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Removal of Aromatic Compounds by a Wet Packed-Bed Scrubber Filled with Waste Edible Oil

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Abstract
Background: The emission of aromatic compounds, particularly BTEX (benzene, toluene, ethylbenzene, and xylene), into the atmosphere poses significant environmental and health issues. Recognizing the limitations of traditional water-based scrubbing methods for hydrophobic compounds, this study aimed to determine the optimal conditions for absorbing BTEX compounds released during the frying process, using waste edible oil as an innovative absorbent in a packed scrubber system.
Methods: The effects of operating parameters such as scrubber bed height, frequency of oil reuse, and pollutant contact time on the removal efficiency and absorption capacity of BTEX were investigated through laboratory-scale experiments and gas chromatography analysis.
Results: The results showed that the removal efficiency of BTEX significantly improved with increased bed height from 64 to 98% for B, from 56 to 87% for T, from 36 to 59% for E, and from 58 to 89% for X. The results also showed a significant decrease in removal efficiency during absorption time. However, the removal efficiencies did not change significantly for all chemicals among the waste oils. The removal efficiency of B related to the use of oil ranged from 80 to 90%, from 63 to 87% for T, from 19 to 58% for E, and from 65 to 88% for X.
Conclusion: This research demonstrates the potential of utilizing waste edible oils in packed scrubbers for effective BTEX removal, highlighting the importance of optimizing operational conditions for improved air pollution control. The findings contribute to the development of more sustainable and environmentally friendly solutions for mitigating VOCs.
Keywords: Benzene, Absorption, Volatile organic compound, Waste, BTEX
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Introduction

Aromatic hydrocarbons, particularly benzene, toluene, ethylbenzene, and xylenes (BTEX), are significant pollutants emitted from various industrial and environmental processes. These chemicals have received extensive attention as they are precursors of toxic radicals in the atmosphere, leading to the formation of secondary pollutants through photochemical reactions, potentially endangering human health indirectly. They can also affect human health directly through inhalation and ingestion routes (1,2). BTEX compounds are released into the environment through both natural and anthropogenic sources, contributing to environmental pollution. They can contaminate air, soil, and water, posing threats to human health and ecosystems (3-5). The U.S. Environmental Protection Agency (USEPA) classified BTEX as priority pollutants due to their toxic and carcinogenic properties. Even at low concentrations, they can cause damage to the kidneys and liver and paralyze the central nervous system (6,7). Various methods, including membrane separation, absorption, adsorption, catalytic oxidation, biodegradation, and combustion, have been studied to control VOCs (8,9). Gas-liquid mass transfer is a complex and important phenomenon frequently occurring in chemical operations, making gas scrubbing applicable for VOC removal (10). Reasonable removal efficiencies can be achieved using water as a scrubbing liquid when the target VOCs are hydrophilic, acidic, or basic (11). Water-packed columns operated at counter-current are often chosen at the industrial scale, although co-current intensified gas-liquid contactors, such as aero-ejectors, static mixers, and venturi scrubbers, could be used effectively (12).

However, many dangerous VOCs in different industrial processes, such as BTEX, are neutral and hydrophobic compounds. Therefore, the replacement of water with a non-aqueous phase liquid (NAPL) to efficiently remove these compounds has recently been reviewed (13). NAPL typically has viscosities one to three orders of magnitude higher than water, making it difficult to use co-current gas-liquid contactors and select a counter-current packed column. Despite the higher viscosity inducing greater mass transfer resistance, liquids with viscosities up to two orders of magnitude higher than water can be successfully implemented in packed columns (14-16).

Several researchers have investigated various synthetic NAPLs for VOC absorption at the laboratory scale, particularly silicone oils (17-19). Recently, Lhuissier et al. have evaluated the partition coefficients of several VOCs in four types of waste oils, indicating lower partition coefficients than those of silicone oils. They demonstrated that lubricants and transformer oils, with their low volatilities, are compatible with the absorption process (20). These waste oils might contain volatile compounds that could cause secondary emissions (21). Polyethylene glycols (PEG) are also investigated by Guo et al. as liquid phases for the absorption of VOCs (22). Ozturk and Yilmaz reported that absorption removal rates ranged from 70% to 95% for methanol, T, B, and carbon tetrachloride when using fresh and waste lubricants and vegetable oils (21). Malhautier et al. (23) investigated a mixture of aromatic, halogenated, and oxygenated compounds in soluble cutting oil. They found that the addition of oil enhanced the absorption of halogenated and aromatic compounds compared to water, but had the opposite effect for oxygenated compounds.

Only two experimental studies have reported T mass transfer in di-2-ethylhexyl adipate and silicone oil on a laboratory scale (24,25). In a previous study, Lhuissier et al. studied the hydrodynamics of lubricant and transformer waste oils in a lab-scale packed column filled with Flexipac® structured packing, demonstrating that it is possible to operate a counter-current packed column with viscous waste oils, particularly transformer oil (14). Margaux Lhuissier (2021) performed VOC absorption in a structured packing fed with waste oils, measuring removal efficiencies of hydrophobic VOCs in silicone and transformer oils up to 80-90%. Nonetheless, owing to its higher viscosity, the removal efficiencies of the lubricant were significantly lower (26).

However, limited investigations have been conducted on the absorption of VOCs, specifically BTEX, in waste oils using a packed column. Consequently, the primary objective of this research was to assess the absorption of BTEX compounds using a packed scrubber filled with waste edible oils.

Materials and Methods

Chemicals

The analytical grade solutions of B, T, E, and X isomers were obtained in the highest purity available from Merck Co. (Darmstadt, Germany). A certain brand of blended cooking oil with the viscosity of 0.04 Pa.s and density of 943 kg/m^3 was chosen from the market as the absorbent for VOC removal.

Sampling and Analytical Methods

A gas-tight syringe (Hamilton Bonaduz, Switzerland) was used every five minutes (0–20 min) for sampling target compounds before and after the absorption column. The samples were measured using a Shimadzu-17A gas chromatograph equipped with a BP-5 (non-polar) capillary column (30 m in length, 0.32 mm in ID, and 0.32 µm in film thickness), a split/splitless injection port, and a flame ionization detector (GC/FID). The programmed temperature was set at 40°C for 3 min, then increased to 140°C at the rate of 8°C min^{-1}, and subsequently, raised to 230°C at a heating rate of 10°C min^{-1}, maintaining this temperature for 5 min. The detector temperature was set at 300°C. The time for measuring the peak of target compounds was considered about 6, 8, 11, and 12 min for B, T, E, and X, respectively.

Quality Control and Quality Assurance

A calibrated rotameter was used to adjust and measure the gas flow. To obtain a calibration curve, specific concentrations of each target compound were prepared in a Tedlar Bag (SKC Inc., the USA) by calculating the designated volume of the liquid and volatilizing it into the sampling bag according to Eq. (1):

$$V = \frac{P \times V_{sb} \times C \times M_w}{\rho \times R \times T \times 1000} \quad (1)$$

Where V is the volume of the target compound (µL), P is the inside pressure of the sampling bag (atm), V_{sb} is the volume of the sampling bag (L), C is the BTEX concentration (ppmv), Mw is the molecular weight of BTEX (g/mol), ρ is the density of BTEX (g/mL), R is the universal gas constant (L atm/mol K), and T is the temperature (K). The prepared concentrations (50–400 ppm) were injected into a GC/FID column, and a calibration curve was generated by plotting the BTEX concentrations against the area under the peak (21).

Laboratory Unit

A semi-packed scrubber filled with waste edible oil was used to absorb BTEX. The experimental setup of the laboratory unit is shown in Figure 1. BTEX compounds were separately injected into a carrier gas (with a flow rate of 200 mL/min) and, subsequently, volatilized (BTEX-N_2 mixture). The mean concentrations (ppm) for the BTEX compounds were 89.87 ± 8.62, 89.80 ± 5.60, 87.53 ± 13.00, and 85.12 ± 7.50, respectively. The volatilized compounds were introduced from the bottom of the absorption column and contacted with the thin film of waste edible oil covering the Raschig rings. In the next step, samples were taken from the top of the column and analyzed. Finally, the removal efficiency was calculated according to Eq. (2):

$$Eff,(\%)=\left(\frac{I-O}{I}\right)\times 100 \quad (2)$$

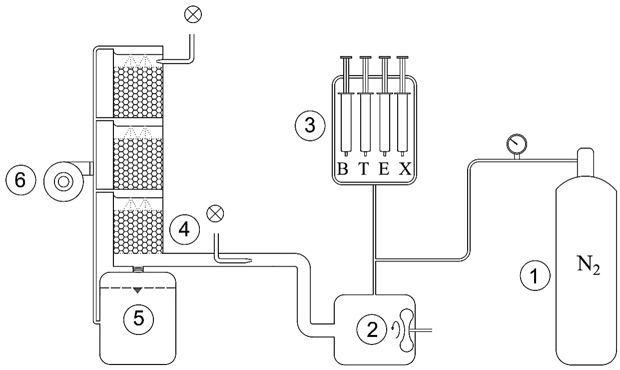

Figure 1. Scheme of the laboratory unit: (1) carrier gas, (2) agitation unit, (3) injection device, (4) absorption column, (5) oil storage chamber, (6) circulating pump, (⊗) sampling points

Where *I* and *O* are inlet and outlet concentrations, respectively. The characteristics of the absorption units are presented in Table 1.

Statistical analysis

Data analysis was conducted using IBM SPSS Statistics 16 software (IBM SPSS Inc., Armonk, NY, USA). The removal efficiencies of BTEX compounds are presented as mean ± standard deviation (SD). The normality of the data was evaluated using the Kolmogorov-Smirnov test, indicating that the efficiencies did not follow a normal distribution among the levels. Consequently, nonparametric methods were applied, and the Kruskal-Wallis test was used to compare the efficiencies across different heights and waste oils, with a significance level of 0.05.

Results

Absorption of BTEX Compounds from Oil by Scrubber

In this study, waste edible oil, a non-polar absorbent, was used to absorb non-polar organic compounds. According to the results, average B showed the highest removal efficiency, followed by X, T, and E.

Effects of Operational Parameters on the Removal of BTEX Compounds

Bed Height

In the present study, we investigated the effect of bed height on the removal efficiency of BTEX compounds. The results indicated a direct correlation between bed height and pollutant removal rates. Moreover, the statistical test revealed a significant difference in efficiency among the heights for all target compounds except E. With a constant inlet flow rate, the EBCT values for bed heights of 15, 30, and 45 cm were 1.19, 2.38, and 3.57 minutes, respectively. As shown in Figure 2B had the highest removal rate at a height of 15 cm, followed by X, T, and E. This order was consistent at bed heights of 30 cm and 45 cm. The findings indicate that column height significantly increases the removal efficiency of BTEX compounds.

Table 1. Characteristics of the absorption unit

Scrubber height	Three heights of 15, 30, and 45 cm with a diameter of 7 cm were considered for the absorption column.
Scrubber pack	A ceramic Raschig ring was used as the packing material, with a depth of 10 cm for each height of the absorption column.  Height: 6 mm Coefficients: a: 771.9 (m^3/m^2) e: 0.62 (m^3/m^3)
Scrubber absorbent	800 mL of edible oil at 230°C was heated 1 to 4 times (W_1, W_2, W_3, and W_4). After cooling, the absorbent was filtered and circulated (200 mL/min) into the column.

Frequency of Oil Reuse

The effect of oil reuse (W1, W2, W3, W4) on target compound removal was innovatively investigated. Changes in the appearance of used oils are shown in Figure 3. The removal efficiency of the target compounds concerning the frequency of oil reuse is shown in Figure 4.

As shown in Figure 4, repeated use of the oil resulted in a slight decrease in efficiency for compounds X and B. In contrast, no clear pattern was observed for compounds T and E. According to the statistical test, the removal efficiencies did not change significantly for any of the chemicals among the waste oils. Generally, W1 exhibited the highest removal efficiency for all compounds. The order of removal was B > X > T > E for W1 and W2, X > B > T > E for W3, and B > T > X > E for W4. B showed the highest removal efficiency, while E showed the lowest. Based on the findings, W1 exhibited the highest removal efficiency, followed by W3, W4, and W2.

Contact Time

The removal efficiency of the target pollutants was measured over time (0, 5, 10, 15, and 20 minutes) by comparing the concentrations before and after the absorption unit. As shown in Figure 5, the total removal efficiencies ranged from 91.60% to 98.84% for B, 86.37% to 97.58% for T, 41.74% to 91.17% for E, and 76.96% to 94.78% for X over the observed periods.

The absorption efficiency of all pollutants decreased

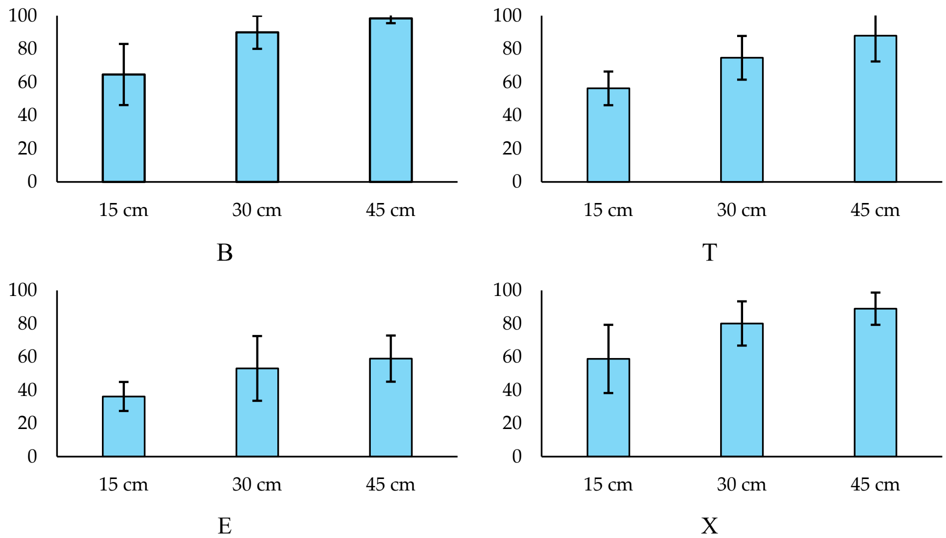

Figure 2. The effect of the height of the absorption unit on the removal of BTEX compounds (W_1, 20 min)

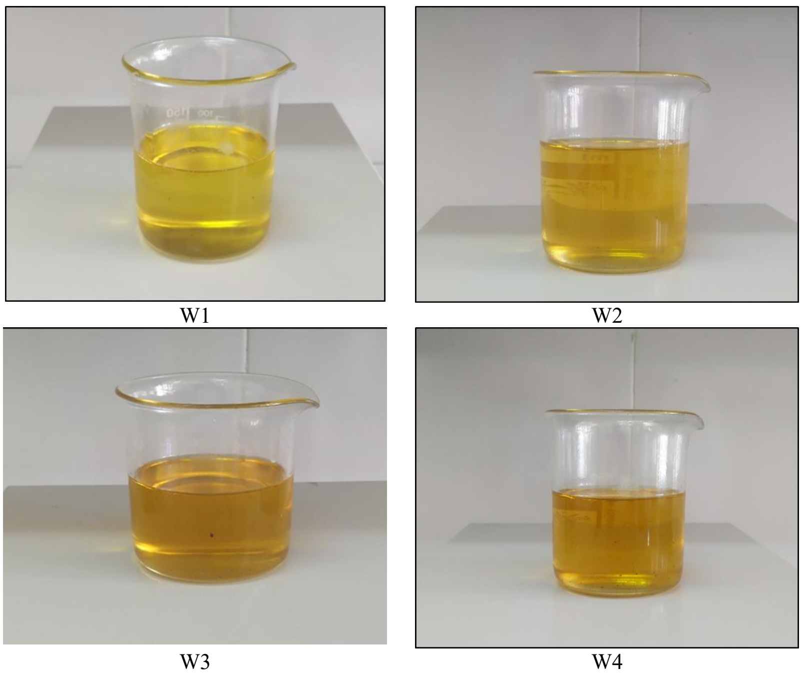

Figure 3. The appearance of the burnt oils used in the scrubbers

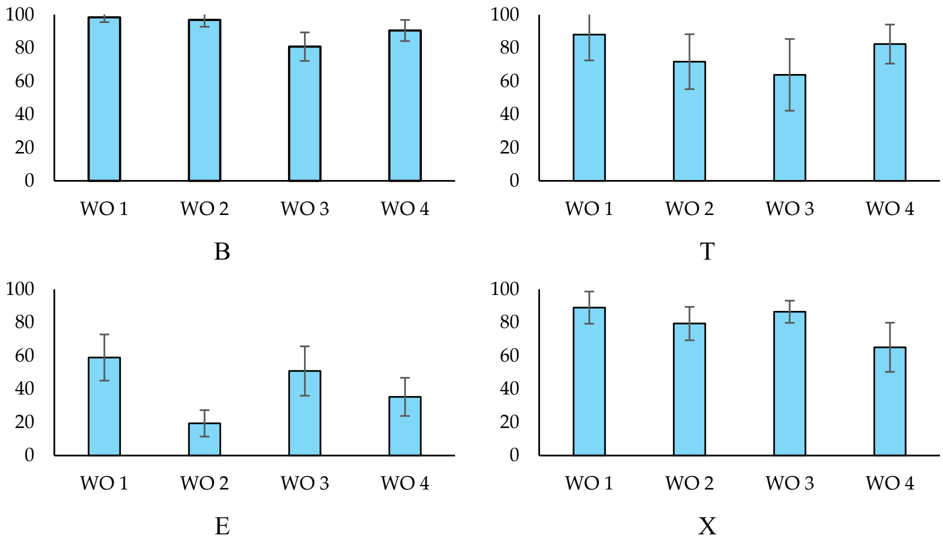

Figure 4. Effect of oil reuse frequency on the removal of BTEX compounds from the frying process (height 45 cm)

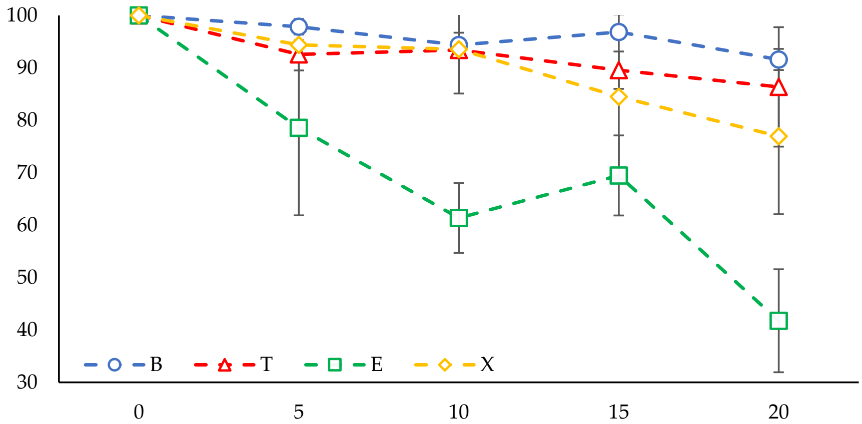

Figure 5. The removal efficiency of BTEX compounds from the frying process over time (W1, height 45 cm)

significantly over time, but at varying rates. According to the results, the highest slope was related to E, followed by X, T, and B. The absorption capacities for pollutants B, T, E, and X were calculated as 0.022, 0.020, 0.017, and 0.018 mg/g of absorbent, respectively.

Discussion

In the absorption process by the scrubber, three main parameters affect the efficiency: pollutant properties (concentration, solubility, chemical structure, and polarity), absorbent properties (flow rate, density, viscosity, chemical structure, polarity, and temperature), and scrubber design parameters (height, diameter, bed properties, and bed pore thickness) (21,27,28). The pollutant flow rate and concentration increase the speed of molecular movement, giving the absorbent less opportunity to contact them. Conversely, an increase in oil flow rate provides more opportunities for absorption due to its abundance. Therefore, theoretically, reducing the flow rate of the pollutant carrier gas or increasing the flow rate of the absorbent material enhances absorption efficiency (21,29).

However, practical increases in absorbent flow rate may not constantly significantly improve efficiency, depending on the chemical properties of the absorbing material. For instance, Chen et al. (29) found that the absorbent flow rate had little effect on NO_2 removal by Na_2SO_3 in a packed scrubber until the ratio of absorbent flow to gas flow increased by three times. Similarly, Chen et al. (30) investigated the effect of pollutant concentration, absorbent flow rate, and gas flow rate on CO_2 removal by amines, identifying gas flow rate as the most influential parameter, followed by pollutant concentration and absorbent flow rate.

According to the principle of "like dissolves like," polar solutes dissolve in polar solvents and non-polar solutes in non-polar solvents (20,31). Shorter alkyl chain lengths can improve the absorption process, as demonstrated by studies showing higher diffusion of aromatic compounds in oil with shorter alkyl chains (32). Biard et al. (33) investigated VOC removal using a water-containing scrubber, finding poor efficiency due to the mixed hydrophilic and hydrophobic nature of VOC. Improved results were obtained using a scrubber with organic solvent in series with the water scrubber. Tarnpradab et al. (31) demonstrated high absorption rates for B, T, styrene, phenol, indene, and naphthalene using waste edible oil.

As mentioned earlier, B showed the highest removal efficiency, followed by X, T, and E. This order can be attributed to the compounds' physical and chemical properties (27). The higher absorption of B may be due to its lower molecular weight compared to the others (34). Additionally, volatility affects absorption, as low boiling temperature and high volatility can reduce absorption potential. Vuong et al. (34) have stated that three organic solvents, due to their low boiling temperatures and high volatilities, have a higher potential for absorbing dimethyl sulfide than T. This study demonstrated that the low boiling temperature and high volatility of dimethyl disulfide led to a reduction in absorption. Similarly, low ethanol absorption was attributed to its low boiling point and the presence of non-polar substances, like glycerol and glycol, in the absorbent (35). Despite high volatility reducing absorption, factors such as low molecular weight and polarity (non-polarity of the absorbent and absorbate) can mitigate this effect. Vegetable oils, containing glycerides with oxygen atoms, are not completely non-polar and can remove compounds with low polarity (20,27).

Scrubber design and related physical parameters can directly affect the efficiency (28). These parameters provide conditions for maximum contact surface and time between contaminated air and the absorbent. For example, Chiang et al. (36) used a rotary-packed scrubber to achieve maximum contact surface by creating a thin layer of oil on the packed media via centrifugal force. Their results showed a direct relationship between circulation speed and the removal efficiency of X and T. The effect of bed height on the removal efficiency of BTEX compounds was surveyed. The results indicated a direct correlation between bed height and pollutant removal rates. Specifically, increasing the bed height increased the contact time between the absorbent and the pollutant (33), which was calculated according to the EBCT values. Two parameters, inlet gas flow rate and column volume, affect the EBCT (37). With a constant inlet flow rate, the EBCT values for bed heights of 15, 30, and 45 cm were 1.19, 2.38, and 3.57 minutes, respectively.

Additionally, increasing the bed height can enhance the absorbent's holding capacity, reduce the column's cross-sectional area, increase the collision rate between gas and liquid, and ultimately improve the mass transfer rate. A taller bed can also enhance mass transfer dynamics, facilitating better distribution and movement of chemicals toward the absorbent particles (38,39). Bhoi et al. (39) investigated the effects of various parameters, including solvent type (rapeseed and soybean oil), column height, absorbent temperature, and absorbent flow rate, on the removal efficiency of compounds such as B, T, and E. The results showed that changes in column height (0.5, 0.8, and 1.1 meters) significantly impacted removal efficiency. Specifically, increasing the height from 0.5 to 1.1 meters improved B removal efficiency from 90% to 97%, although the increase was not significant for T and E. Liu et al. (40) measured the SO_2 absorption rate at different scrubber heights using H_2O, $NaHCO_3$, Na_2CO_3, Na_2SO_3, and $NaHSO_3$ absorbents. They observed that the absorption rate increased with height up to a certain point, then stabilized at heights between 2.5 and 3 meters, which they attributed to absorbent saturation. Dashliborun et al. (38) examined the effect of packing type (M500X and Raschig rings), perpendicular angle (0, 5, 10, and 15 degrees), and column height (0.45 and 0.9 meters) on CO_2 removal using amine. They found that increasing the height at a zero-degree angle improved scrubber efficiency from 75% to 87% with the Raschig ring pack. Liu et al. (40) measured the SO_2 absorption rate at different scrubber heights using H_2O, $NaHCO_3$, Na_2CO_3, Na_2SO_3, and $NaHSO_3$ absorbents. They observed that the absorption rate increased with height up to a point, stabilizing at heights between 2.5 and 3 meters, which they attributed to absorbent saturation.

The effect of oil reuse (W1, W2, W3, W4) on the removal of the target compound was investigated. Waste edible oil exhibits hydrophobic characteristics and possesses a distinctive composition of fatty acids and triglycerides. This attribute may facilitate the solubility of specific aromatic compounds, thereby enhancing their absorption (41). Considering that the oil used in the first phase of the study had been reused multiple times, it is expected that it had changed its chemical properties and accumulated impurities (20,42-44), leading to changes in its appearance (Figure 3). Based on the findings, the removal efficiencies did not change significantly for any of the chemicals among the waste oils. W1 exhibited the highest removal efficiency, followed by W3, W4, and W2. The higher removal efficiency of W1 may be attributed to chemical reactions such as oxidation and hydrogenation that occur during heating (45). Additionally, the presence of food ingredients in the oil can lead to the formation of other pollutants, which can affect absorption efficiency (21). W2, W3, and W4 removed lower concentrations. Generally, oil burning increases the viscosity of the fluid and changes its color. Viscosity is considered an influential parameter for absorption. Higher viscosity prevents the formation of smaller droplets, resulting in weaker diffusion of volatile organic compounds in the oil (27,35,46). Therefore, the most probable reason for the decreased efficiency is the increased viscosity caused by heating, which is associated with chemical changes in the oil (21). In a study (45), the authors compared the T-removal efficiency of sunflower oil with high oleic acid content to that of commercial sunflower oil. The findings indicated that the removal efficiencies of both oils were comparable and not statistically different. This could be attributed to opposing factors: sunflower oil's low resistance to oxidation due to its high oleic acid content, and its high Henry's law constant for T absorption. However, the lower resistance of the oil to oxidation (3.5 times less) results in chemical modifications, reducing its absorption capacity. On the other hand, the higher Henry's law constant of this oil compared to commercial oil allows it to absorb more T.

Changing the absorbent phase from organic to inorganic, and vice versa, alters the rate of pollutant transfer from the gaseous to the liquid phase. Lalanne et al. (29) used a semi-industrial bioscrubber containing different percentages of oil and water to remove three groups of volatile organic compounds, including aromatic, oxygenated, and halogenated compounds. Their results showed that increasing the percentage of oil relative to water increased the efficiency of aromatic compound removal, resulting in faster saturation of the absorbent over time.

The absorption capacity and removal pattern of target chemicals over 20 minutes were considered. According to the results, E had the highest slope attributed to the lower capacity, followed by X, T, and B. The difference can be due to factors such as mass transfer, reaction kinetics, and the characteristics of the sorbate (47). Nakamura et al. (48) studied the absorption of tar by organic oil for 22.5 hours, finding removal efficiencies ranging from 60% to 70%, indicating a lower tendency of the absorbent material due to the presence of various pollutants. In another study (21), the absorption efficiencies of waste vegetable oil for removing compounds B and T were compared. The results indicated that the absorption efficiency for B at 400 minutes and T at 700 minutes was below 10%, showing a higher removal capacity of the oil. Tarnpradab et al. (31) observed a similar pattern after comparing the absorption of B and T by waste edible oil over 600 minutes. However, in the study by Bhoi et al. (49), the absorption of B, T, and E in soybean oil at a temperature of 50°C showed a different order, with E having the lowest slope (highest absorption efficiency), followed by T and B. This difference can be attributed to the volatility of the compounds. Another probable reason is the chemical properties of soybean oil, which contains high levels of glyceride compounds.

Conclusion

This study investigated the potential of waste edible oil for removing BTEX compounds in packed scrubbers. The key findings reveal that the high absorption efficiency of waste edible oil, particularly for B, can be attributed to its non-polarity and compatibility with non-polar BTEX compounds. Additionally, the study highlights the significance of bed height in the packed scrubber system, indicating that increasing the height enhances the removal efficiency of BTEX components for a longer contact time and improves mass transfer. Furthermore, heated oils, especially W1, were found to play a significant role in removal efficiency, suggesting that the properties of the oil and the frequency of its usage affect absorption. The observations regarding the progressive decrease in absorption efficiency over time emphasize the need for periodic replacement or regeneration of the absorbent to maintain the effectiveness of the scrubber system. In conclusion, the research supports the argument that waste edible oil can be an effective absorbent for BTEX compounds, offering a novel approach to addressing environmental pollution. Further optimization of scrubber design parameters, such as bed height and oil reuse strategies, could enhance the economic and ecological benefits of this technology. The study advocates for broader adoption and continuous improvement of waste edible oil absorption systems for the remediation of harmful industrial emissions, contributing to cleaner air and a healthier environment.

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Authors' contributions

Conceptualization: Ali Atamaleki.
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Formal analysis: Ali Atamaleki.
Investigation: Ali Atamaleki.
Methodology: Ali Atamaleki, Saeed Motesaddi Zarandi, Mohamadreza Massoudinejad.
Project administration: Saeed Motesaddi Zarandi.
Software: Ali Atamaleki.
Supervision: Mahdi Ghorbanian.
Validation: Mahdi Ghorbanian, Elham Rahmanzadeh.
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Writing–review & editing: Mahdi Ghorbanian, Amir Mohammad Sheikh Asadi.

Competing interests

The authors declare that they have no conflict of interest regarding the publication of this paper. The authors have not received any financial support or benefits from any organization or individual that could influence the research or the results. Additionally, the authors declare that they have not been involved in any activities or relationships that could create a potential conflict of interest with the subject matter or materials discussed in this paper.

Ethical issues

The project study mentioned in this article has undergone ethical evaluation at Shahid Beheshti University of Medical Sciences (Ethical code: IR.SBMU.PHNS.REC.1399.150). The authors confirm their agreement for the publication of this article.

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Shahid Beheshti University of Medical Sciences founded this study.

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