Heliyon 10 (2024) e27611

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Magnetic stirring in syringe dispersive liquid-liquid microextraction as an effective method for preconcentration of tartrazine dye from food samples: A multivariate analysis approach

Ali Mohammadzadeh [a,c,*], Mahboobeh Amiri [b,d]

[a] Department of Chemistry, Faculty of Science, Islamic Azad University-Arak Branch, Arak, Iran
[b] Department of Microbiology, Faculty of Science, Shiraz Branch, Islamic Azad University, Shiraz, Iran
[c] Young Researchers and Elite Club, Arak Branch, Islamic Azad University, Arak, Iran
[d] Department of Chemistry, Faculty of Science, Shiraz Branch, Islamic Azad University, Shiraz, Iran

ARTICLE INFO

Keywords:
Magnetic stirring in syringe
Tartrazine
Central composite design
Food samples
UV–Vis spectrophotometer

ABSTRACT

A new, rapid, simple, and sensitive preconcentration method and Spectrophotometry determination technique have been presented for the microextraction and determination of trace amount of Tartrazine dye in food samples. In the present system, which is called "Magnetic stirring in syring dispersive liquid-liquid microextraction"(MSIS- DLLME), a cloudy state result is formed in a homemade glass syringe by magnetic agitation. In the MSIS-DLLME system, Tartrazine colour was uprooted into an organic detergent (Toluene) after many twinkles. Subsequently, the organic detergent which was placed on top of the result was transferred into a narrow neck by moving the piston overhead. The effective parameters on the extraction recovery were studied and optimized by Central Composite design (CCD). Under the optimum conditions, the estimation cure is direct in the range of 0.1–1(μg L^{-1}). The limit of detection (LOD), relative standard divagation and enrichment factor were 0.03 μg L^{-1}, ±4.6 (n = 10) and 166, independently. The advanced system was successfully applied for microextraction of Tartrazine in food samples.

1. Introduction

Tartrazine (Tar) is a synthesis dye (Fig. 1) that is substantially used as coloured cumulative in drinks, fruit authorities, ice cream, sweets and goodies [1]. It can be used with colourful synthesized dyes in order to food colouring. The inquiries have shown Asthma, hyperactivity in children and the migraine effect of Tartrazine in mortal [2,3]. In the last decade, the use of food complements, especially coloured artificial colourings gradationally increased in the most commonly used food similar as maquillages, sweets and ice cream. The determination and control of these composites in food are veritably important due to their poisonous goods [4,5]. It is delicate to direct the determination of the trace quantum of Tartrazine. Thus, the preconcentration way is avoidable prior to the necessary analysis. There are various methods for the determination of Tar such as: Ratio spectra first-order derivative UV spectrophotometric [2], Voltammetric system [6], sensors [7], solid phase spectrophotometric determination [3], liquid chromatography

* Corresponding author.
E-mail addresses: a.mohammadzadeh2023@yahoo.com (A. Mohammadzadeh), atefehranjbar68@gmail.com, amirimahsa508@gmail.com (M. Amiri).

https://doi.org/10.1016/j.heliyon.2024.e27611
Received 14 August 2023; Received in revised form 2 March 2024; Accepted 4 March 2024
Available online 5 March 2024

[8–15] photometric discovery and LC-MS [16]. These methods have some advantages, but they have some disadvantages including time-consuming, complex systems and numerous fine computations. The mentioned methods have been significantly replaced by solvent-miniaturized microextraction ways [17]. Dispersive liquid–liquid microextraction [DLLME] was developed in 2006 by Assadi and associates [18]. In traditional DLLME, an admixture of polar dispersive detergent and an organic detergent is used. After fast injection of this mixture into an aqueous solution, a cloudy state is formed. DLLME technique has been widely used in analysis of organic and inorganic species [19]. In the present study, we have used the DLLME system on Magnetic stirring in syringe system for preconcentration and determination of trace quantum of Tartrazine in food samples. This system has good advantages similar to being simple, fast, sensitive and there is no need for centrifugation. Effective variables which perhaps affect the extraction recovery were optimized by the central compound design (CCD).

2. Experimental

2.1. Instrumentation

A double beam UV–Vis spectrophotometer (PerkinElmer Lambda 25, Germany) with a quartz cell and an optical path of 1 cm was used for measure of absorbance (in $\lambda_{max} = 426$ nm). A pH meter model Metrohm Lab-827 was used to solution pH adjustment.

2.2. Reagents

Ultra-pure water (18.2 MΩ) was used to prepare all of the solutions. After being left in a 5 percent (v/v) nitric acid solution for the entire night, the glassware was cleaned with deionized water. Every reagent that is graded as analytical reagent. Acetate buffer solutions (pH 4.0–6.0), phosphate buffer solutions (pH 2.0–3.0, 7.0–9.0), and ammonia buffer solutions (pH, 10.0–12.0) were used to adjust the pH of sample solutions. As extraction solvents for the suggested microextraction method, toluene, 1-octanol, 1-decanol, 1-dodecanol, 1-undecanol, hexane, and heptane were purchased (Merck Darmstadt, Germany).

2.3. Sample preparation

The amount of tartrazine in a variety of food samples, including saffron powder, fruit, pineapple powder, banana dessert, and orange syrup powder, was examined in order to validate the suggested method. The samples were taken in January 2023 from the market in Boushehr, Iran. Each sample weighed two points zero grams, which were dissolved in 2 mL of HCl (2 mol L-1) and sonicated until completely dissolved. Following standard protocol, the generated solution was diluted in a 50 mL conical tube and handled.

2.4. General procedure

Considering Fig. 2a: Phosphate buffer was used to bring the 50 mL sample solution containing sodium chloride (10 percent w/v) and tartrazine (300 μg L^{-1}) to pH = 3. Subsequently, a glass syringe (8.5 cm × 2.09 mm) was filled with the prepared solution. D. The syringe's end was sealed with a retractable septum (Fig. 2. (a). Then, using a Hamilton gas-tight syringe (fig.), 300 point0 μL of toluene was injected into the sample solution through the top of the glass syringe while the solution was being magnetically stirred by a magnet (4 mm*25 mm). 2. b). A cloudy solution resulted from the dispersion of numerous tiny toluene droplets after the solution was vigorously agitated (1200 rpm) (Fig. 2. C). Ten minutes later, the organic phase was gathered at the solution's surface (Fig. 2. (d). the septum was moved to the top of the syringe in the narrow section of the tube to raise it (Fig. 2 f). Subsequently, 200 μL of extraction solvent were manually extracted using a microsyringe and then transferred into the UV–Vis microcell [19–22]. Lastly, the final solution was added to the UV-ViS spectrophotometer for additional examination.

2.5. Experimental design strategy

Four factors were found to have the potential to impact Tartrazine extraction recovery in this study following multiple experiments. The technique of experimental design was employed to optimize these factors. Central Composite Design (CCD) looked into the factors that were effective in extraction recovery. Based on the following benefits, CCD is used widely:

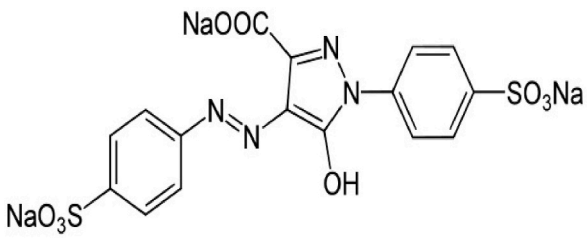

Fig. 1. Structure the tartrazine.

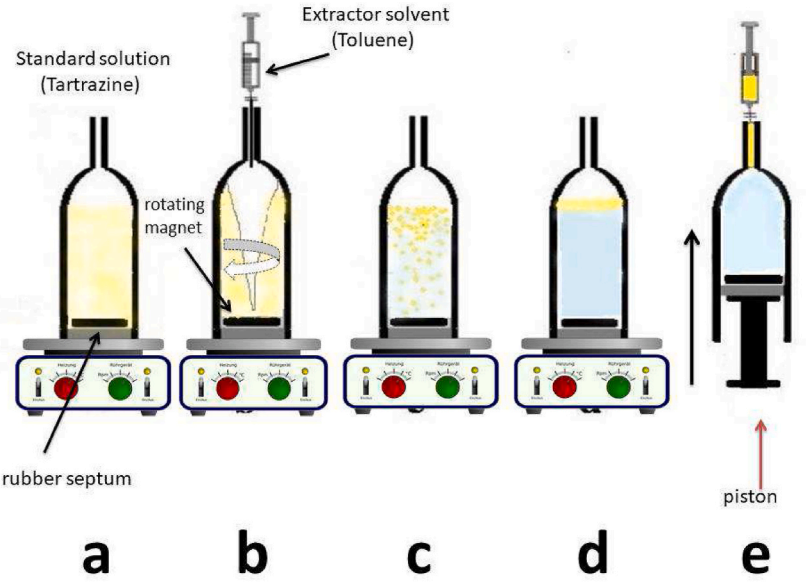

Fig. 2. General process of MSIS-DLLME.

Finding effective factors in the experiment.

1. Studying the factors influencing on responses.
2. Investigating the interactions among the factors.

Table 1
Design matrix by CCD.

Factor		Low	Levels Central	High	Star point α = 2 -α +α
(X_1) pH		3	5	7	1 9
(X_2) Solvent volume (μL)		150	200	250	100 300
(X_3) Stirrer rate (rpm)		600	800	1000	400 1200
(X_4) Extraction time (min)		4	6	8	2 10
Runs	X_1	X_2	X_3	X_4	Absorbance
1	1	200	800	6	0.781
2	3	150	600	4	0.182
3	3	150	600	8	0.323
4	3	150	1000	4	0.413
5	3	150	1000	8	0.590
6	3	250	600	4	0.291
7	3	250	600	8	0.456
8	3	250	1000	4	0.590
9	3	250	1000	8	0.777
10	5	100	800	6	0.192
11	5	200	400	6	0.133
12	5	200	800	2	0.134
13	5	200	800	6	0.245
14	5	200	800	6	0.245
15	5	200	800	6	0.245
16	5	200	800	10	0.327
17	5	200	1200	6	0.423
18	5	300	800	6	0.332
19	7	150	600	4	0.108
20	7	150	600	8	0.156
21	7	150	1000	4	0.184
22	7	150	1000	8	0.215
23	7	250	600	4	0.143
24	7	250	600	8	0.179
25	7	250	1000	4	0.214
26	7	250	1000	8	0.258
27	9	200	800	6	0.167

3. Obtaining the optimum conditions for further studies.

The total number of experimental runs in the CCD is equal to Eq. (1)

$$N = 2^K + 2k + C \quad \text{Eq. (1)}$$

where N, C, and K stand for the number of central points, the number of factors, and the number of experimental runs, respectively. Five levels and a triple central point were used for CCD. Taking into account Eq. (1) there were 27 runs of the experiment (Table 1). In the CCD, trials were carried out at random and each run was duplicated three times in order to reduce the systematic error. The STATISTICA (Version 10.0) program was used to perform all of the statistics.

In this study, four factors were chosen and analyzed by CCD on five levels and at a triplicate central point (Table 1). By considering equation (1), the experimental runs were 27 runs (Table 1). In the CCD, for minimizing the systematic error, experiments were conducted randomly and all runs were replicated three times. According to Table 1, these factors include: the pH of solution (X1), the solvent volume (μL, X2), the stirrer rate (rpm, X3) and the extraction time (min, X4). All Statistics were carried out with Statistica (Version 10.0) software.

3. Results and discussion

3.1. Effect of extraction solvent

Select of a Suitable extraction solvent is a critical step for pre-concentration of Tartrazine by MSIS-DLLME method. For this purpose, several organic solvent (low density than water) were investigated. Accordingly, toluene was the highest extraction recovery (Fig. 3.).

3.2. Salt effect

The effect of ionic strength was investigated in the extraction recovery of Tartrazine. Accordingly, some salts were examined such as NaCl, $NaNO_3$ and $MgSO_4$. As a result, the ionic strength has no significant effect on the extraction recovery of Tartrazine.

3.3. Central composite design (CCD)

The analysis of variance (ANOVA) was shows in Table 2. The p-value is an important parameter in ANOVA table (the significant criteria is $p < 0.05$). Based on the p-value, all factors have significant effect on the extraction recovery of Tartrazine. By referring to the coefficients of R^2 (99.4%) and adjusted R^2 (98.7%), could be concluded that this model has a good relationship between responses and the fitted model. Also, Fig. 4 shows the good fitness of predicted model with experimental responses.

Regression analysis of CCD was carried out and thereby the following equation (Equation No. 2) was obtained which is related to the absorbed Tartrazine.

$$\begin{aligned}\text{Absorbance of Tartrazine}: & -0.3304 - 0.0048X_1 0.0006X_2 + 0.0004X_3 + 0.0620X_4 + 0.0145X_1^2 + 0.000002X_2^2 + 0.00000X_3^2 \\ & - 0.0006X_4^2 - 0.00029X_1X_2 - 0.00013X_1X_3 - 0.0079X_1X_4 + 0.000001X_2X_3 + 0.000022X_2X_4 \\ & + 0.000008X_3X_4 \quad \text{(Eq.2)}\end{aligned}$$

Fig. 5a- c shows the response face plots between the paired factors in CCD. The response face plots are given by brace of significant

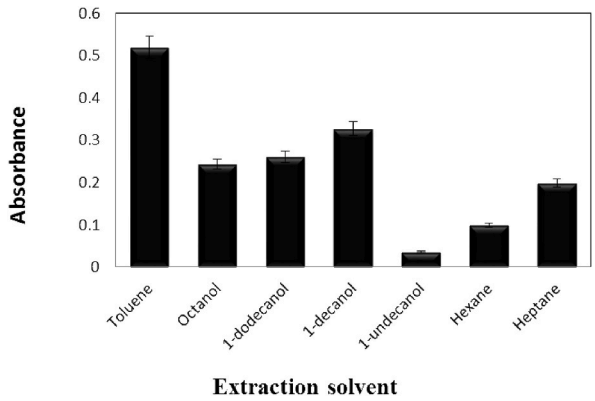

Fig. 3. The Effect of extraction solvent on extraction recovery of Tartrazine
Experimental condition: pH: 3, Solvent volume (μL): 300, Stirring time (min):10 and stirring rate (rpm): 1200.

Table 2
Analysis of variance (ANOVA) for CCD.

Source	Df[a]	SS[b]	MS[c]	F-value[d]	p-value
X_1	1	0.4793	0.4792	1103.928	0.0000
X_2	1	0.0726	0.0726	167.213	0.0000
X_3	1	0.0431	0.0431	99.269	0.0000
X_4	1	0.0006	0.0006	1.431	0.2545
X_1^2	1	0.1635	0.1635	376.629	0.0000
X_2^2	1	0.0018	0.0018	4.367	0.0585
X_3^2	1	0.0615	0.0615	141.704	0.0000
X_4^2	1	0.0001	0.0001	0.292	0.5982
X_1X_2	1	0.0142	0.0142	32.712	0.0000
X_1X_3	1	0.0434	0.0434	99.988	0.0000
X_1X_4	1	0.0163	0.0163	37.609	0.0000
X_2X_3	1	0.0011	0.0011	2.754	0.1228
X_2X_4	1	0.0000	0.0000	0.177	0.6806
X_3X_4	1	0.0001	0.0001	0.348	0.5659
Error	12	0.0052	0.0004		
Total SS	26	0.9142			

[a] **DF**: Degrees of freedom; **SS**[b]: Sum of Square; **MS**.
[c] Mean of Square.
[d] Test for comparing variance of model with variance of residual (error).

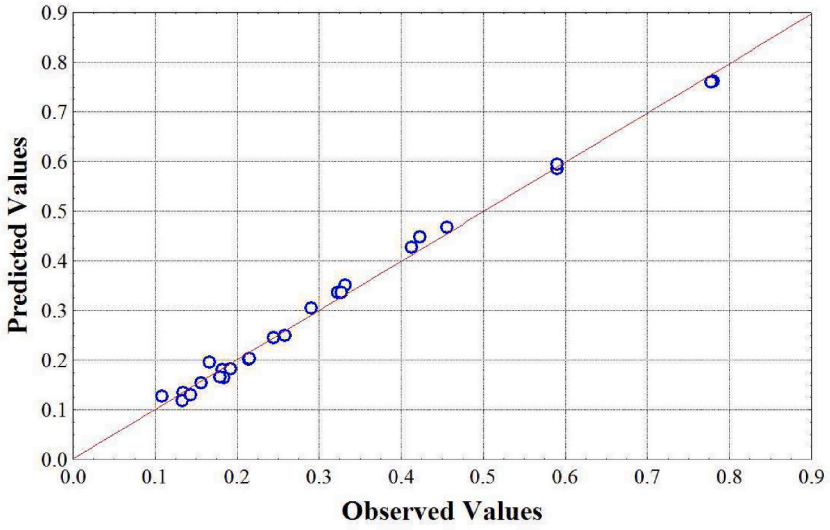

Fig. 4. Plot of predicted value vs. observed values.

factors at the fixed values of other factors. Each factor indicates its effect on the extraction recovery by negative or positive measures. Fig. 5a presents the mated effect of solvent volume (μL) vs. pH of the result. As can be seen, the extraction recovery was increased with an increase in solvent volume and a drop in pH of the result. It was illustrated that an addition in solvent volume could increase the birth recovery. Also, the effect of pH shows that Tartrazine is protonated in an acidic medium and increases the extraction of Tartrazine into the organic phase. Fig. 5b reveals that the increase in the volume of solvent and stirring rate raises the extraction recovery. On the other hand, the commerce between the analyte and extraction detergent was enhanced. As can be seen in Fig. 5c, the increased stirring rate and birth time enhanced the extraction recovery. As a brief result, the extraction recovery of Tartrazine was increased by adding the solvent volume (μL), extraction time (min), stirring rate (rpm) and dropping the pH of the result.

3.4. Optimization of CCD by desirability function (DF)

To optimize extraction recovery, the profile of prognosticated values and advisability option was used. The advisability profile of responses provides the DF value for each factor on the extraction recovery (Fig. 6). The scale of 0.0 (undesirable) to 1.0 (veritably desirable) was used to achieve a global function (D). According to Table 1, the maximum and minimal response for Tartrazine was 0.780 and 0.108, independently. Thus, grounded on Fig. 6, advisability of 1.0, 0.5 and 0.0 were assigned for 0.780, 0.444 and 0.108. also, the advisability value of 1.0 (maximum extraction recovery (0.780) was achieved at optimum conditions Solvent volume of 300 (μL), extraction time of 10 (min), stirring rate = 1200 (rpm) and pH = 3.

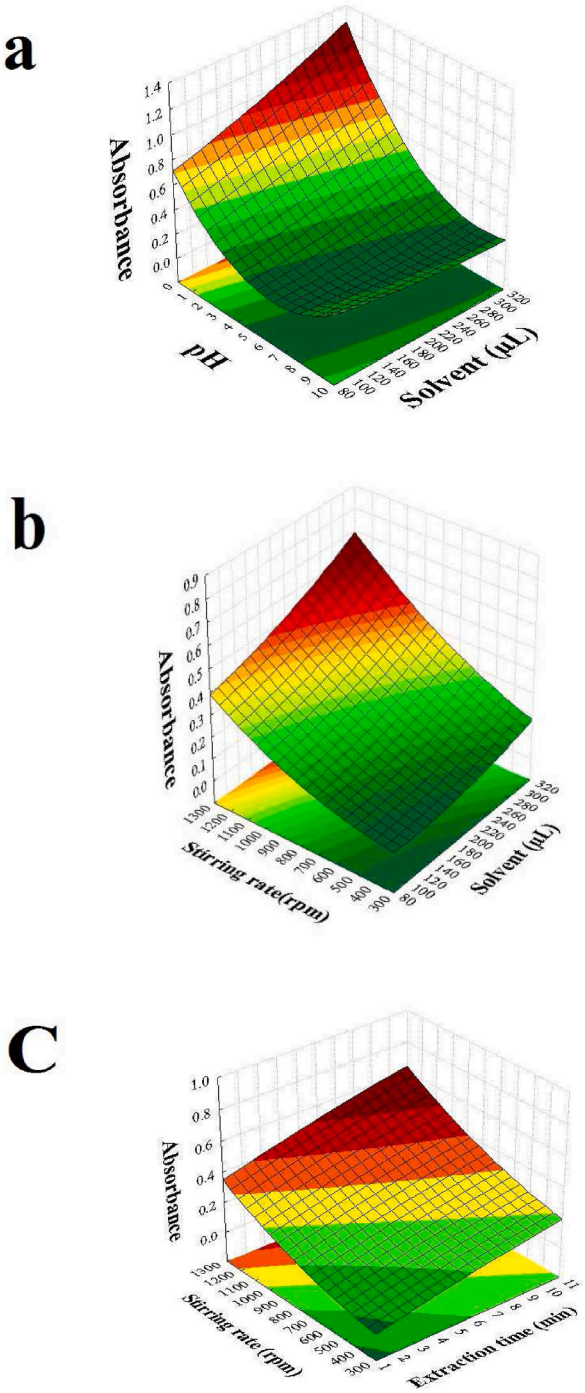

Fig. 5. Response surface plots: (a) pH vs. Solvent volume (μL), (b) stirring rate (rpm) vs. Solvent volume (μL), (c) stirring rate (rpm) vs. Extraction time (min).

3.5. Analytical figures of merits

To evaluate the proposed method, the analytical characteristics were obtained under the optimum conditions. The relative standard deviation (RSD) and dynamic linear range were 4.6 (n = 10) and 0.1–1 μg L^{-1}, respectively. The detection limit (LOD), calculated as 3 S_b/m, where S_b is the standard deviation of the blank and m is the slope of the calibration curve, was 0.03 μg L^{-1}. Analytical characteristics of proposed method are listed in Table 3.

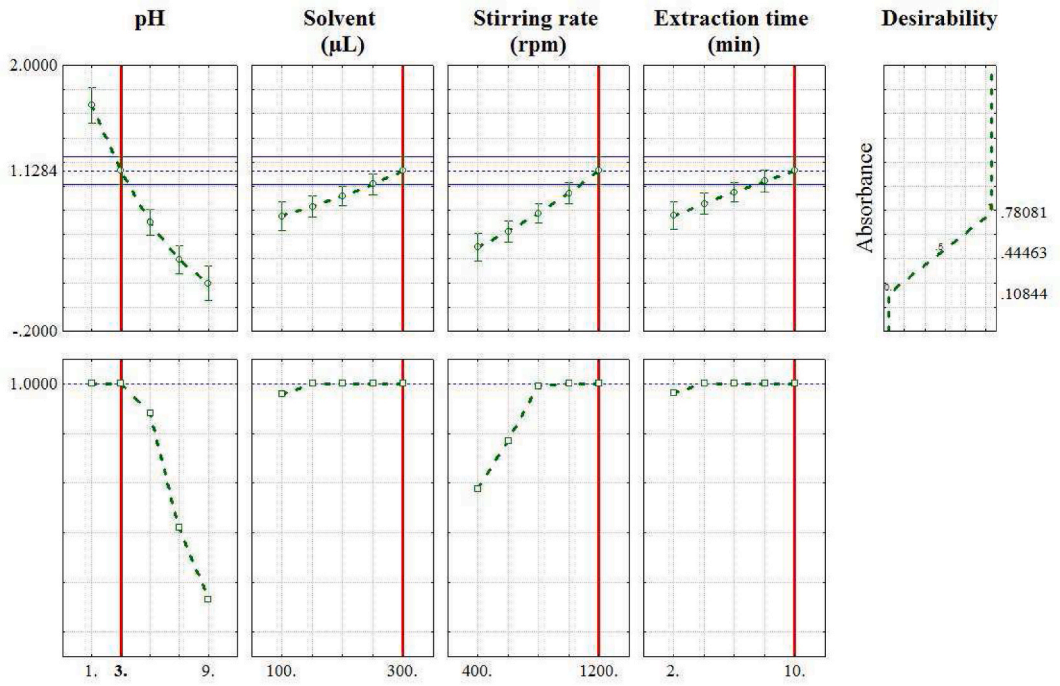

Fig. 6. Desirability function (DF) for CCD.

3.6. Comparison of MSIS-DLLME with other methods

Table 4 compares the characteristic data of the suggested method with other with those of some of the best previously methods for determination of Tartrazine which have been reported in the literature.

3.7. Analysis of real samples

In order to evaluation the proposed method, the determination of Tartrazine were carried out in several food samples (Table 5). The accuracy of the method was assessed by the analysis of the samples spiked with the known amount of Tartrazine. Based on results, the recoveries for the addition of different concentrations of Tartrazine were in the range of 97–101%. it could be comcluded that the matrices of samples has no significant effect on the extraction recovery of Tartrazine by MSIS-DLLME method.

4. Conclusion

In the present study, the magnetic stirring in syringe dispersive liquid-liquid microextraction system was successfully used as a microextraction system for the preconcentration of Tartrazine. The central compound design was successfully used to optimize the affective parameters in birth effectiveness. In This methodology, it is possible to observe the commerce and the main effect of factors on the extraction process. Under the optimum conditions, the system has good logical characteristics. The mentioned advantages and good logical characteristics make this system to be successfully applied to the determination of Tartrazine in food samples. Eventually, Limitations of exploration due to methodological problems can be addressed. For illustration, while this system was primarily concentrated on the birth of poisonous essence, we delved into how this system could apply to the birth and determination of Tartrazine dye. So, we are sure that further parameters should be delved into in order to assess the proposed system.

CRediT authorship contribution statement

Ali Mohammadzadeh: Writing – review & editing, Software, Project administration, Methodology, Investigation, Formal analysis, Data curation. **Atefeh Ranjbar:** Writing – original draft, Software, Investigation, Conceptualization.

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.

Table 3
Analytical characteristic for determination of Tartrazine.

parameter	Value
Enrichment factotor	166
Correlation coefficient (r^2)	0.999
Linear range (μg L^{-1})	0.1–1
Sample volume (mL)	50
R.S.D (%)(n = 10)	4.6
LOD (μg L^{-1})	0.03

Table 4
Comparison of proposed method with other methods for determination of Tartrazine.

Method	LOD (mg L^{-1})	RSD (%)	Linear range (mg L^{-1})	Ref.
SPE	0.04	2.2	0.04–4.8	[3]
IL-ATPSs	0.052	3.2	0.01–50	[23]
MCPE	0.032–0.055	0.99	0.43–10000	[24]
HMDE	3.3	–	6.6–300	[25]
CPE	0.133	–	0.04–4.00	[26]
VALPME	0.019	2.62	–	[27]
VADLLME	0.04	2.1	0.05–1	[28]
MSIS-DLLME	0.03	4.6	0.1–1	This work

Notes: LOD: Limit Of Detection; RSD: Relative Standard Deviation; IL-ATPSs: Ionic liquidbased aqueous two-phase systems; MCPE: modified carbon paste electrode; HMDE: Hanging mercury drop electrode; CPE: Cloud Point Extraction; VALPME: vortex-assisted liquid-phase microextraction; VADLLME: Vortex-assisted dispersive liquid-liquid microextraction; MSISDLLME: magnetic stirring in syringe dispersive liquid-liquid microextraction.

Table 5
Determination of Tartrazine in food samples by MSIS-DLLME method.

Sample	Added (μg L^{-1})	Founded (μg L^{-1})	Recovery (%)
Saffron powder	–	72.3 ± 2.2[a]	–
	100	173.1 ± 2.3	100.8
	150	220.3 ± 4.5	98.6
Fruit juice	–	20.9 ± 1.6	–
	100	119.6 ± 3.4	98.7
	150	172.3 ± 3.5	100.9
Pineapples powder	–	49.4 ± 1.4	–
	100	150.5 ± 3.3	101.1
	150	202.1 ± 4.2	101.8
Dessert bananas	–	124.2 ± 3.2	–
	100	223.8 ± 3.4	99.6
	150	275.3 ± 3.4	100.7
Powder Oranges syrup	–	85.6 ± 2.5	–
	100	184.1 ± 3.7	98.5
	150	232.3 ± 1.3	97.8

N.D: below the detection limit.
[a] Errors correspond to standard deviations of three replicate measurements.

Acknowledgments

We also thank Dr. Majid Ramezani (Department of chemistry, Faculty of science, Arak Branch, Islamic Azad University, Arak, Iran) for providing us technical supports.

References

[1] H. Rahnama, S.M. Mazloomi, E. Berizi, A. Abbasi, Z. Gholami, Food Sci. Nutr. 10 (2022) 3781–3788.
[2] S. Altınoz, S. Toptan, J. Food Compos. Anal. 15 (2002) 667–683.
[3] R. Li, Z.T. Jiang, Y.H. Liu, J. Food Drug Anal. 16 (5) (2008) 6.
[4] Joint FA, World Health Organization, WHO Expert Committee on Food Additives. Evaluation of Certain Food Additives and Contaminants: Seventy-Seventh Report of the Joint FAO/WHO Expert Committee on Food Additives, World Health Organization, 2013.
[5] K. Sathya, H. Jayalakshmi, S.N. Reddy, M.V. Ratnam, D. Bandhu, Biomass Convers. Biorefinery Dec 18 (2023) 1–3.
[6] J. Maslowska, J. Janiak, Chem. Anal. 41 (1996) 855–864.
[7] A.V. Schenone, M.J. Culzoni, N.R. Marsili, H.C. Goicoechea, Food Chem. 138 (2013), 1928-35.
[8] M. Bratu, A.F. Danet, A. Bratu, Revista de Chimie-Bucharest-Original Edition- 56 (2005) 453–458.
[9] M.R. Fuh, K.J. Chia, Talanta 56 (2002) 663–671.

[10] M.S. García-Falcón, C. Pérez-Lamela, E. Martínez-Carballo, J. Simal-Gándara, Food Chem. 105 (2007) 248–259.
[11] A. Husain, W. Sawaya, A. Al-Omair, S. Al-Zenki, H. Al-Amiri, N. Ahmed, M. Al-Sinan, Food Add and Cont. 23 (2006) 245–251.
[12] M. Ma, X. Luo, B. Chen, S. Su, S. Yao, J. Chromatogr., A 1103 (2006) 170–176.
[13] K.S. Minioti, C.F. Sakellariou, N.S. Thomaidis, Anal. Chim. Acta 583 (2007) 103–110.
[14] J.H. Zou, W.D. Chen, J.D. Shao, Chin. J. Anal. Chem. 29 (2001) 1192–1195.
[15] E.A. Rukosueva, G.R. Aliyarova, T.I. Tikhomirova, V.V. Apyari, P.N. Nesterenko, Int J. of Ana Chem. 2 (2020) 1–6.
[16] F.E. Lancaster, J.F. Lawrence, Food Addit. Contam. 16 (1999) 381–390.
[17] M.C. Barciela-Alonso, V. Plata-García, A. Rouco-López, A. Moreda-Piñeiro, P. Bermejo-Barrera, Microw. J. 114 (2014) 106–110.
[18] M. Rezaee, Y. Assadi, M.R. Hosseini, E. Aghaee, F. Ahmadi, S. Berijani, J. Chromatogr., A 1116 (2006) 1–9.
[19] A. Mohammadzadeh, M. Ramezani, A. Niazi, Desalination Water Treat. 57 (2016) 9745–9755.
[20] H. Chen, C. Ruiwen, Shengqing Li, J. Chromatogr. 8 (2010) 1244–1248.
[21] E. Yiantzi, E. Psillakis, K. Tyrovola, N. Kalogerakis, Talanta 80 (2010) 2057–2062.
[22] M. Ezoddin, T. Taghizadeh, B. Majidi, Environ. Technol. 35 (2014) 2401–2409.
[23] O. Sha, X. Zhu, Y. Feng, W. Ma, Food Chem. 174 (2015) 380–386.
[24] H.M. Shawish, N.A. Ghalwa, S.M. Saadeh, H. El Harazeen, Food Chem. 138 (2013) 126–132.
[25] M. Gomez, V. Arancibia, C. Rojas, E. Nagles, Int. J. Electrochem. Sci. 7 (2012) 7493–7502.
[26] A.P. Nambiar, M. Sanyal, P.S. Shrivastav, Food Anal. Methods 10 (2017) 3471–3480.
[27] S. Duman, M. Soylak, J. Iran. Chem. Soc. 20 (2023) 69–78.
[28] Z. Gholami, M.H. Marhamatizadeh, S. Yousefinejad, M. Rashedinia, S.M. Mazloomi, Microchem. J. 170 (2021) 106671.