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Investigation of Element Migration from Aluminum Cooking Pots Using ICP-MS

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Abstract: The present study examined the migration of elements from aluminum cooking pots to foods after the cooking process. This study investigated the impact of pot quality (manufacturer), pot type (traditional or pressure cooker), water supply (tap water/mineral water), food acidity, salt, spices, temperature, and cooking time on the migration of elements into cooked food. The cooking experiments were conducted to simulate the actual cooking conditions. Standard food simulant B, with 3% (*w*/*v*) acetic acid, was used in subsequent cooking trials to confirm the results. Three methods were employed to analyze the elements in the food: ICP-MS, EDS-SEM, and XPS. The cooking pots used in this investigation were examined using a Spectromaxx metal analyzer to characterize their chemical composition. The concentration of aluminum in cooked food samples increased significantly when using an aluminum pressure cooker. Food acidity, cooking duration, and the type of aluminum pot (traditional/pressure cookers) all affected the concentration of elements that migrated into the food. The aluminum level increased from 80.17 to 133.7 µg/g when tomato sauce was added to the food. Increasing the heating time resulted in an increased aluminum content (157.9 µg/g) in the cooked food. Aluminum pressure cookers exhibited the highest amount of aluminum migration into the food. Foods cooked in a pressure cooker made by manufacturer (3) contained the highest aluminum content (252.7 µg/g), which increased the risk of exceeding the daily intake limit of aluminum. The prepared food samples under all conditions showed a safe health profile for daily intake of all elements (Fe, As, Cd, and Pb), except for Al, which exceeded the daily intake limit when using pressure cookers for extended cooking times. The results of element migration into food simulants were consistent with those of food samples. The results confirmed that SEM-EDS and XPS techniques are not suitable for quantifying the elements that migrated into food samples due to their detection limits.

Keywords: aluminum cooking pots; pressure cooker; element migration; ICP-MS

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1. Introduction

Cooking is an essential daily food preparation process. The quality of the pots used for food preparation is a significant factor for ensuring safe and healthy meals. One of the most commonly used types of pots in kitchens is those made from aluminum. When these aluminum pots are subjected to high temperatures during cooking and heating, there is a possibility of chemical elements migrating from the pots to the food in contact with them. The chemical elements that may migrate from these pots include Al, Fe, Ni, Cr, P, K, Ca, Mg, Na, Cu, and Mn [1]. In lower quality products, more dangerous toxic elements such as Pb and Cd [2] may also migrate to the food. Low-quality aluminum cookware, especially those manufactured from metal scraps, have been reported to leach hazardous elements such as Ni, As, Cu, Cd, Pb, and Al into the cooked food [3].

Several studies have examined the migration of elements from aluminum [4–10]. Various factors, such as pot quality, the nature of the food, water type, cooking temperature and time, food acidity, salt, and spice content, have been reported to affect the migration of elements from cooking pots into food. Abderazak et al. [11] conducted a study on the migration of Fe and Al into faba beans from stainless steel and aluminum cooking pots. The results showed that aluminum migrated only into cooked faba beans, increasing Al content from 22.3 to 43.2 mg/kg during the second cooking cycle. Iron also migrated from both pot types, with Fe levels increasing from 53.5 (first cooking cycle) to 66.9 mg/kg (second cycle). Al Zubaidy et al. [5] examined the effect of acidity, salt content, and temperature on aluminum leaching from aluminum cooking pots during food cooking. They used the weight loss method to quantify the aluminum that migrated into food in different food solutions and types of water. Their findings indicated that aluminum leaching increased at low pH levels due to an increased corrosion rate. Leaching was lower when using drinking water compared to tap water due to a lower corrosion rate in the former case. Increasing salt content was found to increase corrosion up to a certain level.

Mohammad et al. [12] investigated aluminum leaching from cooking wares and reported a significant amount of Al leaching from the used pots. They concluded that the addition of citric acid and salt to the food increased aluminum leaching from the cooking pots. Jabeen et al. [13] studied the migration of Al from aluminum wares and foil, finding that the combination of citric acid and tomato juice resulted in the highest Al leaching in beef (292 mg/kg), while the maximum Al level in chicken (210 mg/kg) occurred with a combination of yogurt and lemon juice. Fermo et al. [14] reported on the diffusion of aluminum into meat and fish cooked using commercial aluminum foils, quantifying the results using ICP-MS. Zhou et al. [2] investigated the migration of Pb and Cd from Al, Fe, and ceramic cooking pots to cooked food, finding that the migration of these elements depended considerably on cooking temperature, time, pot materials, and food ingredients. Odularu et al. [15] examined the migration of Al into rice cooked in distilled water using different cooking pots. They found that new Al pots had lower leached Al compared to old Al pots. No significant changes were observed in the leached elements' concentration between new and old stainless-steel pots.

Campos et al. [16] analyzed chicken tissues before and after cooking in iron and aluminum pots, quantifying the content of Al, Fe, Cu, Ca, Cu, Mn, and Ni using microwave-induced plasma optical emission spectrometry and flame atomic absorption spectrometry. Ca, Fe, and Al were the major leached contents in the analyzed samples. Saxena et al. [17] concluded that Al cookware could have a detrimental effect on food quality due to element migration. They reported that Al cookware posed the highest risk among all types of cookware.

The disadvantages of using aluminum cookware for food preparation have been reported [17,18]. These include high levels of element leaching, susceptibility to interaction with acidic foods, migration of toxic elements like Cd and Pb to food, and high levels of Al migration, especially during prolonged cooking cycles. Health problems associated with increased element migration from Al cookware may include Alzheimer's disease, Parkinson's disease, carcinogenicity, chronic renal failure, myalgia, and neurological syndromes [12,15,19–23]. Lead is a toxic element that can have serious health consequences [24,25]. Arsenic can lead to severe health issues known as arsenicosis, causing cancer in the skin, lungs, and kidneys [26–28]. Cadmium is highly toxic to humans, causing kidney problems and chronic toxicity symptoms such as impaired organ function and hepatic dysfunction [21].

The present research aims to investigate the migration of chemical elements from aluminum cooking pots to foods. This study investigated the effect of pot material, pot quality (manufacturer), pot type, water source, food acidity, food salt and spice content, and heating temperature and time on the migration of chemical elements into cooked food. The cooking conditions were designed to simulate actual cooking conditions. Furthermore, additional cooking experiments were performed using standard food simulant B, with 3% (w/v) acetic acid, to validate the results. Moreover, multiple methods, including ICP-MS, SEM-EDS, and XPS, were applied to quantify the elements that migrated to cooked food to confirm the accuracy and reliability of the results and to evaluate the usefulness of these methods for the elemental analysis of the cooked foods.

2. Materials and Methods

2.1. Food Cooking Experiments

The aluminum cooking pots and ingredients used for the food preparation experiments were purchased from the Saudi local market. Aluminum cooking pots from four manufacturers were employed in this study, where one pot from each manufacturer was used to perform the experiments. These included an aluminum cooking pot from manufacturer (1) (code: AC), an aluminum cooking pot from manufacturer (2) (code: AS), an aluminum pressure cooker from manufacturer (3) (code: ACP), and an aluminum pressure cooker from manufacturer (4) (code: APP). These cooking pots were sourced from various suppliers in the Saudi local market to assess the impact of pot quality on element migration. More information about the pots used in the present study is displayed in Tables S1–S4 in the Supplementary Materials. Additionally, both traditional aluminum pots and aluminum pressure cookers were selected to investigate the influence of pressure cooking on element migration into food. The raw food ingredients used included basmati rice, beef meat, tomato sauce, black lemon, table salt, and spices (including cinnamon, cloves, cardamom, and black pepper). Table 1 presents the details of food ingredients and the cooking experiments conducted in this study, along with the pH values measured for the cooked food before and after cooking.

Table 1. The details of food ingredients and food cooking experiments performed using aluminum cooking pots.

Sample Code	Food Ingredients in Grams	Time of Cooking	Temperature of Food during Cooking	pH before Cooking	pH after Cooking (after Boiling)
AC-1	350 g rice + 500 g meat + 1.25 L tap water + 100 mL oil	2 h	Cooking experiment started at room temperature and then the temperature increased till boiling conditions at (100 °C).	8.83	8.33
AC-2	(AC-1) + 60 g tomato sauce + 6 g black lemon			6.48	6.20
AC-3	(AC-2) + 10 g salts + 10 g Kabsa spices + 3 g cinnamon + 3 pieces cloves + 3 pieces Cardamom + 2 g black pepper			6.72	6.17
AC-4	The same (AC-3) except using mineral water (Manhal) instead of tap water			6.80	6.72
AC-5	The same (AC-3) with applying heat for 8 h at 70 °C.	10 h	The same as in the above + heating at 70 °C for 8 h.	6.60	6.48
AS-5	The same (AC-5) but using manufacturer (2) aluminum pot (AS)			6.70	6.32
ACP-5	The same (AC-5) but using manufacturer (3) aluminum pressure pot (ACP)			6.53	6.30
APP-5	The same (AC-5) but using manufacturer (4) aluminum pressure pot (APP)			6.41	6.10

2.2. Food Simulant Preparation

Food simulants were prepared following the guidelines outlined in Regulation (EU) No 10/2011 [29]. Specifically, Food Simulant B, a 3% (*w*/*v*) acetic acid solution, was prepared using the four types of aluminum pots (AC, AS, ACP, and APP). The test numbers used to prepare food simulants are OM4 and OM5 [29]. The OM4 test represents a high-temperature application for all types of foods at temperatures up to 100 °C, with food simulants being subjected to 100 °C for 1 h using all types of pots employed in this study. The OM5 test, on the other hand, simulates high-temperature conditions for all types of foods up to 121 °C, with food simulants being subjected to 100 °C for 2 h using all the types of pots used in this study. The OM5 test is considered one of the worst-case scenarios applied to all food simulants. Table 2 provides details of the conditions for the food simulant tests (OM4 and OM5) applied to aluminum cooking pots, along with the pH values of the food simulants before and after the cooking experiments.

Table 2. The details of food simulant tests (OM4 and OM5) applied to aluminum cooking pots.

Sample Code	Food Constituents	Test Number	Temperature and Time of Cooking Test	pH before Cooking	pH after Cooking (after Boiling)
AC-FS	Food Simulant "B" "3% (*w*/*v*) acetic acid"	OM4	100 °C for 1 h	4.5	4.52
AS-FS					4.56
ACP-FS					4.71
APP-FS					4.64
AC-FS		OM5	100 °C for 2 h		4.76
AS-FS					4.78
ACP-FS					4.93
APP-FS					4.92

2.3. Chemical Composition Analysis of Cooking Pots

The chemical composition of the cooking pots used in this study was analyzed using an advanced benchtop spark-optical emission spectrometer (Spectromaxx metal analyzer, model Q4-Tasman, Bruker, Germany). Optical emission spectroscopy via arc and spark excitement is considered as one of the ideal methods for chemical composition analysis of metals and alloys. The calibration was carried out using a certified reference material (CRM) of a reference aluminum sample. The "Al-10-M" spectromaxx program was applied for system calibration which is frequently used when examining pure aluminum/low alloy aluminum. The cooking pots were carefully cut using an 'angle grinder' at both bottom and side positions to collect four samples from each pot for chemical analysis. The dimensions of each specimen are 3 × 3 cm. Three test replicates were performed for each specimen, resulting in a total of 12 tests for each pot. The average chemical analysis from these 12 replicates was considered for each cooking pot.

2.4. Characterization of the Elements Migrated to the Cooked Food and Food Simulants

The cooked food samples were digested using a microwave digestion system (Milestone, START D, Leutkirch im Allgau, Germany). Each sample from the cooked food, weighing between 210 mg and 260 mg, was digested in the START D microwave digestion system in a closed vessel. The heating program in the microwave digestion system consisted of three phases: (1) irradiation for 10 min at 800 W and 100 °C; (2) irradiation for 10 min at 800 W and 130 °C; and (3) irradiation for 15 min at 800 W and 180 °C. Finally, a 30 min period of forced ventilation was used to cool the system. The containers were then allowed to cool down, emptied, filtered, and quantitatively transferred into 100 mL volumetric plastic flasks. The volumes were adjusted to the mark with purified water. The digestion procedures were carried out in triplicate for each sample and blank, and the digested samples were subsequently analyzed using the ICP-MS technique. ASTM Type II purified water (0.055 µS/cm) was obtained using the Barnstead water purification system (Thermo Electron LED GmbH, Langenselbold, Germany). This purified water was used for the digestion of food samples and for diluting the standard solutions used for calibration. The food samples were decomposed using a mixture of 3 mL HNO_3 and 1 mL H_2O_2. The used nitric acid (HNO_3) is 70% concentration analytical reagent grade, while hydrogen peroxide (H_2O_2) is 30% w/v (100 vol) stabilized pure (the reagents were supplied from Panreac Applichem, Darmstadt, Germany). An ICP-MS stock tuning solution containing Ce, Co, Li, Tl, and Y (10 µg/L) was used for system setup and calibration. Multielement calibration standard solutions (10 µg/mL) containing the metals of interest (supplied by Agilent Technologies, Palo Alto, CA, USA) were used to construct calibration curves where the method was verified using spiked samples at three different concentrations in triplicate. Moreover, several measures were taken to ensure the validity and reliability of the obtained results such as using a dynamic reaction cell (DRC), which can reduce or eliminate the polyatomic interferences by introducing a collision or reaction gas into the cell. Elemental analysis and quantification were performed using the Agilent ICP-MS 7800 technique (Model G8421A, USA). Table 3 shows the operating conditions of ICP-MS technique in the present study.

Table 3. Operating conditions of the ICP-MS technique used in the present study.

ICP-MS Condition	Value
RF power (W)	1550
Sample Depth (mm)	8.0
Sample up-take (s)	30
Nebulizer type	Micro-Mist
Carrier gas (L/min)	1.05
Make up gas (L/min)	0.15
Plasma gas (L/min)	15
Nebulizer Pump (rps)	0.1
He gas (mL/min)	5
Energy Discrimination (V)	3
Points/peak	3
Repetitions	3
Integration time/mass (s)	0.3

The same procedures were applied to examine the raw materials used in preparing the food experiments before cooking to quantify the elemental concentration in each food ingredient including rice, meat, lemon, cloves, black pepper, cardamom, spices, cinnamon, and tomato sauce. Furthermore, the same procedures, except digestion, were applied to examine the food simulant samples using the ICP-MS technique.

For the elemental analysis of food samples, an SEM (Apreo Lovac FEGSEM, Thermo Fisher Scientific, Waltham, MA, USA) equipped with an EDS system (OCTANE PRO, USA) and XPS (Thermo Fisher Scientific, Waltham, MA, USA) were used. Approximately 20 g of food samples was collected from each food experiment. These samples were initially dried in the oven at 60 °C and left overnight to ensure complete drying. The dried food samples were placed in 50–100 mL porcelain crucibles and then transferred to the oven. After drying, the samples were removed from the furnace and subjected to a grinding process. The dried food samples were manually ground into fine powders using a mortar and pestle, with each sample being ground for 10 min to achieve fine powders of the dried food samples. These food powder samples were used for SEM-EDS and XPS elemental analysis.

3. Results and Discussion

3.1. Chemical Analysis of Aluminum Pots Used in the Present Study

The chemical composition of the cooking pots used in this study was analyzed using a Spectromaxx metal analyzer to confirm their chemical composition. The results for each pot represent the average obtained from 12 tests conducted on four specimens cut from various locations on each pot. The results of the chemical analysis, presented in Table 4, show the composition of the aluminum cooking pots in weight percent. It is evident that the primary element in the pots is aluminum (Al). However, the AC and ACP pots contain a lower amount of aluminum compared to the AS and APP pots.

Table 4. The average chemical composition of the aluminum cooking pots according to spectromaxx metal analysis.

Element	Chemical Composition, Weight Percent (wt.%)			
	Pot Codes			
	AC	AS	ACP	APP
Al	99.24	99.60	98.59	99.53
Fe	0.3782	0.2483	0.6036	0.2889
Ni	0.0029	0.0008	0.0010	0.0011
Mg	0.0030	<0.0005	<0.0005	<0.0005
P	0.0040	0.0037	0.0041	0.0039
Cr	0.0038	0.0016	0.0085	0.0025
Mn	0.0219	0.0012	0.6528	0.0144
Si	0.2207	0.0859	0.0782	0.0753
Ti	0.0267	0.0137	0.0155	0.0091
Cd	0.0006	0.0005	0.0006	0.0005
Pb	<0.0065	<0.0065	<0.0065	<0.0065
Sn	<0.0005	<0.0005	<0.0005	<0.0005
Sr	<0.0010	<0.0010	<0.0010	<0.0010
Zr	0.0002	0.0001	0.0004	0.0003
Cu	0.0045	<0.0010	<0.0010	<0.0010
Zn	0.0347	0.0032	0.0089	0.0149
V	0.0105	0.0089	0.0255	0.0143
Sb	0.0035	0.0037	0.0021	0.0017
B	0.0026	0.0019	0.0018	0.0016
Co	0.0008	<0.0005	0.0006	<0.0005
Ca	0.0007	0.0006	<0.0005	<0.0005

3.2. The Results of Elemental Analysis Using ICP-MS Technique for Cooked Food

The raw materials used in preparing the food experiments, including rice, meat, lemon, cloves, black pepper, cardamom, spices, cinnamon, and tomato sauce, were analyzed using the ICP-MS technique before cooking to quantify the elemental concentration in each food ingredient. Three replicates for each raw ingredient were performed, and the average of the elemental concentrations in the raw food ingredients is presented in Table 5. The total elemental concentration in the cooked food can be attributed to two sources: the first is the elemental content derived from the raw food ingredients (see Table 1 for the list of food ingredients), and the second source is the elements that migrated from the cooking pots.

The samples obtained from the cooked food in aluminum pots were analyzed using the ICP-MS technique, with six samples examined for each cooking experiment, and the averages are presented in Table 6. The results displayed in Table 6 depict the elements that migrated from the aluminum cooking pots to the cooked food, after subtracting the contribution of elements from the ingredients/raw materials based on the data in Table 5. Additionally, the elements analyzed included the major elements found in the aluminum cooking pots, namely Al and Fe, as determined by the results of the Spectromaxx metal analysis (as presented in Table 4). Furthermore, common toxic elements such as As, Cd, and Pb were analyzed to confirm their presence in the cooked food. Table 6 provides the results for the metals that migrated into the cooked food in aluminum pots under various cooking conditions. It is important to note that the total elemental concentration in food samples is the sum of the elemental concentrations from the food's raw ingredients and the elemental concentrations that migrated from the aluminum pots. Table 6 shows the concentration of elements migrated from cooking pots only after deducting the contribution of elements sourced from raw ingredients.

Table 5. The average elemental concentration in raw ingredients.

Raw Materials	Elements Concentration (μg/g)									
	Al		Fe		As		Cd		Pb	
	* AVE	* SD	AVE	SD	AVE	SD	AVE	SD	AVE	SD
Rice	15.20	1.017	10.94	0.5971	0.0892	0.0064	0.0313	0.0027	0.1261	0.0029
Dry Lemon	58.61	3.876	84.06	5.303	0.0262	0.0012	0.0137	0.0013	0.2101	0.0192
Clove	83.48	7.047	79.39	4.953	0.0173	0.0009	0.0062	0.0005	0.4919	0.0253
Black pepper	134.4	9.630	131.6	7.834	0.0436	0.0017	0.0169	0.0013	0.2819	0.0171
Cardamom	48.22	2.549	52.62	2.405	0.2691	0.0199	0.0716	0.0061	0.6583	0.0346
Kabsa Spices	110.2	5.696	192.2	8.893	0.0696	0.0053	0.0374	0.0002	0.3814	0.0235
Cinnamon	55.53	3.006	40.17	2.472	0.2284	0.0061	0.0543	0.0034	1.1688	0.0746
Meat	11.64	0.487	24.12	1.463	0.0071	0.0003	0.0092	0.0007	0.0360	0.0024
Tomato Sauce	16.10	0.5096	15.32	1.204	0.0150	0.0011	0.0203	0.0016	0.0158	0.0014
Table Salt	21.55	1.361	14.54	1.005	0.1330	0.0014	0.1663	0.0134	0.3698	0.0307

* AVE refers to an average of three replicates and SD indicates the standard deviation.

Table 6. The ICP-MS elemental analysis results of food prepared by aluminum pots.

Sample Code	Elements Concentration (μg/g)									
	Al		Fe		As		Cd		Pb	
	* AVE	* SD	AVE	SD	AVE	SD	AVE	SD	AVE	SD
AC-1	80.17	5.997	6.493	0.4941	0.0053	0.0007	0.0005	0.00004	0.0056	0.00084
AC-2	133.7	8.501	8.922	0.6759	0.0172	0.0014	0.0008	0.00009	0.0087	0.00092
AC-3	129.4	6.277	9.296	0.8298	0.0113	0.0011	0.0002	0.000019	0.0078	0.00089
AC-4	130.1	9.304	11.98	0.9419	0.0279	0.0026	0.0001	0.000012	0.0084	0.00091
AC-5	157.9	8.722	14.45	1.1573	0.0265	0.0012	0.0010	0.00013	0.0039	0.00049
AS-5	145.4	7.697	9.171	0.7209	0.0267	0.0031	0.0007	0.00011	0.0054	0.00078
ACP-5	252.7	7.793	17.22	1.3158	0.0292	0.0021	0.0002	0.00003	0.0034	0.00053
APP-5	221.0	6.085	9.401	0.3199	0.0182	0.0017	0.0009	0.00014	0.0025	0.00037

* AVE refers to an average of three replicates and SD indicates the standard deviation.

The same results as those presented in Table 6 are depicted in Figures 1 and 2. Figure 1a illustrates that the addition of tomato sauce and lemon (AC-2 sample) led to an increase in the content of Al and Fe that migrated to the food compared to the AC-1 sample. Notably, aluminum migration is more pronounced compared to the other elements; the pots used in the present study contain more than 99 wt.% (weight percent) aluminum, so the migration of aluminum was observed to be higher than the remaining minor elements in the cooking pots where the driving force for metal ion migration is strongly dependent on the metal ions' concentration gradients. The concentration of toxic elements (As, Cd, and Pb) in the cooked food appears to be relatively small, and any variations observed from one run to another can be disregarded due to the low concentrations of these elements.

Figure 1b reveals insignificant changes in the content of migrated elements to the food when spices were added (AC-3 sample) compared to the AC-2 sample. Figure 1c demonstrates that there is no significant change in the elemental concentration that migrated into the cooked food when cooking with tap water (AC-3) vs. mineral water (AC-4). Figure 1d shows a significant increase in the concentration of Al and Fe in the cooked food when the heating time is extended to 8 h at 70 °C (AC-5 sample). The results consistently indicate that aluminum is the primary element that migrates from aluminum cooking pots, while the remaining elements such as Fe migrate with lower concentrations in comparison to aluminum. The toxic elements (As, Cd, and Pb) are present in minor concentrations in the cooked food under all cooking conditions, remaining below the acceptable limits.

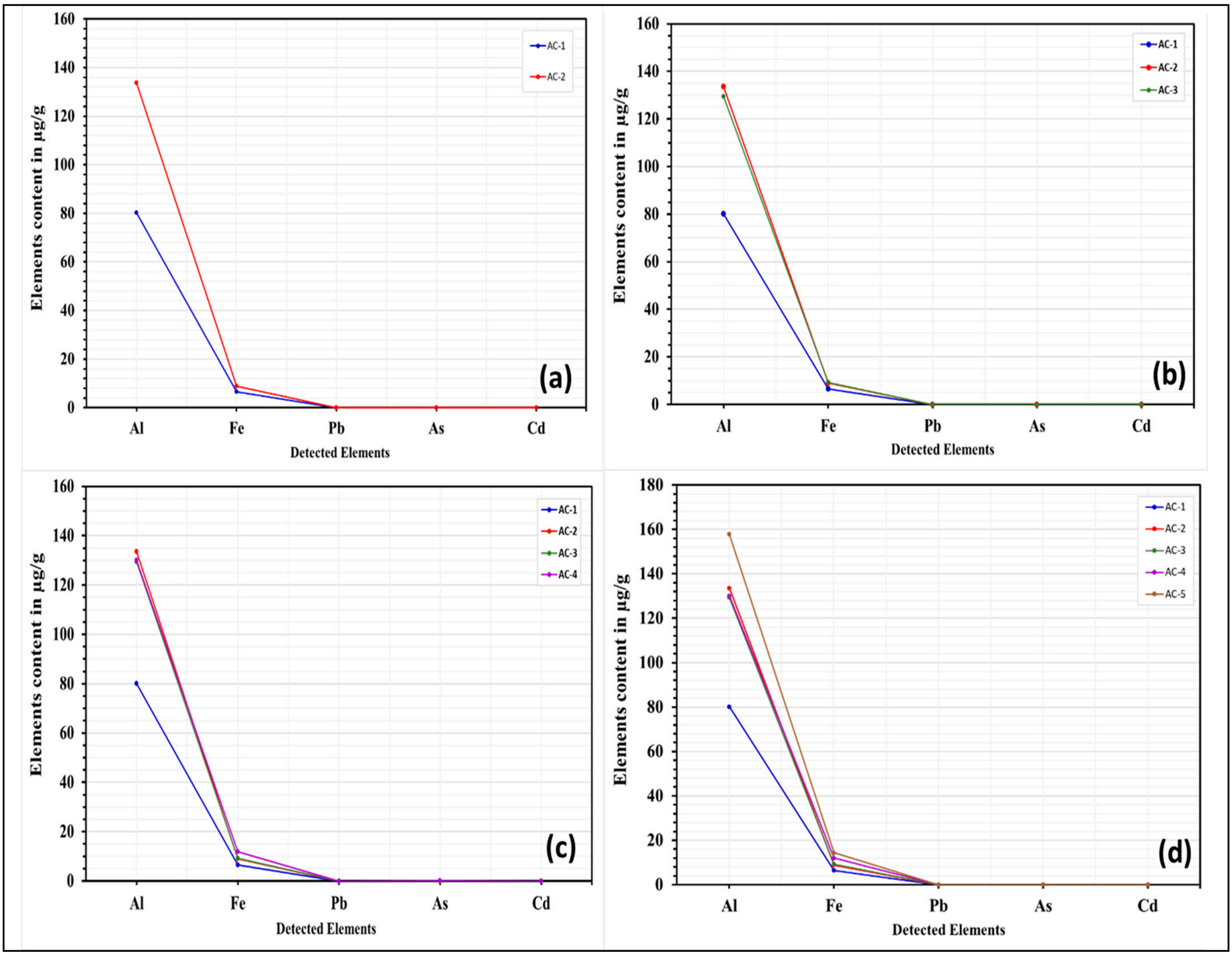

Figure 1. The elements migrated to the cooked food from manufacturer (1) aluminum cooking pot (AC) under various cooking conditions: (**a**) the effect of tomato sauce (AC2 vs. AC1); (**b**) the effect of added spices (AC3 vs. AC2); (**c**) the effect of mineral water (AC4 vs. AC3); and (**d**) the effect of long-time heating (8 h @ 70 °C) (AC5 vs. AC3).

Figure 2 illustrates the impact of different types of aluminum cooking pots on the concentration of elements that migrate into the food. A substantial increase in aluminum concentration in the cooked food is observed when using aluminum pressure cookers. The highest aluminum concentration (252.7 µg/g) was observed in the food cooked with the ACP pressure cooker (ACP-5 sample). Food acidity, heating time, and the type of aluminum pot (traditional or pressure cookers) played a significant role in the concentration of elements that migrated into the food. Based on the present results, special attention is needed for aluminum concentration due to its considerable variation under various processing variables. The aluminum content increased from 80.17 μg/g (AC-1 sample) to 133.7 μg/g when tomato sauce was added (AC-2 sample). Further increases in aluminum concentration (157.9 μg/g) in cooked food were observed with extended heating times, as seen in the AC-5 sample. AC and AS aluminum cooking pots showed a nearly similar effect on the concentration of elements that migrated into the food, as indicated by the results for AC-5 (157.9 μg/g) and AS-5 (145.4 μg/g), which are presented in Table 6 and Figure 1. The pressure cookers ACP and APP exhibited the highest amount of aluminum migration into the food, with the maximum aluminum concentration detected in the food prepared with the ACP pressure cooker, reaching 252.7 μg/g. The effect of mineral water (AC-4 sample) compared to tap water (AC-3 sample) appears to have an insignificant impact on the level of elements that migrate into the food. This is likely due to minor changes in the food's acidity (pH) in both cases. Additionally, the addition of salt and spices had a minor effect on the content of elements that migrated into the food (AC-3 vs. AC-2 samples). Since the addition of various spices to the food can have conflicting effects on element migration, the net impact appears to be neutral, as shown in the results presented in Table 1 and Figure 1.

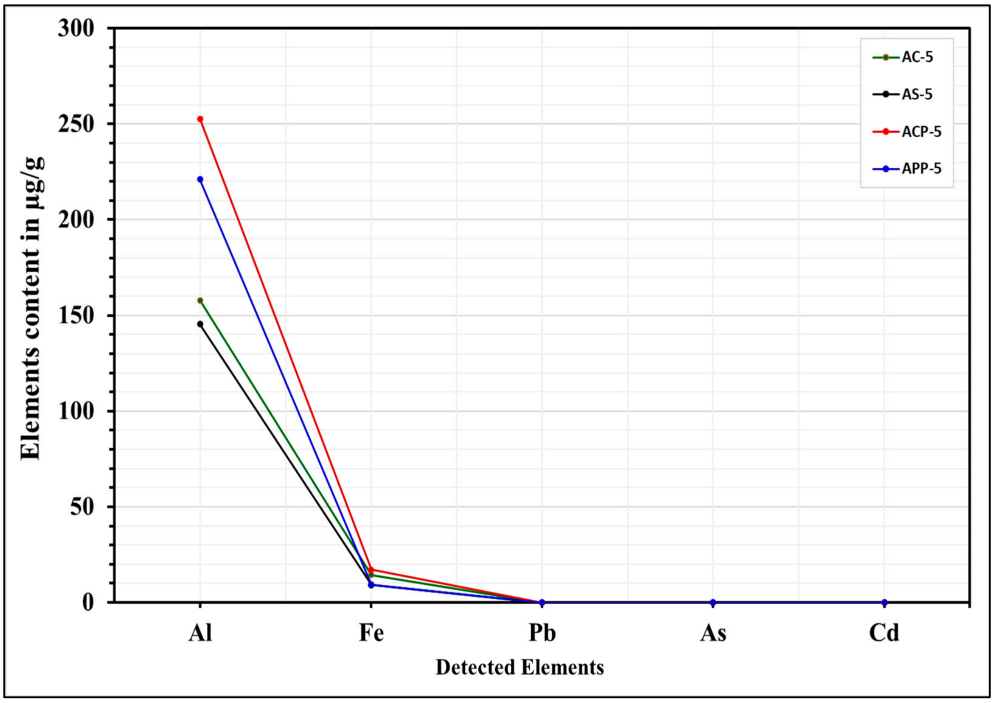

Figure 2. The elements migrated to the cooked food using different cooking pots: manufacturer (1) aluminum cooking pots (AC-5); manufacturer (2) aluminum cooking pots (AS-5); manufacturer (3) aluminum pressure cooker (ACP-5); and manufacturer (4) aluminum pressure cooker (APP-5). The experiments presented in this figure were carried out according to AC-5 cooking conditions.

3.3. The Results of Elemental Analysis Using ICP-MS Technique for Food Simulants

The elements that migrated to food simulants were analyzed using the ICP-MS technique. Food simulants were prepared following Regulation (EU) No 10/2011, where food simulant B, consisting of 3% (*w*/*v*) acetic acid, was prepared using the four types of aluminum pots. The tests applied to prepare food simulants are OM4 and OM5. OM4 represents the high-temperature application for all types of foods at temperatures up to 100 °C. Food simulants were subjected to 100 °C for 1 h for all types of aluminum pots used in this study. OM5 represents the high-temperature application for all types of foods, up to 121 °C, with food simulants subjected to 100 °C for 2 h for all types of aluminum pots used in this study. According to the standard, OM5 represents one of the worst-case conditions applied to all food simulants. The reference sample (R0) in Table 7 refers to the prepared food simulant as it is (water with 3% (*w*/*v*) acetic acid), without applying any cooking conditions. Table 7 presents the elements that migrated to food simulants from different types of aluminum pots under OM4 and OM5 test conditions. The values listed in Table 7 represent the averages of three trials conducted using the ICP-MS technique.

Table 7. The ICP-MS elemental analysis results of food simulants prepared by aluminum pots.

Test Number	Sample Code	Elements Concentration (μg/g)									
		Al		Fe		As		Cd		Pb	
		* AVE	* SD	AVE	SD	AVE	SD	AVE	SD	AVE	SD
Reference Sample	R0	0.0067	0.0005	0.0363	0.0025	0.0014	0.00008	0.00009	0.000011	0.0009	0.00010
OM4 Test	AC-FS	11.75	0.2930	0.9413	0.0317	0.0072	0.00060	0.00032	0.000028	0.0011	0.00009
	AS-FS	9.148	0.1637	0.4725	0.0259	0.0029	0.00030	0.00007	0.000010	0.0006	0.00006
	ACP-FS	61.76	1.6927	0.2143	0.0171	0.0102	0.00070	0.00005	0.000006	0.0005	0.00004
	APP-FS	35.05	0.9137	1.064	0.0586	0.0076	0.00040	0.00005	0.000005	0.0010	0.00008
OM5 Test	AC-FS	121.7	4.6288	5.397	0.3272	0.0110	0.00090	0.00007	0.000009	0.0013	0.00010
	AS-FS	117.6	2.0345	4.050	0.1794	0.0175	0.00172	0.00018	0.000024	0.0017	0.00009
	ACP-FS	249.6	3.3540	9.868	0.1886	0.0151	0.00103	0.00008	0.000011	0.0016	0.00021
	APP-FS	211.8	5.8550	5.843	0.3712	0.0149	0.00106	0.00011	0.000013	0.0021	0.00019

* AVE refers to an average of three replicates and SD indicates the standard deviation.

Figure 3 illustrates the concentration of aluminum that migrated to the food simulant when OM4 and OM5 tests were applied using different cooking pots (manufacturer (1) aluminum cooking pots (AC-FS); manufacturer (2) aluminum cooking pots (AS-FS); manufacturer (3) aluminum pressure cooker (ACP-FS); and manufacturer (4) aluminum pressure cooker (APP-FS)). The suffix "FS" refers to the food simulant. Figure 3 shows that the aluminum (Al) concentration in the food after applying the OM4 test was higher than that in the reference sample (R0) but lower than that after applying the OM5 test for all types of aluminum cooking pots. The OM5 test involved high temperatures at 100 °C for two hours, and the heating time was found to have a significant effect on the concentration of aluminum in the food simulant. The food prepared using the aluminum pressure cooker (ACP-5) showed the highest amount of aluminum that migrated to the food simulant (249.6 μg/g). Figure 4 displays the content of iron (Fe) that migrated to the food simulant when OM4 and OM5 tests were applied using different aluminum cooking pots. Fe migration significantly increased when applying the OM5 test compared to OM4 due to prolonged heating at 100 °C. The food simulant prepared with the ACP pressure cooker under OM5 test conditions revealed the highest Fe content (9.868 μg/g). The concentration of the migrated toxic elements, namely As, Cd, and Pb, in the food simulant when applying the OM4 and OM5 tests using different aluminum cooking pots is shown in Table 7. Overall, the concentration of the migrated toxic elements was observed to increase in the food simulant prepared with all types of aluminum cooking pots under prolonged heating conditions (OM5 test). The concentration of toxic elements is minor, and the changes observed after applying OM4 and OM5 tests can be disregarded. The greatest attention should be given to the migration of aluminum, which showed an increased content when applying the OM5 test, and aluminum migration was further increased when using the aluminum pressure cooker.

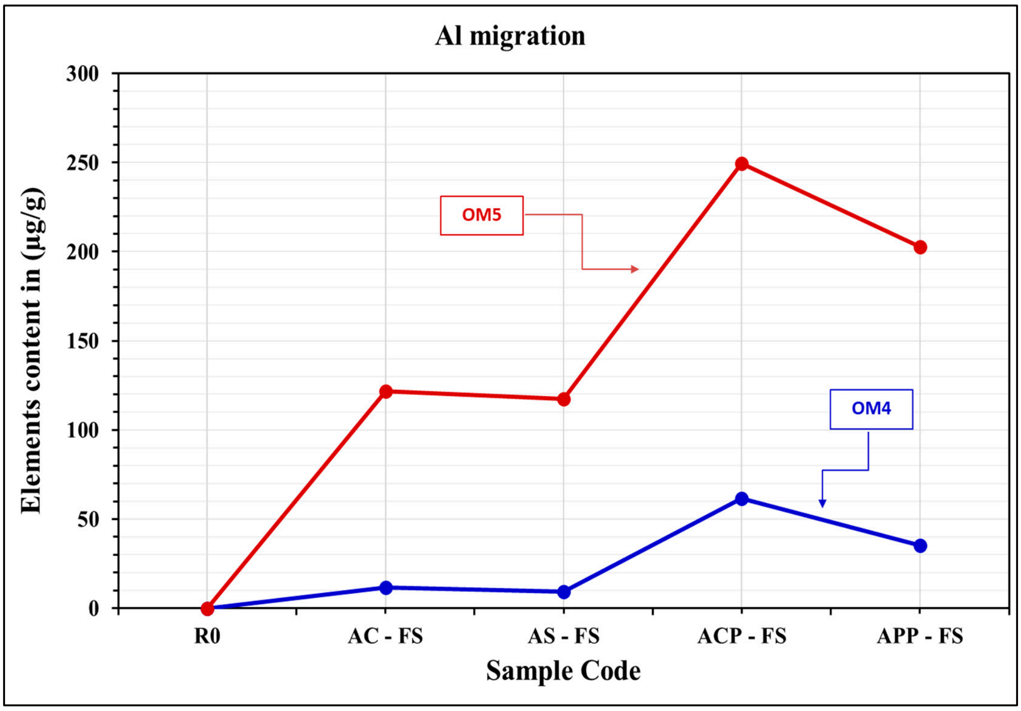

Figure 3. The content of aluminum migrated to food simulant when applying OM4 and OM5 tests using different aluminum cooking pots.

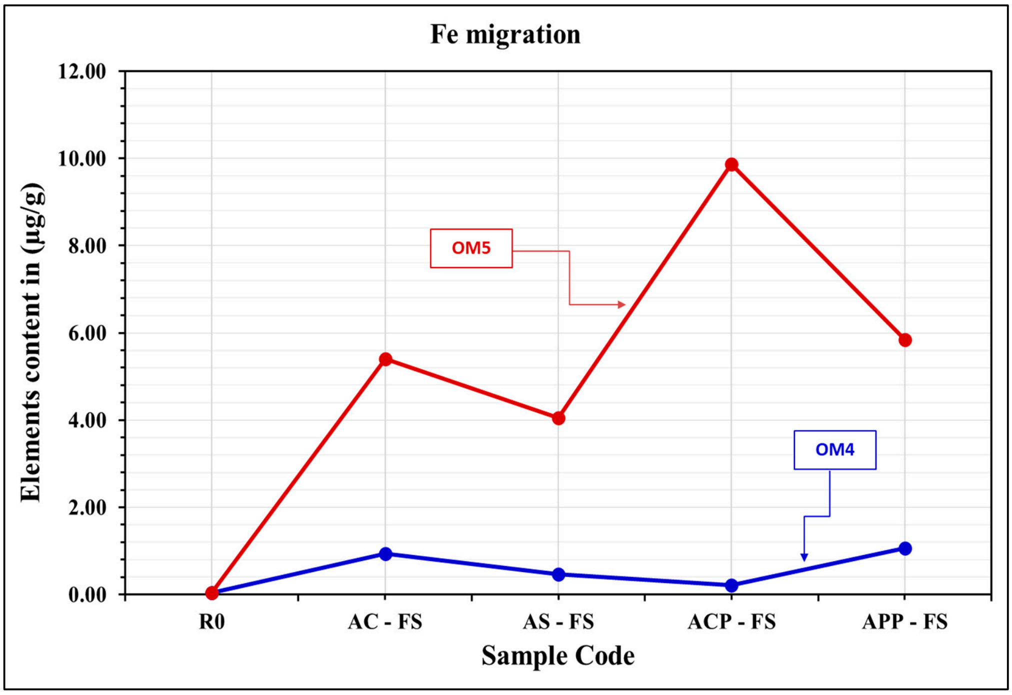

Figure 4. The content of iron migrated to food simulant when applying OM4 and OM5 tests using aluminum cooking pots.

Based on the obtained results, it was observed that aluminum is the primary element that migrates from the pots to the cooked food and food simulants. Iron was also detected in significant quantities. The major factors affecting the migration of elements in the present study were food acidity [2], pot quality (manufacturer), the temperature and duration of the cooking process, and the type of pot (traditional pot or pressure cooker), with pressure cookers significantly affecting element migration. The impact of acidity on the migration of metal ions can be explained by considering electrode reactions and the tendency for ionic migration, which is influenced by the solution's pH, determined by its acidity [30]. Metal ion solubility is influenced by the acidity of the medium, which affects the concentration gradient and the driving force for ionic migration. The coordination of metal ions with water molecules can also be influenced by the medium's acidity, which in turn affects Bronsted acidity and the release of protons from hydrated metal ions [31]. Several processes explain the effect of acidity on metal ion migration: (i) Acidic constituents in food, such as tomato products, accelerate the transfer of aluminum from food contact materials to food. (ii) The concentration of the acetic acid solution was positively correlated with the transport of metal ions. (iii) Cations such as Al3+ and Fe3+, which have a high charge-to-size ratio, migrated into the solution and acted as Bronsted acids, polarizing the attached water molecules and increasing acidity [32].

Temperature (heat) was observed to have a significant impact on the amount of Al and Fe that migrated into food from the cooking pots. As the metal surface and solution temperature rise during the cooking process, they affect the kinetic energy and diffusion rate of metal atoms and ions. Electron transfer and ionic migration are more likely to occur when metal atoms and ions move and collide at higher temperatures. The electro-migration of metal atoms and ions is influenced by the temperature of the metal conductor and current density during cooking, resulting in a process known as electro-migration [33,34]. Heat can accelerate various chemical processes, such as oxidation or hydrolysis, leading to the formation of more metal oxides or hydroxides, which can dissolve the metal surface. Consequently, more metal ions may be released into the solution and moved to other areas based on the established concentration gradient [2,35]. High temperature is a known accelerator of metal corrosion, where chemical interactions between a metal's surface and its surroundings, including oxygen, water, acid, or salt, are accelerated by the application of heat. Under these conditions, oxides, hydroxides, and salts of metals are expected to form, releasing even more metal ions into the food. For example, alumina (Al_2O_3) and aluminum hydroxide $Al(OH)_3$ are expected products from aluminum's interaction with the environment. Similarly, iron oxide (Fe_2O_3) can form due to iron's interactions with its surroundings. The corrosion of metals can be influenced by several factors, including the metal's chemistry, the pH of the solution, and the presence of inhibitors or catalysts [36]. When cooking with an acidic medium, aluminum undergoes chemical migration according to Equation (1):

$$Al + 3H^+ \rightarrow Al^{3+} + \frac{3}{2}H_2 \quad (1)$$

When an acidic food interacts with aluminum metal, it produces aluminum ions and hydrogen gas, as shown in Equation (1) above. Hydrogen gas escapes in the form of bubbles, while the aluminum ions remain in the cooked food. The migration of aluminum ions can be influenced by factors such as the food's acidity, cooking temperature, and duration [37]. Iron can also corrode in acidic food, releasing iron ions into the food when it reacts with oxygen and water to form rust, as described in Equation (2):

$$4Fe + 3O_2 + 6H_2O \rightarrow 4Fe(OH)_3 \quad (2)$$

However, rust can dissolve more extensively and release additional iron ions into the food when it is acidic, as in the case of tomato sauce. This chemical reaction is represented by Equation (3):

$$Fe(OH)_3 + 3H^+ \rightarrow Fe^{3+} + 3H_2O \quad (3)$$

Cooking at higher temperatures and/or for longer durations promotes forward reactions, leading to the increased release of metal into the food [38].

Extended cooking in metal containers increases the likelihood of metal ions corroding from the surface into the food, with the metal, the food, and the heat all contributing to this process. Elevated temperatures and longer cooking times can further increase the release of metal ions [39]. When using pressure cookers, the migration of metal ions such as aluminum and iron can be affected. Generally, metal corrosion and dissolution depend on the system's pressure [40,41]. The migration of ions from the metal surface to the food can be accelerated by increasing the system's pressure during cooking, as is the case with pressure cookers.

3.4. The Results of SEM-EDS Analysis

SEM-EDS analysis was conducted to confirm the detected element concentrations in the cooked food samples, mainly consisting of rice that was dried and ground into powder. It should be mentioned that SEM-EDS analysis was performed on all samples from various food experiments, as shown in Table 1; however, only one example related to the ACP-5 condition will be presented to avoid repetition. Figure 5 presents SEM-EDS results for food samples prepared under the conditions of ACP-5 as outlined in Table 1. In Figure 5a,b, secondary electron images at low (200×) and high magnification (500×) display the size and morphology of the prepared food samples. Figure 5c highlights the selected area for chemical analysis, outlined in red in the same images. Figure 5d displays the EDS spectra of the examined area, and the top-right insert in Figure 5d provides a table of chemical analysis listing the quantitative values of elements in atomic percent. The results of EDS spectra and quantitative chemical analysis showed that only carbon and oxygen were detected in the examined samples. During EDS analysis, five elements of interest, namely Al, Fe, As, Cd, and Pb, were selected to check their concentrations in the cooked food samples. However, the EDS technique did not detect any of these elements in the food samples prepared using aluminum cooking pots. No peaks for these elements were observed in the obtained EDS spectra, and quantitative chemical analysis did not yield any values for the selected elements. The reason for this is that the concentrations of the detected elements, as per the ICP-MS results presented in Table 1, as well as Figures 1 and 2, fall below the detection limit of the EDS technique. EDS cannot detect trace elements with concentrations below 0.1 wt.%, and its reported detection limit is 0.1 wt.% (1000 ppm or 1000 μg/g) [42–44].

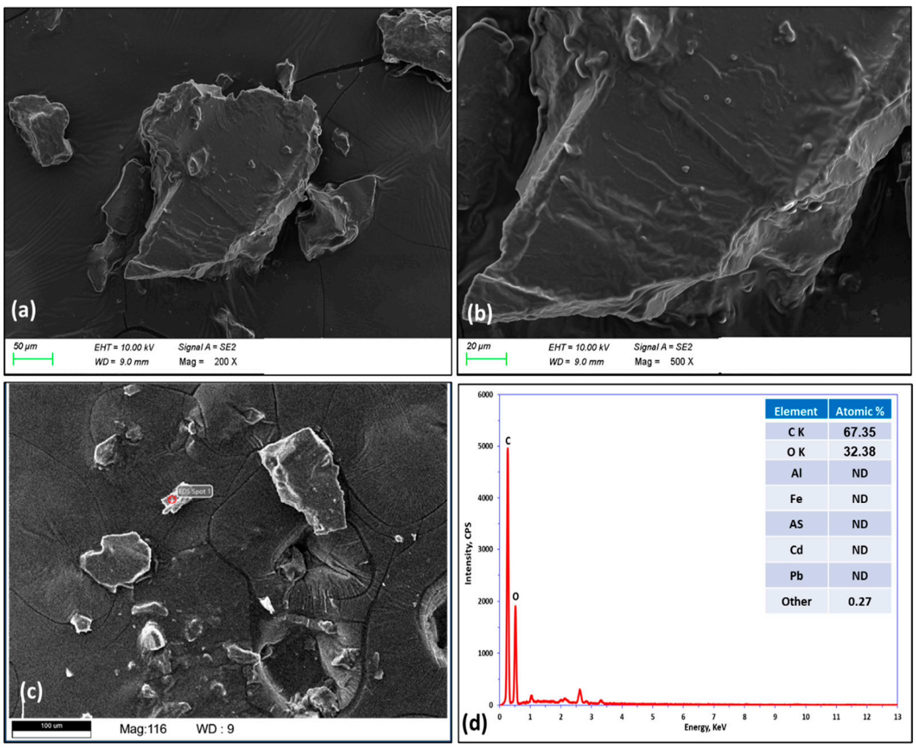

Figure 5. SEM-EDS results collected from food samples prepared using an aluminum cooking pot according to ACP-5 experiment conditions: (**a**) secondary electron image of the morphology and size of the prepared food sample at 200×; (**b**) secondary electron image of the prepared food sample at 500×; (**c**) the selected area for chemical analysis (outlined in a red color); (**d**) the EDS spectrum and chemical analysis results of the selected area in (**c**).

3.5. The Results of XPS Analysis

An additional chemical analysis technique was employed to further verify the presence of detected elements in food samples. X-ray photoelectron spectroscopy (XPS) is a surface-sensitive technique used for the quantitative analysis of elemental composition, particularly at the top surface layers of samples. XPS analysis was conducted to confirm the concentration of detected elements in the cooked food samples. As mentioned earlier in the previous subsection, the prepared food samples primarily consist of rice, which was dried and ground into a powder form for XPS analysis. It is worth mentioning that XPS analysis was performed on all samples collected from various food experiments, as shown in Table 1; however, only one example related to the ACP-5 condition will be presented to avoid repetition. Figure 6 displays the XPS survey (full) scan spectrum collected from food samples prepared according to the ACP-5 condition outlined in Table 1. The x-axis of this spectrum covers the binding energies of all possible elements that could be detected in the food samples prepared using aluminum cooking pots, including Al, Fe, As, Cd, and Pb. However, only carbon and oxygen were detected and quantified in the examined food samples, as indicated by the XPS survey scan presented in Figure 6. The corresponding quantitative chemical analysis is inserted in the upper-left corners of the same figure. Figure 7 presents the XPS high-resolution spectra related to the survey scan in Figure 6, specifically for the ACP-5 experiment conditions. The high-resolution scan was performed to identify the elements of interest. However, Figure 7 shows that none of these elements, including C 1s, O 1s, Al 2p, Fe 2p, As 3d, Cd 3d, and Pb 4f, were detected except for carbon and oxygen. The XPS technique did not detect any of the elements of interest in the prepared food samples using aluminum cooking pots because the content of these elements fell below the detection limit of the XPS technique. The reported detection limit for XPS is typically below 1 at.% to 0.1 at.% [45–47], meaning that trace elements below this concentration cannot be detected by XPS. Based on the ICP-MS results presented in Table 1, the concentration of all detected elements is indeed below the detection limit of the XPS technique.

In the present study, three analytical methods were applied to quantify the elements migrated to food to ensure the accuracy and reliability of the results. Different methods may display different levels of sensitivity and detectability of different elements which allow results validation and confirmation. Applying these three methods provided a comprehensive analysis and confidence with the results. Table 8 shows the principle, advantages, disadvantages, and detection limits of the three techniques applied in the current study.

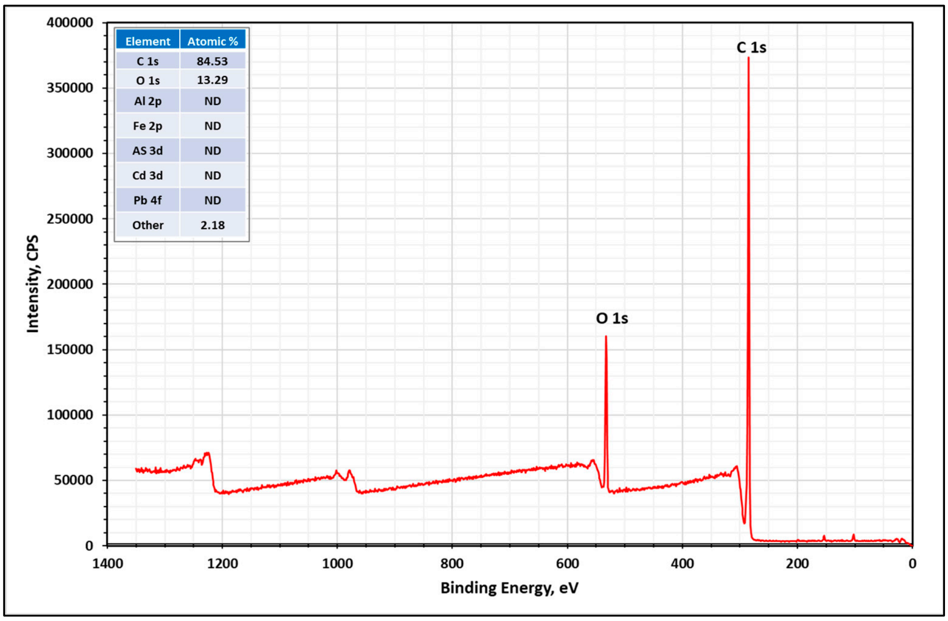

Figure 6. XPS survey scan spectrum collected from food samples prepared by aluminum cooking pot according to ACP-5 experiment conditions.

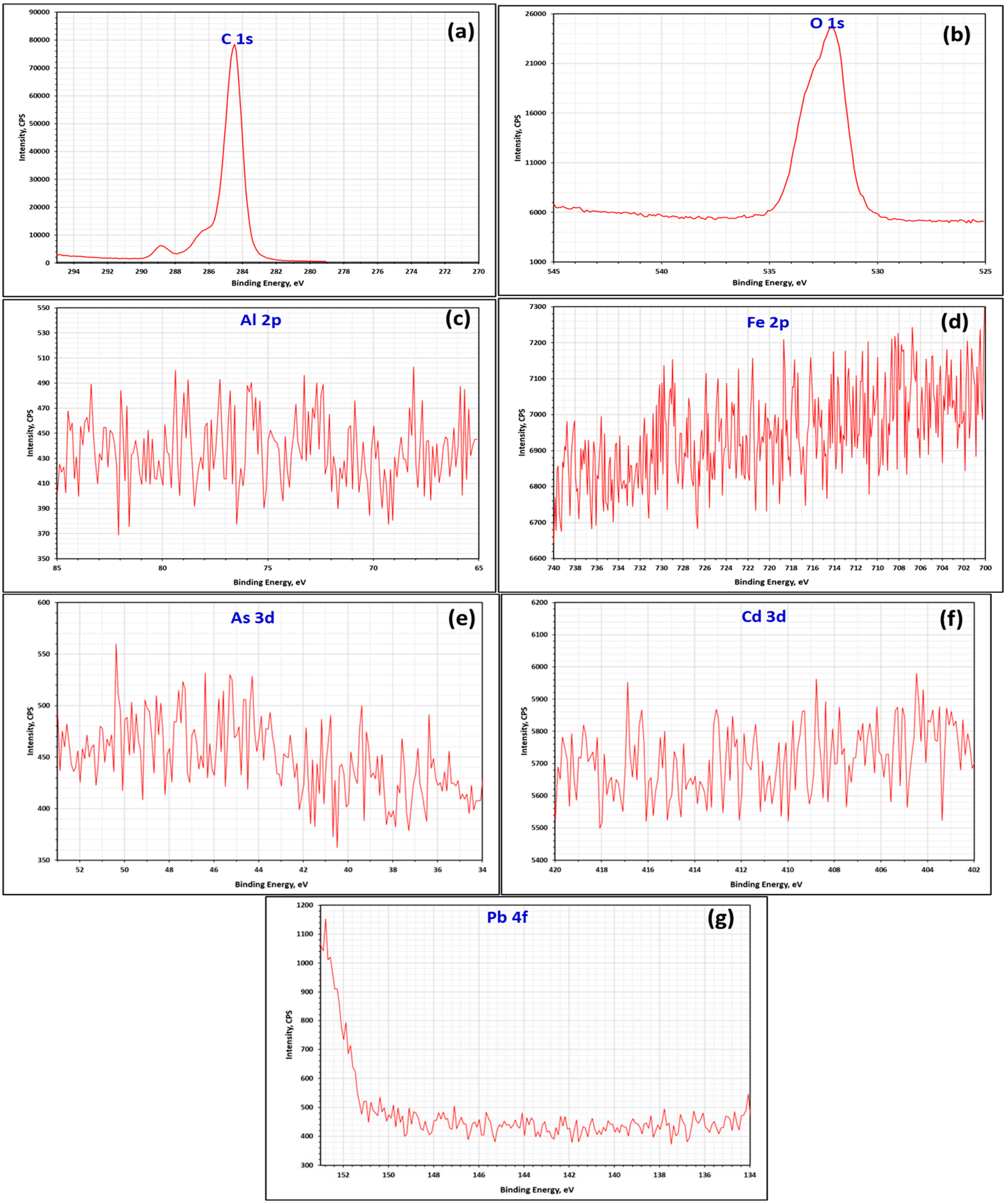

Figure 7. XPS high resolution spectra related to the survey scan in Figure 6 (ACP-5 experiment conditions): (**a**) C 1s; (**b**) O 1s; (**c**) Al 2p; (**d**) Fe 2p; (**e**) As 3d; (**f**) Cd 3d; and (**g**) Pb 4f.

Table 8. Advantages, disadvantages, and detection limits of ICP-MS, EDS, and XPS techniques used in the present study [42–49].

	ICP-MS	EDS	XPS
Principle	• It analyses the mass-to-charge ratio of ions generated from a sample by inductively coupled plasma. • It is primarily used for elemental analysis, especially for trace and ultra-trace elements.	• It is linked with scanning electron microscopy (SEM) and detects characteristic X-rays emitted from a sample when bombarded with electrons. • It is used for elemental analysis in the form of point scan, line scan, and area mapping.	• It measures the kinetic energy of electrons ejected from a sample when irradiated with X-rays. • It provides information about the elemental composition, chemical state, and electronic state of the surface.
Advantages	• It is highly sensitive. • It can detect elements at trace and ultra-trace levels. • It detects a wide range of elements. • It provides accurate and precise quantitative results for elemental concentrations.	• It provides elemental distribution maps. • It is relatively fast and easy for use.	• It is highly surface-sensitive and provides information about the chemical composition and electronic states of elements on the sample surface. • It provides information about the oxidation state and chemical bonding of elements.
Disadvantages	• Sample preparation needs digestion of solid samples. • Interference from the sample matrix can affect the accuracy of results.	• Lower sensitivity compared to ICP-MS, making it less suitable for trace element analysis. • It is surface-sensitive, providing information about the top few micrometers of the sample. • Overlapping peaks can make it challenging to identify certain elements accurately.	• Lower sensitivity compared to ICP-MS, making it less suitable for trace element analysis. • It is limited to the top few nanometers of the sample surface. • Operate under ultra-high vacuum conditions, limiting the types of samples that can be analyzed.
Detection limit	• Below 1–100 ng/L (ppt, part per trillion).	• Below 0.1 wt.%.	• Below 0.1–1 at.%.

3.6. The Permissible Daily Intake Limit of Chemical Elements

The Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) have reported the acceptable daily intake levels for aluminum (21 mg/day), iron (18 mg/day), arsenic (0.030 mg/day), cadmium (0.058 mg/day), and lead (0.252 mg/day) [50–52]. Table 9 displays the calculated daily intake quantities (mg/day) based on the results obtained from the actual food experiments presented in Table 6. The daily intake quantity was determined by multiplying the proposed amount of food (100 g) consumed per day by the concentration of metals present in that food. According to the results provided in Table 9, the food samples prepared under all conditions demonstrated a safe health status regarding the daily intake of iron, arsenic, cadmium, and lead. However, for aluminum, it exceeded the acceptable daily intake limit when pressure cookers were used for extended cooking periods. Nevertheless, the daily intake of aluminum remained within safe health limits for the AC-1, AC-2, AC-3, AC-4, AC-5, and AS-5 cooking conditions. The health risk associated with aluminum intake becomes more significant when the daily food consumption by an individual increases and/or when pressure cookers (ACP5 and APP5) are employed. It is worth noting that the AC5, AS-5, ACP-5, and APP-5 cooking conditions represent extreme cooking scenarios, where the food was cooked for 2 h at 100 °C and then kept at 70 °C for 8 h, as outlined in Table 1.

Table 9. The calculated intake quantity per day (mg/day) for the results of actual food experiments.

Sample Code	Calculated Intake Quantity per Day (mg/Day)				
	Al	**Fe**	**As**	**Cd**	**Pb**
AC-1	9.327	2.417	0.0046	0.0019	0.0079
AC-2	14.73	2.782	0.0056	0.0019	0.0079
AC-3	14.44	3.029	0.0052	0.0020	0.0087
AC-4	14.52	3.298	0.0069	0.0020	0.0088
AC-5	17.29	3.544	0.0067	0.0021	0.0084
AS-5	16.05	3.017	0.0068	0.0021	0.0085
ACP-5	26.78	3.821	0.0070	0.0020	0.0083
APP-5	23.61	3.040	0.0059	0.0021	0.0082

The present study may reveal an impact at local, national, and international levels. Understanding element migration from aluminum pots is crucial for local communities as it directly impacts the health of individuals. The present study attracts the attention of local communities regarding the extensive use of aluminum pots, which may lead to elevated concentrations of aluminum in cooked food and cause health risks. Furthermore, there is a relevant contribution of the present research that is expected to attract the attention of local communities, especially regarding health concerns related to aluminum pressure cookers. The outcomes of the current study reveal an important contribution on the national level through the potential development or revision of national public health policies. The national standard organization may establish or update the policies related to the materials and type of pots used in cookware to protect consumers through minimizing health risks associated with element migration. Concerning the international level, the present research may offer a contribution to the global standards related to international trade regulations and agreements about cookware materials to ensure the safety and quality of cookware products across borders. The research outcomes of the current study can contribute to educational campaigns at local, national, and international levels to raise awareness among consumers about the recommended conditions of using aluminum cookware and the associated potential health risks.

The strengths of the current study include several aspects, such the protection of public health by assessing the potential migration of heavy elements to food; the contribution to consumer awareness about potential risk of aluminium cookware; the possible contribution to the development of policies and regulations at local, national, and international levels to ensure the safety of cookware materials; and confirming the unsuitability of EDS and XPS methods for quantifying the elements that have migrated to food. On the other hand, the weakness of the present study is mainly related to the fact that the migration of elements is a complex process influenced by various factors such as the type of pot, food ingredients, pH, temperature, and cooking time. Understanding the interplay of these factors is challenging. Furthermore, the diversity of cookware materials, coupled with variations in cooking techniques, is a great challenge to capture and report the full range of potential exposures and related health issues.

The novel aspects of the present study are summarized as follows:

- Examining element migration from aluminium cooking pots to foods, covering almost all possible variables that meet real-life cooking conditions, such as pot quality, pot type (traditional pots or pressure cookers), water supply (tape or mineral water), food acidity, the addition of salt, spices, and the effect of cooking time.
- Using actual food experiments like real food preparation and validating the results with standard food simulant experiments.
- Studying the effect of extreme cooking conditions on element migration to food such as experiments AC-5; AS-5; ACP-5; APP-5.

- The investigation of element migration from aluminium pressure cookers.
- Element migration was characterized using three techniques (ICP-MS, EDS, and XPS) for evaluating the results, which provided a more comprehensive analysis and more confidence in the results.
- This study confirmed the unsuitability of EDS and XPS techniques for quantifying elements that have migrated to food.
- This study can contribute to the protection of public health by assessing the migration of heavy elements to food and the awareness of consumers about the potential risk of aluminium cookware, especially aluminium pressure cookers.

4. Conclusions

In the present study, we investigated the migration of elements from aluminum cooking pots under various variables. Based on the results obtained in this study, the following conclusions can be drawn:

- The concentration of elements that migrated to food in actual food samples was significantly influenced by factors such as food acidity, heating time, and the type of aluminum pot used (traditional or pressure cookers).
- Aluminum was identified as the major element that migrated from aluminum cooking pots, while other elements, such as iron (Fe), exhibited lower concentrations compared to aluminum. Toxic elements like arsenic (As), cadmium (Cd), and lead (Pb) were present in minor concentrations in the cooked food under all cooking conditions, and their levels were below acceptable limits.
- The concentration of aluminum deserves special attention due to its significant variation under various processing variables. Aluminum content increased from 80.17 μg/g to 133.7 μg/g when tomato sauce was added to the food, and a further increase in aluminum concentration (157.9 μg/g) was observed with longer heating times.
- There was a noticeable increase in aluminum concentration in the cooked food when aluminum pressure cookers were used, with the highest concentration of aluminum (252.7 μg/g) observed in food cooked with an ACP pressure cooker.
- The results obtained for element migration content in food simulants were consistent with those obtained from the actual food samples.
- Element migration was observed to significantly increase when applying OM5 tests compared to OM4 due to prolonged heating at 100 °C.
- Food simulants prepared using the ACP pressure cooker with OM5 test conditions exhibited the highest concentrations of aluminum (249.6 μg/g) and iron (9.868 μg/g).
- The concentration of toxic elements in food simulants was minor, and changes after applying OM4 and OM5 tests could be disregarded.
- Based on the results of the analysis conducted in this study, the SEM-EDS and XPS techniques are not suitable for quantifying elements that migrated to food samples due to their detection limits.
- For daily intake, all elements (Fe, As, Cd, and Pb) in the prepared food samples under various conditions were found to be within safe health limits, except for aluminum, which exceeded the daily intake limit when pressure cookers were used for extended cooking times. It should be noted that experimental conditions such as AC5, AS-5, ACP-5, and APP-5 represented extreme cooking scenarios involving 2 h of cooking at 100 °C followed by 8 h at 70 °C.
- To minimize element migration to food, it is recommended to use traditional aluminum cooking pots rather than aluminum pressure cookers for cooking. Furthermore, each cooking cycle using traditional Al pots should not exceed two hours.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/app132413119/s1, Table S1: Technical specifications of the pot (code: AC) received from manufacturer (1); Table S2: Technical specifications of the pot (code: AS) received from manufacturer (2); Table S3: Technical specifications of the pot (code: ACP) received from manufacturer (3); Table S4: Technical specifications of the pot (code: APP) received from manufacturer (4).

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Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to contract conditions with the funding organization.

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Conflicts of Interest: The authors declare no conflict of interest.

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