Research Article

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One-factor-at-a-time method combined with ICP-MS for determining 11 elements in soy sauce and their migration from the containing glass bottles

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Abstract: This study was designed to determine the 11 metal elements (Al, Cr, Mn, Fe, As, Ni, Cu, Zn, Cd, Sb, and Pb) in soy sauce and their migration from the containing glass bottles. Inductively coupled plasma mass spectrometry (ICP-MS) was applied for the determination of the elements and one-factor-at-a-time method was employed for optimizing the ICP-MS parameters in migration experiment and microwave digestion experiment. The developed method was successfully applied to determine the content of 11 elements. The results showed that the experiment had excellent correlation and sensitivity. The accuracy of the elements in the migration study and test of soy sauce itself ranging from 84.25% to 118.75% was satisfied, and the precision of the method was validated and the RSD was no more than 15.5%. The concentration of all the detected metal elements migrated from the glass bottles were between 0.3450 and 2.398 ng·mL⁻¹, and the risk assessment indicated that the metal elements in soy sauce had no risk to the public health. The proposed methodology in this study was successfully applied for the quality control for metal elements in soy sauce and the containing glass bottles for the first time, and a research method suitable for soy sauce consumption process control and risk assessment has been established.

Keywords: elements, soy sauce, migration, ICP-MS, glass bottles

1 Introduction

As one of the popular condiments, soy sauce originated from Asia and spread all over the world. It is mainly made from soybeans and flour via fermentation using microbial enzymes. From the analytical point of view, soy sauce is a complex matrix, and contains about 8% proteins, 5% carbohydrates, 0.8% fiber, 0.6% lipids, 5.5% sodium, and 0.4% potassium [1,2]. However, the undesired pollution in soy sauce may result in high level of metal elements which could be a potential risk to human health [2]. Metal elements such as lead and copper in soy sauce are mainly derived from soil, raw materials, environment, production process, and packaging materials [3,4]. The packaging materials play an important role in food safety and shelf life by protecting the products from external light, microorganisms, and air.

Chemical migration mainly depends on the type of material contacting the food products [5]. Studies showed that packaging materials can contaminate food with some harmful elements [6,7]. Glass is widely used in packaging materials for soy sauce because of its eco-friendly property and low cost. However, it was reported that at least 18 different elements were found in glass (may be as stabilizers), including aluminum, antimony, arsenic, barium, bismuth, boron, calcium, germanium, hafnium, lanthanum, magnesium, niobium, strontium, tantalum, titanium, yttrium, zinc, and zirconium [8–10]. Previous research [11,12] reports showed that glass bottles contaminated bottled water with heavy metals. Therefore, the investigation of the unwanted elements which migrate from soy sauce packaging materials is of great importance.

The toxicity of elements is closely related to their exposure concentration. Chemical elements are essential for biological processes and play an important role in normal growth and development of the organism, but

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can be toxic when taken in excess [13,14]. Nowadays, soy sauce is consumed worldwide, the amount of soy sauce made in China was around ten million tons in 2016 [2]. When studying the migration of elements from the glass bottles, it is also necessary to evaluate the health risk of chemical elements intake from the daily consumed soy sauce. Target hazard quotient (THQ) is an effective tool to evaluate the health risk of different elements, which is calculated based on the ratio of estimated daily intake (EDI) to oral reference dose (RfD) [15] of soy sauce. This method is based on the Region III risk-based concentration table from United States Environmental Protection Agency (USEPA) [16]. Furthermore, RfD refers to Integrated Risk Information System from USEPA [17]. Simultaneously, Food and Agriculture Organization of the United Nations/World Health Organization/the Joint Expert Committee on food additives evaluated the safety of contaminants and toxins in food, and finally revised the concept of provisional tolerable weekly intake (PTWI). Accordingly, risk assessment was carried out for the chemical elements on human health through PTWI and THQ.

Currently, inductively coupled plasma mass spectrometry (ICP-MS) is widely used for the determination of chemical elements [3] at ppt levels and it is recommended by China National Food Safety Standard for the determination of elements in condiments, such as soy sauce [18,19]. International Organization for Standardization (ISO) 7,086-1:2,000 requires that a glass bottle should be briefly washed with a solution containing a nonacidic detergent at a temperature of approximately 40°C, followed by rinsing with tap water, and then distilled water, and finally filled with a 4% aqueous acetic acid (CH_3COOH) solution.

This study optimized the microwave digestion method for soy sauce sample pretreatment, selected the optimal system, and established an ICP-MS method for the simultaneous determination of 11 metal elements. This research aims to determine the chemical elements in soy sauce and their possible migration from glass bottles. And the health risk assessment on the chemical elements' migration from the glass bottles into soy sauce has been carried out for the first time. This study provided a reference to further control quality of chemical elements in condiments and their packaging materials.

2 Experimental methods

2.1 Materials and reagents

All chemicals and reagents were of chromatographic grade. Nitric acid (69% HNO_3) was purchased from Beijing Chemical Works (Beijing, China). Ethanol ($\text{CH}_3\text{CH}_2\text{OH}$)

was obtained from Tianjin Concord Technology Co., Ltd (Tianjing, China). Acetic acid (CH_3COOH) was acquired from Sinopharm Group Chemical Reagent Co., Ltd (Shanghai, China). The water used for the experiment came from Shenyang Wahaha Rongtai Food Co., Ltd (Shenyang, China). Hydrogen peroxide (H_2O_2) was obtained from Shenyang Yuwang Chemical Glass Instrument Co., Ltd (Shenyang, China).

The standards of Al(III) , Cr(III) , Mn(II) , Fe(III) , Ni(II) , Zn(II) , As(III) , Cd(II) , Sb(III) , and Pb(II) were purchased from Guobiao Testing & Certification Co., Ltd (Beijing, China) and Cu(II) were purchased from AnPu Experimental Technology Co., Ltd (Shanghai, China). The working standard solutions were prepared daily by diluting each single-element standard stock solution. All standard solutions need to be sealed and stored below 4°C in dark before analysis.

2.2 Apparatus

The constant temperature and humidity incubator, LRHS-300F-II, was purchased from Shanghai Longyue Instrument Equipment Co., Ltd (Shanghai, China). The Multiwave Pro microwave digester (Anton Paar, Austria) and the BHW-09A45 acid-driven processor (Anton Paar, Austria) were used for sample digestion. All the elements were measured by an Agilent 7800 ICP-MS (Agilent, USA) with a MicroMist nebulizer. The instrumental conditions are set in Table 1.

2.3 Sample preparation

In the experiment, seven different brands of soy sauce with different packagings were randomly selected from the local supermarket in Shenyang, Liaoning. The glass bottles of three different brands were studied for the migration of elements.

Before test, all the glass bottles were rinsed with distilled water and soaked in 4% (v/v) CH_3COOH at 22°C for 24 h. Then, 10 mL of fresh extraction solution was transferred to a microwave digestion tank and heated at 150°C for 60 min in order to digest CH_3COOH . After cooling down, the remaining solution in the microwave digestion tank was transferred into a 50 mL volumetric flask and diluted to the volume with 2% (v/v) HNO_3 . All the sample solutions were prepared in duplicate.

The purpose of the digestion experiment was to make it easier to extract the chemical elements in soy sauce. To test elements in soy sauce, 1 mL of soy sauce sample,

Table 1: Instrumental conditions (Agilent 7800 ICP-MS)

Instrumental conditions	Parameters
Helium gas flow rate (mL·min ⁻¹)	0.8
Carrier gas flow rate (mL·min ⁻¹)	1
Plasma gas flow rate (mL·min ⁻¹)	15
Auxiliary gas flow rate (mL·min ⁻¹)	1
Makeup gas flow rate (mL·min ⁻¹)	1
Spray chamber temperature (°C)	3
Peripump (rps)	0.1
Point of peak	3
Replicates	2–3

6 mL of HNO₃, and 2 mL of H₂O₂ were added into the microwave digestion tank and digested in a microwave

system at 180°C for 15 min. After the completion of digestion, the tank was placed in a hot plate for 30 min to evaporate the solvents. Then, the cooled and dried sample was diluted with 2% (v/v) HNO₃ and transferred to a 50 mL volumetric flask before analyzing using ICP-MS.

2.4 Method validation

The linear regression analysis was verified by the correlation coefficient (R^2) of the calibration curve for each element. The limit of detection (LOD) of 11 metal elements was estimated to be three times of the standard deviation (SD) of ten blank samples. The limit of quantification

Table 2: Regression equation, linear range, sensitivity, instrument precision, and accuracy for migration experiments

Elements	Regression equation	R^2	Range (ng·mL ⁻¹)	LOD (ng·mL ⁻¹)	LOQ (ng·mL ⁻¹)	RSD of instrument precision (%)	Standard solution (ng·mL ⁻¹)	Average recovery (%)	RSD (%)
Al	$y = 166.8x + 904.04$	0.9992	0.10–200	0.089	0.297	0.8	8.00 10.00 12.00	106.52 101.05 99.59	1.4 5.2 6.1
Cr	$y = 8,001x + 16,195$	0.9994	0.10–200	0.047	0.155	0.8	0.40 0.50 0.60	84.25 87.87 97.83	1.1 7.4 5.6
Mn	$y = 3895.8x + 1706.4$	0.9999	0.05–100	0.013	0.044	0.8	0.40 0.50 0.60	106.83 110.33 108.83	0.4 2.1 4.0
Fe	$y = 6696.5x + 47,277$	0.9880	0.10–200	0.087	0.290	0.9	8.00 10.00 12.00	95.33 101.34 105.20	0.8 1.3 2.1
Ni	$y = 3897.7x + 1,068$	1.0000	0.10–200	0.021	0.071	1.2	0.40 0.50 0.60	93.67 101.33 100.94	2.2 3.5 5.1
Cu	$y = 10823x + 7194.9$	1.0000	0.10–200	0.060	0.200	1.1	4.00 5.00 6.00	102.22 107.59 103.06	4.6 4.2 2.7
Zn	$y = 1488.3x + 17,625$	0.9782	0.10–200	0.158	0.528	1.6	8.00 10.00 12.00	111.57 108.16 85.03	4.3 4.8 9.5
As	$y = 926.43x + 166.57$	0.9999	0.05–100	0.033	0.110	1.2	0.16 0.20 0.24	118.75 110.33 113.89	4.8 4.7 5.6
Cd	$y = 2316.2x + 266.93$	1.0000	0.05–100	0.006	0.020	0.7	0.16 0.20 0.24	92.50 97.67 114.17	11.1 4.6 1.3
Sb	$y = 5813.6x + 1885.2$	0.9999	0.05–100	0.019	0.064	0.5	0.16 0.20 0.24	102.08 110.00 102.78	6.0 2.7 5.0
Pb	$y = 22,564x + 12,233$	1.0000	0.05–100	0.012	0.039	0.5	0.16 0.20 0.24	118.54 109.17 96.25	6.0 5.5 4.8

Figure 1: Effect of time on the concentration of each element in migration.

(LOQ) was determined to be ten times of the above SD. To assess recovery, each chemical element was added to the samples at concentrations of 80%, 100%, 120% in triplicate.

To validate the precision and accuracy for migration study and test the elements in the soy sauce itself, the samples were spiked with mixed standards at three levels and then analyzed as described for normal samples in triplicate.

Figure 2: Effect of different acidity solvent on the concentration of each element in migration.

Figure 3: Effect of different levels of H₂O₂ in digestion solvent on the recovery of each element.

2.5 Extraction, migration, and digestion tests

To confirm the effect of extraction time [20], extraction with 4% (v/v) CH₃COOH for 48 h resulted in a near-logarithmic profile. 10 mL of samples were withdrawn at 1, 2, 4, 8, 24,

and 48 h, and heated at 150°C for 60 min to remove the organic solvents. After cooling down to ambient temperature, samples were diluted to 50 mL with 2% (v/v) HNO₃.

In order to study the effect of different acids on the migration, two mixed standards (1.0 and 20.0 ng·mL⁻¹) were prepared according to the sample preparation

Figure 4: Effect of different digestion times on the recovery of each element.

Figure 5: Effect of temperature on the recovery of each element in digestion.

method. The influence of different organic solvents on the element migration was investigated by changing the solvent type with fixed concentration of 4% (v/v) CH_3COOH and 2% (v/v) HNO_3 .

Different digestion solvents in microwave digestion of soy sauce was evaluated. The recovery of the elements in three different solvents (8 mL of HNO_3 , 7 mL of HNO_3 with 1 mL of H_2O_2 , and 6 mL of HNO_3 with 2 mL of H_2O_2) was compared. In order to verify whether the acid concentration will affect the results, a blank digestion solution was prepared.

3 Results and discussion

3.1 Optimization of experiment conditions

3.1.1 Condition optimization for migration experiment

The migration experiment condition of the elements from glass bottles was optimized (Table 2). Concentration ratios of the metals in different leaching solutions vary significantly from acid to acid [21]. Preliminary data showed that the type of the organic solvent and incubating time were critical for the migration of 11 elements and the concentration of element ions was examined in the eluate after

different solvent erosions. The different leaching solutions were evaluated in microwave digestion including 10% (v/v in water) $\text{CH}_3\text{CH}_2\text{OH}$, 20% (v/v in water) $\text{CH}_3\text{CH}_2\text{OH}$, and 4% (v/v in water) CH_3COOH . The results showed that 4% (v/v in water) CH_3COOH had the best migration effect and indicated that the presence of H_2O_2 made the digestion more complete because they could make the digestion system to have stronger oxidation ability [22].

The effect of migration time is shown in Figure 1. After 24 h, the increasing rate of concentration of each element in 4% (v/v) CH_3COOH started to slow down, indicating that the migration reached stable state.

The effect of different acidity on the migration is shown in Figure 2, and the results were in the range of 0.8–1.2. There was no interference with the organic solvent in the analytical values.

3.1.2 Optimization of digestion condition

In most cases, the decomposition of samples could be affected by a combination of different parameters, such as the microwave power, digestion time, pressure, and temperature in the sealed digestive tube [3,23].

HNO_3 , HNO_3 -perchloric acid (HClO_4), and HNO_3 - H_2O_2 . HClO_4 are highly explosive and oxidizing acids which may have a potential safety risk. Due to the strong oxidizing property of H_2O_2 , it is often used as a digestion reagent

Table 3: Organic solvent interference in migration experiment

Elements	1.0 ng·mL ⁻¹			20.0 ng·mL ⁻¹		
	Concentration in dilute nitric acid (ng·mL ⁻¹)	Concentration in 4% acetic acid (ng·mL ⁻¹)	Ratio	Concentration in dilute nitric acid (ng·mL ⁻¹)	Concentration in 4% acetic acid (ng·mL ⁻¹)	Ratio
Al	1.020	1.141	1.119	21.996	20.819	0.946
Cr	0.839	0.951	1.134	20.357	20.212	0.993
Mn	0.966	1.130	1.170	21.688	21.078	0.972
Fe	1.113	1.055	0.948	22.476	24.491	1.090
Ni	0.975	1.027	1.053	21.365	22.563	1.056
Cu	0.905	1.061	1.172	21.150	21.610	1.022
Zn	1.086	1.156	1.064	20.933	23.612	1.128
As	0.955	1.113	1.165	21.751	23.495	1.080
Cd	0.964	1.111	1.153	21.414	21.143	0.987
Sb	0.967	1.085	1.123	21.753	21.375	0.983
Pb	1.022	0.921	0.901	21.501	20.982	0.976

with HNO₃ [24]. H₂O₂ may increase the solubility of Pb [25]. Therefore, different levels of H₂O₂ in the digestion solvent were investigated. Results showed that 2 mL of H₂O₂ with 6 mL of HNO₃ as the digestion solution had the highest recovery (Figure 3).

The effect of the digestion time was also studied, and the recovery of the elements at different digestion times (5, 10, 15, and 20 min) was compared. According to the results in Figure 4, the highest recovery was found at 15–20 min.

As to the digestion temperature, it was reported that temperature changes in microwave digestion might affect the results [4]. Efficient digestion for all chemical elements is presented in Figure 5. Moreover, the analytical results showed that 180°C gave a statistically higher recovery than the other temperatures.

The mixed standard solutions (5.0 and 100.0 ng·mL⁻¹) were diluted using the blank solution with a fixed 2% (v/v) HNO₃. The results are shown in Table 3. All the concentrations were in the range of 0.8–1.2%, and there was no significant change in the recovery.

3.2 Method validation

The analytical method employed for 11 chemical elements detection was validated on linearity, LOD, LOQ, accuracy, and precision. The results from validation studies indicated the method had a satisfactory performance.

The linearity of the method using the least squares method was evaluated by calculating the regression coefficients. The regression equations were derived from statistical tests and the linear range of the migration experiment was within 0.05–200 ng·mL⁻¹. In this range, the regression model was considered acceptable for most

tested elements ($R^2 > 0.978$). The LOD levels were between 0.006 ng·mL⁻¹ (LOD Cd) and 0.158 ng·mL⁻¹ (LOD Zn). The LOQ levels were between 0.020 ng·mL⁻¹ (LOQ Cd) and 0.528 ng·mL⁻¹ (LOQ Zn). The experiment had excellent correlation and sensitivity.

Meanwhile, the linearity test gave a correlation coefficient ($R^2 > 0.994$) for every element with a linearity range of 0.25–100 or 0.50–200 ng·mL⁻¹. The LOD and LOQ of the method are between 0.015 ng·mL⁻¹ (LOD Cd) or 0.025 ng·mL⁻¹ (LOQ Cd) and 0.490 ng·mL⁻¹ (LOD Al) or 1.632 ng·mL⁻¹ (LOQ Al).

The instrument precision of this method was evaluated using RSD and the results are shown in Table 4. All the values for the elements migrated from glass ranged from 0.5% to 1.6%.

Table 4: Trace elements migration in different brands of soy sauce glass bottles

Elements	Haitian's migration (ng·mL ⁻¹)	Donggu's migration (ng·mL ⁻¹)	Qinghua's migration (ng·mL ⁻¹)	National standard limit (ng·mL ⁻¹)
Al	26.96	26.93	20.9675	None
Cr	0.9425	—	—	None
Mn	0.2101	—	—	None
Fe	17.14	16.1325	20.165	None
Ni	13.73	13.565	13.16	None
Cu	11.5925	12.17	9.3775	None
Zn	17.8225	34.0175	26.065	None
As	—	—	—	200
Cd	—	—	—	500
Sb	—	—	—	1,200
Pb	0.408	0.345	0.353	1,500

Note: — means the measured value is less than the LOQ.

Table 6: Concentrations and ratios of 5.0 and 100.0 ng·mL⁻¹ mixed standard solution in two solutions

Elements	Content in 7 brands (μg·mL ⁻¹)	5.0 ng·mL ⁻¹			100.0 ng·mL ⁻¹		
		Concentration in 2% dilute nitric acid (ng·mL ⁻¹)	Concentration in blank digestion solution (ng·mL ⁻¹)	Ratios	Concentration in 2% dilute nitric acid (ng·mL ⁻¹)	Concentration in blank digestion solution (ng·mL ⁻¹)	Ratios
Al	0.043–0.203	5.494	5.822	1.060	99.382	107.201	1.079
Cr	0.082–0.162	5.514	5.535	1.004	94.208	104.751	1.112
Mn	0.359–0.886	5.664	5.727	1.011	97.067	98.125	1.011
Fe	0.857–2.398	5.517	4.669	0.846	94.732	104.888	1.107
Ni	0.078–0.202	4.993	5.729	1.147	96.934	99.148	1.023
Cu	0.006–0.021	5.227	5.703	1.091	96.144	100.095	1.041
Zn	1.199–1.953	5.070	5.089	1.004	97.438	107.017	1.098
As	—	5.655	5.474	0.968	99.542	97.694	0.981
Cd	—	5.404	5.180	0.959	97.852	96.063	0.982
Sb	—	5.266	5.345	1.015	98.677	97.108	0.984
Pb	0.006–0.168	5.400	5.752	1.065	98.902	102.109	1.032

Note: — means the element was not detected.

The precision and accuracy of the elements in the migration study and test of soy sauce itself were validated. The accuracy was found to be between 84.25% and 118.75% for 11 chemical elements, and the RSDs were within 0.4–11.1%. For analyzing elements in soy sauce, the accuracy ranged from 93.57% to 114.65%, and the RSDs were all less than 15.5% (Table 5). The accuracy and precision of the method were reliable and suitable.

3.3 Analysis of real samples

Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sb, and Pb in seven brands of soy sauce samples were tested using the validated method, and the contents of 11 elements are shown in Table 6.

Samples from three different brands of glass bottles were analyzed by the validated migration method (Table 7). Several elements with different concentrations were detected in all the samples. The results showed that the content of each element in the soy sauce in glass bottle did not exceed the national standard [26].

3.4 Safety evaluation

Risk assessment was conducted separately for essential and toxic chemical elements. THQ and the EDI of 11 elements were analyzed and compared with PTWI and RfD. The daily intake of soy sauce for Chinese residents (9 g per person per day) was obtained from China Health and Nutrition Survey (CHNS, 2002). The THQ of ingested 11 elements was calculated using the following equation:

Table 7: Result of 11 trace elements in seven kinds of soy sauce samples

Elements	Haitian's plastic bag (μg·mL ⁻¹)	Haitian's plastic bottle (μg·mL ⁻¹)	Haitian's glass bottle (μg·mL ⁻¹)	Donggu's plastic bag (μg·mL ⁻¹)	Donggu's glass bottle (μg·mL ⁻¹)	Qinghua's plastic bottle (μg·mL ⁻¹)	Qinghua's glass bottle (μg·mL ⁻¹)	National standard limit (ng·mL ⁻¹)
Al	0.187	0.182	0.123	0.043	0.085	0.069	0.203	None
Cr	—	—	—	—	0.082	—	0.162	None
Mn	0.432	0.886	0.866	0.465	0.542	—	0.359	None
Fe	0.857	1.371	1.005	0.992	0.947	2.398	1.946	None
Ni	0.082	0.202	0.170	0.078	0.084	0.146	0.135	None
Cu	0.019	0.014	0.019	0.020	0.021	0.014	0.006	None
Zn	1.199	1.288	1.454	1.153	1.632	1.953	1.586	None
As	—	—	—	—	—	—	—	0.5
Cd	—	—	—	—	—	—	—	None
Sb	—	—	—	—	—	—	—	None
Pb	0.024	0.019	0.007	0.006	0.106	0.020	0.168	1.0

Note: — means the measured value is less than the LOQ.

Table 8: EDI and THQ of trace elements due to the soy sauce and glass bottles

Elements	RfDo (μg.kg ⁻¹ .day ⁻¹)	Maximum exposure level in the soy sauce		Maximum exposure level in the soy sauce glass bottles	
		EDI	THQ	EDI	THQ
Essential					
Al	1,000	0.2400 × 10 ⁻⁴	0.2400 × 10 ⁻⁷	3.187 × 10 ⁻⁶	3.187 × 10 ⁻⁹
Cr(III)	1,500	0.1915 × 10 ⁻⁴	0.1276 × 10 ⁻⁷	0.1111 × 10 ⁻⁶	0.7407 × 10 ⁻¹⁰
Mn	140	0.1047 × 10 ⁻³	0.7479 × 10 ⁻⁶	0.2483 × 10 ⁻⁷	0.1774 × 10 ⁻⁹
Fe	700	0.2834 × 10 ⁻³	0.4049 × 10 ⁻⁶	2.383 × 10 ⁻⁶	0.3404 × 10 ⁻⁸
Ni	20	0.2388 × 10 ⁻⁴	0.1194 × 10 ⁻⁵	1.623 × 10 ⁻⁶	0.8115 × 10 ⁻⁷
Cu	40	0.2483 × 10 ⁻⁵	0.6208 × 10 ⁻⁷	1.438 × 10 ⁻⁶	0.3595 × 10 ⁻⁷
Zn	300	0.2309 × 10 ⁻³	0.7697 × 10 ⁻⁶	4.022 × 10 ⁻⁶	0.1341 × 10 ⁻⁷
Toxic					
As	0.3	—	—	—	—
Cd	1	—	—	—	—
Sb	0.4	—	—	—	—
Pb	357*	0.1986 × 10 ⁻⁴	0.5563 × 10 ⁻⁷	0.4847 × 10 ⁻⁷	0.1358 × 10 ⁻⁹

Note: — represents the undetected value; *represents the safety evaluation of Pb use the PTWI established by JECFA; EDI means “estimated daily intakes;” THQ means “target hazard quotient.”

$$\text{EDI} = [(\text{IR} \times C)/\text{BWa}] \times 10^{-3} \quad (1)$$

where IR is the daily intake of soy sauce per person ($\text{g}\cdot\text{day}^{-1}$); C is the content of every element in the soy sauce ($\text{g}\cdot\text{kg}^{-1}$); and BWa stands for average of body weight (kg).

$$\text{THQ} = \text{EDI}/\text{RfD} \quad (2)$$

where THQ represents the value of risk (if value of THQ is less than 1, it means that this level is unlikely to cause any obvious deleterious effects to the general health).

According to the database of the Nutrition and Chronic Diseases of Chinese residents, the average body weight of Chinese residents is 66.2 kg [27]. The density of soy sauce is regarded as $1.15 \times 10^3 \text{ kg}\cdot\text{m}^{-3}$.

The THQ value of each chemical element and the maximum allowed levels in the soy sauce were far less than 1 (Table 8), suggesting that the listed ingested metal elements in soy sauce is within the safe range. It is critical that humans may underestimate or overestimate the health hazard due to the variety of interactions among the mixture components [28]. Chemical elements intake under real conditions will be lower than that resulting from migration into the food simulant.

4 Conclusion

In this study, a novel method, one-factor-at-a-time method combined with ICP-MS, for determining Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sb, and Pb in soy sauce and their migration

from the containing glass bottles was developed. By optimizing the migration experiment and digestion experiment conditions, determining the extraction time, the effect of different acids, and different digestion solvents, the ICP-MS methods were formed. The chemicals in the glass migrate to the soy sauce at very low levels. Combined with the risk assessment, it was confirmed that the soy sauce and glass bottles used in this study were safe. The methods for determining the content of 11 chemical elements are accurate and reliable. Accurate and fair results provide theoretical basis for long-term safe storage of soy sauce. This experiment provided a new direction on the quality control of condiments.

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