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Continuous sample drop flow-based microextraction method as a microextraction technique for determination of organic compounds in water sample

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ABSTRACT

Continuous sample drop flow-based microextraction (CSDF-ME) is an improved version of continuous-flow microextraction (CFME) and a novel technique developed for extraction and preconcentration of benzene, toluene, ethyl benzene, m-xylene and o-xylene (BTEXs) from aqueous samples prior to gas chromatography–flame ionization detection (GC–FID). In this technique, a small amount (a few microliters) of organic solvent is transferred to the bottom of a conical bottom test tube and a few mL of aqueous solution is moved through the organic solvent at relatively slow flow rate. The aqueous solution transforms into fine droplets while passing through the organic solvent. After extraction, the enriched analyte in the extraction solvent is determined by GC–FID. The type of extraction solvent, its volume, needle diameter, and aqueous sample flow rate were investigated. The enrichment factor was 221–269 under optimum conditions and the recovery was 89–102%. The linear ranges and limits of detection for BTEXs were 2–500 and 1.4–3.1 μg L^{-1}, respectively. The relative standard deviations for 10 μg L^{-1} of BTEXs in water were 1.8–6.2% ($n=5$). The advantages of CSDF-ME are its low cost, relatively short sample preparation time, low solvent consumption, high recovery, and high enrichment factor.

1. Introduction

The sample preparation step in an analytical process typically consists of extraction to isolate and enrich the components of interest from a sample matrix [1]. Modern trends in analytical chemistry have moved toward the simplification and miniaturization of sample preparation and minimization of organic solvent and sample volumes [2]. Traditional solvent extraction has been applied to a variety of compounds; however, shortcomings such as the need for large amounts of hazardous organic solvents, large volume of samples, generation of large amounts of pollutants, and the lengthy process make it expensive, environmentally unfriendly, tedious, and labor intensive [3]. Simple, rapid, and environmentally friendly sample preparation methods are essential. Solid phase micro-extraction (SPME) and liquid phase micro-extraction (LPME) are new miniaturized sample preparation techniques that are simple, fast, and either solvent-free or require slight amounts of organic solvent [4]. SPME is a solvent-free process developed by Arthur and Pawliszyn in 1990 [5].

Different approaches have been developed for LPME since its inception [6], including single drop microextraction (SDME) [7], hollow fiber–liquid phase microextraction [8], dispersive liquid–liquid microextraction (DLLME) [1], and solidification of floating organic drop microextraction [9]. SDME [7,10,11] offers advantages such as low cost and low solvent consumption, and the possibility of carry-over between analyses is negligible. Disadvantages to SDME include it being time-consuming and significantly affected by stirring rate. SDME can be classified into several approaches. Jeannot and Cantwell [7] suggested a direct-immersion SDME method in 1997; a disadvantage of this method is the instability of the droplets at high stirring speeds. Static SDME provides a good reproducibility but results in limited enrichment with a long extraction time [12].

Liu and Lee [13] reported a novel liquid–liquid microextraction technique which was termed continuous-flow microextraction (CFME) to provide higher enrichment in a much shorter time. In this method, the extraction solvent drop is injected into a glass chamber using a conventional microsyringe and held at the outlet tip of a PTFE connection tube; the solvent drop interacts continuously with the sample solution and extraction proceeds simultaneously. CFME differs from other SDME approaches in that a drop of solvent fully and continuously makes contact with fresh, flowing sample solution [12]. Another advantage is the high

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preconcentration factor that requires smaller volumes of aqueous samples for extraction [12].

CFME also has disadvantages. The microdroplet requires support or a microsyringe and the droplet is instable. The volume of the microdroplet is limited to ~5 μL. Another disadvantage is the difficulty of removing the droplet from the solution. Water may also be transferred into the syringe and may result in problems in some instruments. This study presents a new version of the CFME method called continuous sample drop flow-based mcroextraction (CSDF-ME). Like CFME, a few microliters of organic solvent is used as the extraction solvent. In contrast to CFME, the fine droplets of sample solution pass through the organic solvent in CSDF-ME instead of experiencing the continuous contact of the flowing aqueous solution with the outer surface of the solvent droplet. In CSDF-ME, organic solvent that is water immiscible and has higher density than water is placed at the bottom of a conical bottom test tube and a continuous flow of aqueous sample solution is passed through the organic solvent using a peristaltic pump. The extraction solvent is very stable and easily transferred to analytical instruments by microsyringe after extraction. The performance of CSDF-ME method is tested by the determination of benzene, toluene, ethyl benzene, and xylenes (BTEXs) in the water samples using gas chromatography flame–ionization detection (GC–FID).

2. Experimental

2.1. Chemicals and reagents

Methanol, benzene, toluene, ethylbenzene, m-xylene and o-xylene were purchased from Merck (Germany). Stock solutions of BTEXs (1000 mg L^{-1}) were prepared by dissolving specific amounts of each in methanol. Stock solutions were stored at 4 °C in the refrigerator. Fresh working solutions (10 mg L^{-1}) were prepared daily by diluting the standard stock solutions with doubly distilled water to the required concentrations. Chloroform (suprasolvent for gas chromatography), carbon tetrachloride (GR), and carbon disulfide (GR) were purchased from Merck (Germany).

2.2. Apparatus

GC analysis was carried out on an Agilent 6890 N gas chromatograph with a flame ionization detector (Agilent Technologies, CA, USA). Helium (99.999%, Gulf Cryo, UAE) was used as the carrier gas at a constant linear velocity of 34 cm s^{-1} and make-up gas (45 mL min^{-1}). The injector temperature was constant at 150 °C. Injections were done in splitless/split mode with a split ratio of 1:10. Separation was carried out in a BP-5 30 m × 0.32 mm capillary column with a 0.25 μm stationary film thickness and 95% methyl–5% phenyl copolymer column (Agilent Technologies).

The oven temperature was programmed as follows: the initial temperature of 40 °C (was held for 3 min), increased at 20 °C min^{-1} to 80 °C and held there for 1 min, gain increased at 50 °C min^{-1} to 150 °C and held at 150 °C for 2 min. The total time for one GC run was 9.40 min. The FID temperature was maintained at 300 °C and hydrogen gas was generated using a hydrogen generator (CFH200, Peak Scientific) for FID at a flow of 40 mL min^{-1}. The flow of zero air (99.999, Air Products) for FID was 450 mL min^{-1}. The laboratory-made peristaltic pump had a flow rate of 0.45–3 mL min^{-1}.

2.3. CSDF-ME procedure

A 4.00 mL sample of doubly distilled water was spiked with BTEXs at a concentration of 10 μg L^{-1}. Fig. 1 shows the insertion of 20 μL of chloroform (extraction solvent) into the bottom of a small conical bottom test tube using 25 μL syringe. A narrow needle with a 0.3 mm external diameter connected to a narrow tube was placed in the chloroform. The spiked aqueous solution was then passed through the chloroform in the bottom of the conical bottom test tube through the narrow needle at a flow rate of 0.45 mL min^{-1} using a peristaltic pump. Since water density is lower than that of organic solvent, fine droplets of aqueous sample formed in the chloroform and rose up through the conical bottom test tube. During this step, BTEXs was extracted into the chloroform.

The 1.00 μL of remaining chloroform in the bottom of the conical bottom test tube was removed using a 1.00 μL microsyringe (zero dead volume, SGE) and injected into the GC. The volume of the remaining organic phase (chloroform) was determined using a 25 μL microsyringe to be 15 ± 0.2 μL.

2.4. Calculation of enrichment factor and extraction recovery

The enrichment factor (EF) is defined as the ratio of the remaining analyte concentration (C_{rem}) to the initial analyte concentration (C_0):

$$EF = \frac{C_{rem}}{C_0}$$

The C_{rem} was obtained from the calibration graph of the direct injection of BTEXs standard solution into the chloroform at 0.5–3 mg L^{-1}.

Extraction recovery (ER) is defined as the percentage of total analyte (n_0) extracted from the remaining phase (n_{rem}).

$$ER = \frac{n_{rem}}{n_0} \times 100 = \frac{C_{rem} \times V_{rem}}{C_0 \times V_{aq}} \times 100\%$$

$$ER = (V_{rem}/V_{aq})EF \times 100\%$$

where V_{rem} and V_{aq} are the volumes of the remaining phase and sample solution, respectively.

3. Results and discussion

Different factors in CSDF-ME can affect the microextraction method: the extraction solvent type and volume, the aqueous solution flow rate (extraction time), the diameter of the needle which determines the size of the aqueous droplets that form in the organic solvent, and volume of the aqueous solution. It is very important to optimize these factors to obtain good ERs, high EFs, and short processing times.

3.1. Selection of extraction solvent

The organic solvent should be immiscible in water, be capable of extracting the required compounds, have a density that is higher than water, and show good GC behavior. The solvents used and compared in the extraction of BTEXs were carbon tetrachloride, carbon disulfide, and chloroform.

To produce the same volume at the end of extraction (15 ± 0.2 μL), 20 μL of chloroform, 23 μL of carbon tetrachloride and 26 μL of carbon disulfide were used to extract BTEXs from the aqueous solution. Extraction conditions were fixed for all three solvents: 4 mL of aqueous solution, a needle with a 0.3 mm external diameter, and 0.45 mL min^{-1} aqueous solution flow rate. The average recovery (n=3) for all three solvents is shown in Table 1. The extraction recoveries for the solvents were similar. Chloroform was chosen as the best extraction solvent because when carbon tetrachloride was the extraction solvent, the peak for benzene occurred in the solvent peak region and poor repeatability was observed when carbon disulfide was used.

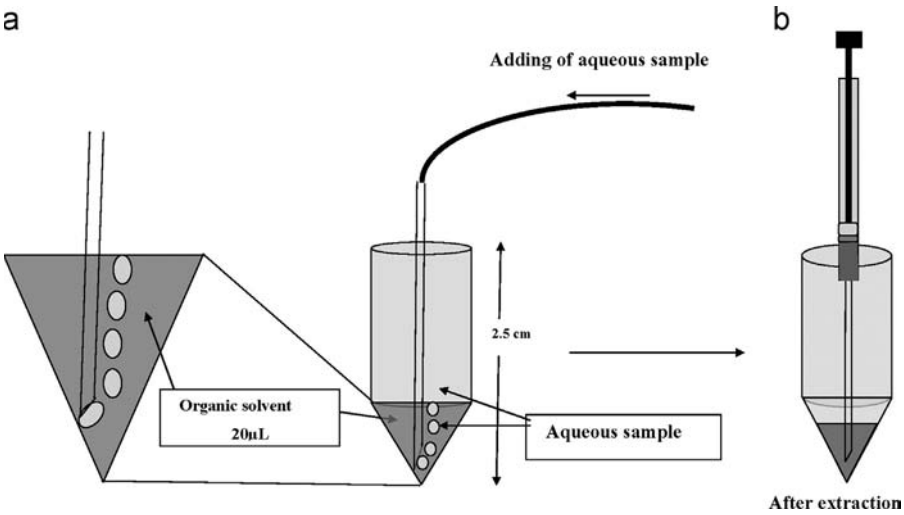

Fig. 1. CSDF-ME procedure: (a) introduction of aqueous sample into extraction solvent (chloroform), (b) removal of a portion of remaining organic phase (15 ± 0.2 μL) by a 1 μL syringe in order to inject into GC–FID.

Table 1
Extraction efficiency of various extraction solvents[a].

Compounds	Recovery (%)		
	Carbon tetrachloride, mean ± SD (n=3)	Carbon disulfide, mean ± SD (n=3)	Chloroform, mean ± SD (n=3)
Benzene	–	100.5 ± 1.9	89.4 ± 1.2
Toluene	104.6 ± 1.4	95.4 ± 2.4	102.3 ± 1.7
Ethylbenzene	84.1 ± 1.6	97.4 ± 2.1	89.6 ± 2.0
m-Xylene	80.4 ± 1.0	88.8 ± 2.6	99.4 ± 1.6
o-Xylene	94.6 ± 1.7	86.5 ± 3.3	91.2 ± 1.9

[a] Extraction conditions: aqueous sample volume, 4 mL; analytes concentrations, 10 μg L^{-1} of each BTEXs; extraction solvent volume (chloroform), 20 μL; aqueous sample flow rate, 0.45 mL min^{-1}; needle external diameter, 0.3 mm.

3.2. Effect of extraction solvent volume

For liquid phase microextraction, the volume of the extraction solvent must be controlled to control its effect on the analytical signals. Commonly, the volume of extraction solvent is kept small so that it can achieve the highest possible EF and least toxicity to the environment. On the other hand, the maximum possible amount should be used to extract the maximum analyte [4] and ensure that the volume of solvent extraction is sufficient for chromatographic analysis.

To study the effect of the volume of extraction solvent, testing was done using different volumes (20, 30, 40, 50 and 60 μL) of chloroform and the same extraction procedure. Fig. 2 shows that increasing the volume of chloroform from 20 to 60 μL resulted in a fixed extraction recovery for most compounds, which indicates the quantitative extraction of BTEXs. Increasing the volume of chloroform increased the volume of the remaining organic solvent at the end of microextraction. Increasing the volume of extraction solvent decreased the enrichment factor because the volume of remaining organic solvent increased (Fig. 3). High EF and good ER were obtained by decreasing the volume of extraction solvent. The highest sensitivity was achieved using 20 μL of chloroform.

3.3. Effect of needle diameter

The size of the aqueous droplets forming in the organic solvent depends upon the diameter of the needle. It plays an important role in the rate of achieving equilibrium state and mass transfer.

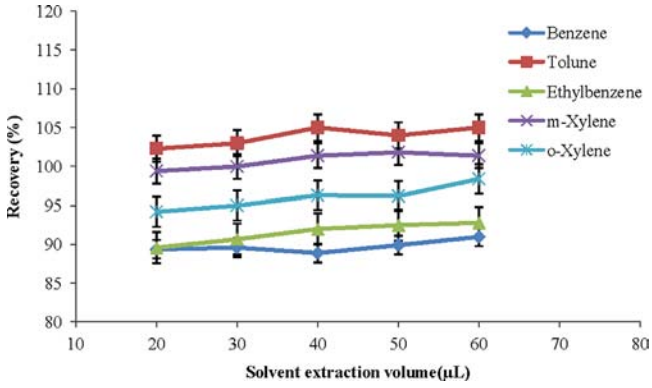

Fig. 2. Effect of the volume of chloroform on the recovery of BTEXs obtained from CSDF-ME. Extraction conditions: aqueous sample volume, 4.00 mL; extraction solvent (chloroform) volume, 20.0 μL; analytes concentrations, 10 μg L^{-1} of each BTEXs; needle external diameter, 0.3 mm.

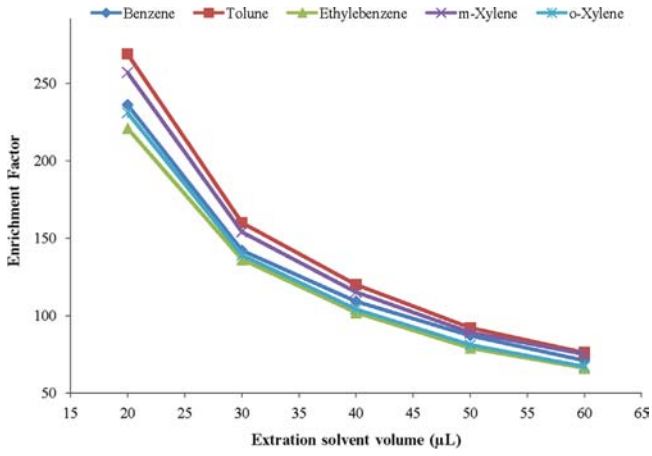

Fig. 3. Effect of the volume of chloroform on the enrichment factor of BTEXs obtained from CSDF-ME. Extraction conditions, as with Fig. 2; aqueous sample flow rate, 0.45 mL min^{-1}.

Decreasing the needle diameter decreased the size of the aqueous droplets, which resulted in higher surface contact between the two phases' higher efficiency for mass transfer. Droplets from a thinner needle had a higher surface area–volume ratio compared to those from a thicker needle resulting in higher extraction efficiency and shorter extraction time. The effect of needle

diameter on recovery and EF was tested for two external needle diameters (0.3 and 1.3 mm) under constant experimental conditions. The results showed that the highest sensitivity was achieved using a needle with an external diameter of 0.3 mm.

The volume of organic solvent (chloroform) in this test was 30 μL for both needles. A volume of 20 μL of chloroform was not used because the thicker needle created large aqueous droplets.

3.4. Effect of aqueous sample flow rate

Increasing flow rate of the aqueous sample increased the rate of forming the aqueous droplets in the organic solvent in the present study. The effect of flow rate of the aqueous sample was studied at 0.45 to 1.4 mL min^{-1}. It was found that increasing the flow rate of the sample decreased the contact time between aqueous droplets and the organic solvent. When the flow rate increased, the ER decreased because mass transfer decreased (Fig. 4). A flow rate of 0.45 mL min^{-1} was selected to provide high ER and EF. The peristaltic pump was not capable of generating flow rates of less than 0.45 mL min^{-1}, but since ER at this flow rate was nearly 100%, flow rates of less than 0.45 mL min^{-1} were not considered for optimization.

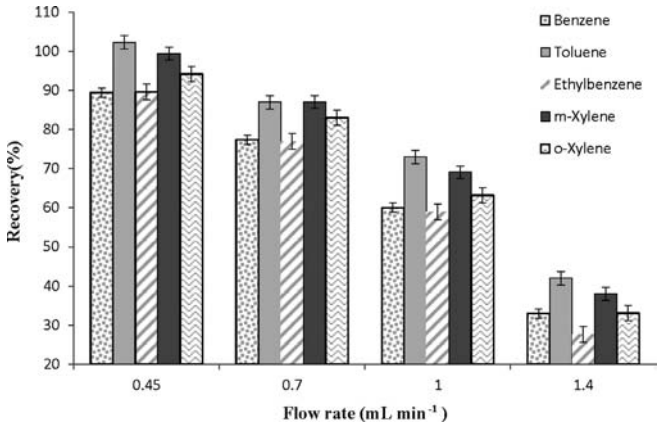

Fig. 4. Effect of the aqueous sample flow rate on the recovery of BTEXs obtained from CSDF-ME. Extraction conditions: aqueous sample volume, 4.00 mL; extraction solvent (chloroform) volume, 20.0 μL; analytes concentrations, 10 μg L^{-1} of each BTEXs.

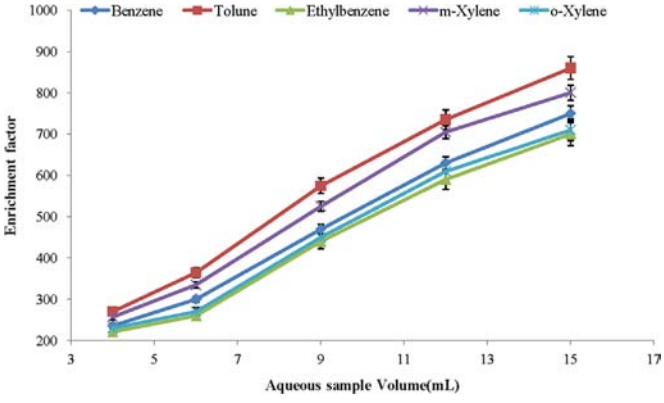

Fig. 5. Effect of the aqueous sample volume on the enrichment factor of BTEXs obtained from CSDF-ME. Extraction conditions: extraction solvent (chloroform) volume, 20.0 μL; analytes concentrations, 10 μg L^{-1} of each BTEXs; needle external diameter, 0.3 mm; aqueous sample flow rate, 0.45 mL min^{-1}.

3.5. Effect of volume of aqueous sample

Increasing the volume of the aqueous sample increased the analytical signals. To investigate the effect of volume of the aqueous sample, 4–15 mL of aqueous sample were injected into 20 μL of extraction solvent under constant experimental conditions. The results are shown in Fig. 5. Increasing the volume of the aqueous sample increased EF; however, the extraction time (time required to move the aqueous sample through the extraction solvent at a flow rate of 0.45 mL min^{-1}) also increased from 8.9 to 33 min. As a result, a volume of 4 mL was selected to avoid lengthening the extraction time.

Table 2
Quantitative result of CSDF-ME and GC–FID of BTEXs from aqueous sample[a].

Analytes	LOD[b] (μg L^{-1})	EF[c]	RSD %[d], $n=5$	LR[e] (μg L^{-1})	r^{2}[f]
Benzene	2.4	236	1.8	10–500	0.999
Toluene	1.4	269	2.9	2–500	0.999
Ethyl benzene	3.1	221	6.2	15–500	0.999
m-Xylene	1.9	257	3.4	5–500	0.999
o-Xylene	2.0	231	4.5	5–500	0.999

[a] Extraction conditions: aqueous sample volume, 4.00 mL; extraction solvent (chloroform) volume, 20.0 μL; analytes concentrations, 10 μg L^{-1} of each BTEXs; aqueous sample flow rate, 0.45 mL min^{-1}; needle external diameter, 0.3 mm.
[b] LOD, limit of detection for a S/N=3.
[c] EF, enrichment factor.
[d] RSD % without using internal standard at a concentration of 10 μg L^{-1} of each BTEXs.
[e] LR, linear range.
[f] r^2 without using internal standard.

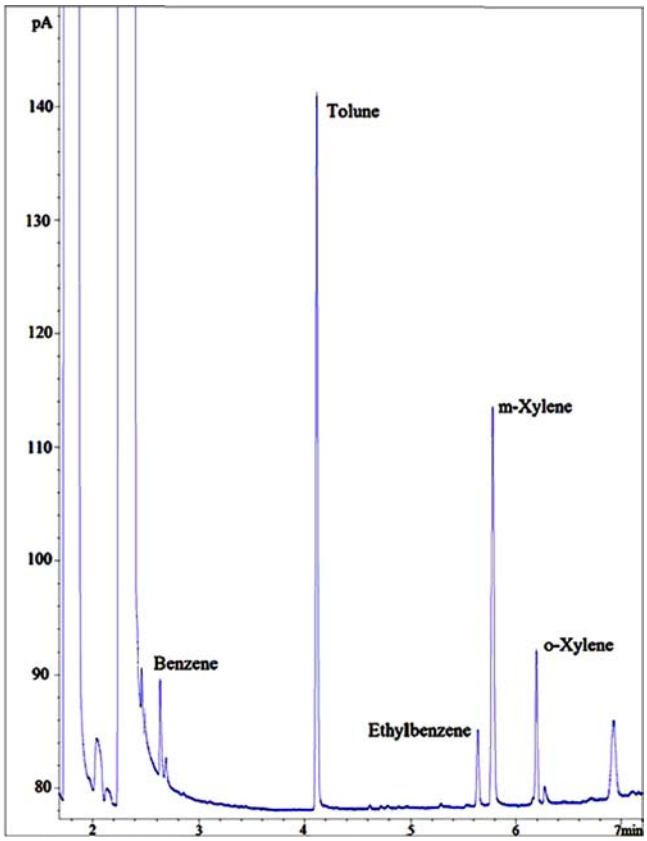

Fig. 6. Chromatogram of river water spiked at concentration level of 10 μg L^{-1} of BTEXs obtained by using CSDF-ME combined GC–FID.

Table3
Recovery values obtained for the determination of BTEXs in river water sample[a].

Compounds	River water sample concentration ($\mu g\ L^{-1}$)	Added concentration ($\mu g\ L^{-1}$)	Found concentration (SD, $n=3$)[b] ($\mu g\ L^{-1}$)	Relative recovery (%)
Benzene	nd[c]	10	8.9 ± 0.2	89.0 ± 1.2
Toluene	Nd	10	10.2 ± 0.3	102.0 ± 1.7
Ethylbenzene	Nd	10	8.9 ± 0.4	89.0 ± 2.0
m-Xylene	Nd	10	9.9 ± 0.2	99.0 ± 1.6
o-Xylene	Nd	10	9.1 ± 0.3	91.0 ± 1.9

[a] Extraction conditions: aqueous sample volume, 4.00 mL; extraction solvent (chloroform) volume, 20.0 μL; analytes concentrations, 10 $\mu g\ L^{-1}$ of each BTEXs; aqueous sample flow rate, 0.45 mL min^{-1}; needle external diameter, 0.3 mm.
[b] SD, standard deviation.
[c] nd, not detected.

Table 4
Comparison of the CSDF-ME–GC–FID method with other related methods for determination of BTEX.

Analytes	Methods	LOD ($\mu g\ L^{-1}$)	RSD %	Time (min)	Reference
BTEXs	HF–LPME–GC–FID	2.2–4	1.3–5.1	30	[14]
BTEXs	HS–LPME–GC–FID	3.2–12.9	2.5–9.3	20	[15]
BTEXs	HS–SPME–GC–FID	100–800	3.5–10	30	[16]
BTEXs	DSDME–GC–FID	0.8–7	1.8–2.4	25	[17]
BTEXs	DI–SDME–GC–FID	5–10	4.7–8.7	25	[18]
PAHs	CFME–GC–MS	0.001–0.01	6.7–25	15	[19]
Me- and Et-chlorides	CFME–GC–μECD	0.01–0.02	< 4.3	10	[20]
Phenols	CFME–GC–FID	2.3–3	< 6.9	15	[21]
BTEX	DLLME–GC–FID	0.1–0.2	0.9–6.4	5	[22]
BTEX	CSDF–ME–GC–FID	1.4–3.1	1.8–6.2	9	This work

3.6. Quantitative analysis

The analytical characteristics of CSDF-ME were evaluated for the BTEXs. The estimated figures of merits under optimized conditions are shown in Table 2. The limits of detection (LOD), based on a signal-to-noise ratio (S/N) of 3 were 1.4–3.1 $\mu g\ L^{-1}$. The reproducibility of the peak responses was tested using 5 replicate experiments under optimized conditions. The relative standard deviation (RSD %) of the BTEXs (10 $\mu g\ L^{-1}$) was 1.8–6.2% without internal standards. The coefficient of correlation (R^2) was 0.999 without internal standards. The enrichment factors of the BTEXs were 221–269. The linearity of calibration curve was 2–500 $\mu g\ L^{-1}$.

3.7. Analysis of real sample

River water was collected from the city of Mahabad in Iran, extracted using the CSDF-ME method and analyzed by GC–FID. The results showed that it was free of BTEXs contamination. The river water was then spiked with BTEXs at a concentration of 10.0 $\mu g\ L^{-1}$ to assess matrix effects. Fig. 6 shows the chromatogram obtained for river water spiked with BTEXs. The relative recovery from the river water was 89–102% for all BTEXs samples (Table 3).

3.8. Comparison of CSDF-ME and other related methods

The efficiency of the proposed CSDF-ME for the selected analytes was compared with those of previously reported methods for relative standard deviation, extraction time, and limit of detection. Table 4 shows that CSDF-ME produced RSDs that were better than or comparable to those of other extraction methods. As seen, the extraction time of CSDF-ME is almost shorter than for all other methods because of the large surface area of contact between the extraction solvent and the sample solution and the elimination of the requirement for stirring. This method solved the main problems encountered with CFME of the microdroplet being lost from its support and the smaller volume of extracting solvent not matching the requirements of the instrumentation for a higher injection volume.

The proposed method has several advantages over conventional DLLME. In contrast to conventional DLLME, no centrifuge is required for the collection of extraction solvent. There is no need for dispenser solvent, so less organic solvent is used and the on-line automation of this method makes it simpler than DLLME. In comparison with on-line sequential injection liquid–liquid microextraction, it appears that the proposed method is simpler and requires less equipment.

4. Conclusion

A novel approach for liquid phase microextraction (LPME) is proposed which requires no support for microdroplets and no disperser solvent. The proposed CSDF-ME is a high-performance preconcentration method with a short extraction time because of the high surface area of contact between the sample solution and microextraction solvent. The other advantages of CSDF-ME are its simplicity of operation, low cost, high recovery, and high enrichment factor.

The limitations of the CSDF-ME method are that it usually requires the use of organic solvents that are heavier than water. It is hoped that CSDF-ME can be adapted in future studies to use solvents that are lighter than water. CSDF-ME can also be applied for determination of other compounds that can be extracted using organic solvents.

Acknowledgments

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