

Research Article

Simultaneous Removal of MTBE and Benzene from Contaminated Groundwater Using Ultraviolet-Based Ozone and Hydrogen Peroxide

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Efficiency of ultraviolet-ozone (UV/O₃) and ultraviolet-hydrogen peroxide (UV/H₂O₂) processes was investigated for simultaneous removal of methyl tertiary butyl ether (MTBE) and benzene from contaminated ground water. The photoreactor employed housed 15-watt low pressure (LP) and 150-watt medium pressure (MP) mercury UV lamps. Oxidation of contaminants was studied at two different levels of ozone and hydrogen peroxide. Brackish groundwater samples were spiked with MTBE and benzene up to a concentration of 500 µg L⁻¹. Removal potential was evaluated at different parameters such as UV type and intensity and peroxide and ozone dosages, as well as contact time. Results indicated that no removal of the contaminants was attained when treated with hydrogen peroxide or ozone alone. However, about 50% and 30% removal of MTBE were achieved in 30 minutes when irradiated with MP-UV and LP-UV lamps, respectively. On the other hand, UV/H₂O₂ process was found to be superior in removal of MTBE (90% in 10 min.) and benzene (95% in 5 min.) compared to UV/O₃ process. Furthermore, removal of benzene was comparatively easier than MTBE in both approaches. It is hence concluded that higher UV intensities and elevated doses of H₂O₂ accelerate simultaneous removal of MTBE and benzene from water.

1. Introduction

In oil producing countries, contamination of water resources is generally attributed to fuel additives and different fuel products. These contaminants reach water bodies through different routes such as the process of transportation, mixing, refining, and filling operations. Fuel products such as benzene, toluene ethyl benzene, and xylenes (BTEX) and fuel additives such as methyl tert-butyl ether (MTBE) are among the organic pollutants commonly detected in water resources. MTBE is added to fuel to enhance the octane number and reduce air emissions of carbon monoxide since the issuance of US Clean Air Act (USCAA) in 1970s [1]. Major causes of MTBE pollution are leakages in underground storage tanks and pipelines and accidental spills during refining processes. Many studies have reported their presence and impacts in atmosphere [2] as well as in groundwater [3]. Toxicity analysis conducted on test animals has shown that direct exposure to MTBE, even for shorter periods of time, can cause nausea, vomiting, and sleepiness [3].

On the other hand, benzene has received a lot of attention due to its carcinogenic nature [4]. It has been reported that, in drinking water, more than 0.005 mg/L of benzene can seriously increase the risk of cancer [5]. Benzene concentration in gasoline is being regulated at 1% by volume that renders it harmful carcinogen for living organism. Studies conducted on animal models show that benzene toxicity can increase the chances of cancer mutagenesis at p53 gene which results into uncontrolled growth of cells [6].

Despite the fact that both MTBE and benzene may exist together, they have different physical and chemical characteristics which affect their transportation and fate in the environment. For example, solubility of MTBE in water bodies is 20 times higher (43,000–54,300 mg L⁻¹) than benzene (1780 mg L⁻¹) and other compounds in BTEX [7]. This enhances the movement of MTBE in underground formations and renders it difficult to trace. Furthermore, lower K_{oc} and K_{ow} values of MTBE indicate its lesser ability for adsorption to solid particles than benzene [8].

A number of techniques have been employed for the removal of MTBE and benzene from contaminated water. Bioremediation has not been very successful due to low biomass production and slow growth kinetics of bacteria [9]. Furthermore, air stripping has been reported by some researchers as a noneconomical technique for MTBE removal from water [9] and adsorption to granular activated carbon (GAC) or synthetic resins only leads to transfer of MTBE from one phase to another instead of remediation. However, superior results in removal of MTBE have been reported by employing advanced oxidation processes (AOPs) [10, 11]. Commonly used AOPs include the use of ozone-hydrogen peroxide oxidation (O_3/H_2O_2), UV-hydrogen peroxide oxidation UV/ H_2O_2 , ultrasonic ozonation (O_3), Fenton and photo-Fenton reactions, and photocatalysis with titanium dioxide (TiO_2) [9, 12, 13]. These processes generate highly reactive hydroxyl radicals (OH^\bullet) that in turn react with the contaminants and degrade them. Under optimum conditions, these hydroxyl radicals can completely mineralize the organic pollutants to their basic constituents, carbon dioxide and water [9]. Systems coupled with O_3/H_2O_2 possess great potential to degrade high amount of MTBE from water than O_3 and H_2O_2 . However, the only disadvantage found in the system is the capability for bromate formation through adjustment of H_2O_2/O_3 ratio [9]. Benzene has also been removed by using modern techniques such as UV/ H_2O_2 [14] and photocatalytic process of UV/ TiO_2 [15]. Likewise, other studies have also reported the photocatalytic degradation of gaseous benzene over UV-irradiated TiO_2 nanoballs [16, 17]. A system possesses higher efficiency of degradation than O_3 or UV alone but requires high cost and energy sometimes [9].

However, there is lack of convincing data for simultaneous removal of both benzene and MTBE using AOPs, despite the fact that both contaminants have quite often been found together in industrial wastes [18]. Therefore, the main objective of this study was to evaluate the efficacy of AOPs in simultaneous removal of benzene and MTBE from contaminated water.

2. Materials and Methods

2.1. Solutions. A control solution was prepared from deionized water. Highly pure (>99.5%) MTBE and benzene were obtained from Sigma-Aldrich (UK). Stock standard solutions of 100 ppm of MTBE and benzene were prepared in deionized water and spiked to initial concentration of about 500 ppb.

A test solution was prepared from groundwater samples that were collected from a well located in Dhahran, Saudi Arabia. The well has a total depth of 180 m and taps Umm er Radhuma (UER) aquifer. The well, being pumped at an average rate of about 1000 gallons min^{-1} , is mainly used for domestic cleansing and toilet flushing purposes. Chemical analysis of groundwater was performed to determine levels of dissolved salts (Table 1). Both MTBE and benzene were not detected in the raw water samples and hence were spiked at concentrations of 500 ppb.

Hydrogen peroxide (H_2O_2) of approximately 35% purity, purchased from local market, was used to prepare the stock

TABLE 1: Characteristics of groundwater samples.

Parameters	Value
pH	7.09
TDS, mg/L	3645
D.O., mg/L	5.78
Turbidity, NTU	0.1
TSS mg/L	5.0
TOC mg/L	5.36
MTBE $\mu g/L$	<1.0
VOC $\mu g/L$	<1.0
BTEX $\mu g/L$	<1.0
Alkalinity-T ($CaCO_3$)	433.5
Cl^- mg/L	1220.38
NO_3^{--} mg/L	3.809
Br^- mg/L	4.212
SO_4^{--} mg/L	545.69
Ca^{++} mg/L	255.1
Fe^+ mg/L	<0.009
K^+ mg/L	34.2
Mg^{++} mg/L	82.5
Na^+ mg/L	631.2

solution at a concentration of 3750 ppm. Actual concentration of H_2O_2 solution was determined by titration method using standardizing solution (0.1N) of potassium permanganate ($KMnO_4$).

Ozone oxidation was facilitated by ozone generator model C-Lasky C-L1010 (Air Tree Ozone Technology Co., Taiwan). The unit can produce O_3 at the rate of $10 g h^{-1}$ if pure oxygen is used while at $2 g h^{-1}$ if air is used at flow rate of $6 L min^{-1}$ ($25^\circ C$). Stock solution of O_3 in water (30 ppm) was prepared on daily basis. The amount of ozone dissolved in the aqueous phase was determined by indigo method by Hoigne and Bader [19]. Ozone was spiked into previously MTBE-benzene spiked waters at concentrations of 1 and 5 ppm.

2.2. UV Photoreactor. Spiked water samples (500 mL) with MTBE and benzene were transferred to the bench-scale UV photoreactor (Figure 1) (NORMAG Co., Hessenstraße, Hofheim, Germany). Two types of ultraviolet lamps were housed in the photoreactor: (i) 15 W low pressure (LP) Hg lamp which emits light at 254 nm with intensity of $6.5 \times 10^{-3} W/cm^2$ and (ii) 150 W medium pressure (MP) Hg lamp which emits light at a wider spectrum ranging from 200 to 400 nm with intensity of $5.3 \times 10^{-2} W cm^{-2}$. Due to volatile nature of MTBE and benzene, the temperature of solutions was kept constant at room temperature during treatment.

2.3. Experimental Layout. Three separate sets of experiments were designed for each type of spiked water.

- (I) In the first set of experiments, MTBE-benzene spiked waters were treated with two levels of each of H_2O_2 (20 and 100 ppm), O_3 (1 and 5 ppm), and UV radiation



FIGURE 1: The NORMAG photoreactor.

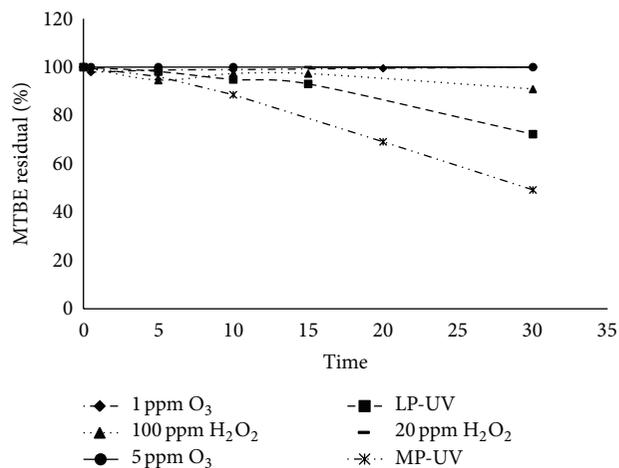
(LP and MP). Separate experiments were performed with each treatment.

- (II) In the second set of experiments, MTBE-benzene spiked waters were treated with a combination of different concentrations of H_2O_2 (20 and 100 ppm) and UV radiation (254 and 600 nm).
- (III) In the third set of experiments, MTBE-benzene spiked waters were treated with a combination of different concentrations of O_3 (1 and 5 ppm) and UV radiation (LP and MP).

In all treatments, water samples were collected after runtimes of 0, 5, 10, 20, and 30 min and analyzed for residual amount of MTBE, benzene, and by-products. During the rigorous bench-scale testing of the process, operational factors such as concentration of H_2O_2 and O_3 and contact time were optimized for the removal of MTBE and benzene.

2.4. Chemical Analyses of Samples. Effluent samples from the UV photoreactor were collected periodically and analyzed for its concentration levels of MTBE and benzene using an ISQ single quadrupole GC/MS system (Thermo Scientific) equipped with TriPlus for headspace injection system. A 60 m long, 0.32 mm i.d. Rtx-502.2 (Restek Corp., USA) capillary column was used for the separation purpose. The carrier gas was helium flowing at the rate of 1.7 mL min^{-1} . The column temperature was programmed to rise from 50 to 220°C at the rate of $20^\circ\text{C min}^{-1}$. The mass spectrometer was operated in the selected ion mode (SIM). Calibration curves were prepared for MTBE and benzene and some by-products.

2.5. Calculation of EEO Values for UV/ H_2O_2 and UV/ O_3 . Electrical energy per order (EEO) of contaminant provides a convenient way to assess the effectiveness of removal of various organic compounds for a given water sample using UV irradiation. It is estimated in terms of kWh/1000 gal/order of removal, which is determined at the optimum

FIGURE 2: Removal of MTBE using H_2O_2 , O_3 , and UV only.

H_2O_2 or O_3 concentrations. Higher values of EEO of a contaminant indicate that the contaminant is more difficult to remove [20]. The EEO ($\text{kWh/m}^3/\text{order}$) was calculated from the following equation:

$$EEO = \frac{P \times t \times 1000}{V \times 60 \times \log(C_i/C_f)}, \quad (1)$$

where P is the power input (kW), t is the irradiation time (min), V is the volume (L) of the water in the reactor, and C_i and C_f are the initial and final pollutant concentrations. For one order of removal, concentration should be reduced to $C_i/C_f = 10$ and $\log(C_i/C_f)$ would be equal to 1 [21].

3. Results and Discussion

3.1. Treatment of MTBE-Benzene Spiked Waters by H_2O_2 , O_3 , or UV Alone. As listed in Table 1, the chemical analyses of groundwater revealed its brackish nature with a total of dissolved solids of over 3600 ppm. Moreover, levels of chloride, bromide, and sulfate ions are on the higher side at 1220, 4.2, and 546 ppm, respectively. High levels of these anions were expected to compete with the target pollutants (MTBE and benzene) for the hydroxyl radicals [22–25]. In addition, the effect of water quality on the UV exposure and irradiation efficiency is low due to the low turbidity level of 0.1 NTU. The scavenging activities of the anions especially chlorides, carbonates, bicarbonate, and other species has also been reported by other researchers [23, 25]. Classical Fenton's system also supports this behavior of anionic species in terms of nonproductive reactions [23, 26, 27].

Figures 2 and 3 show that both H_2O_2 and O_3 were not able to influence the removal of MTBE or benzene from contaminated groundwater under the conditions used in the study. This can be attributed to the strong bonding within molecular structure of MTBE and benzene [28, 29]. The second reason could be the relatively low oxidative potential of H_2O_2 and O_3 [30].

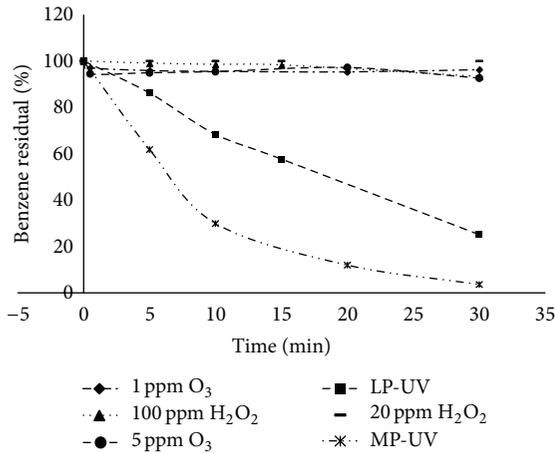


FIGURE 3: Removal of benzene using H_2O_2 , O_3 , and UV alone.

However, significant removal of MTBE was observed when the spiked water was treated by UV radiation. Approximately, 50% of MTBE was removed within 30 minutes of exposure to 150 W MP UV lamp and about 30% in samples exposed to 15 W LP UV lamp. These findings support the results of previously conducted studies that reported partial degradation of MTBE by UV photolysis [31].

Similarly, removal of benzene was substantial in samples exposed to UV photolysis (Figure 3). Approximately 75% of benzene was removed by 15 W LP-UV lamp within 30 minutes and 95% benzene was removed by 150 W MP-UV irradiation. This accelerated removal of benzene was apparently due to the fact that benzene is slightly less stable against UV radiation than MTBE [32].

3.2. Treatment of MTBE-Benzene Spiked Waters by UV/ H_2O_2 Process. Results of remediation of MTBE-benzene spiked waters by UV/ H_2O_2 process are presented in Figures 4 and 5. Removal of the MTBE by UV/ H_2O_2 was significantly enhanced as compared to removal by H_2O_2 or UV alone (Figure 4). All four possible combinations of UV types (15 W LP and 150 W MP) and H_2O_2 concentrations (20 and 100 ppm) were tested, namely, (i) LP/20 ppm, (ii) LP/100 ppm, (iii) MP/20 ppm, (iv) and MP/100 ppm. The removal efficacy was found to be influenced by the concentration of H_2O_2 as well as by the intensity of UV radiation. With highest removal efficiency at MP/100 ppm and LP/100 ppm followed by MP/20 ppm and LP/20 ppm, the concentration of H_2O_2 seems to be the dominating factor as irradiating with both LP-UV and MP-UV lamps removed >95% of MTBE at 100 ppm H_2O_2 within 20 minutes of exposure (Figure 4). Results of this study are consistent with previous studies that have also reported additive effect of coupled UV/ H_2O_2 techniques in removal of MTBE from contaminated waters [33].

Similarly, superior efficacy for benzene removal was observed in samples treated with UV/ H_2O_2 process compared to the samples treated separately with UV and H_2O_2 (Figure 5). Benzene was completely removed within 10 min by all four possible combinations of UV intensities (LP and MP)

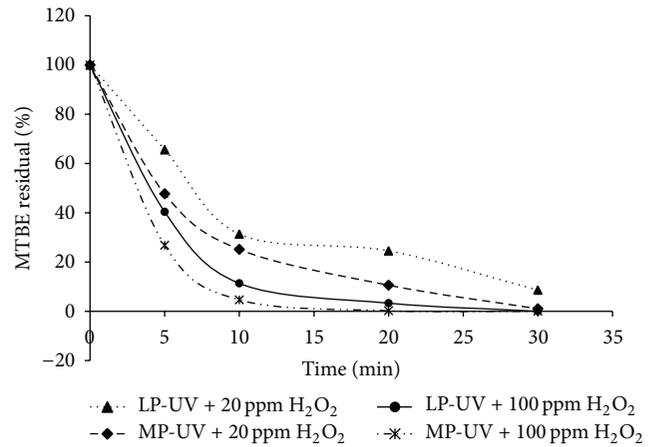


FIGURE 4: Removal of MTBE using UV/ H_2O_2 process.

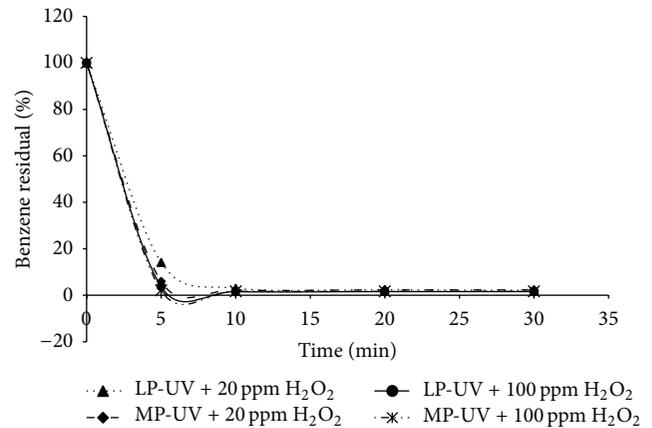


FIGURE 5: Removal of benzene using UV/ H_2O_2 process.

and H_2O_2 concentrations (20 and 100 ppm). More than 95% of benzene was removed within 5 minutes of treatment time in MP/100 ppm and LP/100 ppm. Hence, use of UV/ H_2O_2 is one of the most efficient methods for remediation of benzene contaminated water. It would be interesting to compare benzene removal by this treatment with alternative remediation methods, such as air stripping and activated carbon adsorption. These results are in tendon with earlier studies in which the successful degradation of benzene using UV/ H_2O_2 coupled reactions has been reported [14].

3.3. Treatment of MTBE-Benzene Spiked Waters by UV/ O_3 Process. This study was designed to evaluate combined effect of UV irradiation and ozone oxidation (UV/ O_3) on remediation of MTBE-benzene spiked waters. Again, all four possible combinations of UV intensities (LP and MP) and O_3 concentrations (1 and 5 ppm) were tested, namely, (i) LP/1 ppm, (ii) LP/5 ppm, (iii) MP/1 ppm, (iv) and MP/5 ppm. Results of the experiment are shown in Figures 5 and 6.

Remediated process seemed to be influenced mainly by O_3 concentration as maximum removal of MTBE was recorded in the samples treated with MP/5 ppm O_3 as 80%

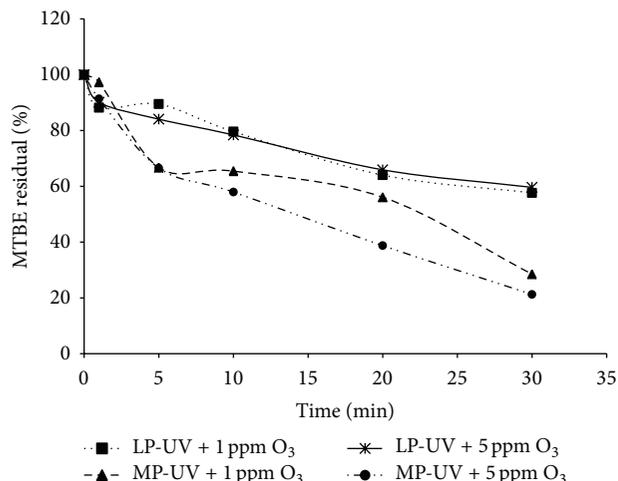


FIGURE 6: Removal of MTBE using UV/O₃ process.

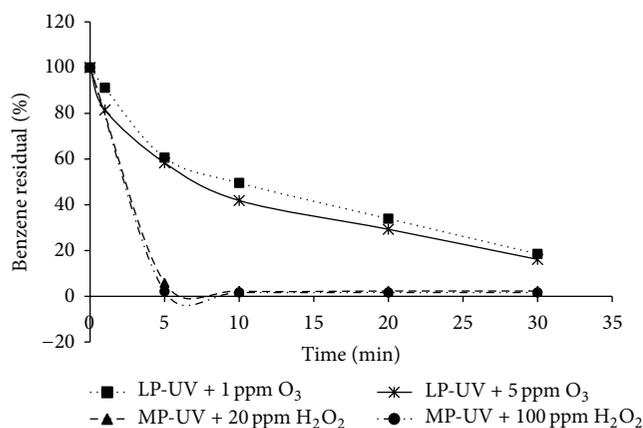


FIGURE 7: Removal of benzene using UV/O₃ process.

followed by LP/5 ppm O₃ as 70% in 30 min, while only a little amount of MTBE (40%) was removed at 1 ppm of O₃ concentration in both LP and MP treated samples (Figure 6).

Benzene removal by UV/O₃ process was chiefly influenced by the type of UV radiation (Figure 7) as more than 95% of benzene was removed after 30 minutes in samples treated with MP/5 ppm and MP/1 ppm compared to 80% removal in samples treated with LP/1 ppm and LP/5 ppm. Hence, removal of benzene was superior at higher UV intensities.

Results of the remediation assays clearly demonstrate superiority of using MP over LP radiation and H₂O₂ over O₃ in an AOP. In all studied treatments benzene was removed relatively quickly compared to MTBE. Hence, MTBE may become a slight limiting factor when both contaminants are treated simultaneously by AOPs.

The MTBE degradation by UV/H₂O₂ process is demonstrated by the formation of by-products (i.e., tertiary butyl formate (TBF) and tertiary butyl alcohol (TBA)) as presented in Figure 8 which shows the formation of TBF and TBA when MTBE is treated with LPUV lamp with 50 ppm H₂O₂.

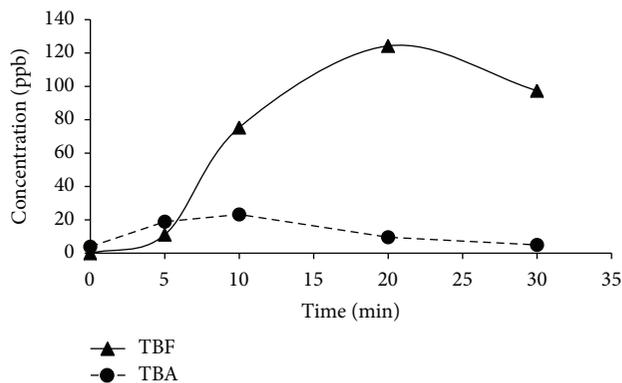


FIGURE 8: By-products formed due to degradation of MTBE.

TABLE 2: The EEO values for MTBE and benzene by LP-UV and MP-UV at 100 ppm of H₂O₂.

Process	Pollutant	EEO = KW-hr/1000 gal
LP-UV—100 ppm H ₂ O ₂	MTBE	21.03
LP-UV—100 ppm H ₂ O ₂	Benzene	6.31
MP-UV—100 ppm H ₂ O ₂	MTBE	15.77
MP-UV—100 ppm H ₂ O ₂	Benzene	8.41

3.4. *The EEO for Benzene Removal by UV/H₂O₂.* The optimum conditions for removal of MTBE using the 15 W LP-UV lamp were peroxide (H₂O₂) of 100 ppm (Figure 4). Table 2 summarizes the calculations of the EEO values for MTBE and benzene when treating contaminated groundwater and reducing the concentration of MTBE and benzene by 10 orders of magnitude using both LP 15 W and MP 150 W combined with 100 ppm H₂O₂. From the data in Table 2, it is estimated that 10 and 2 minutes are needed to irradiate the water spiked with MTBE and benzene, respectively, when using LP-UV + 100 ppm H₂O₂. The values of EEO for MTBE and benzene are 21 kW·hr/1000 gal and 6.3 kW·hr/1000 gal, respectively, while the EEO values for the 2 contaminants calculated for the 150 W MP-UV lamp were 15.7 and 8.41, respectively. Hence, benzene is relatively easier to remove from water. These results are slightly higher than the EEO levels reported in previously published work (*Calgon AOT Handbook*, 1996) which stated EEO values of 10 kWh/1000 gal for MTBE and 2–5 kWh/1000 gal for benzene. The main reason for the high values of EEO is attributed to the quality of the groundwater treated which consists of high levels of radical scavengers as discussed above.

The EEO values for MTBE removal by UV/O₃ process were not calculated since 10 orders of removal of MTBE were not attained within the 30 minutes of reaction time used in the study.

4. Conclusion

Both MTBE and benzene can be removed simultaneously from contaminated water using advanced oxidation processes employing different combinations of UV, H₂O₂, and O₃. Remediation assays clearly demonstrate superiority of

using MP over LP radiation and H_2O_2 over O_3 . Superiority of using UV/ H_2O_2 over UV/ O_3 process and also its efficiency in removing benzene as compared to MTBE, from contaminated water, were established. It was also confirmed by lower EEO values for UV/ H_2O_2 as compared to that of UV/ O_3 . The efficacy of these processes clearly depends upon the concentrations of H_2O_2 and O_3 and intensity of UV radiation. Furthermore, benzene was comparatively easier to degrade than MTBE. Hence, MTBE may become a slight limiting factor when both contaminants are targeted simultaneously. Detailed studies are required to unearth exact mechanism behind variable behaviors of benzene and MTBE towards AOPs.

Conflict of Interests

The author declares that there is no conflict of interests regarding the publication of this paper.

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