

Evaluation of Wastewater Treatment Plant in Pollution Control of Petrochemical Industries: A Case Study: of Abadan Petrochemical Company

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Abstract

The function of petrochemical wastewater treatment plants should be evaluated intermittently, due to the increase in production and new effluent flow and organic load to the wastewater. In this study, the efficiency of a petrochemical wastewater treatment plant has been evaluated by sampling and determination of major pollutants of wastewater during six months. The results showed that physical, chemical and biological processes used in the plant, succeeded to decrease the COD mean value of wastewater to 80 mg L^{-1} level. The average overall yields of the total hydrocarbons, volatile organic compounds, phenols and sulfides removal were measured as 77%, 97%, 99.8% and 94.2%, respectively. The majority of volatile organic compounds (88.5%) and sulfides (78.5%) were decreased by stripping and chemical oxidation. The results showed that the existing wastewater treatment plant was successfully assessed to decrease the organic pollutants, while total suspended solids increased slightly during treatment processes although remained within the authorized limit.

Keywords: Activated sludge, Petrochemicals, Phenols, PVC, Wastewater.

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چکیدہ

با توجه به تغییرات کمی و کیفی پساب تولیدی در واحدهای پتروشیمیایی که ناشی از تغییر ظرفیت واحد و تخلیه ترکیبات جدید به واحدهای تصفیه یساب است، عملکرد واحدهای تصفیه یساب آنها می باید در بازههای زمانی مختلف مورد بررسی قرار گیرد. در این تحقیق، بازدهی یکی از واحدهای تصفیه پساب صنعت پتروشیمی با نمونه برداری از پساب و اندازه گیری آلاینده های مهم در مدت شش ماه مورد ارزیابی قرار گرفته است. نتایج نشان داد که فر آیندهای فیزیکی، شیمیایی و بیولوژیکی مورد استفاده در واحد تصفیه پساب، باعث کاهش مقدار اکسیژنخواهی شیمیایی (COD) یساب تا حد ۸۰ mgL⁻¹ شده است. مقادیر متوسط بازدهی کلی حذف هیدرو کربن ها، مواد فرار آلی، فنل ها و سولفیدها از پساب به ترتیب ۷۷، ۹۷، ۹۹/۸ و ۹۴/۲ درصد اندازه گیری شد. فر آیندهای عریانسازی و اکسیداسیون شیمیایی قادر به حذف عمده ترکیبات آلی فرار (۸۸/۵ درصد) و ترکیبات سولفیدی (۷۸/۵ درصد) از پساب بودهاند. در حالی که سیستم تصفیه پساب موجود قادر به کاهش آلایندههای آلی بوده ولی مواد جامد معلق پساب طی مراحل مختلف تصفیه کمی افزایش یافته است، اگرچه مقادیر آن همچنان در محدوده استاندارد قرار گرفته است.

كلمات كليدى: پساب، بىوىسى، لجن فعال، پتروشيمى، فنل ها.

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Introduction

Wastewater (WW), generated by catalytic and thermal processes of petrochemical plants in addition to suspended solids, contains hydrocarbons (paraffinic and aromatic), chlorinated hydrocarbons, and sulfides. One of the major problems the industrialized world is facing today is the contamination of soil, groundwater, sediments, surface water and air with hazardous and toxic chemicals. The long-term production and use of different petroleum derivatives have caused widespread contamination of water and soil around facilities handling those (Morelli et al., 2005). Petrochemical plants are typically large and complex, where the combination and sequence of processes is usually very specific to the characteristics of the products manufactured. Due to the toxic effects of some chemical compounds existing in many petrochemical industry effluents, a combination of physicochemical and biological treatments of such wastewater is usually used for reliable efficiencies (Wang et al., 2009). The levels of COD and some chemical contents of wastewater are important in the effluents of treatment plants. Gravity separation and coagulation methods are usually used for removing oil and grease in the wastewater, which requires further treatment by chemical or biological oxidation for complete mineralization (Jördening and Winter, 2005). Chemical oxidation methods are fast, but they are expensive and may result in the formation of undesirable by-products. Biodegradation of petrochemical compounds by aerobic or anaerobic biological treatments is more specific and relatively inexpensive (Pophali et al., 2007). Activated sludge is a well-established biological process in treatment of petrochemical WWs. Most of the petrochemical plants in Iran use "activated sludge process" for treating their WWs.

Abadan Petrochemical Company (APC) is a petrochemical complex, located in the South of Iran, producing mainly poly-vinylchloride (PVC). APC plant consists of different sub-units, namely: Olefin, Ethylene dichloride, Vinyl chloride, Tetramer, Chloralkali and PVC polymerization (Abadan Petrochem. Co. website). The wastewater produced by APC contains chlorinated chemicals in addition to different hydrocarbons. It also contains some sulfides which are mostly due to sour feed of the plant. After passing different treatments, the effluent of APC wastewater discharges into Arvand Rood, a river which flows to the Persian Gulf. The water flow rate of Arvand Rood at 450 $m^3 S^{-1}$, makes it the largest river in Iran.

WW discharges into international waters have been gaining considerable attention during recent years. In Iran, the majority of petrochemical plants are located in the vicinity of the Persian Gulf or rivers flowing into the Persian Gulf. The Abadan petrochemical plant producing polyvinyl-chloride faced an increase in WW production due to the plant's growth in recent years. For this reason, the present work has been done to evaluate the efficiency of its wastewater treatment plant (WWTP) and to check the conformity of WW effluent characteristics to environmental standards. In this regard, some modifications might be suggested for further improvements.

Materials and Methods

The field site (Abadan Petrochemical Company) is located in Abadan, a city in the South of Iran. The mean annual maximum temperature and atmospheric humidity of the site were 46.5 °C and 63%, respectively. The wastewater treatment plant (WWTP) of APC consisting of primary (oil separation, equalization, stripping), secondary (biological treatment, settling), and tertiary (chlorination) sections, treats an average wastewater flow of 3600 m³ day⁻¹ (Fig. 1). The industrial wastewater mainly consists of two streams which are treated separately: (a) The effluents of PVC production and Chlor-Alkali units (flow rate: 65 $m^{3}h^{-1}$) that is characterized by a low concentration of organic content (COD $<100 \text{ mgL}^{-1}$), and (b) effluents from other production units (flow rate: 85 m³h⁻¹) that are characterized by a higher concentration of organic compounds (COD≈ 20000 mgL^{-1}). Stream (a) contains PVC suspended particles which are settled in a separation tank. Due to high pollution load, the b-Stream is treated physically and biologically before being diluted by the a-stream. The majority of oil and grease of the (b)-stream is removed by a physical purification system using two-stage gravity separators (namely API), resulting in a COD reduction from 20000 to around 1000 mgL⁻¹. An equalization tank permits homogenization of different WW streams as well as the stripping of volatile compounds using surface aerators. The biological stage of WWTP consists of a single-step activated sludge tank and the sludge clarifier. The activated sludge process operates at 25-37 °C during the year. Diverse species of prokaryotic and eukaryotic organisms act on wastewater under aerobic conditions for biodegradation of contaminant compounds. The hydraulic residence time and mean cell residence time of the activated sludge tank are 30 hours and 15 days, respectively. Chlorination is the final treatment of WW after being clarified in clarifiers. The effluents of chlorination

process (b-stream), Chlor-Alkali unit and clarified WW of PVC unit (a-stream) are mixed and homogenized before discharging to Arvand Rood (See Fig. 1).

Sampling

Six sampling runs were operated in February, March, May, June, July and August of the year 2009. Air temperatures were, correspondingly, 30, 26, 45, 43, 44 and 45 °C. Composite samples were taken from wastewater effluent of the API oil separator (point 1, in Fig. 1), while a simple sampling method was used for effluents of the equalization tank (point 2), clarifier of activated sludge (point 3), chlorination (point 4), and final disposal to the Arvand Rood (point 5). Average values for the composite samples have been reported. Sampling methods and preservation of the samples were performed based on standard methods (Eaton *et al.*, 1995).

Materials

Solvents (n-Hexane, Ethylene dichloride, Benzene and



Figure 1- Schematic diagram of WWTP of APC. VCI: Vinyl chloride; EDC: Ethylene dichloride; PVC: Polyvinyl chloride; DDB: Dodecyl benzene; Tetramer: Propylene tetramer. The circles show the sampling points.

MTBE) were purchased from Merck Co. COD vials were prepared from HACH Co. Helium gas was 99.99% pure and obtained from Roham Gas Co., Tehran, Iran. All chemicals were analytical grade and obtained from commercial sources.

Analysis

Dissolved oxygen (DO) and temperature of WWs were measured by HACH DO-meter model SENSION 6. Total suspended solids (TSS), sulfides and phenolic compounds were measured by a photometric method using HACH spectrophotometer model DR/2400 compared to standard curves. Total dissolved solids (TDS) were measured by conductivity determination of the solution by HACH conductometer model 50150.

COD analysis was conducted by reactor digestion HACH method 8000, which was approved by USEPA as per Federal Register, April 21, 1980, 45 (78) 26811-26812, using a low range vial (0-150 mg/L) and a high range vial (0-1500 mgL⁻¹). HACH methods are described in detail in the HACH' Odyssey DR/2400 spectrophotometer manual. Chloride is the most important interference when determining COD Each COD vial contains mercuric concentration. sulfate that eliminated chloride interference up to level specified by the method. Total hydrocarbons were quantified gravimetrically as solvent soluble fraction and are reported as oil and grease content (Eaton et al., 1995). The extraction solvent was consisted of 80 vol% n-Hexane and 20 vol% MTBE.

Concentration of vinyl chloride monomer (VCl) was measured by a gas chromatograph Shimadzu 14A equipped with FID detector. The GC operated under isothermal program at 50 °C. The injection port and detector temperatures were 120 and 150 °C, respectively. The carrier gas was Helium 99.99%. The column was DB1 (25 m X 0.53 mm, film thickness: 1µm) bounded phase fused silica. After solvent extraction, the concentrations of ethylene dichloride (EDC) and benzene were analyzed by GC Chrompack Model CP-9001 using Carbowax 600 packed column. The column was maintained at 50–65 °C with increase rate of 0.7 °C min⁻¹. The injection port and detector temperatures were 170 °C.

Results

Six sampling runs were carried out during February to August to monitor the variation of the pollution load of industrial wastewaters of APC and the efficiency of its WWTP in removal of contaminants. Five different points have been allocated for taking samples (See Fig. 1). The effluent of the APC to Arvand Rood has been compared to the standards of the Department of the Environment, Islamic Republic of Iran (Alavi Nasab, 2002).

Variation of temperature and dissolved oxygen

Table 1 presents the temperature of sampling points during six months, which clearly shows two different temperature regimes: February-March, and May-August. The highest temperatures were measured at sample point 1 where the average temperature was 41.1 °C during May-August. This average temperature reduced to 36.9 and 32.3 °C for sample points 2 and 3, respectively, as a consequence of aeration in the equalization and activated sludge tanks. The average temperature of WW in activated sludge tank was measured as 31.7 °C.

Variation of dissolved oxygen in streams of WWTP is demonstrated in Fig. 2. The DO concentration is an environmental factor that may inhibit the biological reactions as the growth rate of aerobic microorganisms is reduced at low DO. By using surface aerators, DO values at equalization tank and biological treatment were kept above 4 mgL⁻¹, which was necessary for efficient metabolism of microbial communities.

Variation of suspended and dissolved solids

As the solid particles have a substantial effect on the amount of organic matter, the concentrations of total suspended solids (TSS) were measured (Fig. 3). At all runs, TSS content of wastewater increased from API

Sampling Run	Sampling Point Temperature (°C)					
	1	2	3	4	5	AST*
February	30.0	29.0	24.0	24.0	31.0	27
March	29.0	26.0	21.6	21.2	29.5	25
May	40.8	35.6	31.5	30.8	38.2	35
June	41.9	38.2	33.3	33.7	37.5	34
July	39.5	35.5	31.3	29.6	35.0	32
August	42.2	38.4	33.1	33.0	39.8	37

Table 1- Variation of wastewater temperature at different runs and sampling points.

*AST: Activated sludge tank

separators to the equalization stage which was due to the addition of other WW streams to the equalization reservoir. In the February, March and June samples, suspended solids of clarified WW (point 3) showed an increase compared to point 2, which was the result of clarifier malfunctioning. In other sampling runs, the clarifiers significantly decreased the TSS values. A considerable increase in TSS content of WWTP effluent to the Arvand Rood was observed, which obviously originated from mixing different streams in the mixing reservoir (please refer to Fig. 1), as well as sludge losses. Quantities of TSS values in WWTP effluent were measured as 43 to 143 mgL⁻¹, which in all cases were out of standard limits (i.e. 40 mgL⁻¹) (Alavi Nasab, 2002).



Figure 2. Dissolved oxygen concentrations in effluents of each WW treatment.



Figure 3- Total suspended solids of WW streams after each treatment.

The variation of total dissolved solute (TDS) during six sampling runs is shown in Fig. 4. The TDS concentrations in WWTP effluent ranged 2800-7800 mgL⁻¹. In most sampling runs, TDS values of WW decreased slightly in equalization tank because of dilution in other streams. The solute concentrations remained nearly unchanged during further processes. Mixing of a- and b-streams resulted in increase of 36, 54, 64, 31, 33, and 216 percent in TDS compared to treated b-stream (i.e.chlorination effluent).



Figure 4 - Total dissolved solute of WW streams in different sampling runs.



Figure 5- Chemical oxygen demand (COD) of sampling points in six runs.

Removal of organic compounds

Organic compounds constitute the main pollutant fraction in most petrochemical plants. The majority of heavy hydrocarbons were separated in API separators which resulted in a considerable decrease of COD from 20000 to less than 1400 mgL⁻¹. Chemical oxygen demand (Fig. 5) and total hydrocarbons (Fig. 6) are determined to evaluate the efficiency of WWTP in the degradation of organic pollutants. As can be seen from the figures, COD has been decreased from the API separators to the effluent of the equalization tank,

which is attributed to dilution of the WW streams, as well as stripping of volatile compounds by aeration in equalization reservoir (see Fig. 7). During February to August, the biodegradation yields of activated sludge were determined as 66.4, 63.4, 87.8, 89.1, 57.9, and 60.8% in regard to COD reduction, where the temperature was measured in the range of 25 to 37 °C. Also, different ciliated protozoan and rotifer communities were observed in microscopic examination of the activated sludge.



Figure 6 - Total hydrocarbon Concentrations of different WW streams in six sampling runs.

The concentrations of volatile compounds (ethylene dichloride, vinyl chloride and benzene) are plotted in Fig. 7. At all sampling runs, the volatile compounds were decreased considerably after equalization/stripping stage due to aeration (point 2). The remainder volatiles in WW were ceased after biological treatment where biodegradation as well as air stripping were occurred. An increase in vinyl chloride concentration was observed at sampling point 5, where effluent of PVC production unit mixed with the biologically-treated stream. The effluent of PVC unit

contains higher concentrations of vinyl chloride, as it is used as the monomer of polymerization process.

The decreasing trend of total hydrocarbons was similar to COD variation (Fig. 6). The overall hydrocarbons removal in WWTP was in the range 70–90%. The biological treatment was the stirring process in hydrocarbon removal, where biodegradation yields of 63, 53, 41, 80, 30, and 63% were assessed at six sampling runs. Total hydrocarbon contents on WWTP effluent were less than 10 mgL⁻¹, in most of the sampling runs.



Figure 7- Variation in concentrations of volatile compounds during WW treatments at five sampling points (refer to Fig. 1) during six months.

The removal of phenols at different stages of WWTP of APC during March-August is shown in Fig. 8. The concentrations of phenols were very low, at level of 1 mgL⁻¹, however, they were traced at different WW streams because of high toxicity and severe environmental regulations ($<1 \text{ mgL}^{-1}$ in local standards). Reduction of phenolic compounds in the equalization process was observed because of WW dilution. Biodegradation yields of phenolic compounds in sampling run March-August were measured as 95.6, 96.4, 95.8, 95.9, and 95.0%, respectively.

Abatement of sulfides

Sulfides are toxic and corrosive compounds that cause environmental and economic problems in a variety of sectors, such as petrochemical industries. The concentration profiles for sulfides in the effluent of different streams are shown in Fig. 9. The highest amount of sulfides was determined in June at around 7600 mgL⁻¹. Due to sour feed of the plant (i.e. propane and butane produced from the cracking of crude petroleum), relatively high concentrations of sulfides were produced in Olefin unit, which were discharged into the WW.



Figure 8 - Variation of phenol concentrations at five sampling points.



Figure 9 - Variation of sulfide concentrations at five sampling points during six months.

The equalization tank was the predominant stage in sulfide reduction. The results showed that 44-98% of sulfides were removed in the equalization tank. Higher sulfide reduction yields (more than 83%) were observed in May, June, and July, when the WW temperatures within the equalization tank were above 35°C. Evidence denoted the occurrence of oxidation of sulfides to a more benign form of sulfate in the equalization tank in the presence of low concentrations of oxidizing cations such as manganese and cobalt in addition to a high rate of aeration. The concentration of sulfides in the WW effluent of activated sludge treatment were in the range 2-42 mgL⁻¹, respectively, which were higher than permitted values (3 mgL^{-1}) (Alavi Nasab, 2002). Adding traces of manganese and cobalt compounds to the equalization reservoir has been proposed which may accelerate chemical oxidation of sulfides and keep them within the permitted concentrations.

Discussion

Temperature and dissolved oxygen

The average temperature of WW within the activated sludge tank was measured as 31.7 °C during the whole sampling period of this research (Table 1), which favored the growth and activity of mesophilic microorganisms. An increased WW temperature has a positive influence on biological WW treatment methods, since an increase in temperature also increases the activity of microorganisms. On the other hand, any temperature increase results in a higher volatilization rate of volatile compounds, and lower oxygen input capacity of the aeration system (Rosenwinkel et al., 2005). In previous studies, the temperature transients is reported to be associated with poor sludge settleability, more negatively charged sludge, increased filament abundance, and decreased concentrations of protozoa and metazoa. Although, the controlled, periodic temperature oscillations (such as seasonal changes in this case) was shown to have a slight impact on COD removal efficiency, and was not seemed to affect robust microorganisms that withstood

the temperature shift (Morgan-Sagastume and Allen, 2003). Wastewater temperature in an activated sludge processes varies much less than some other biological processes, e.g. trickling filters and lagoons. A temperature variation of ± 6 °C from the average value of 31.6 °C was observed, the variation which is expected for activated sludge processes in most treatment plants (Wang et al., 2009).

At all sampling runs, the DO values in effluent of WWTP were measured above 3 mgL^{-1} (Fig. 2) which satisfied the local environmental standard (i.e. 2 mgL^{-1}) (Alavi Nasab, 2002).

Suspended and dissolved solids

A considerable increase in the TSS content of WWTP effluent to the Arvand Rood was observed (Fig. 3), which obviously originated from mixing of different streams in mixing reservoir, as well as sludge losses. TSS values in WWTP effluent ranged 43-143 mgL⁻¹ that in all cases were outside standard limits (i.e. 40 mgL⁻¹). The suspended solids have been originated from biological sources (biomass losses of activated sludge tank) and mineral sources (sand, silt etc. caused by washing of plant area). Use of coagulation aids before sludge clarifier and addition of another settling tank before final discharge to the river has been proposed to solve the problem.

The dissolved solid concentrations in WWTP effluent ranged 2800-7800 mgL⁻¹ (Fig. 4). Majority of the solutes consisted sodium chloride which was drained off from the Chlor-Alkali unit. Comparison of the flow rates of Arvand Rood and WWTP effluent (450 vs. 0.05 m³S⁻¹) indicates 9000-times dilution of WWTP effluents in the river. Based on the environmental regulations, the river TDS should not be changed more than 10% in a distance of 200 m from the plant effluent discharging to the river (Alavi Nasab, 2002). The APC WW effluent has not exceeded the authorized value, since the TDS value of the river changes within the range of 5000-8000 mgL⁻ ¹ and considering very high dilution of WW into the

river flow.

Organic compounds

The measurements show that the microorganisms of WW are adapted to the temperature range 25-37 °C. A microscopic examination of the activated sludge showed different ciliated protozoan and rotifer communities, which reveals the high efficiency of WWTP (Bitton, 1999). However, refinery and petrochemical wastewaters contain refractory organics which are hardly biodegraded. In addition, some refractory organics are highly toxic and inhibitory to microbial activity which may lead to poor effluent quality and unstable operation. Therefore, despite the high residence time of WW in biological stage, i.e. 30 h, COD quantities lower than 72 mgL^{-1} were not attained (Fig. 5).

The Values of COD reduction (61-89%) obtained in this study were comparable to the custom activated sludge processes (Wang et al., 2009). Transient, nonsteady state conditions in biological WW treatment are common, and can be caused by changes in substrate and nutrient characteristics or concentration, and by changes in the environmental temperature. The overall COD removal yields during WW treatments (i.e. from the effluent of API separators to the discharge to the river) were 90.6, 92.2, 92.3, 92.4, 82.5, and 88.7% at six runs. The effluent COD of WWTP was lower than 100 mgL⁻¹ which met the local standard regulations (Alavi Nasab, 2002).

The average reduction in volatile constituents in stripping process of WWTP was measured as 88.5% (Fig. 7). A higher stripping rate was achieved for vinyl chloride due to its higher Henry's law constant (i.e. 2.4 atm-m³mol⁻¹) (Bitton, 1998). Overall mean values of EDC, VCl and benzene removal yields during six months were 100, 91.3, and 100%, respectively. However, suitable treatment should be considered for the volatile compounds stripped to the air.

Considerable reduction in phenols in activated sludge treatment was observed (more than 95%) in this study (Fig. 8) which reveals the presence of microorganisms having the enzymatic pathway of phenol degradation. Many microbial strains capable of degrading phenol have been reported: Pseudomonas

putida, Pseudomonas fluorescens, Acinetobacter, Trichosporon cutaneum and Candida tropicalis. Most of them are capable of degrading phenol at low concentrations. However, phenol is toxic to most type of microorganisms at a sufficiently high concentration and can be a growth rate inhibitor. Also, fungi strains have been reported to be active in phenol biodegradation. Nocardia hydrocarbonoxydans, an actinomycetes, was found to effectively degrade phenol and showed higher inhibitory concentration level (Busca et al., 2008). Phenol biodegradation yield in seawater was previously reported as 35 and 67% using activated sludge and two species of Acenitobacter at an initial concentration of 100 mgL^{-1} (Kobayashi et al., 2004). Complete biodegradation of phenol in seawater using Serratia sp. a novel bacterium Aplysia kurodai has been also reported (Kobayashi et al., 2007). However, biosorption of organic compounds, e.g. phenol, on activated sludge should not be ignored. Slight oxidation of phenols by chemical oxidation was seen in chlorination process (Fig. 8). Using chlorine as an oxidant was also reported in the literature as an alternative oxidation procedure for phenol removal (Busca et al., 2008).

The results reveal that the primary treatment of APC wastewater reduces the majority of heavy hydrocarbons and volatile organic compounds. Considerable removal of COD, total hydrocarbons and phenolic compounds was achieved in the secondary stage (bio-treatment). Tertiary treatment (chlorination) affected oxidation of phenols, COD and hydrocarbons.

Sulfides

The outcomes of present study showed that 44-98% of sulfides were removed in equalization tank. In an alkaline aqueous solution (as in the conditions of equalization tank, where the pH value varies between 8.8-11.9), sulfide compounds can be photooxodized to form sulfate in the presence of some photo-active agents which are present in the wastewater (Iliev and Mihaylova, 2002). However, it should be noted that the concentration of sulfide is pH-dependent, and at a slightly alkaline pH, sulfides are precipitated as heavy metal sulfides (Jordening and Winter, 2005). The residence time of WW in equalization reservoir, 6 h, was enough for oxidation/precipitation of sulfides.

In conclusion, the present study finds that all environmental parameters of wastewater were below the authorized values of local standard regulations except TSS and sulfides (Alavi Nasab, 2002). To solve the problem, addition of chemical oxidants to the equalization tank for reduction of sulfides and use of coagulation aids before sludge clarifier for decreasing TSS quantities has been suggested.

Although, sophisticated wastewater treatment processes have been developed during recent years, the results of this study showed the effectiveness of well-established traditional wastewater treatment process (activated sludge) in reduction of organic pollution load in petrochemical industries.

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