Bull. Chem. Soc. Ethiop. **2024**, 38(6), 1509-1519. ISSN 1011-3924

© 2024 Chemical Society of Ethiopia and The Authors Printed in Ethiopia \circ 2024 Chemical Society of Ethiopia and The Authors Printed in Ethiopia DOI: https://dx.doi.org/10.4314/bcse.v38i6.1 Online ISSN 1726-801X DOI: https://dx.doi.org/10.4314/bcse.v38i6.1

QUANTIFICATION OF SELECTED HEAVY METALS THROUGH INDUCTIVELY COUPLED PLASMA-OPTICAL EMISSION SPECTROMETRY IN CONTAINERS OF YOGURTS USED IN ERBIL CITY, KRG, IRAQ

Hawraz Sami Khalid, Aveen Faidhalla Jalal* , Hawar Fakhir Mohammed, Huda Yousif Sharef and Nabil Adil Fakhre

Department of Chemistry, College of Education, Salahaddin University-Erbil, Erbil, Kurdistan Region, Iraq

(Received January 22, 2024; Revised June 6, 2024; Accepted August 6, 2024)

ABSTRACT. Heavy metals are recently introduced in the manufacturing of various household utensils. The objective of this study was to investigate different heavy metals contents in the composition of yogurts containers collected from the markets of Erbil City. The collected containers were aluminum containers and plastic containers with several colors. An inductively coupled plasma optical emission spectrometer was used to analyse the contents of nine heavy metals including cadmium, cobalt, chromium, copper, iron, lead, manganese, nickel, and zinc. The level of total detected metal load found in the analyzed containers. The recorded level for total detected metal load in the overall aluminum containers sample was very high and approximately 27 times more than that level in the overall plastic containers sample. Lead and cadmium were known as toxic heavy metals and detected with a high amount in the aluminum containers composition. The total contamination in the aluminum containers composition was noticeably exceeded the permissible world limits for heavy metals in packaging materials. Results obtained clarified that the white color of plastic containers is more preferable and safer than the dark or other multi-color plastic containers items in terms of heavy metals.

KEY WORDS: Aluminum and plastic container, Heavy metal, ICP-OES, Yogurt

INTRODUCTION

Heavy metals (HMs) are generally known as most environmental contamination and natural elements with a high atomic weight and relatively high density (more than 6.0 g/cm^3) compared to water [1, 2]. In recent years, industrial, geogenic, agricultural, domestic effluents, pharmaceutical, and atmospheric sources are commonly reported and classified as primary sources of HMs in the environment [3, 4]. Cd, Hg, Pb, As, and Cr were known as toxic elements and play a vital role in developing varying diseases. They are classified as human carcinogens, known to persuade multiple organ damage, and are considered systemic toxicants, even at lower exposure [5-7]. The degree of HMs toxicity depends on various factors, such as the route of exposure, the dose, chemical species, the nutritional, genetics, gender, and age status of exposed individuals [7].

Several HMs and their organic forms are exposed to humans in different ways, including the occupational workplace, environment, water, food supply, and industrial products [8-12]. Metals and alloys are widely used to produce various household utensils and food containers as a safety barrier between the food and the environment [13]. The presence of toxic HMs from the starting raw materials can contaminate packing/container compositions [14-17]. Consequently, food containers can release HMs ions in their composition [13], transferring HMs into foods by the leaching process and causing food contamination [18]. The leaching process is mainly linked to the foods' chemical or physical conditions, including pH and temperature [19-21]. Releasing metal ions may high amount, unacceptable, exceed the world toxicological reference values, and endanger the consumers' health. As a result, several technical guidance was recently established to regulate metals and alloys used in food contact materials and articles [13].

 $\overline{}$

^{*}Corresponding authors. E-mail: aveen.jalal@su.edu.krd

This work is licensed under the Creative Commons Attribution 4.0 International License

According to recent studies, different household items used as food contact materials recorded varying concentrations of toxic HMs. Several recent studies have been conducted to assess any significant level of HMs contents or leaching from samples of cooking utensils [22], aluminum cookware [23, 24], aluminum foil [25], and stainless steel kitchenware and tableware [26] items. Many kinds of researches have also applied on various clay and glass items, including glass dinnerware [27], glass-clay containers [28], traditional clay pots [29], domestic ceramic wares [30] and glass and enamels of consumer container bottles [15]. In recent years, scientists also investigated paper and paperboard food packaging [31], packaging papers, and paperboards [14] in terms of HMs contents. Many researchers also studied on toxic HMs leaching or composition contents of plastic food containers [17], recycled plastic food packaging [32], colored plastic table dishes [33], bottles of drinking water [34] and plastic bags [35].

Recent studies have shown that varying levels of different toxic HMs were detected in several household items' composition. Weidenhamer and Fitzpatrick [24] stated that lead content was detected in 86% (36 out of 42) samples of aluminum cookware collected from different developing countries. Results also showed ranges for Pb content were below the detection limit (BDL) - 610 mg/kg in Kenya, BDL - 620 mg/kg in Tanzania, 240 - 850 mg/kg in Ivory Coast, 270 - 860 mg/kg in the Philippines, 460-1000 mg/kg in Guatemala, 280-1570 mg/kg Bangladesh, BDL-2620 mg/kg in India, BDL-2690 mg/kg in Indonesia, 340-3020 mg/kg in Nepal, BDL-7070 mg/kg in Viet Nam aluminum cookware items [24].

The contents of Pb, Cd, and Cr were 32.38, 0.41, 101.57 in mg/kg unit, respectively in pot ceramic; 140.91, 1.14, and 175.68 in mg/kg unit, respectively in bowl ceramic [30]; 51.20, 20.50, and 133.00 in mg/kg unit, respectively in brown glass; 202.00, 21.70, and 2020.00 in mg/kg unit, respectively in green glass; 73.30, 18.30, and BDL in mg/kg unit, respectively in clear glass containers; and 45.70, BDL, and 22.20 in mg/kg unit, respectively in a clear drinking glass container [15]. The contents of Pb, Cd, and Cr were 12.77, 0.16, and 6.14 in mg/kg unit, respectively in Lahmacun box packaging [31]; and 0.34, not tested (NT), and 0.45 in mg/kg unit, respectively in Pizza box [14] composition containers.

Various new and commercial analytical methods have been adopted for heavy ions detection including single-element techniques (AFS, spectrometry and colorimetry, immuno-chemical methods), or as simultaneous multielement analyses (FAAS, ICP-AES, ICP-MS, NAA, GC, LC, IC, and CE) and electrochemical [36].

The European Union (EU) Directive 94/62/EC (amended by 2004/12/EC) on Packaging and Packaging Waste and the State of California Health and Safety Code (the State of California Toxics in Packaging Prevention Act of 2006) set out a maximum limit of 100 mg/kg by weight for all Pb, Cd, Cr (VI), and Hg in packaging materials [37-39]. To the best of our knowledge, there is no previous study addressed the contents of HMs in yogurt containers in Erbil City, Kurdistan Region, Iraq. Therefore, this study was conducted to assess some toxic HMs within the composition of local aluminum and plastic containers used to store local yogurt products.

EXPERIMENTAL

Chemicals and instruments

All the chemicals used in this study were of analytical reagent grade, including concentrated sulfuric acid (H₂SO₄ 97%), perchloric acid (HClO₄ 70%), hydrochloric acid (HCl 37%), and nitric acid (HNO3 65%) (Scharlau, Spain, Extra Pure). Distilled water was also used during sample preparation and dilution. Classical Digestion-Heater (Gerhardt) with digestive Kjeldal's flask was used during sample digestion. Inductively coupled plasma - optical emission spectrometer (ICP-OES, Spectro Arcos FHS22, GmbH, Kleve, Germany) was used during metal analysis.

Sample collection

Plastic and aluminum pot containers were commonly used for the storage of local yogurt products by local cattle herder in Erbil city and its suburbs, Kurdistan Region (Iraq). For this study, new samples of yogurts aluminum containers (ALC) and plastic containers (PCs) were purchased from various local markets. The PCs samples were seven different colors: white, off-white, red, orange, green, pink, and dark green. In a worse case, there were no labels and publication of a prior information notice on the local yogurts containers.

Sample preparation and digestion

Before the digestion process, all glassware was cleaned, washed well with nitric acid, and distilled water to prevent contamination. Strong oxidizing acids were used to digest plastic samples as previously published [40-42]. A weight of 1.00 g plastic sample was placed into a conical digestion flask containing 15.0 mL of a mixture of concentrated nitric acid (HNO₃ 65%), sulfuric acid (H₂SO₄ 97%), and perchloric acid (HClO₄ 70%) with a volume ratio (1:1:1). The digestion was performed with digestive Kjeldahl flask using a classic digestion-heater under a hood until the mixture becomes clear. The digested solution was then allowed to cool, transferred, and diluted to 25.0 mL in a volumetric flask using distilled water.

The proposed method by Norris [43] was easily applied to digest aluminum samples at room temperature. 0.5 g of aluminum sample was added to digestive Kjeldahl flask with 12.0 mL of $H₂O$: HCl (1:1) with water being added first, and the HCl (37%) then added slowly. The sample digestion was continuing to complete without heating support until the mixture became clear. The digested solution was then allowed to cool, and the volume was made up to 25.0 mL in a volumetric flask using distilled water. The above steps were individually repeated three times for each type of sample and a blank solution. A blank solution that contains only the digested acids or the used reagents used to digest samples were individually prepared for each of the analyzed samples. The blank solution is mainly used for calibration purposes or zeroed the absorbance of all the other presented components in the sample solution except the interest component.

Metal analysis

Inductively coupled plasma optical emission spectrometer (ICP-OES) was utilized to investigate the contents of nine heavy metals (HMs) such as Cd, Co, Cr, Cu, Fe, Pb, Mn, Ni, and Zn in yogurts container. Optimum operating conditions for the instrument were well selected and utilized because all operating parameters are software controlled. Detailed information on the fundamental features of the tool, appropriate operating condition (Table 1), and selected wavelengths (lines) with a limit of detection (LOD) for the selected metals during the analysis (Table 2) were applied according to the instruments manufacture guideline [44]. The instrument was also calibrated against multi-element standards. A limit of detection (LOD) for each of the analyzed metal is calculated Eq. (1); the method's precision and accuracy were studied in excellent agreement for the whole of the analyzed elements. The standard reference material (NIST, SRM 1640a, trace elements in natural waters) was used based on the instruments manufacture instruction [44].

$$
LOD = \frac{3RSDb \cdot c}{SBR} \tag{1}
$$

LOD, RSDb, SBR, and c denote detection limit, a relative standard deviation of ten replicates of the blank, signal to background ratio, and the standard's concentration.

Hawraz Sami Khalid *et al.*

Table 1**.** Shows ICP operating conditions.

Table 2. LOD of the selected lines (nm).

The estimated contents of the selected HMs were measured in $\mu g/L$ (parts per billion) in whole the digested solutions and then converted to mg/kg (parts per million) wet weight for analyzed samples using the dilution factor equation Eq. (2) as follows:

The estimated contents $(mg/kg) = \frac{\mu g/L \times 25.0 \text{ mL}}{ \arctan a \cdot \text{mass of a sample (g)} \times 1000}$ (2)

Permissible limits in packaging materials and packaging composition set by The EU Directive 94/62/EC [37], and the State of California Health and Safety Code [38, 39] were employed in comparisons with detected HMs in container samples of the present study.

Statistical analysis

Microsoft Excel 2016 and GraphPad Prism 8 software programs were used to construct tables and compare the obtained results. Statistical analysis was employed using Student's t-test, and the significance level was set to 0.05. Results data were calculated and presented as average with standard deviation (SD) and below the method detection limit or not detectable (BDL) of triplicate measurements.

RESULTS AND DISCUSSION

In the present study, both yogurts plastic container (PC) and aluminum container (ALC) samples were collected from local markets at Erbil City and analyzed for targeted heavy metals (HMs) including Cd, Co, Cr, Cu, Fe, Pb, Mn, Ni, and Zn. The HMs contents in the whole yogurt container samples were analyzed by ICP–OES after acid digestion. The summarized results of HMs concentrations, the average values of triplicate samples, standard deviation (SD), and level of total detected metals load (TDML) in the examined samples are shown in Table 3. The TDML in the ALC samples was very high and equal to 627.36 mg/kg, while the load in an overall PC was 23.13

Bull. Chem. Soc. Ethiop. **2024**, 38(6)

1512

mg/kg. Detailed loads (TDML) in each of the ALC and PC components represent the sum contents calculated for all detected HMs (Figure 1 and Table 3).

Metal concentration in plastic container (PC)

During this investigation, seven different colors of the available yogurt PCs, such as white, offwhite, red, orange, green, pink, and dark green color, were investigated for the presence of the selected nine HMs levels. These colors of the PCs have been mainly used for the storage of local yogurts products.

The contents of the recorded HMs varied in the range; Cd (DBL - 0.37 mg/kg), Co (0.22 – 0.79 mg/kg), Cr (BDL – 5.46 mg/kg), Cu (BDL – 2.92 mg/kg), Fe (1.34 – 26.87 mg/kg), Mn (0.03 – 2.56 mg/kg), Ni (0.06 – 0.31mg/kg), Pb (BDL – 8.93 mg/kg), and Zn (1.75 – 13.56 mg/kg). In this study, the order of the average \pm SD for targeted HMs contents of overall examined PCs were as follows; 10.98 ± 7.99 mg/kg for Fe > 6.47 ± 4.10 mg/kg for $Zn > 1.83\pm3.21$ mg/kg for Pb > 1.66 ± 1.98 mg/kg for Cr $> 0.80\pm1.08$ mg/kg for Cu $> 0.65\pm0.87$ mg/kg for Mn $> 0.47\pm0.19$ mg/kg for $Co > 0.14 \pm 0.10$ mg/kg for $Ni > 0.13 \pm 0.15$ mg/kg for Cd (Table 3).

The obtained results indicated that the contents of Fe in 5 out of 7 of the colorful PCs samples were higher than the contents of the other targeted HMs. The results obtained also confirmed that Co, Fe, Mn, Ni, and Zn metals were detected in the whole of the examined PC colors. However, Cd, Pb, and Cr toxic HMs were detected in 4, 5, and 6 out of 7 of the examined colorful PC samples, respectively. In terms of the TDML, the order of HM contents in the PCs with different colors were as dark green (51.26 mg/kg) > orange (30.28 mg/kg) > green (29.03 mg/kg) > red (16.82 mg/kg) > pink (16.63 mg/kg) > off-white (14.43 mg/kg) > white (3.45 mg/kg) . Statistical data (One-Sample t-test) confirmed that the recorded TDML level in white PC was significantly (p-value = 0.005) lower than that level in the whole other examined PC colors. The obtained results also indicated that dark or multi-colors containers contained higher levels of HMs than light colors. For the presence of high HM levels in dark colors plastic, the values recorded in this study were in a good agreement with the previous work of Alam; Yang [35], who investigated HMs contents in various types of plastic bags. In this study, toxic HMs, including Cd, Pb, Cr, and Cu, were not detected in the white PC sample. The obtained results confirmed that the light plastic color (white color) could be seen as a safer yogurt PC in terms of HMs.

Metal concentration in aluminum container (ALC)

The results for the targeted HMs contents in the ALC sample are shown in Table 3 and Figure 2. The obtained results indicated that all of the targeted HMs were detected in the ALC sample. The contents (average±SD) of the selected HMs in the examined overall ALC sample were recorded and ordered as follows; 393.05±19.92 mg/kg for Pb > 187.62±4.75 mg/kg for Fe > 21.47±1.45 mg/kg for Cd > 9.86±0.21 mg/kg for Ni > 7.79±1.75 mg/kg for Zn > 5.58±0.05 mg/kg for Mn > 1.15±0.39 mg/kg for Cr > 0.42±0.04 mg/kg for Co > 0.41±0.19 mg/kg for Cu. Results also indicated that lead, iron, and cadmium recorded the highest average metal contents in the ALC sample. Statistical data (paired t-test) showed that there is no significant difference (p-value $=$ 0.172) between the recorded contents of the targeted HMs among overall ALC and PC samples (Table 3). However, the obtained results revealed that the average contents of Pb, Fe, Cd, Ni, and Mn metals in the overall ALC sample were noticeably higher than their corresponding average levels in the overall PC sample (Table 3). Thus, the recorded level for TDML in the overall ALC sample (627.36 mg/kg) was approximately 27 times more than that level in the overall PC sample (23.13 mg/kg).

Hawraz Sami Khalid *et al.*

Types of analysed container

Figure 1. Levels of TDML in ALC and PC samples.

Detected metals in aluminum container

Figure 2. Contents of HMs in ALC sample.

The recorded quantities of the targeted HMs in the samples were compared with permitted values in packaging materials regulated by the European Council [37] and the State of California Health and Safety Code [38, 39]. Based on these standards regulation, the sum of the detected contents of metals such as Cd, Cr (VI), Pb, and Hg present in packaging or packaging components shall not exceed 100 mg/kg by weight. The highest contents (average \pm SD) of Pb, Cd, and Cr in this study were 8.93 ± 1.92 , 0.37 ± 0.04 , and 5.46 ± 0.83 in mg/kg unit, respectively in examined PC samples; and 393.05 ± 19.92 , 21.47 ± 1.45 , and 1.15 ± 0.39 in mg/kg unit, respectively in ALC samples. The obtained results indicated that the sum of content levels of the detected HMs present in components of the PC samples did not reached the world's permissible limits. However, the sum of content level of the mentioned toxic HMs present in the composition of the examined ALC sample was noticeably reported to exceed the maximum permissible limit (100 mg/kg) regulated by the European Council [37] and State of California Health and Safety Code [38, 39].

Types of	Data	The concentrations of some heavy metals (mg/kg)										
container		Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	(mg/kg)	
White PC	Average	BDL	0.27	BDL	BDL	1.34	0.03	0.06	BDL	1.75	3.45	
	SD	BDL	0.03	BDL	BDL	0.51	0.01	0.02	BDL	0.07		
Off-white PC	Average	BDL	0.58	0.09	0.20	7.85	0.24	0.12	0.13	5.23	14.43	
	SD	BDL	0.04	0.04	0.01	0.45	0.01	0.06	0.17	0.46		
Pink PC	Average	BDL	0.79	0.11	0.37	8.98	0.33	0.07	BDL	6.00	16.63	
	SD	BDL	0.05	0.07	0.05	0.11	0.03	0.04	BDL	0.60		
Red PC	Average	0.37	0.42	1.28	0.21	9.45	0.40	0.06	0.44	4.19	16.82	
	SD	0.04	0.08	0.20	0.09	1.82	0.10	0.05	0.07	0.74		
Green PC	Average	0.20	0.22	5.46	1.64	7.90	0.23	0.31	8.93	4.15	29.03	
	SD	0.05	0.04	0.83	0.15	0.43	0.02	0.15	1.92	2.30		
Orange PC	Average	0.07	0.56	1.91	0.28	14.44	0.77	0.13	1.68	10.44	30.28	
	SD	0.01	0.03	0.23	0.02	1.50	0.08	0.02	0.80	1.69		
Dark green PC	Average	0.27	0.46	2.76	2.92	26.87	2.56	0.26	1.60	13.56	51.26	
	SD	0.06	0.06	0.23	0.35	3.64	0.39	0.16	0.67	1.66		
Overall PC	Average	0.13	0.47	1.66	0.80	10.98	0.65	0.14	1.83	6.47	23.13	
	SD	0.15	0.19	1.98	1.08	7.99	0.87	0.10	3.21	4.10		
Overall ALC	Average	21.47	0.42	1.15	0.41	187.62	5.58	9.86	393.05	7.79		
	SD	1.45	0.04	0.39	0.19	4.75	0.05	0.21	19.92	1.75	627.36	

Table 3. The concentrations of the nine targeted heavy metals in local yogurts plastic and aluminum containers.

BDL: below the method detection limit, SD: standard deviation, PC: plastic container, ALC: aluminum container, TDML: total detected metals load.

The detected average for HMs contents in the examined yogurts container was compared with that previously reported data in the other different containers (packaging or packaging components) samples and are shown in Table 4. According to results in the present study, the detected average for Cd quantity in the investigated PC sample was lower than that previously reported quantity from green and black low-density polyethylene bags [45], polyethylene, highdensity polyethylene, low-density polyethylene, and polyvinyl chloride bags [35], recycled polyethylene terephthalate food packaging [32], and recycled polyethylene terephthalate foodproduct containers [17]. However, the detected average for Cd content in the PC sample was higher than that previously stated from colored plastic tables [33] and polyethylene terephthalate packing of bottled water [34]. Results confirmed that the detected average for Pb quantity in the PC sample was lower than that previously described quantity (Table 4) from green and black lowdensity polyethylene bags [45], polyethylene, high-density polyethylene, low-density polyethylene, and polyvinyl chloride bags [35], and polyethylene terephthalate packing of bottled water [34]. However, results verify a good agreement between the recorded average of Pb content in the PC sample with that content previously examined and reported from colored plastic tables in Iraq [33].

In terms of the differences in Cd content in packing materials, the results showed that level content of Cd in the studied ALC sample was higher than previously reported level contents (Table 4) from stainless steel cup, pot, and bowl containers [26]. Note that the average Pb content in the ALC sample was reported to be 393.05 mg/kg, and close to the Pb content (364.00 mg/kg) in the investigated aluminum cookware samples in developing countries [16]. The recorded Pb content in the examined ALC sample was higher than that previously reported levels content from aluminum foil [25], stainless steel cup, pot, and bowl containers [26] and Syria and China aluminum cookware [23]. However, detected results on the alloying of aluminum cookware collected from Saudi and India showed Pb contents of 700.00 mg/kg and 510.00 mg/kg, respectively [23]. Furthermore, the recently recorded data verified that lead content in 26% (10

out of 42) of the collected aluminum cookware items from different developing countries was above 1000 mg/kg ($> 0.100 \%$ per weight) [24].

Table 4. HMs concentrations (mg/kg) in digests of previously investigated containers compared to detected results in the present study.

Samples		The concentration of some heavy metals (mg/kg)									References	
location	Types of Container	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn		
Erbil, Iraq	Plastic yogurt container (PC)	0.13	0.47	1.66	0.80	10.98	0.65	0.14	1.83	6.47	This study	
Baghdad, Iraq	Colored plastic table	BDL	NT	0.97	NT	NT	NT	NT	1.61	NT	$[33]$	
California, USA & China	RPET food-product containers	10.92	NT	8.18	NT	NT	NT	11.59	0.15	NT	$[17]$	
USA	RPET food packaging 11.09			8.36	NT	NT	NT	11.77	0.17	NT	$[32]$	
Shanghai, China	PE plastic bags	3.50	NT	18.58	23.53	NT	8.03	NT	13.38	45.47		
	HDPE plastic bags	10.52	NT	20.88	99.01	NT	6.82	NT	16.17	98.42	$[35]$	
	LDPE plastic bags	7.33	NT	9.69	28.62	NT	6.11	NT	6.29	22.50		
	PVC plastic bags	102.1 1	NT	4.76	17.01	NT	3.81	NT	4.90	31.02		
Kampala, Uganda	Black LDPE bags			88.00 25.00 35.00	NT	NT	NT	NT	1333.0	NT	[45]	
	Green LDPE bags	112.0 $\boldsymbol{0}$		30.00 45.00	NT	NT	NT	NT	1725.0	NT		
Sarajevo, Bosnia	PET packing of bottled water	0.01	0.42	NT	NT	0.721	NT	NT	3.29	0.10	$[34]$	
Erbil, Iraq	Aluminum yogurt container (ALC)	21.47 0.42		1.15	0.41	187.62	5.58		9.86 393.05	7.79	This study	
Owerri, Nigeria	Aluminum foil	NT	NT	0.08	NT	5.39	0.50	0.59	BDL	1.15	$[25]$	
Kumba, Cameroon	Aluminum cookware	NT	NT	NT	NT	NT	NT	NT	364.0	NT	[16]	
Shinjuku, Japan	Stainless steel cup	BDL	$\overline{\text{NT}}$	15.27 $\frac{0}{0}$	$\overline{\text{NT}}$	73.48 $\%$	10.19 $\frac{0}{0}$	0.82 $\%$	BDL	NT		
	Stainless steel pot	BDL	NT	13.45 $\%$	NT	72.19 $\%$	10.41 $\%$	1.04 $\frac{0}{0}$	BDL	NT	$[26]$	
	Stainless steel bowl	BDL	NT	12.06 $\frac{0}{0}$	NT	72.75 $\%$	8.88%	0.21 $\frac{0}{0}$	BDL	NT		
Riyadh, Saudi Arabia	China aluminum cookware	NT	NT			300.00 690.0 5220.0 3510.0		NT	300.0	1310. $\mathbf{0}$		
	India aluminum cookware	NT	NT			210.00 3520 2060.0 800.0		NT	510.0	1400. $\mathbf{0}$	$[23]$	
	Saudi aluminum cookware	NT	NT	410.00 3800			1030.0 1900.0	NT	700.0	380.0		
	Syria aluminum cookware	NT	NT			610.00 1470 2230.0 2300.0		NT	150.0	770.0		

PE: polyethylene, HDPE: high-density polyethylene, LDPE: low-density polyethylene, PVC: polyvinyl chloride, PET: polyethylene terephthalate, RPET: recycled polyethylene terephthalate, NT: not tested, BDL: below the methods detection limit, %: detected contents as per weight percent.

According to the obtained results, Pb (393.05 mg/kg) and Cd (21.47 mg/kg) contents were detected with a high amount in the ALC composition and exceeded the permissible world limit (100.00 mg/kg by weight for the sum of content levels of HMs). Low pH of yogurt can enhance the leaching process of metals from the ALC composition. It is also consistent with several researchers findings that the acidity of foods can enhance the leaching process of HMs from the household utensils [19, 20, 24]. Thus, the yogurts ALC may be a potential source of HMs

contamination of yogurt products due to the leaching process. Based on the results, it is not suggested to use aluminum containers to prepare and store local yogurt products, even for a short period. Additionally, findings suggest that using white plastic containers (PC) to store local yogurt is preferable and safer than aluminum container (ALC) items. Further study prioritized to assess the level of each toxic HMs that would leach from the containers to contaminate yogurt products during normal yogurts processing, preparation, and storage. Based on previous works result, foodstuffs and simulated food methods were recommended to assess of HMs leachability level from local ALC and PC containers, respectively [46, 47].

CONCLUSION

The results obtained in this study reveal that all the local yogurt container items investigated in Erbil city included several toxic HMs with varying contents and, in some cases, at levels above the world permissible limits. The results also showed that aluminum containers contaminated by Pb and Cd with a high amount, which can be easily released in an acidic medium and transferred to yogurt by the leaching process. Aluminum containers are not preferred to use in the preparation and storage of yogurt and can cause a risk to the consumer's health. This study has also indicated that the frequent use of the white plastic container in the preparation and storage of local yogurt products is preferable and safer than using aluminum container items in terms of HMs.

REFERENCES

- 1. El-Amier, Y.; Haroun, S.; Mowafy, A.; Abd-Elwahed, M. Environmental risk assessment and chemical contamination with heavy metals in the sediments of three drains, south of Manzala Lake (Egypt)*. Bull. Chem. Soc. Ethiop.* **2022**, 36, 315-327.
- 2. Fergusson, J.E. *The Heavy Elements: Chemistry, Environmental Impact and Health Effects*. Pergamon Press: Maxwell House, Fairview Park, Elmsford; **1990**.
- 3. He, Z.L.; Yang, X.E.; Stoffella, P.J. Trace elements in agroecosystems and impacts on the environment*. J. Trace Elem. Med. Biol.* **2005**, 19, 125-140.
- 4. Mohammed, A.; Orosun, M.; Okeola, F.; Raji, M.; Tesi, G.; Yusuph, O. Risk assessment of heavy metals and polycyclic aromatic hydrocarbons in ground water samples around the vicinity of an asphalt plant in north central, Nigeria*. Bull. Chem. Soc. Ethiop.* **2023**, 37, 1337- 1349.
- 5. Ibrahim, B.M.; Fakhre, N.A.; Jalhoom, M.G.; Qader, I.N.; Shareef, H.Y.; Jalal, A.F. Removal of lead ions from aqueous solutions by modified cellulose*. Environ. Technol.* **2022**, 45, 2335- 2347.
- 6. Sharef, H.Y.; Jalal, A.F.; Ibrahim, B.M.; Fakhre, N.A.; Qader, I.N. New ion-imprinted polymer for selective removal of Cu2+ ion in aqueous solution using extracted *Aloe vera* leaves as a monomer*. Int. J. Biol. Macromol.*, **2023**, 239, 124318.
- 7. Tchounwou, P.B.; Yedjou, C.G.; Patlolla, A.K.; Sutton, D.J. Heavy metal toxicity and the environment. *EXS* **2012**, 101, 133-164.
- 8. Dorne, J.; Kass, G.; Bordajandi, L.R.; Amzal, B.; Bertelsen, U.; Castoldi, A.F.; Heppner, C.; Eskola, M.; Fabiansson, S.; Ferrari, P. Human risk assessment of heavy metals: principles and applications*. Metal Ions Life Sci*. **2010**, 8, 27-60.
- 9. Jalal, A.F.; Khalid, H.S.; Yasin, I.T.; Xanamir, M.R. Estimation of some metals in children's colorful modeling clay sold in the markets of Erbil City, Kurdistan Region, Iraq*. Zanco J. Pure Appl. Sci.* **2020**, 32, 134-145.
- 10. Khalid, H.S.; Jalal, A.F.; Khdr, K.W.; Ahmad, M.O. Determination of some heavy metals in environment of bakery and samoon furnaces at Erbil City, Kurdistan Region, Iraq*. Zanco J. Pure Appl. Sci.* **2020**, 32, 176-186.

Hawraz Sami Khalid *et al.*

- 11. Krishna, Y.S.; Sandhya, G.; Babu, R.R. Removal of heavy metals Pb(II), Cd(II) and Cu(II) from waste waters using synthesized chromium doped nickel oxide nano particles*. Bull. Chem. Soc. Ethiop.* **2018**, 32, 225-238.
- 12. Mousavian, N.A.; Mansouri, N.; Nezhadkurki, F. Estimation of heavy metal exposure in workplace and health risk exposure assessment in steel industries in Iran*. Measurement* **2017**, 102, 286-290.
- 13. European Directorate for the Quality of Medicines & Health Care (EDQM). *Metals and Alloys Used in Food Contact Materials and Articles: A Practical Guide for Manufacturers and Regulators*; **2013**.
- 14. Sood, S.; Sharma, C. Levels of selected heavy metals in food packaging papers and paperboards used in India*. J. Environ. Prot.* **2019**, 10, 360-368.
- 15. Turner, A. Heavy metals in the glass and enamels of consumer container bottles*. Environ. Sci. Technol.* **2019**, 53, 8398-8404.
- 16. Weidenhamer, J.D.; Kobunski, P.A.; Kuepouo, G.; Corbin, R.W.; Gottesfeld, P. Lead exposure from aluminum cookware in Cameroon*. Sci. Total Environ.* **2014**, 496, 339-347.
- 17. Whitt, M.; Vorst, K.; Brown, W.; Baker, S.; Gorman, L. Survey of heavy metal contamination in recycled polyethylene terephthalate used for food packaging*. J. Plast. Film Sheeting.* **2013**, 29, 163-173.
- 18. Mania, M.; Szynal, T.; Rebeniak, M.; Postupolski, J. Exposure assessment to lead, cadmium, zinc and copper released from ceramic and glass wares intended to come into contact with food*. Rocz. Panstw. Zakl. Hig.* **2018**, 69, 405-411.
- 19. Dong, Z.; Lu, L.; Liu, Z.; Tang, Y.; Wang, J. Migration of toxic metals from ceramic food packaging materials into acid food simulants*. Math. Probl. Eng.*, **2014**, 2014.
- 20. Semwal, A.D.; Padmashree, A.; Khan, M.A.; Sharma, G.K.; Bawa, A.S. Leaching of aluminium from utensils during cooking of food*. J. Sci. Food Agric.* **2006**, 86, 2425-2430.
- 21. Zhou, L.; Rui, H.; Wang, Z.; Wu, F.; Fang, J.; Li, K.; Liu, X. Migration law of lead and cadmium from Chinese pots during the cooking process*. Int. J. Food Prop.* **2017**, 20, S3301- S3310.
- 22. Rittirong, A.; Saenboonruang, K. Quantification of aluminum and heavy metal contents in cooked rice samples from Thailand markets using inductively coupled plasma mass spectrometry (ICP-MS) and potential health risk assessment*. Emir. J. Food Agric.* **2018,** 372- 380.
- 23. Al Juhaiman, L.A. Estimating aluminum leaching from aluminum cookware in different vegetable extracts*. Int. J. Electrochem. Sci.* **2012**, 7, 7283-7294.
- 24. Weidenhamer, J.D.; Fitzpatrick, M.P.; Biro, A.M.; Kobunski, P.A.; Hudson, M.R.; Corbin, R. W.; Gottesfeld, P. Metal exposures from aluminum cookware: an unrecognized public health risk in developing countries*. Sci. Total Environ.* **2017**, 579, 805-813.
- 25. Duru, C.E.; Duru, I.A. Mobility of aluminum and mineral elements between aluminum foil and bean cake (Moimoi) mediated by pH and salinity during cooking*. SN Appl. Sci.* **2020**, 2, 348.
- 26. Shiozawa, Y.; Haneishi, N.; Suzuki, K.; Ogimoto, M.; Takanashi, M.; Tomioka, N.; Uematsu, Y.; Monma, K. Survey on metals contained in stainless steel kitchenware and tableware*. Shokuhin Eiseigaku Zasshi, J. Food Hyg. Soc. Jpn.* **2017**, 58, 166-171.
- 27. Sinha, P.K.; Mandal, S.; Kundu, D. Leaching of lead and cadmium from glass dinnerware*. J. Environ. Health Sci. Eng.* **2007**, 49, 58-61.
- 28. Valadez-Vega, C.; Zúñiga-Pérez, C.; Quintanar-Gómez, S.; Morales-González, J.A.; Madrigal-Santillán, E.; Villagómez-Ibarra, J.R.; Sumaya-Martínez, M.T.; García-Paredes, J. D. Lead, cadmium and cobalt (Pb, Cd, and Co) leaching of glass-clay containers by pH effect of food*. Int. J. Mol. Sci.*,**2011**, 12, 2336-2350.

1518

- 29. Nsengimana, H.; Munyentwali, A.; Muhayimana, P.; Muhizi, T. Assessment of heavy metals leachability from traditional clay pots "inkono" and "ibibindi" used as food contact materials*. Rwanda J.* **2012**, 25, 52-65.
- 30. Idris, M.; Oyewale, A.; Lawal, A. Assessment of leaching of some heavy metals from domestic ceramic wares*. Niger. J. Chem. Res.* **2015**, 20, 29-38.
- 31. Elmas, G.M.; Çınar, G. Toxic metals in paper and paperboard food packagings*. BioResour.* **2018**, 13, 7560-7580.
- 32. Whitt, M.; Brown, W.; Danes, J.E.; Vorst, K.L. Migration of heavy metals from recycled polyethylene terephthalate during storage and microwave heating*. J. Plast. Film Sheeting.* **2016**, 32, 189-207.
- 33. Hussein, H.J. Investigation content of some heavy metals in plastic table dishes*. IJMRCP* **2017**, 9, 92-97.
- 34. Pehlić, E.; Šapčanin, A.; Nanić, H.; Ćehajić, A. *The Content of Heavy Metals in "PET" Bottles of Drinking Water and Its Electrical Conductivity*, in *International Conference "New Technologies, Development and Applications"*, June 14-16, 2018 – Sarajevo, Bosnia and Herzegovina; **2018**.
- 35. Alam, O.; Yang, L.; Yanchun, X. Determination of the selected heavy metal and metalloid contents in various types of plastic bags*. J. Environ. Health Sci. Eng*. **2019**, 17, 161-170.
- 36. Mourya, A.; Mazumdar, B.; Sinha, S.K. Determination and quantification of heavy metal ion by electrochemical method*. J. Environ. Chem. Eng.* **2019**, 7, 103459.
- 37. Hänsch, K.; Kinkel, K. European Parliament and Council Directive 94/62/EC of 20 December 1994 on packaging and packaging waste*. J. Environ. Law* **1995**, 7, 323-337.
- 38. DTSC. *Toxics in Packaging Expiration of Exemptions January 1, 2010, Fact Sheet: Information for Manufacturers and Suppliers*. **2009**; Available at: https://dtsc.ca.gov/toxicsin-packaging-expiration-of-exemptions-january-1-2010-fact-sheet/.
- 39. HSC. *Toxics in Packaging Prevention Act: Hazardous Waste Control*. **2009** ; Available at: https://leginfo.legislature.ca.gov/faces/codes_displayText.xhtml?lawCode=HSC&division=2 0.&title=&part=&chapter=6.5.&article=10.4.
- 40. Livingstone, K.; Leticia, B.; Emmanuel, O. Assessment of level of lead and cadmium in selected plastic toys imported from china on the Ghanaian market*. Chem. Mater. Res.* **2014**, 6, 62-68.
- 41. Lobo, H.; Bonilla, J.V. *Handbook of Plastics Analysis*. Vol. 68, Crc Press: Boca Raton, Florida; **2003**.
- 42. Sharef, H.Y.; Fakhre, N.A. Designing, characterization, and application of cross-linked chitosan possessing Schiff base and ion-imprinted network polymer for selective removal of Cu(II) from aqueous solution*. Desalin. Water Treat.* **2022**, 276, 160-174.
- 43. Norris, S.J.E. *CRM Solutions: How to Prepare Metal Alloy Samples for Analysis by ICP-OES or ICP-MS*. **2017**; Available at: https://www.armi.com/blog/how-to-prepare-metal-alloysamples-for-analysis-by-icp-oes-or-icp-ms.
- 44. AMETEK. *Spectro ICP Report; Analysis of Aqueous Solutions by ICP-OES with Radial Plasma Observation. Spectro Arcos, Spectro Analytical Instruments GmbH*. FHS22; Available at: https://extranet.spectro.com/-/media/31793ADA-B987-4D37-B597- 04AFB66C7C22.pdf.
- 45. Musoke, L.; Banadda, N.; Sempala, C.; Kigozi, J. The migration of chemical contaminants from polyethylene bags into food during cooking*. Open Food Sci. J.* **2015**, 9, 14-18.
- 46. Ojezele, O.J.; Ojezele, M.O.; Adeosun, A.M. Cooking utensils as probable source of heavy metal toxicity*. Middle East J. Sci. Res.* **2016**, 24, 2216-2220.
- 47. Khan, S.; Khan, A.R. Contamination of toxic heavy metal in locally made plastic food packaging containers*. Global J. Sci. Front. Res. B Chem.* **2015**, 15, 19-24.