Assessment of Volatile Organic Compounds Removal in Kuwait Wastewater Treatment Plants

Abdullah Almatouq, Mohd Elmuntasir Ahmed, Mishari Khajah, and Rashed Al-Yaseen

***Abstract*—This study presents the results of an analysis of the most common Volatile Organic Compounds (VOCs) in wastewater samples from one of the largest wastewater treatment plants in Kuwait. Samples from four different locations (plant influent, aeration tank inlet, aeration tank, and plant effluent) were collected weekly and were analyzed for toluene, benzene, xylene, 1, 3, 5 trimethylbenzene, dichloromethane and chloroform. The results showed that all the selected VOCs were detected except benzene. The average concentrations of VOCs that entered the treatment plant were as follows: chloroform 0.22 ±0.03 µg/mL, dichloromethane 0.18 ±0.05 µg/mL, toluene 0.08 ±0.01 µg/mL, O-Xylene 0.03 µg/mL, M-Xylene 0.01 µg/mL, P-Xylene 0.02 µg/ml, and 1,3,5-trimethylbenzene 0.02 µg/mL. The concentration of VOCs during the treatment processes decreased for all the selected VOCs (treatment efficiency >98%), except chloroform, which has increased. The disinfection process in the treatment plant might have generated byproducts (such as chloroform) which could have increased chloroform concentration. In conclusion, all of the analyzed VOCs in this study were lower than the maximum level of VOCs in treated wastewater.**

***Index Terms*—Benzene, cancer, wastewater treatment plant.**

I. Introduction

Wastewater contains different types of toxic and nontoxic pollutants, such as heavy metals, pesticides, and Volatile Organic Compounds (VOCs). VOCs are manufactured and/or naturally occurring highly reactive hydrocarbons that are characterized by low water solubility. VOCs are organic chemicals that mainly contain carbon and can easily escape into the atmosphere as gases due to high vapor pressures and low boiling point. VOCs can be a potential risk to the environment and human health. Exposure to high concentrations of VOCs, such as benzene and toluene which can lead to serious health risks (immune, nervous, and reproductive systems, and lead to several types of cancers) [1]. There are different sources of VOCs, however, concerns over VOCs emissions from wastewater treatment plants (WWTPs) into the environment have increased because WWTPs are considered to be one of the major sources of VOC emissions in different countries [2]. VOC emissions can transport long distances from the source and it is expected that people living in the areas even more than 4 km away from the source were still potentially exposed to cancer and other serious risks [3].

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In wastewater treatment plants, VOCs can be removed by different mechanisms, however, stripping, biodegradation, and sorption are the main processes affecting VOC. VOCs can be divided into different types and compounds; however, there are common compounds that can be found in every wastewater treatment plant around the world. Toluene, benzene, xylene, 1, 3, 5 trimethylbenzene, 1, 4 dichlorobenzene, dichloromethane, chloroform, and tetrachloroethene are the most common compounds in wastewater treatment plants [4]. VOCs concentrations and availability can be determined by the source of wastewater, treatment plant's processes, operational conditions, and temperature. In addition, emission rates of VOCs in wastewater treatment plants vary and are affected by different parameters, such as wastewater quality, operational parameters, chemical species, and temperature. Due to the toxicity of VOCs to human health, different organizations, such as the World Health Organization (WHO) [5], have established maximum acceptable concentrations for some VOCs in drinking water.

Worldwide studies on VOCs in wastewater treatment plants have focused on VOCs removal processes from wastewater and evaluated the health effects of VOCs on human health. However, there is a lack of studies on VOCs fate in Kuwait wastewater treatment plants. In Kuwait, the activated sludge process (biological treatment) is the main treatment process in Kuwait WWTPs and VOCs concentrations in conventional WWTPs vary depending on the treatment stage and process. Studies showed that VOC concentrations usually are below the limits of quantification after the activated sludge process [2] where volatilization and air stripping are the main removal mechanisms in the activated sludge process. In addition, there are some VOCs produced during the physical, chemical, and biological reactions in the treatment plant. Researchers have detected high concentrations of VOCs in some processes of the treatment plant, which could possibly be due to the difficulties in removing VOCs through biodegradation, volatilization, and air stripping [2]. Furthermore, the chlorination process in the WWTPs and the use of chemicals for specific treatments can both result in generating more VOC species.

Different studies have reported the emissions of VOCs from wastewater treatment processes. Cheng *et al.* [6] studied VOCs emissions from five wastewater treatment plants in Taiwan. The study concluded that the primary treatment stage, including inlet station, neutralization, and primary clarifier, had the largest aqueous concentration in the treatment units of WWTPs and the secondary treatment unit (activated sludge) emitted most of the VOCs. Furthermore, Chen *et al.* [7] investigated the fates of three aromatic hydrocarbons (benzene, toluene, and xylenes) in the activated sludge process. The results showed that the effect of aeration on the fates of the VOCs was complex, and affected by different factors such as Henry's law coefficients and the treatment process, as these factors had positive and negative impacts on volatilization and biosorption/biodegradation of the VOCs in wastewater treatment plants. Therefore, the objectives of this study were: to identify and quantify the most common volatile organic compounds at Kabd WWTP, and to assess the efficiency of VOC removal at Kabd WWTP.

II. Material and Methods

In this study, the most common VOCs in Kabd WWTP have been identified and quantified. Kabd wastewater treatment plant has been selected for this study because it is one of the largest treatment plants in Kuwait that receives wastewater from highly populated areas. All laboratory analyses were conducted at Kuwait Institute for Scientific Research laboratories, which are ISO 9001-2015 certified in the field of Quality Assurance (QA) and Quality Control (QC). The targeted VOCs were toluene, benzene, xylene, 1, 3, 5 trimethylbenzene, 1, 4 dichlorobenzene, dichloromethane, chloroform, and tetrachloroethene. A custom standard solution including toluene, benzene, xylene, 1, 3, 5 trimethylbenzene, 1, 4 dichlorobenzene, dichloromethane, chloroform, and tetrachloroethene was purchased and used to establish the calibration curves. Sodium chloride (1 g) was added to sample vials before collection to increase the ionic strength of the solution and reduce the VOC solubilities. Wastewater samples from different processes (influent, aeration tank inlet, aeration tanks, and effluent) have been collected once a week from Kabd WWTP. For VOCs analysis, 40-mL wastewater samples were collected and sealed in 40-mL headspace vials for later analysis. Shimadzu Gas Chromatography coupled with Mass Spectrometry (GC/MS) was used to identify and determine the VOC concentrations in the wastewater. All wastewater samples were stored at 4 ℃ and analyzed 72 h after the sampling. The GC was equipped with (30 m–0.25 mm internal diameter) Rxi-624il MS column (Restek company). The samples were analyzed in duplicates, and blank samples were added in each batch to confirm data quality and reliability. The calibration curves of the selected VOCs were developed and checked before the analysis ($R^2 > 0.99$). Furthermore, wastewater samples were analyzed also for wastewater quality parameters (Analysis Methods for Wastewater Examination [8]) including temperature, pH, Electrical Conductivity (EC), Dissolved Oxygen (DO), Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), Total Dissolved Solids (TDS), Nitrite (NO_2), Nitrate (NO_3), Ammonia (NH_3), total nitrogen, and Total Phosphorous (TP).

III. Results and Discussion

A. VOCs Identification and Quantification

The most common VOCs in wastewater were identified and quantified in Kabd WWTP. The VOCs were monitored in 4 different locations (inlet, aeration tank inlet, aeration tank, effluent). In total, ten VOCs including toluene, benzene, M-xylene, P-xylene, O-xylene 1, 3, 5 trimethylbenzene, 1, 4 dichlorobenzene, dichloromethane, chloroform, and tetrachloroethene were analyzed in wastewater only. The concentrations of VOCs in the gas phase have not been measured. The calibrations curves were drawn using 6 concentrations (0.1, 0.25, 0.5, 0.75, 1, 2 μg/mL), with $R^2 \geq 0.99$. However, the calibration curves of tetrachloroethene and 1,4-dichlorobenzene had low R^2 value (<0.6); therefore, the results of these 2 compounds are not reliable and will not be discussed in this paper. The average concentrations of VOCs that entered the treatment plant are as follows: chloroform 0.22 μg/mL, dichloromethane 0.18 μg/mL , toluene 0.08 μg/mL, O-Xylene 0.03 μg/mL, M-Xylene 0.01 μg/mL, P-Xylene 0.02 μg/mL, and 1,3,5-trimethylbenzene 0.02 μg/mL. Benzene was not detected at any point in the treatment plant because benzene was rapidly volatilized from the wastewater to the gaseous phase. Similar results were found by Yang *et al.* [3], where more than 90% of benzene was volatilized from the aqueous phase at the beginning of the experiment. Moreover, the concentrations of VOCs were monitored at the outlet, and the concentrations were as follows: chloroform 1.45 μg/mL, dichloromethane 0.1 μg/mL; and the rest of the VOCs were not detected.

Further samples have been collected to understand the removal of VOCs in the treatment plant. Samples from the aeration tank inlet and in the middle of the aeration tank were collected and analyzed. Results showed that VOCs concentration in the inlet of aeration tank were as follows: dichloromethane 0.08 μg/mL, chloroform 0.01 μg/mL, toluene 0.03 μg/mL, O-Xylene 0.02 μg/mL, M-Xylene 0.01 μg/mL, P-Xylene 0.01 μg/mL, and 1,3,5-trimethylbenzene 0.01 μg/mL, where the concentrations of VOCs in the middle of aeration tank were as follows: chloroform 0.18 μg/mL, dichloromethane 0.12 μg/mL, and toluene 0.03 μg/mL (Table I).

TABLE I: VOCs Average Concentrations in KABD Wastewater Treatment Plant

VOCs	Inlet (μg/mL)	Aeration Tank Inlet (μg/mL)	Aeration Tank (μg/mL)	Effluent (μg/mL)
Dichloromethane	0.18	0.08	0.12	0.10
Chloroform	0.22	0.01	0.18	1.45
Benzene	0.0	0.0	0.0	0.0
Toluene	0.08	0.03	0.03	0.0
M-Xylene	0.01	0.01	0.0	0.0
P-Xylene	0.02	0.01	0.0	0.0
O-Xylene	0.03	0.02	0.0	0.0
Trimethylbenzene	0.02	0.01	0.0	0.0

Different studies have investigated the concentration of VOCs in WWTPs. Similar results were obtained by Fatone *et al.* [9], where the study analyzed more than 20 compounds in 4 different treatment plants. The study showed that 1,2,4-trimethylbenzene, toluene, ethylbenzene, xylene, styrene, and 4-chlorotoluene were the most common compounds in the selected treatment plants, and the concentrations of these compounds are similar to the concentrations in this study. Furthermore, Escalas *et al.* [10] studied the concentration of different VOCs at 4 different locations (influent, pretreatment effluent, primary effluent, and plant effluent) in a treatment plant in Spain. Around 34 VOCs were detected and analyzed in the treatment plant. The concentration of toluene was the highest among the VOCs (23 µg/mL) followed by chloroform (9.5 µg/mL). The concentrations of all the VOCs in the effluent were lower than the plant influent. Toluene has been found in most of the studies with high concentrations because it is widely used as an industrial feedstock and a solvent [9].

Results showed that all of the analyzed VOCs in this study were lower than the maximum level of VOCs in drinking water [11]. The concentration of VOCs during the treatment processes decreased for all the selected VOCs, except chloroform, which increased sixfold (0.22 to 1.45 µg/mL). Fig. 1 shows the average concentrations of VOCs in each process in Kabd treatment plant. Similar phenomena have been found in different studies, where chlorinated VOCs have increased during the treatment process [12]. At the last process (disinfection process) in the treatment plant, chlorine was added to the treated wastewater and it could have reacted to the natural organic matters in water to form large amounts of Disinfection Byproducts (DBPs), such as chloroform and dichloromethane. Studies have confirmed that the disinfection process, as well as the improper storage and disposal methods, are the main sources of chloroform [12]. Furthermore, the chlorination process might increase the concentration of dichloromethane in treated wastewater. Another potential reason for increasing the concentration of chloroform in the treatment plant is the circulation ratio of wastewater, as increasing the circulation ratio could lead to an increase in the concentration of chloroform in the liquid effluent.

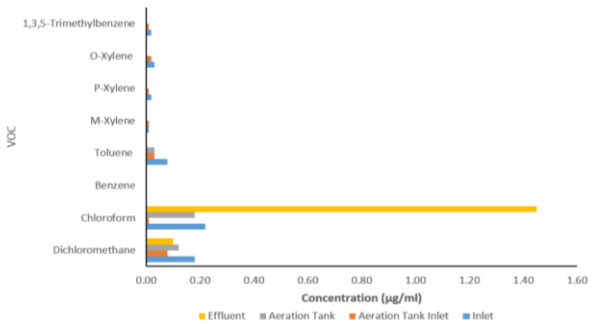

Fig. 1. Concentrations of VOCs in different locations in the treatment plant.

B. *Chlorinated and Non-chlorinated VOCs*

Chlorinated volatile organic compounds, including Polychloromethanes (PCMs), Polychloroethanes (PCAs), and Polychloroethylenes (PCEs) are a group of ubiquitous contaminants that have been widely detected in environmental media and possess high volatility and strong recalcitrance to degradation [12]. Chlorinated VOCs have been introduced into the environment through human activities, such as the production of pharmaceuticals and pesticides. Furthermore, the disinfection process in water and wastewater treatment using chlorination is another source of VOCs. Chlorine in water and wastewater reacts with organic matters to form large amounts of disinfection byproducts (such as chloroform). Fig. 1 shows the concentrations of VOCs in different processes at Kabd wastewater treatment plant. A significant difference ($P<0.05$) was noted in the removal rate between the chlorinated (dichloromethane and chloroform) and non-chlorinated compounds (benzene, toluene, M-Xylene, P-Xylene, O-Xylene, and 1,3,5-trimethylbenzene). The major removal rate of the non-chlorinated compounds mostly occurred in the activated sludge process (> 95%); whereas, the removal rate of the chlorinated compounds was low (< 35%) in the activated sludge tank. The biodegradation process is the major mechanism for the removal of non-chlorinated VOCs including toluene and benzene, and stripping is the main removal mechanism for chlorinated compounds, such as chloroform, and trichloroethylene [2]. Stripping can be defined as a process by which water contaminated with slightly soluble VOCs may be purified by transferring the contaminants from the water to the air.

The non-chlorinated compounds were completely removed in the aeration tank (Fig. 2 and Fig. 3), indicative that the biodegradation process is occurring successfully in the aeration tank. However, the removal of chlorinated compounds in the aeration tank was low (< 35%). A deterioration in the stripping process led to a low removal rate of the chlorinated VOCs, as different factors might affect the efficiency of the stripping process. Studies have demonstrated that the efficiency of diffused air system (activated sludge process) may range from 50% to 85% due to the small interfacial area and short contact time between the wastewater and air [13]. In addition, at low concentrations (below 0.5 ppb), stripping process becomes difficult and inefficient.

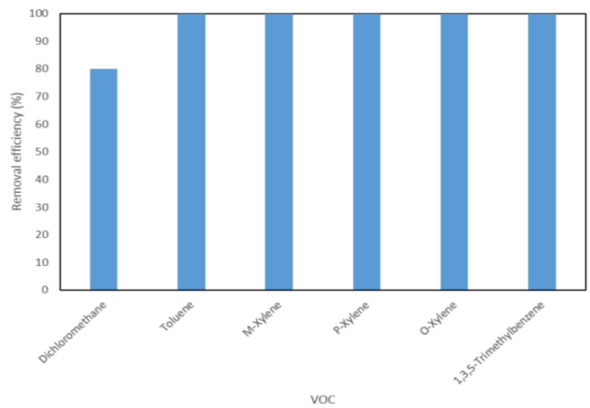

Fig. 2. Overall removal efficiency of VOCs in Kabd WWTP.

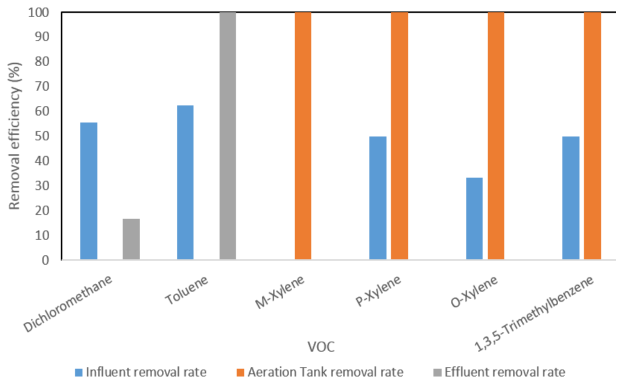

Fig. 3. Removal efficiency of VOCs at each stage in Kabd WWTP.

Alternatively, effluent samples showed that the concentrations of the chlorinated compounds were higher than the concentration of non-chlorinated compounds. For example, the average concentration of chloroform in the effluent was higher than the average of the influent. In some cases, the concentrations of chloroform in the effluent were high (> 10 µg/mL), and that leads to an increase in the average concentration of chloroform in the effluent. According to the centres for disease control and prevention [14], different disinfection byproducts, such as chloroform, dibromochloromethane, and bromoform might be created due to the reaction of hypochlorous acid and hypobromous acid with the organic matter. Thus, the concentration of chloroform in the effluent has increased. The WHO has classified chloroform as a possible human carcinogen [5]. Nevertheless, there still is inadequate evidence relative to the concentration that could cause human carcinogenicity.

What is more, variations in the removal rates for the selected compounds have been detected as to whether these could be due to the physicochemical properties of the selected compounds, biodegradability, and the water-phase concentrations [2].

C. The Effect of Treatment Process on VOCs Removal

The removal efficiency of VOCs in the treatment plants may vary from one to another, as the removal rate depends on the treatment technology, physicochemical properties of the selected compounds, biodegradability, and the water-phase concentrations. Wastewater samples were collected from four different locations (plant influent, aeration tank inlet, aeration tanks, and plant effluent). The removal rate of the VOCs in the primary stage was low (between 30–45 %), except for chloroform (95 %); whereas, the activated sludge process had the highest VOC removal rates. M-Xylene, P-Xylene, O-Xylene, and 1,3,5-trimethylbenzene were completely removed in the activated sludge process (Fig. 3). Similar results were found in the study of Yang *et al.* [2], where a high removal rate was observed in high surface area processes, such as activated sludge process and high water-phase concentrations. Furthermore, Chen *et al.* [7] reported the importance of aeration and activated sludge in the transformation of the VOCs from air to water and from water to sludge, as the concentration of VOCs that have large Henry's law coefficients were reduced more significantly in the air and water phases, resulting in positive and negative impacts on the volatilization and biodegradation of the VOCs in WWTPs. In aeration tanks, different VOCs removal mechanisms (air stripping, biodegradation, and/or adsorption to suspended solids) might occur simultaneously; thus, the removal rate was high [15]. The air stripping removal mechanism is a function of air volume or concentration in the aeration tank. Therefore, obtaining the optimal air volume in the aeration tank is crucial to obtaining high VOCs removal efficiency. On the other hand, the biodegradation mechanism in the aeration tank occurs rapidly before air stripping takes place. Different VOCs are removed through the biodegradation mechanism, as compound biodegradability is a function of the first-order rate constant (k1). Then again, the limited availability of k1 values in literature is a major disadvantage in the evaluation of VOC removal by biodegradation [15].

D. The Effect of Temperature on VOCs Removal

Wastewater samples from Kabd WWTP were collected once a week between January 2020 to September 2021. The results showed that there was a significant correlation (P<0.05) between VOCs removal and the temperature. The volatilization rate of the VOCs varied through the seasons, where higher volatilization was noticed in high-temperature seasons. Table II shows the Pearson's correlation coefficients between wastewater temperature and VOCs concentrations in treatment plant influent. Wastewater temperature did not show to have a significant effect on chloroform, toluene and P-Xylene concentrations. However, dichloromethane, O-Xylene and 1,3,5-trimethylbenzene had a significant correlation with wastewater temperature, where dichloromethane, O-Xylene and 1,3,5-trimethylbenzene concentrations decreased with the increase in wastewater temperature. Different studies have determined the effect of meteorological parameters, such as temperature, relative humidity, solar radiation, wind direction and speed on VOCs removal. Lin *et al.* [16] studied the effect of wind speed, temperature, and sunlight on VOCs removal and distribution. The study concluded that wind speed and direction influenced the distribution of concentrations of VOCs, where high wind speeds resulted in decreased VOCs concentrations. Henry's constant plays a crucial role in the removal of VOCs by air stripping. Henry's constant is directly proportional to the temperature of the system, where an increase in Henry's constant would show an increase in the stripping rate and as a result, a decrease in the biodegradability of the compound. This is indicative that with an increase in Henry's constant, the volatility of the compounds will shift, and the compound will be more vulnerable to removal by stripping [15].

TABLE II: PEARSON'S CORRELATION COEFFICIENTS BETWEEN WASTEWATER TEMPERATURE AND VOCS CONCENTRATIONS IN TREATMENT PLANT INFLUENT

VOC	Pearson's correlation coefficients	P value
Dichloromethane	-0.4797	0.0011
Chloroform	0.0539	0.7310
Benzene	-	-
Toluene	-0.1633	0.2950
M-Xylene	-0.3862	0.0100
P-Xylene	-0.1174	0.4530
O-Xylene	-0.5365	0.0002
Trimethylbenzene	-0.6216	0.0008

Kuwait has a hot summer season (May to September) with a long daytime (sun hours) and high wind speed. July and August are the hottest months in Kuwait, as well as the longest sunshine hours. The meteorological results are in agreement with the study results which were obtained throughout the year. This can explain the low concentrations of VOCs in the treatment plant influent through the summer, as well as the high removal efficiency. Yang *et al.* [3] showed that the strongest emissions of aromatic hydrocarbons were observed in summer, followed by spring, fall, and winter. However, the opposite trend of the gaseous concentrations were noticed in the winter, as the highest aqueous concentrations of VOCs in wastewater were detected in winter.

IV. Conclusion

The main findings from this study showed that all the selected VOCs were identified and quantified except benzene and all the analyzed VOCs were lower than the maximum level of VOCs in drinking water. The concentration of VOCs during the treatment processes decreased for all the selected VOCs, except chloroform, which increased sixfold due to the reaction of hypochlorous acid and hypobromous acid with the organic matter. In addition, the concentrations of the chlorinated compounds were higher than the concentration of non-chlorinated compounds and the major removal rate of the non-chlorinated compounds mostly occurred in the activated sludge process (> 95%); whereas the removal rate of the chlorinated compounds was low (< 35%) in the activated sludge tank.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Rashed Al-Yaseen was responsible for collecting wastewater samples from the wastewater treatment plant; Mishari Khajah was responsible for Preliminary data collection; Abdullah Almatouq and Mohd Elmuntasir Ahmed analyzed the data and wrote the paper; all authors approved the final version.

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