



## First report on BTEX leaching from waterpipe tobacco wastes (WTWs) into aquatic environment

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### ABSTRACT

Waterpipe tobacco wastes (WTWs) may contain considerable levels of hazardous contaminants such as BTEX (benzene, toluene, ethylbenzene, o-xylene, and m/p-xylene). However, no research has been carried out on BTEX levels in WTWs and the release of these pollutants into the water environment. This research examined the levels of BTEX in WTWs of flavored/local tobacco and also the release rate of these toxins into three kinds of water, including seawater (SW), tap water (TW), and distilled water (DW) with different leaching times (15, 30 min, 1.2, 4, 8 h, and 1, 2, and 4 days). The mean contents of BTEX in TW samples of Al-Mahmoud, Al-Fakher, Mazaya, Al-Ayan brands, and local tobacco samples were  $17.0 \pm 4.14$ ,  $19.1 \pm 4.65$ ,  $19.6 \pm 4.19$ ,  $18.8 \pm 4.14$ , and  $3.16 \pm 0.63$   $\mu\text{g}/\text{kg}$ , respectively. The mean BTEX levels in flavored tobacco samples were considerably greater than that of local tobacco ( $p < 0.05$ ). The WTWs leaching experiments showed that the levels of BTEX ranged from 5.26 to 6.12, 5.02–5.60, and 3.83–5.46  $\mu\text{g}/\text{L}$  in DW, TW, and SW, respectively. All target compounds were found for all exposure times in DW, TW, and SW samples. After adding sodium azide as an antibacterial agent to water samples (simulating biodegradation processes), higher levels of BTEX compounds were detected in SW. Further

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research is needed to address the potential environmental hazards due to WTWs leaching into aquatic environments.

## 1. Introduction

Tobacco smoking remains one of the biggest risk factors for morbidity/mortality at the global level. Based on the report of World Health Organization (WHO), nearly 7 million people die each year because of tobacco smoking [1,2], and this estimate will increase to 8.3 million annual deaths by 2030 [3]. Tobacco is consumed in various forms in different countries. In many countries around the world, one of the oldest ways of tobacco consumption is waterpipe tobacco smoking [4,5]. Waterpipe smoking involves passing air heated with charcoal over tobacco (either local or flavored) to create smoke [6]. This smoke is passed through water to cool and moisten the smoke to make it more facile for the consumer to inhale [7]. Waterpipe (their equivalent names include narghile, waterpipe, hubble-bubble, arghile, arguile, and shisha) are now commonly used to smoke tobacco across the Eastern Mediterranean, Africa, Asia, the United States, and other Western countries, especially among youth and young adults [8–11].

Recent epidemiological research has shown that tobacco consumption by waterpipe has negative effects on human health, including esophageal cancer, lung and oral cancers, infectious diseases, impaired respiratory function, cardiovascular disease, low birth weight, and dental problems [4,12,13]. The increasing trend of waterpipe smoking not only provides risks to more people who smoke but may also lead to increased environmental pollution from the disposal of waterpipe tobacco wastes (WTWs) [10,14]. Unlike the waste from discarded cigarettes, toxic chemicals from WTWs can be released into the aquatic and terrestrial environments through waste produced from different parts of the waterpipe system [12]. Toxic pollutants such as heavy metals, BTEX (benzene, toluene, ethylbenzene and xylene), toxic carbonyls, formaldehyde, acrolein, acetaldehyde, polycyclic aromatic hydrocarbons (PAHs), phenol and its derivatives, volatile organic compounds (VOCs), primary aromatic amines, carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), nicotine, etc., are present in considerable quantities in un-smoked waterpipe tobacco and its resulting combustion products [10,12,15,16]. These hazardous chemicals may release of improperly thrown away WTWs and contaminate water bodies (e.g., coastal, river and possibly groundwater around landfills without suitable covering to control its leachates) [17–19]. Terrestrial animals such as goats, cats, sheep, cows, dogs, etc. may also be dangerously get hurt after eating or exposure to WTWs [18,20,21]. Thus, hazardous leachates deriving from WTWs leached into water ecosystems may have detrimental impacts on aquatic animals (e.g., shrimp, fish, octopuses, mollusk), or amphibians such as frogs and newts [22–26].

To date, leachates containing toxic chemicals such as heavy metals [27–29], aromatic amines [30,31], PAHs [22,32], BTEX [33], via cigarette butts [16] have been studied, and contamination by heavy metals [34], and PAHs [35] through WTWs contamination in aquatic environments has been documented. However, no previous research has been carried out on the leaching of BTEX compounds from WTWs into aquatic environments. The current research is the first attempt to quantify the BTEX content in burnt tobacco wastes from WTWs and their leachates in three types of water including seawater (SW, collected from the Persian Gulf), tap water (TP), and distilled water (DW). The specific goals of this study are to.

- o Quantify BTEX levels in fresh flavored/local tobacco samples used in hokkah smoking;
- o Quantify BTEX levels in freshly smoked flavored/local WTW samples;
- o Determine leaching rates of BTEX from freshly smoked flavored/local WTWs into different water types (distilled water [DW], tap water [TW], and seawater [SW]);
- o Assess the eco-toxicological hazard of BTEX leached from WTWs for aquatic life.

## 2. Material and methods

### 2.1. Reagents and standards

The mix standard solution of BTEX (2 mg/mL in methanol, 1 mL) was purchased from Supelco™ and used as the standard. *N*-hexane (Merck, Germany) and GFL analytical water purification systems (GFL, Germany) were used for lab water sources to prepare the sample and stock/standard solutions. A BTEX stock solution (10 mg/L) was procured using *n*-hexane, and then the desired diluted solution were procured from the stock solution in ultrapure water. Ethylbenzene-d10 (2000 µg/mL in methanol, CAS No.25837-05-2) purchased from Supelco™ was used as internal standard.

### 2.2. Study design and sample collection

Four most commonly consumed tobacco brands in Iran, including Al-Ayan, Mazaya, Al-Fakher and Al-Mahmoud, and the 4 most popular flavors, including two-apple, mint gum, blueberry and orange-cream, were provided (5 samples of each brand/flavor). Moreover, local unflavored tobacco samples (five samples) were also provided (Fig. S1). Also, fresh flavored/local tobacco was procured to a regular waterpipe smoker and remaining WTWs in the head of the hookah was collected after smoking. These wastes were put in amber glass bottle, foil-wrapped, transferred to the laboratory in a cold box, and stored in a suitable laboratory conditions (−4 °C) until analysis.

### 2.3. Leaching experiments of BTEX from WTWs

To evaluate the leaching rate of BTEX compounds from WTWs, the established protocol by Dobaradaran et al. [33] was used. Briefly, three kinds of water experiments including DW, TW, and SW were prepared. After quantifying the of BTEX concentrations in WTWs from collected brands/flavors, the WTWs with the maximum level of  $\sum$ BTEX (WTWs of Mazaya brand with the flavor of orange-cream) was selected for leaching tests. For this purpose, 1 gr of WTWs was soaked in a 20-mL amber vial (WHEATON® Screw Thread Headspace, Canada) containing 10 mL of the target water (DW, TW, and SW, separately). The BTEX concentration levels in the leachate were then quantified using gas chromatography–mass spectrometry (GC-MS). All experiments were replicated 3 times and the average BTEX concentrations were presented in  $\mu\text{g/L}$ . To evaluate the effect of biodegradation mechanisms on BTEX compounds, 40 mg/L of sodium azide was added to aqueous samples and BTEX content was analyzed after a contact time of four days [32]. All BTEX leaching tests were conducted at laboratory temperature via WTWs and control samples.

### 2.4. Extraction of BTEX compounds and analysis procedures

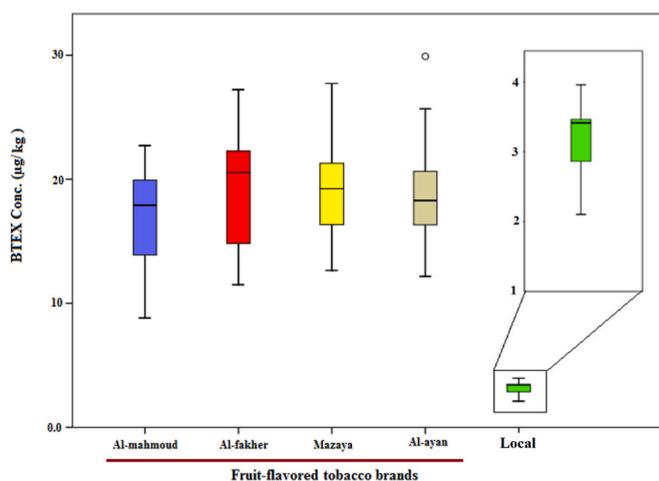
After homogenizing each type of tobacco sample (flavored/local unsmoked tobacco, flavored/local smoked tobacco waste), 1 gr of sample was placed in an appropriate flask (25 mL) and spiked with 10  $\mu\text{g/kg}$  of internal standard. Thereafter, 5 mL of *n*-hexane was added to them and sonicated for 20 min. The ultra-sonicated solution was passed over  $\text{Na}_2\text{SO}_4$  until dryness and concentrated to 1 mL utilizing a rotary evaporator (RE) at 45 °C.

### 2.5. Gas Chromatography-Mass Spectroscopy (GC/MS) instrumentation

BTEX compounds in the samples were quantified utilizing an Agilent 7820 A gas chromatograph (Palo Alto, CA) equipped with a 5977 E mass spectrometer (Gas Chromatography-Mass Spectroscopy (GC/MS)). Separation of BTEX was achieved on a HP-5MS UI, 30 m, 0.25 mm inner diameter, 0.25  $\mu\text{m}$  film thickness column (J&W Scientific, Folsom, CA). The flow rate helium carrier gas was 1.2 mL/min 1  $\mu\text{L}$  the each sample was injected in split-less mode at 310 °C. The oven temperature was first kept at 80 °C for 2 min and then raised to 280 °C with a ramp of 30 °C/min; it was kept at this temperature for 1.83 min. The temperature of the mass detector was 280 °C. The mass spectrometer was set at 70 eV electron impact and placed in selective ion monitoring (SIM) mode. The temperature of both MS transfer line and ion source was set to 230 °C and quadrupole was 150 °C.

### 2.6. Quality control (QC) and quality assurance (QA)

The analytical method for BTEX compounds was validated by determining the limit of detection (LOD) and limit of quantification (LOQ). For this purpose, control samples were prepared in parallel with the experiments on the extraction of analytes from WTWs and the leaching of BTEX compounds from WTWs into water bodies using a similar procedure (fresh tobacco/unsmoked tobacco as control matrix in the analysis of WTWs samples and water samples without WTWs as blank matrix in the analysis of liquid samples from leaching tests). In addition, the standard solution of BTEX (1000  $\mu\text{g/mL}$  each in dimethyl sulfoxide solution (DMSO) purchased from Sigma Aldrich Company was used to prepare the standard solution. The LOD and LOQ values in WTWs samples were determined based on the method presented by Ripp et al. [36]. More precisely, the accuracy/precision of the analytical method was determined by spiking 3 concentrations of internal standard (5, 10, and 20  $\mu\text{g/kg}$  in WTWs samples and 5, 10, and 20  $\mu\text{g/L}$  in liquid samples) into the blank matrix, with 3 repetitions for each concentration. The validation results including accuracy, precision, LOD and LOQ are given in



**Fig. 1.** Comparison of BTEX compounds concentration in waterpipe tobacco wastes (WTWs) of different fruit-flavored brands (Al-Mahmoud, Al-Ayan, Mazaya, and Al-Fakher) and local tobacco samples.

Table S1.

## 2.7. Data analysis

SPSS Statistics version 22 software was used to statistical analysis of the data. The Kolmogorov-Smirnov test was utilized to evaluate the normality of the observed data. The significant difference between BTEX concentrations in different categories was assessed by ANOVA and Turkey post hoc tests.  $P < 0.05$  was utilized as the criterion for differences significance.

## 3. Results and discussion

### 3.1. BTEX concentrations in WTWs of different brands

The BTEX levels in WTWs of flavored tobacco of Al-Mahmoud, Al-Ayan, Mazaya, and Al-Fakhe brands, as well as in local tobacco samples were measured and depicted in Fig. 1. The average  $\pm$  SD levels of total BTEX compounds in WTWs tobacco samples of Al-Mahmoud, Al-Fakher, Mazaya, Al-Ayan brands and local tobacco were  $17.0 \pm 4.14$ ,  $19.1 \pm 4.65$ ,  $19.6 \pm 4.19$ ,  $18.8 \pm 4.14$ , and  $3.16 \pm 0.63$   $\mu\text{g}/\text{kg}$ , respectively. The maximum level was found in Mazaya tobacco brand and the lowest value was observed in local tobacco samples. Statistical analysis showed that the BTEX concentrations in flavored tobacco samples of different brands were significantly higher ( $p < 0.05$ ) than in local tobacco samples (Table 1) while this differences was not significant among different brands of flavored tobacco. The high concentrations of BTEX in flavored tobacco samples compared to local samples can be attributed to the addition of large amounts of flavors, aroma compounds, aromatic substances, and other additives added during tobacco processing [37,38]. In addition, the level of each BTEX in WTWs tobacco samples of the four mentioned brands was higher than in local tobacco samples. As indicated in Table 1, among the BTEX compounds, the average level of toluene in all fruit-flavored and local tobacco samples were greater than other compounds. The mean concentrations of this compound in Al-Fakher, Mazaya, Al-Ayan, Al-Mahmoud, brands and local tobacco samples were 8.3, 7.62, 7.23, 7.11 and 2.3  $\mu\text{g}/\text{kg}$ , respectively. It should be noted that toluene is a toxic compound that can cause adverse human health endpoints such as headaches, central nervous system damage, loss of control and consciousness, and even death from short-term exposure [39]. Therefore, the discharge of tobacco waste containing large amounts of toluene and/or other BTEX compounds into the environment may have negative impacts on human health as well as aquatic organisms [7]. Unfortunately, no similar reports were found that examined the BTEX concentrations in the waste products of flavored and local tobacco samples to compare the findings of the current study with. In our previous research, greater BTEX level was reported in indoor air of hookah cafés than cigarette cafés [7]. In a study conducted by Ghobadi et al. [40], the BTEX content in urine of waterpipe smokers (99 smokers) and non-smokers (31 non-smokers) was analyzed. They report that the mean levels of benzene, toluene, ethylbenzene, o-xylene, and m/p-xylene in the hookah tobacco smokers were 471, 671, 127.91, 90.6, and 46.0 ng/g, respectively, with toluene and benzene having the highest concentrations [40].

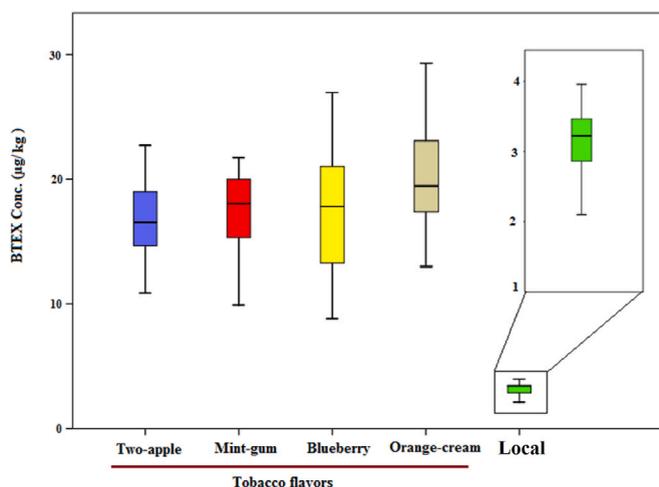
### 3.2. The BTEX levels in WTWs from different flavored and local tobacco samples

The mean  $\pm$  SD levels of BTEX in the WTWs of four flavored (Two-apple, Mint-gum, Blueberry, and Orange-cream) and local tobacco samples were determined (Fig. 2 and Table 2). As seen, the average levels of  $\Sigma$ BTEX compounds in the WTWs of various flavors including Two-apple, Mint-gum, Blueberry, Orange-cream, and local tobacco samples were  $16.7 \pm 3.04$ ,  $17.4 \pm 3.27$ ,  $17.5 \pm 4.72$ ,  $22.9 \pm 3.29$  and  $3.16 \pm 0.63$   $\mu\text{g}/\text{kg}$ , respectively. The average concentrations of  $\Sigma$ BTEX compounds in tobacco samples followed the order of Orange-cream > Blueberry > Mint-gum > Two-apple > local. Based on the results, the maximum levels of BTEX in WTWs samples was observed in Orange-cream flavored tobacco and the lowest concentration was observed in local tobacco samples (Fig. 2). The higher BTEX levels in orange-cream tobacco could be attributed to the cream in such samples which includes fat as part of the

Table 1

BTEX concentration levels (mean  $\pm$  SD,  $\mu\text{g}/\text{kg}$ ) in HTWs samples collected from different brands.

Tobacco brands	Statistical analysis	Benzene	Toluene	Ethylbenzene	O-Xylene	P-Xylene	$\Sigma$ BTEX
Al-Mahmoud	Mean $\pm$ S.D	$1.76 \pm 0.34$	$7.11 \pm 1.75$	$2.62 \pm 0.98$	$1.83 \pm 0.61$	$3.66 \pm 1.22$	$16.98 \pm 4.14$
	Comparison with Al-Fakher	$P = 0.62$	$P = 0.39$	$P = 0.74$	$P = 0.59$	$P = 0.45$	$P = 0.57$
	Comparison with Mazaya	$P = 0.48$	$P = 0.47$	$P = 0.53$	$P = 0.40$	$P = 0.38$	$P = 0.46$
	Comparison with Al-ayan	$P = 0.41$	$P = 0.73$	$P = 0.81$	$P = 0.49$	$P = 0.31$	$P = 0.54$
Al-Fakher	Mean $\pm$ S.D	$1.91 \pm 0.41$	$8.3 \pm 2.6$	$2.79 \pm 1.31$	$2.03 \pm 0.85$	$4.11 \pm 1.47$	$19.14 \pm 4.65$
	Comparison with Mazaya	$P = 0.71$	$P = 0.62$	$P = 0.63$	$P = 0.76$	$P = 0.52$	$P = 0.68$
	Comparison with Al-ayan	$P = 0.65$	$P = 0.42$	$P = 0.90$	$P = 0.91$	$P = 0.44$	$P = 0.67$
	Comparison with local	$P < 0.05$					
Mazaya	Mean $\pm$ S.D	$2.08 \pm 0.54$	$7.62 \pm 1.3$	$3.21 \pm 1.32$	$2.12 \pm 0.98$	$4.52 \pm 1.97$	$19.55 \pm 4.19$
	Comparison with Al-ayan	$P = 0.86$	$P = 0.64$	$P = 0.52$	$P = 0.93$	$P = 0.88$	$P = 0.68$
	Comparison with local	$P < 0.05$					
Al-Ayan	Mean $\pm$ S.D	$2.12 \pm 0.53$	$7.23 \pm 1.29$	$2.78 \pm 1.5$	$2.07 \pm 0.86$	$4.62 \pm 1.66$	$18.82 \pm 4.14$
	Comparison with local	$P < 0.05$					
Local	Mean $\pm$ S.D	$0.19 \pm 0.07$	$2.3 \pm 0.55$	$0.27 \pm 0.1$	$0.21 \pm 0.1$	$0.4 \pm 0.17$	$3.16 \pm 0.63$



**Fig. 2.** Comparisons of BTEX compound levels in WTWs of different flavors (Orange-cream, Blueberry, Mint-gum, and Two-apple) and local tobacco samples.

**Table 2**

BTEX concentration levels (mean  $\pm$  SD,  $\mu\text{g}/\text{kg}$ ) in HTWs samples of different flavors.

Tobacco brands	Statistical analysis	Benzene	Toluene	Ethylbenzene	O-Xylene	P-Xylene	$\Sigma$ BTEX
<b>Two-apple</b>	Mean $\pm$ S.D	1.76 $\pm$ 0.29	7.16 $\pm$ 1.75	2.32 $\pm$ 1.06	1.81 $\pm$ 0.7	3.67 $\pm$ 1.25	16.72 $\pm$ 3.04
	Comparison with Mint-gum	P = 0.98	P = 0.91	P = 0.59	P = 0.98	P = 0.41	P = 0.83
	Comparison with Blueberry	P = 0.61	P = 0.86	P = 0.51	P = 0.86	P = 0.42	P = 0.75
	Comparison with Orange-cream	P = 0.35	P = 0.39	P = 0.18	P = 0.32	P = 0.08	P = 0.23
	Comparison with local	P < 0.05					
<b>Mint-gum</b>	Mean $\pm$ S.D	1.75 $\pm$ 0.34	7.22 $\pm$ 1.35	2.68 $\pm$ 0.9	1.80 $\pm$ 0.68	3.95 $\pm$ 1.02	17.41 $\pm$ 3.27
	Comparison with Blueberry	P = 0.60	P = 0.81	P = 0.77	P = 0.85	P = 0.98	P = 0.84
	Comparison with Orange-cream	P = 0.34	P = 0.41	P = 0.28	P = 0.32	P = 0.15	P = 0.26
	Comparison with local	P < 0.05					
<b>Blueberry</b>	Mean $\pm$ S.D	1.96 $\pm$ 0.56	6.85 $\pm$ 1.86	2.80 $\pm$ 1.28	1.92 $\pm$ 0.79	3.95 $\pm$ 1.76	17.48 $\pm$ 4.72
	Comparison with Orange-cream	P = 0.34	P = 0.23	P = 0.39	P = 0.36	P = 0.14	P = 0.24
	Comparison with local	P < 0.05					
<b>Orange-cream</b>	Mean $\pm$ S.D	2.4 $\pm$ 0.38	9.03 $\pm$ 1.67	3.61 $\pm$ 1.55	2.51 $\pm$ 0.97	5.33 $\pm$ 0.86	22.87 $\pm$ 3.29
	Comparison with local	P < 0.05					
<b>Local</b>	Mean $\pm$ S.D	0.19 $\pm$ 0.07	2.3 $\pm$ 0.55	0.27 $\pm$ 0.1	0.21 $\pm$ 0.1	0.4 $\pm$ 0.17	3.16 $\pm$ 0.63

flavoring. Previous research suggests that fat in waste product combustion may generate organic pollutants (e.g., BTEX, PAHs etc.) [41, 42]. According to previous reports, fat content may be associated with BTEX emission [43–45]. Indeed, the oxidation of fatty acids may be accelerated at the high temperatures measured in the waterpipe head, leading to more BTEX generation [43,46].

The concentration level of each BTEX compound in WTWs samples are given in Table 2. The highest level of BTEX compounds in all flavored and local tobacco samples were related to toluene. The averages concentration of this pollutant in flavored tobacco samples including Orange-cream, Mint-gum, Two-apple, Blueberry, and local tobacco samples were 9.03  $\pm$  1.67, 7.22  $\pm$  1.35, 7.16  $\pm$  1.75, 6.85  $\pm$  1.86, and 2.3  $\pm$  0.55, respectively. The findings of the statistical test also indicated that the BTEX concentrations in different fruit-flavored tobacco samples were considerably ( $p < 0.05$ ) greater than local tobacco samples. Unfortunately, there has been no previous research on the BTEX levels in post-consumption waste of flavored and local tobacco samples to compare to our results. In a project done by Polzin et al. [47], the maximum and minimum concentrations of BTEX compounds in 41 cigarette brands sold in the United States were found for toluene (4.5–82.4  $\mu\text{g}/\text{cig}$ ) and o-xylene (0.2–3  $\mu\text{g}/\text{cig}$ ), respectively. Also, the average concentration levels of benzene, toluene, ethylbenzene, m/p-xylene, and o-xylene in cigarette mainstream smoke were 44.1, 57.4, 4.4, 9.9, and 1.7  $\mu\text{g}/\text{cig}$ , respectively [47].

### 3.3. Leachates of BTEX from WTWs into water environments

To evaluate the release of BTEX compounds from WTWs into water, three water samples including DW, TW, and SW were analyzed at specific contact times (15, 30 min, 1.2, 4, 8 h, and 1, 2, and 4 days) (Fig. 3, Tables S2–S4). All BTEX compounds were detected in all three DW, TP, and SW samples, and the highest concentration levels were for toluene in three water samples. As shown, total concentration levels of BTEX compounds at different contact times ranged from 5.26 to 6.12, 5.02–5.6, and 3.83–5.46  $\mu\text{g}/\text{L}$  in DW, TW, and SW samples, respectively. The BTEX concentrations in DW and TW leachate samples (5.26–6.12  $\mu\text{g}/\text{L}$ ) were higher than in SW

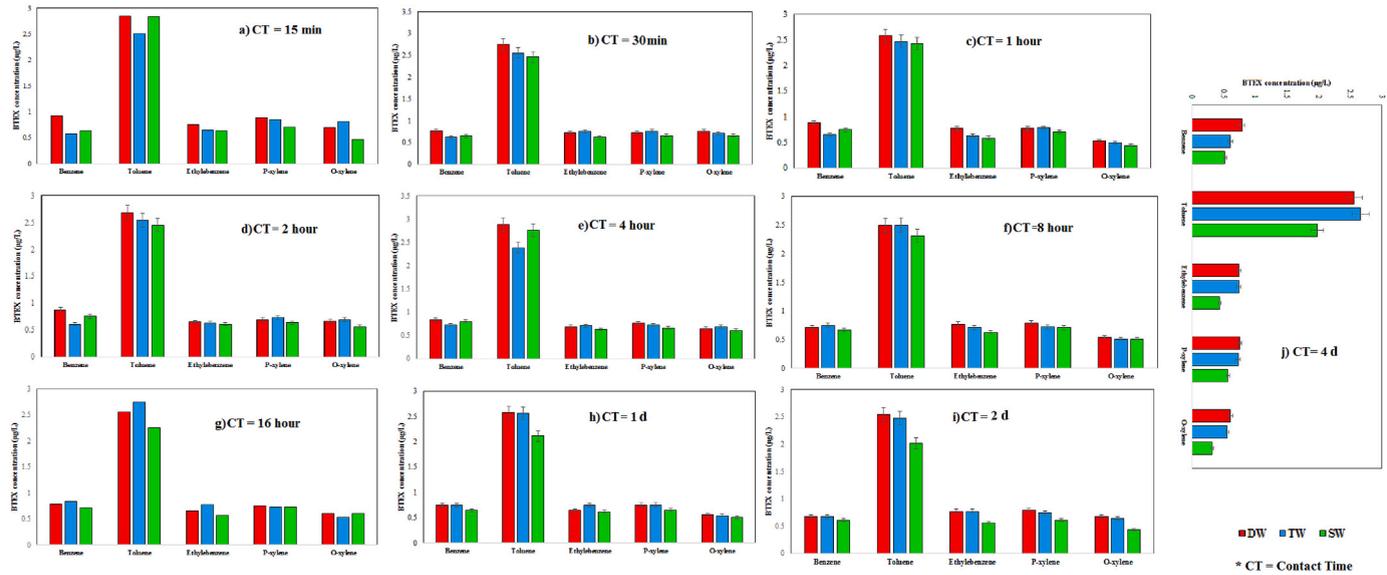
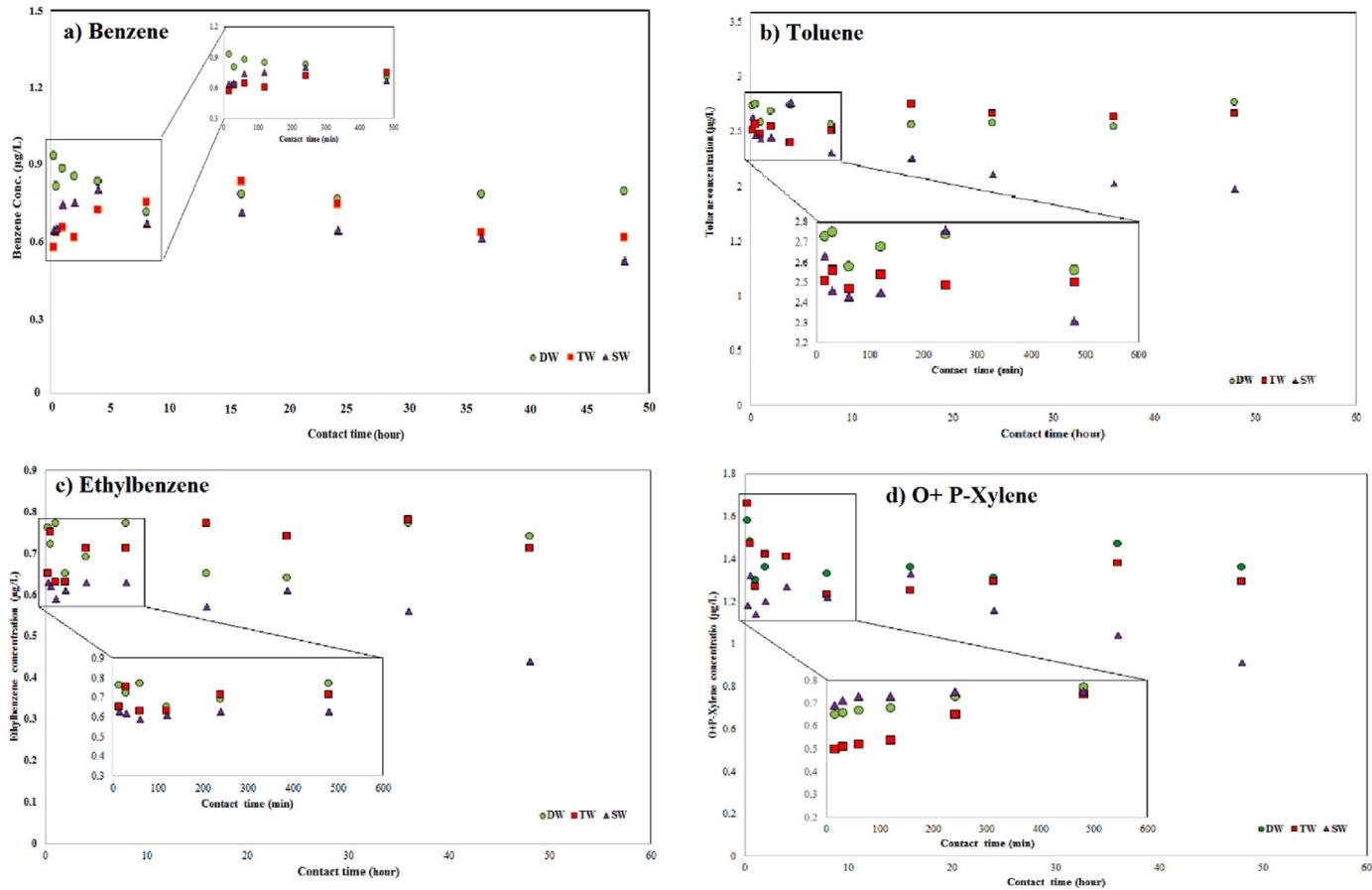


Fig. 3. Comparison of BTEX leachates compounds ( $\mu\text{g/L}$ ) from WTWs into different types of water (distilled water (DW), tap water (TW) and sea water (SW)) at different contact times (a: 15 min, b: 30 min, c: 1 h, d: 2 h, e: 4 h, f: 8 h, g: 16 h, h: 1 day, i: 2 days and j: 4 days).



**Fig. 4.** The release rate of a) benzene, b) toluene, c) ethylbenzene and d) O + P/M-xylene into different types of water (distilled water (DW), tap water (TW) and sea water (SW)) at different contact times.

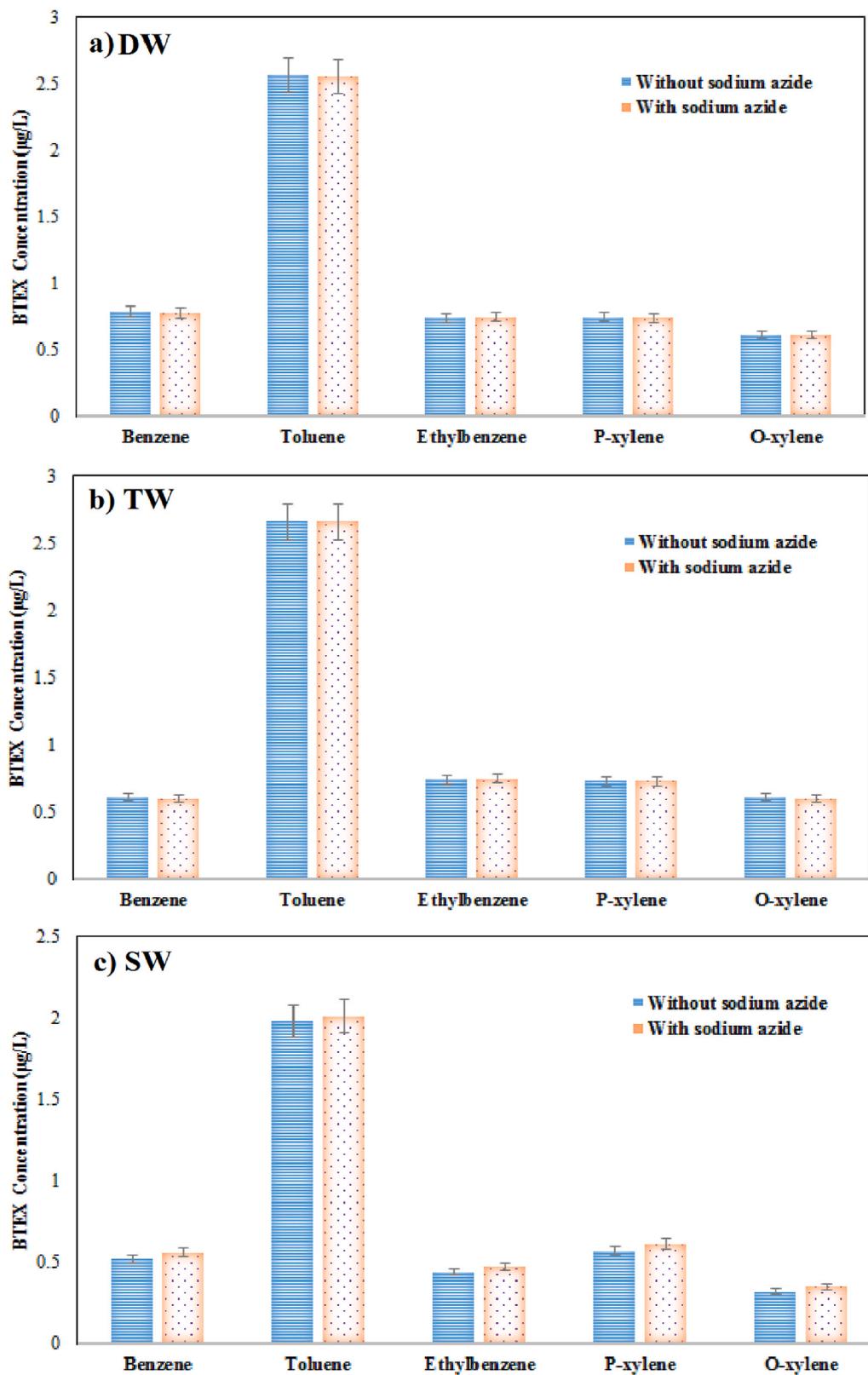


Fig. 5. Concentration levels of benzene, toluene, ethylbenzene, m/p-xylene, and o-xylene in different water samples (a: DW, b: TW and c: SW) with/without adding sodium azide.

leachate samples. However, the differences were observed among the levels of BTEX compounds in DW and TW samples at all contact times was not significant (Fig. 4). In line with a previous study by Dobaradaran [33] for cigarette butts, the highest concentration of BTEX analyts in the leachate samples was for toluene. They also found that the difference between the concentration level of each BTEX compound in DW and river water (RW) samples at the studied contact times was not significant, which is in agreement with the observations of the current work [33]. Our previous study showed that the leachate levels of PAHs from WTWs into water bodies including DW, TW, and RW ranged from 0.13 to 3.51, 0.12 to 3.63 and 0.11–3.64  $\mu\text{g/L}$ , respectively, at contact times of 15 min to 2 months [35]. There were also no considerable differences between the PAHs concentrations in all three leachate samples [35]. It is noteworthy that, benzene has greater vapor pressure in comparison with toluene and tends to become airborne, and toluene has high solubility in solution compared to other BTEXs. Therefore, it is not surprising that the concentrations of toluene in WTWs leachates was greater than other BTEX compounds [33,48]. In a study similar to this research, the urinary levels of BTEX in the smoker (as exposure biomarkers of toxics) were studied. The urinary levels of benzene/toluene of smokers was 4.6 and 1.2 times higher than for non-smokers [49].

### 3.4. The effect of sodium azide

Sodium azide was used as a chemical preservative to investigate the effects of biodegradation of BTEX compounds in leachates. The results indicated that after a period of two months, there was no significant difference between the concentration level of each BTEX compound in selected water environments (DW, TW and SW) without and with addition of sodium azide (Fig. 5 a-c). The higher amount of BTEX compounds in water without addition of sodium azide can be assumed to be similar to microbial degradation in seawater [50,51], justified in part by the significant microorganism populations in sea water [50]. Similar observations have been reported in previous research on the release of PAHs from burnt tobacco wastes [35] and cigarette butts [32]. Also, the findings regarding PAHs distribution in other matrixes (e.g., soil, sediment, etc.) [52,53] were in accordance with our observations.

### 3.5. Ecological risk effects of leachable BTEX

There is no established regulation regarding BTEX compounds in WTWs leachates to protect aquatic ecosystems and human health from these hazardous chemicals. Thus, we compared the final BTEX levels in different water samples in our study to European environmental quality standards (EQSs) as Water Framework Directive (WFD) standards [54]. It should be noted that this standard only establishes guidance for benzene, whose annual mean level for inland surface waters and other surface waters is given as 10 and 8  $\mu\text{g/L}$ , respectively. It has not established any limits or guidelines for other BTEX compounds such as toluene, ethylbenzene, m/p-xylene and o-xylene. Our findings indicate that benzene concentrations in WTWs leachates are lower than recommended WFD standards by EQSs values for inland and other surface waters. A similar study by Dobaradaran et al. reported similar results for cigarette butt leachates [33]. Due to the increasing consumption of tobacco worldwide, a large amount of waste is generated each year that can be released into the environment and water bodies. This waste may contain various toxic compounds including BTEX, which have serious detrimental impacts on human health and aquatic organisms. Moreover, tobacco waste may also contain large amounts of hazardous pollutants such as toxic elements, PAHs, and other toxic pollutants [32]. Due to these facts, regulatory measures are needed to mitigate the environmental impacts of all tobacco wastes (cigarettes butts, discarded e-cigarettes, and WTWs), which are persistent and ubiquitous environmental problems. To reduce the tobacco wastes in the environment, it is proposed to set up and adopt appropriate waste management options based on the regulations and strategies established for tobacco wastes [12]. In addition, the environmental-friendly techniques using  $\text{CO}_2$ -mediated pyrolysis process can be another useful option to minimize the generation of hazardous matters and for energy recovery from these wastes [55].

## 4. Conclusion

The findings of this work suggest that the wastes of burned tobacco from water pipe smoking contain hazardous pollutants and may result in the disbursement of these hazardous pollutants into various media, including water bodies, as well as negative impacts on human health and the environment. The study showed that the BTEX levels in different flavored tobacco samples was significantly higher than in local tobacco samples used in waterpipe smoking. Also, the average level of toluene in the WTWs and in local tobacco samples was highest among all BTEX compounds studied. In addition, we observed that after a period of two months, there was no significant difference between the concentration levels of BTEX compounds in water samples with and without addition of sodium azide, indicating no change due to possible biodegradation conditions. Because of the high prevalence of waterpipe tobacco use and the generation of large volumes of tobacco waste, this waste can potentially enter the aquatic environment and release high loads of BTEX compounds. Consequently, BTEX compounds may pose serious hazards to aquatic ecosystems and also threaten human health if they enter the food chain. Regulatory regimes should carefully consider options to require the safe disposal of WTWs or even to change waterpipe tobacco content to prevent the production of higher levels of BTEX compounds such as found in the flavored products. As with discarded cigarette butts, regulatory approaches for source reduction of post-consumption tobacco product waste are lacking. More public information is needed to secure such regulations, and more attention in general should be paid to the tobacco industry as a source of environmental contamination, no matter the type of tobacco product used.

## Ethics approval and consent to participate

“Not applicable”.

## Availability of data and materials

Additional data from the study are available by request to the corresponding author by email.

## CRedit authorship contribution statement

**Mohammad Reza Masjedi:** Data curation, Writing – original draft, Writing – review & editing. **Zahra Torkshavand:** Methodology, Software, Validation, Writing – original draft. **Hossein Arfaeinia:** Conceptualization, Supervision, Writing – original draft, Writing – review & editing. **Sina Dobaradaran:** Conceptualization, Supervision, Writing – original draft, Writing – review & editing. **Farshid Soleimani:** Data curation, Formal analysis, Methodology, Writing – original draft. **Akram Farhadi:** Funding acquisition, Writing – review & editing. **Roshana Rashidi:** Data curation, Formal analysis, Investigation. **Thomas E. Novotny:** Methodology, Writing – review & editing. **Sara Dadipoor:** Methodology, Software, Writing – review & editing. **Torsten C. Schmidt:** Methodology, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.heliyon.2023.e21946>.

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