styrene, ACN, acrylamide and acrylic acid in WWTP.

Discussion

In line with the results of this study, the average influent flow rate during the study was significantly lower than design and long-term values. The lower flow rate increases the hydraulic retention time (HRT) in all treatment units except for the biological reactor. Higher HRT values in treatment units and simultaneously high temperature of wastewater along with the turbulence in the tanks, increase potential emissions of VOCs to the atmosphere (24,25).

According to the results, the COD and BOD_5 in the influent were 1345±414 and 783±473 mg/L, respectively, in which both were higher than the WWTP design values. Consequently, the BOD_5/COD ratio in the influent was calculated as 0.58 which indicates a good potential for biodegradability of the wastewater (26,27). It should be noted that the presence of some toxic and refractory compounds such as cyanide, ACN, etc., in the influent, may adversely affect the performance of biological system (28,29).

The overall COD and BOD_5 removal efficiency in WWTP were 80 and 90%, respectively. As can be seen, 52% (344 kg/d) removal of COD and 69% (266 kg/d) removal of BOD_5 have occurred in preliminary and primary treatment units while; these units are usually designed for

Table 5. Overall mass balance calculations (The positive and negative signs indicate inflows and outflows, respectively)

Streams (m³/d)	Measured data		Balanced data		
	Average ± SD	RSD (%)	Average ± SD	RSD (%)	Estimated error (%)
Influent flow	469 ± 57	12.2	487 ± 14	2.9	3.7
Effluent flow	− 489 ± 63	12.9	− 487 ± 13	7.4	− 0.42
Treated effluent	− 486 ± 62	12.8	− 484 ± 12	2.5	− 0.41
Waste activated sludge	− 2.2 ± 0.2	9.1	− 2.2 ± 0.2	9.1	0
Grit chamber effluent	− 0.5 ± 0.1	20	− 0.5 ± 0.1	20	0
Error in measurements	− 19.7	-	0.00		-
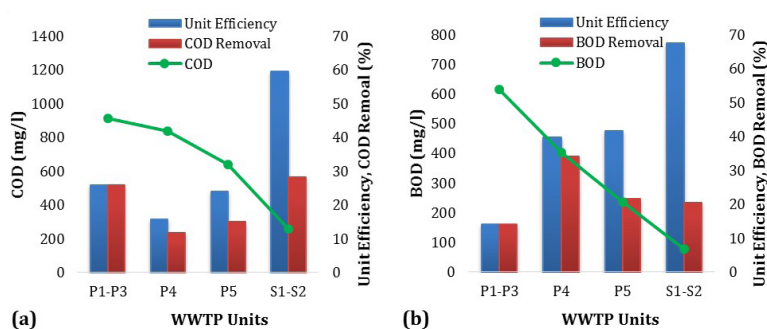
Figure 2. Removal efficiencies of individual units of WWTP as COD and BOD_5.

Table 6. Operational characteristics of biological reactor

Measurements	Units	Design data	Measured data	(26)
DO	mg/l	-	3.04 ± 0.47	-
pH	-	6.5-7.5	7.05 ± 0.13	-
Temperature	˚C	-	31.2 ± 2.8	-
HRT	h	22.5	19.4	20-30
SRT	d	-	21.2	20-40
SVI	ml/g	80-120	277 ± 42	-
MLSS	mg/L	2500-3000	1816 ± 236	2000-5000
MLVSS	mg/L	1500-1800	1408 ± 206	-
F/M	kgBOD/kgMLVSS.d	0.15	0.15	0.04-0.1
OLR	kgBOD/m³.d	0.45	0.28	0.1-0.3
RAS	% of influent	-	99.1	50-150
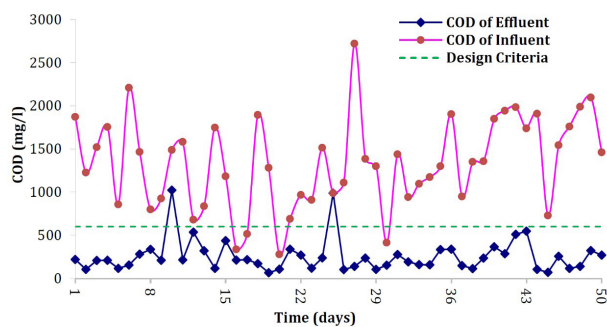
Figure 3. Daily variation of COD in the influent and effluent of WWTP.
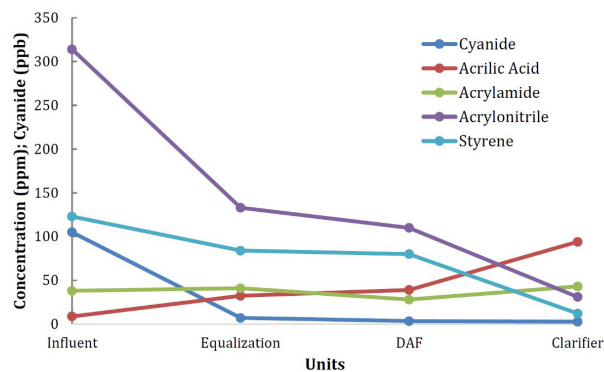
Figure 4. Variations of styrene, acrylonitrile, acrylamide, acrylic acid and cyanide in different units of WWTP.

Table 7. Characteristics of treated effluent

Measurements	Units	Design value	Measured data
DO	mg/l	2	2.1 ± 1.1
Temperature	˚C	-	30.3 ± 1.9
COD	mg/l	≤ 600	260 ± 45
BOD$_5$	mg/l	≤ 200	77 ± 13
TSS	mg/l	≤ 50	74 ± 37
Acrylonitrile	mg/l	-	31 ± 4
Styrene	mg/l	-	12 ± 2.5
Cyanide	ppb	2	2.7 ± 0.8

eliminating suspended solids as well as equalizing quantitative and qualitative parameters. Reduction in COD and BOD$_5$ prior to biological unit can be due to the removal of TSS in previous units such as fine screen and DAF and also VOCs stripping of volatile compounds into the atmo-

sphere. Since the removal rate of TSS as the main probable separated constituents of COD was fairly small before the aeration basin, it can be concluded that a major portion of COD removal at the upstream units of the biological reactor has probably occurred as a result of VOCs stripping to the atmosphere. It is estimated that about 84% of total VOCs in a petrochemical wastewater can be released into the atmosphere from the WWTP through stripping process (30). These undesirable emissions can be considered as one of the sources of air pollution, having serious adverse health effects on human (31,32).

Figure 2 shows average BOD$_5$ and COD removal of 68 and 59%, respectively, across the biological reactor at an average organic loading rate (OLR) of 0.236 kgBOD/m³.d. Generally, the efficiency of biological treatment process severely depends on the reactor design and operational conditions. The operating data of the biological reactor (Table 6) shows that some parameters are out of the range when compared to the WWTP design and literature recommended values.

The mean value of MLSS within the activated sludge was 1816 ± 236 mg/L, while it should be in the range of 2500-3000 mg/L in line with the plant design. Also, the mean value of MLSS measured in returned activated sludge (RAS) was 3410 ± 564 mg/L. According to literature, the MLSS concentration of RAS from clarifiers should typically be in the range of 4000 to 12 000 mg/L (26). However, the RAS pumping rate was 99.1% at an average flow rate; the lower values of MLSS in RAS have failed the return stream to maintain a sufficient concentration of activated sludge in the aeration tank to obtain the necessary degree of treatment in the desired time interval. It seems that it can be the cause of insufficient removal rates of BOD$_5$ and COD across the bioreactor. The low values of MLSS have led to the increase of the food to microorganism (F/M) ratio (up to 0.168 kgBOD/kgMLVSS.d) within the bioreactor.

As shown in Table 6, the HRT in aeration tank was 19.4 hours at average flow, while the corresponding design value was 22.5 hours. The reduction in HRT was as a result of the RAS flow rate from clarifier. The decrease in MLSS concentration and HRT in the bioreactor may severely affect the performance of organic compounds oxidation and nitrification processes, which can lead to process un-

steadiness and poor quality of the effluent.

The results indicated poor removal of TSS in DAF system (54±15%). Also, the COD and BOD$_5$ removal efficiencies in DAF system were 24±9 and 42±6%, respectively, which show variations in the unit's performance over the operating period. Unsteady characteristics of the influent, shows the inadequacy of the chemical additions and the full-influent pressurization in the DAF system which can be considered as the main reasons for the unstable performance of DAF system.

Generally, the WWTP demonstrated sufficient removal efficiency with respect to COD, BOD$_5$, ACN and styrene. Although TSS removal efficiency of the plant was 90%, TSS concentration in effluent was higher than design value which was mainly due to the higher concentration of TSS in the influent and the poor removal performance of screening and DAF.

Nitrile compounds and their derivatives are cyanide-substituted carboxylic acids that have an R-CN structure. Most nitriles are highly toxic and some are mutagenic and carcinogenic. ACN contains a –CN group which is toxic to organisms and it is not easily decomposed biologically (33). As shown in Figure 4, preliminary treatment units (grit chamber and equalization tank) removed the major portion of ACN (58%) and cyanide (94%). The high removal rate in equalization tank may be due to the turbulence effect created by high aeration rate (32).

As shown in Figure 4, the acrylic acid concentration increased and accumulated with the removal of ACN. Wang and Lee (34) indicated that the microorganisms within the biological reactor use ACN as a substrate which leads to the accumulation of acrylic acid in the system. After the complete removal of ACN, the acrylic acid is consumed. Therefore, incomplete removal of ACN in the biological treatment can be as a result of the low HRT. In the case of styrene, 31% was removed in preliminary treatment units. The styrene removal efficiency of the biological reactor was 85%. As earlier mentioned, the large portion of styrene and ACN can be emitted into the atmosphere from the surfaces. All of the styrene and ACN removal (90%) throughout the WWTP are not related to biodegradation or elimination by other effluents such as granular or dewatered sludge.

Toxchem model (Environmega), which was designed to model the fate of toxic organic chemicals in wastewater treatment processes and regarded by USEPA as appropriate models for air emissions estimation (35-37), was employed to estimate the portion of air emission of each component. The results revealed showed that 43% (65 kg/d) and 85% (51 kg/d) of entered ACN and styrene were emitted into the air, respectively, in which the equalization basin had the major portion of the air emission. The high emission rate from equalization basin is as a result of high concentration of pollutants in the basin and its large surface area for evaporation (25,38). Cheng and Chou (39) indicted that the turbulence effect could result in total VOC emission, increasing from 46 to 90%.

Conclusion

A full-scale evaluation of the ABS WWTP was conducted in this study. The method of error detection and data reconciliation was employed to correct raw flow rate data and fulfil the mass balance. The evaluation results, using balanced data showed that 52% (344 kg/d) and 69% (266 kg/d) loss of COD and BOD$_5$, respectively, occurred in the upstream units of the biological reactor. Nevertheless, removal efficiency of biological reactor was 59 and 68% for COD and BOD$_5$, respectively. The results showed inadequate TSS removal of DAF system, which was mainly due to inadequacy of the chemical additions as well as the full-influent pressurization of the system. Investigating the fate of styrene and ACN by means of Toxchem model revealed that a large portion of these were emitted into the atmosphere. The high emission rates from low height area sources can lead to high concentration of considered toxic pollutants in the ambient air around WWTP.

Ethical issues

We certify that all data collected during the current study was presented in this manuscript; no data from the study has been or will be published separately. The local ethical review committee of the Tabriz University of Medical Sciences approved the study (Ethical No. B/255).

Competing interests

The authors declare they have no competing interests.

Authors' contributions

All authors were involved in study design, data collection, and article approval.

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