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Impact of Cookware Types on Leaking Heavy Metals into Food based on pH Values, and their Potential Impact on the Health Status of a Saudi Sample using a Survey Study Design

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Abstract: This study aimed to measure the leaked metal concentrations in cooked food using three types of cookware (stainless steel (18/10), cast iron, and aluminum). Two types of vegetables were used based on their pH values, tomatoes (pH 4.30-4.90) and zucchini (pH 9), as acidic and basic food materials. In addition, a survey study was performed on a randomly chosen Saudi sample to investigate the impact of their income, age, and awareness level in choosing between the different cookware available in the market, as well as to investigate the impact of the used cookware on their health status. The results showed that cooking acidic food (e.g., tomatoes) in aluminum pots can lead to an increase in the concentrations of leaked metals into the food such as leaking copper (0.27mg/kg), iron (3.048mg/kg), aluminum (0.91mg/kg) and magnesium (95.13 mg/kg) but no change in the concentrations of leaked metals into the food such as Chromium and Nickle (<0.4mg/kg). Results revealed that the only cookware that had the least leaked metals into the acidic food was cast iron. For the basic food (e.g., zucchini), results showed that zucchini turned into an acidic medium after cooking which increased the leaking of different metals into the cooked zucchini. The only cookware that had the least leaked metals into the zucchini was the stainless steel (18/10). The leaked metals into the food may affect the health; the results of this study's survey showed a potential correlation between using cast iron and stainless steel with the incidence of diabetes, arthritis, and heart diseases. Results also showed that some individuals are aware of choosing between the different cookware available in the market based on the health impact. However, their income affected their choices significantly regardless of their awareness. In conclusion, it can be recommended based on the results of this study that cast iron is more preferable to be used for cooking acidic food, whereas aluminum is not preferable for cooking basic foods.

Keywords: cookware, heavy metals, pH, zucchini, tomatoes, aluminum, cast iron, stainless steel.

تأثير أنواع أواني الطهي على تسرب المعادن الثقيلة الى الغذاء بناء على قيم pH وتأثير المحتمل على الحالة الصحية لعينة سعودية باستخدام الدراسة المسحية

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المستخلص: هدفت هذه الدراسة إلى قياس تركيزات المعادن المتسربة في الطعام المطبوخ باستخدام ثلاثة أنواع من أواني الطهي (الفولاذ المقاوم للصدأ (10/18) والحديد الزهر والألمنيوم). تم استخدام نوعين من الخضار على أساس قيم الأس الهيدروجيني، الطماطم (الرقم الهيدروجيني 4.30-4.90) والكوسا (الرقم الهيدروجيني 9)، كمواد غذائية حمضية وقاعدية. بالإضافة إلى ذلك، تم إجراء دراسة استقصائية على عينة سعودية تم اختيارها عشوائيًا للتحقق من تأثير دخلهم وعمرهم ومستوى وعيهم في الاختيار بين أواني الطهي المختلفة المتوفرة في السوق، وكذلك لمعرفة تأثير أواني الطهي المستخدمة على حالتهم الصحية. أظهرت النتائج أن طهي الطعام الحمضي (مثل الطماطم) في أواني الألمنيوم يمكن أن يؤدي إلى زيادة تركيزات المعادن المتسربة في الطعام مثل تسرب النحاس (0.27 مجم / كجم) والحديد (3.048 مجم / كجم) والألمنيوم (0.91 مجم/كلجم) والمغنيسيوم(95.13مجم/كجم) ولكن لم تتغير تركيزات المعادن المتسربة للطعام مثل الكروم والنيكل (<0.4 مجم/كجم). أظهرت النتائج أن أواني الطهي الوحيدة التي تحتوي على أقل المعادن المتسربة في الطعام الحمضي هي الحديد الزهر. بالنسبة للغذاء القاعدي (مثل الكوسة)، أظهرت النتائج أن الكوسة تحولت إلى وسط حمضي بعد الطهي مما زاد من تسرب المعادن المختلفة إلى الكوسة المطبوخة. كانت أواني الطهي الوحيدة التي تحتوي على أقل المعادن تسربًا في الكوسة هي الفولاذ المقاوم للصدأ (10/18). قد تؤثر المعادن المتسربة في الطعام على الصحة؛ أظهرت نتائج الدراسة الاستقصائية هذه وجود علاقة محتملة بين استخدام الحديد الزهر والفولاذ المقاوم للصدأ مع الإصابة بمرض السكري والتهاب المفاصل وأمراض القلب. كما أظهرت النتائج أن بعض الأفراد على دراية بالاختيار بين أواني الطهي المختلفة المتوفرة في السوق بناءً على التأثير الصحي. ومع ذلك، أثر دخلهم على اختياراتهم بشكل كبير بغض النظر عن وعيهم. في الختام، يمكن التوصية بناءً على نتائج هذه الدراسة أن الحديد الزهر هو الأفضل لاستخدامه في طهي الأطعمة الحمضية، في حين أن الألمنيوم ليس مفضلًا لطهي الأطعمة القاعدي.

الكلمات المفتاحية: تجهيزات المطابخ، المعادن الثقيلة، الكوسة، الطماطم، الألمنيوم، الحديد الزهر، الفولاذ المقاوم للصدأ.

Introduction

Minerals such as calcium, chromium, copper, and iodine are earth-originated elements that are essential for the being health and cannot be synthesized by living organisms[1]. In contrast, heavy metals are also naturally occurring elements but are characterized by high atomic weight and a higher density at least five times greater than that of water[2]. Heavy metals include titanium, vanadium, chromium, manganese, and iron[3]. These metals are widely used in manufacturing several types of cookware. For instance, stainless steel cookware is a mixture of iron, carbon, and chromium and is one of the most common cookware used in food preparations worldwide. It is widely known that stainless steel cookware is preferable for all sorts of ingredients and cooking methods. It has been reported that the best choice of stainless steel is that marked as 8/10 or 10/18, indicating the ideal chromium-nickel ratio for corrosion resistance[4]. Stainless steel cookware is mostly used due to its durability, (It does not interact with food, and is not scratchable) stainless steel is a great option for non-toxic, long lasting durable cookware ideal for boiling, sauteing, and baking. It is especially good for small batch baking as it retains heat well and cooks' food evenly[5]. However, high ratios of nickel may cause allergic reactions if stainless steel were used in the preparation and storage of acidic foods[6]. Another type of metal-made cookware is cast iron, a hard and relatively brittle alloy that contains a mixture of iron, carbon, and silicon. Carbon content in cast iron is higher than that of stainless steel, ranging from 2 to 4%, whereas silicon and other metals (e.g., manganese and trace amounts of sulfur and phosphorous) range from 1 to 3%. Due to the high content of carbon, cast iron hardens as a heterogeneous alloy. Cast iron is characterized by a lower melting temperature than other iron alloys[7]. It can be preferable in food preparations due to its ability to hold high temperatures for a long time than any other cookware, as well as due to its naturally non-stick property. However, it may affect the flavor of acidic foods[8]. Another common type of metal cookware is aluminum, a composition of metals mainly consisting of aluminum with the addition of little rations of other metals. Aluminum cookware is mostly manufactured by mixing solid metals with liquid aluminum and then cooling it down to form a homogeneous solid solution. The added solid metals may compromise 15% of the alloys' total mass, including iron, copper, magnesium, silicon, and zinc[9]. Aluminum cookware is preferable in food preparations due to its lightweight, making it a good heat conductor. But green leafy vegetables and citrus fruits absorb it[6].

Heavy metals involved in metal-made cookware such as stainless steel, cast iron, and aluminum may leak into food and drinks during cooking. Therefore, consuming food or drinks prepared in this metal cookware may cause some health risks due to the presence of these heavy metals. For instance[10] found that urinary levels of metals such as arsenic, chromium, aluminum and zinc showed an association with diabetes, while arsenic and zinc showed an association with pre-diabetes and atherosclerosis. Thus, this study aims to discover the potential effect of heavy metal on human health in Saudi sample by analyzing some samples and using Survey Study.

Methods and Materials

Cookware and Food Materials

Three types of cookware (stainless steel made in Germany, cast iron made in UA, and aluminum made in Egypt) were purchased from the local market in Buraydah, Saudi Arabia. Two types of vegetables Tomato (*Solanum lycopersicum*) and zucchini (*Cucurbita pepo*) were purchased from the local markets of Buraydah, Saudi Arabia. The vegetables were selected based on their pH values, acidic and basic. Tomatoes, as an acidic sample (pH 4.90–4.30), and Zucchini, basic in the raw phase (pH 9) and acidic when cooked (pH 6–4.1).

Preparation of Vegetable Samples

The three cookware were prepared by washing them with distilled water. Then, freshly bought tomatoes and zucchini were washed with distilled water and cut into small weighed portions (442 g) to prepare eight samples; for both tomatoes and zucchini, two samples were raw (control), two samples were cooked in cast iron, two samples cooked in an aluminum pot, and two samples cooked in a stainless-steel pot.

The weighed samples, except for the control, were minced thoroughly and mixed with 100 mL of distilled water to form a homogenous mixture. The different pots were heated to 110 °C, the prepared mixtures were then placed into the heated pots and cooked for 15 min with regular stirring using a polyamide spoon. After 15 min of cooking, the mixtures were taken out and the temperature for each pot was measured.

Determination of Leaked Metals Concentration in Cooked Vegetable mixture Samples

The concentrations of different metals, including aluminum (Al), chromium (Cr), copper (Cu), iron (Fe), magnesium (Mg), nickel (Ni), and zinc (Zn) for the eight samples were measured using an ICB-MS device at the Saud Ajal Laboratory (Riyadh, Saudi Arabia).

Survey Study Design

A survey study was performed from 16 January 2022 to 16 June 2022 to investigate the potential adverse effects of using different metal cookware on the health status of the chosen Saudi sample. The study sample was chosen randomly from different regions in Saudi Arabia (The Riyadh Province, The Makkah Province, The Eastern Province, The Madinah Province, The Al Baha Province, The Al Jawf Province, The Northern Borders Province, The Qassim Province, The Ha'il Province, The Tabuk Province, The 'Aseer Province, The Jizan Province and The Najran Province); 1,406 participants were included in this study. The survey was designed to include two main sections; first, data regarding the participant's demographic, economic, and health status, and second, data regarding the types of cookware used by the participants and their awareness of choosing between the different cookware available in the market. This study was performed under the approval of the Committee of Research Ethics (Institutional Review Board, IRB) of Qassim University, Saudi Arabia (approval no. 21-24-01).

Statistical analysis

Statistical analysis was performed using SPSS (Ver. 21). One-way analysis of variance (ANOVA) was applied, followed by Tukey's test, and the results were considered significant if (p<0.05) according to (Steel, 1997).

Results and Discussion

Concentrations of Leaked Metals in Cooked Tomatoes

The results as shown concentrations of different metals (mg/kg) in cooked tomatoes using three different metal pots in **Table (1)** demonstrate that Al could leak into food during cooking acidic food in Al pots. After 15 min of cooking tomatoes at 110 °C in an Al pot, the concentrations of Al increased in the cooked tomatoes compared to the raw tomatoes (<0.002) mg/kg in row tomatoes *vs.* 0.002 -0.91 mg/kg in cooked tomatoes. Whereas cooking tomatoes in the other two pots, stainless steel and cast iron, did not lead to any leaks in metals into the cooked tomatoes; results were similar to the raw tomatoes (p<0.05). Indicating that only Al pots, compared to stainless steel and cast iron, can lead to a leak in the Al into food. Higher temperatures are required for to dissolve more easily because Al pots is good conductor of heat[10]. Al can dissolve in hot alkalized conditions[11], and thus, it showed an increase in the cooked tomatoes, as an example of acidic foods. The temperatures of the pots after the expiry time were 175 °C, 161 °C, and 138 °C, respectively, Concentrations of different metals (mg/kg) in cooked tomatoes using three different metal pots.

The concentration of Cu in the cooked tomatoes using all three pots also showed an increase (0.27-0.201- 0.226 mg/kg) compared to the raw tomatoes (0.171). However, the highest concentration of Cu was observed in samples cooked in the Al pot (0.27 mg/kg). Whereas cooking tomatoes in stainless steel pots showed the least concentration of leaked Cu (0.201 mg/kg). The high concentration of Cu after cooking in the Al pots is due to the fact that Higher temperatures are required for cu to dissolve more easily[18]. Cu can dissolve in dilute acids[12]. Similarly, Fe showed an increase after cooking tomatoes in Al pots and stainless compared to the raw tomatoes (3.048 – 2.002 mg/kg *vs<1.0.*, respectively). Whereas cooking cast-iron pots did not show any increase in the concentrations of Fe. Higher temperatures are required for Fe to dissolve more easily[10]. Fe can also dissolve in heated dilute acids[13]. The reason for the stability is that cast iron pots are iron and carbon (steel)[7], and steel does not react with acids[14].

As for Mg concentrations, results showed an increase after cooking tomatoes in all pots compared to raw tomatoes. Consistently, the highest concentrations were observed after cooking in the Al pot (95.13 mg/kg), whereas the least concentrations were observed after cooking in cast iron (70.973 mg/kg). Higher temperatures are required for Mg to dissolve more easily from Al pots[10]. The rise of Mg in Al pots is due to the fact that magnesium dissolves in acidic media[15]. Zn concentrations were also screened, in contrast to the other results, it showed a decrease after cooking in the different pots compared to the raw data. The most notable decrease was observed in tomatoes cooked in cast iron (3.414 mg/kg). Zn is insoluble in water; therefore, it showed a decrease after cooking [16]

Table 1. Concentrations of different metals (mg/kg) in cooked tomatoes using three different metal pots.

Heavy Metals (mg/kg)	Cookware Type / (The temperatures of the pots after the expiry time)			
	Uncooked (Control)/pH 4.30 – 4.90	Aluminum/ 138°C	Stainless steel (18/10)/ 161°C	Cast iron/ 175°C
Al	<0.002	0.91*	<0.002	<0.002
Cr	<0.4	<0.4	<0.4	<0.4
Cu	0.171	0.27*	0.201	0.226
Fe	<1.0	3.048*	2.002	<1.0
Mg	62.185	95.13*	74.039	70.973
Ni	<0.4	<0.4	<0.4	<0.4
Zn	5.777	3.63	3.579	3.414#

***Represents the height from the raw sample - #Represents the decrease from the raw sample. Al: Aluminum; Cr: Chromium; Cu: Copper; Fe: Iron; Mg: Magnesium; Ni: Nickle; Zn: Zink.**

Concentrations of Leaked Metals in Cooked Zucchini

After 15 min of cooking zucchini in the three metal pots, results showed that Al concentrations decreased in the cooked zucchini as compared to the raw sample (0.45-0.93 mg/kg *vs.* 0.93, respectively), as shown in **Table (2)**. The most decrease was observed in samples cooked in the stainless-steel pot (0.45mg/kg). The cooked zucchini turns into an acidic medium because when a green vegetable is cooked, the plant's natural acid is leached into the cooking liquid and is trapped there creating an acidic cooking medium[17].

, and thus, Al cannot dissolve in acidic media[11]. Results also showed a decrease in Cu concentrations in the zucchini cooked using the Al pot compared to the raw zucchini (0.489 mg/kg *vs.* 0.522, respectively). Whereas in the other pots, Cu concentrations showed an increase. The highest increase was observed in zucchini cooked in cast iron (0.578 mg/kg), due to the increase in water loss in pots[18]. As for Fe concentrations, zucchini cooked in Al pots showed the highest increase as compared to the raw zucchini (13.88 mg/kg *vs.* 3.785, respectively), which is due to the ability of Fe in dissolving in acidic media[13], and as mentioned earlier, zucchini turns into an acidic medium after cooking. For the other pots, Fe showed a decrease mostly in samples cooked in cast iron (3.656 mg/kg).

Mg concentrations also showed a notable decrease in zucchini cooked in Al pot compared to the raw data (188.326 mg/kg *vs.* 215.21, respectively). The other pots did not show a notable effect on the Mg concentration. Higher temperatures are required for Mg to dissolve more easily[19]. The temperature of the Al pot after 15 min of cooking was 138 °C (**Table 1**), which is lower than that of the other pots because it is not good conductor for heat. However, for cast iron, Mg concentrations showed an increase (229.344 mg/kg), could be due to the ability of Mg in dissolving in acidic media as zucchini turns into such media after cooking[6]. For Zn concentrations, all samples showed a decrease compared to the raw data (5.522-7.383 mg/kg *vs.* 8.588 mg/kg, respectively). The most notable decrease was observed in zucchini cooked in Al pot (5.522 mg/kg), which is due to the fact that Zn is water-insoluble[16]. Where the study agreed with the study[19], which showed that aluminum pots filtered the largest number of heavy metals that were filtered from some pots that exceeded the standard limits: ammonium (Al), nickel (Ni)) and iron (Fe). Transferred materials move from official materials to published materials The study also indicates that stainless steel did not result in leaching of nickel and chromium, perhaps due to the short cooking time. This was confirmed by a study[20] that stainless steel leaches nickel and chromium from foods during cooking, depending on the cooking time.

Table 2. Concentrations of different metals (mg/kg) in cooked zucchini using three different metal pots.

Heavy Metals (mg/kg)	Cookware Types			
	Uncooked (Control)/pH 9.0	Aluminum	Stainless steel (18/10)	Cast iron
Al	0.93	0.49	0.45#	0.72
Cr	<0.4	<0.4	<0.4	<0.4
Cu	0.522	0.489#	0.557	0.578*
Fe	3.785	13.88*	3.766	3.656#
Mg	215.21	188.326#	217.096	229.344*

Heavy Metals (mg/kg)	Cookware Types			
	Uncooked (Control)/pH 9.0	Aluminum	Stainless steel (18/10)	Cast iron
Ni	<0.4	<0.4	<0.4	<0.4
Zn	8.588	5.522#	6.412	7.383

***Represents the height from the raw sample - #Represents the decrease from the raw sample. Al: Aluminum; Cr: Chromium; Cu: Copper; Fe: Iron; Mg: Magnesium; Ni: Nickle; Zn: Zink.**

The Most Common Types of Cookware used by the Study's Sample

In this study, a survey was applied to a randomly chosen Saudi sample to determine the most used cookware among Saudi population(The Riyadh Province, The Makkah Province, The Eastern Province, The Madinah Province, The Al Baha Province, The Al Jawf Province, The Northern Borders Province, The Qassim Province, The Ha'il Province, The Tabuk Province, The 'Aseer Province, The Jizan Province and The Najran Province). The results as shown in **Table (3)**, demonstrated that the most used cookware was stainless steel, ffollowed by granite, aluminum, glass, cast iron, pride, stones, tefal, ceramic, iron, and melanin. (Stainless steel is a versatile material for cookware. You can easily find stainless steel pots and fringe pans, as well as griddles, lasagna pans, roasting trays, muffin tins, and baking sheets[5]

Table 3. Most common types of cookware used among a Saudi sample.

Cookware Types	Repeat	%	Rank
Stainless Steel	709	22.7025	1
Granite	603	19.3084	2
Aluminum	550	17.6113	3
Glass	538	17.2270	4
Cast Iron	294	9.4140	5
Pride	263	8.4214	6
Stone (Magash)	148	4.7390	7
Tefal	11	0.3522	8
Ceramic	3	0.0961	9
Iron &Melamine	2	0.0640	10

Relationship between the Preferable or Chosen Cookware and the Samples' Characteristics

In this study, we investigated the relationship between the used cookware and the samples' income, age, and awareness level, as shown in **Table (4)**. The results demonstrated that with the increase of the samples' income the use of stainless steel and cast-iron cookware increased because it is expensive. There was a significant positive correlation between the use of these types of cookware and the increase in the samples' income (p-values= 0.000 and 0.021, for stainless steel and cast iron, respectively). In contrast, the other types of cookware were inversely correlated with the samples' income; with the decrease in income, the use of the other cookware increased.

Regarding the samples' age, the results showed that the use of cast iron, stainless steel, aluminum, granite, and stone was correlated with the samples' age ($p \leq 0.05$). The use of aluminum and stainless steel was significantly correlated with the individuals' awareness (p-values= 0,000 and 0.002, for aluminum and stainless steel, respectively). This correlation is related to the samples' choice in preferring stainless steel over aluminum due to their beliefs that aluminum can cause adverse effects on the health whereas stainless steel has no potential adverse effects.

Table 4. The correlation between the Saudi sample's income, age, and awareness level and their choice in using between the different cookware.

Heavy Metals	Correlation of Household Income to the Type of Cookware used		Correlation of Age to the Type of Cookware used		Correlation of Awareness Level to the Choice of Cookware	
	Degree	*p*-value*	Degree	*p*-value*	Degree	*p*-value*
Cast Iron	9.823	0.021	15.637	0.001	0.338	0.845
Aluminum	0.952	0.813	7.927	0.048	27.501	0.000
Granite	3.271	0.352	10.073	0.018	4.294	0.117
Glass	5.704	0.127	5.944	0.114	4.817	0.090
Pride	0.733	0.865	2.370	0.499	5.371	0.068
Stone (Magash)	3.941	0.268	9.027	0.029	2.299	0.192
Stainless Steel	17.924	0.000	52.823	0.000	12.857	0.002

****p*-values ≤0.05 indicates a significant correlation.**

Relationship between the Types of Cookware used and the Samples' Health Status

The relationship between the different types of cookware used by the study sample and their health status was investigated as well, **Table (5)**. The results demonstrated that those who use cast iron had a higher incidence of diabetes, arthritis, and heart diseases ($p \leq 0.05$). Cooking with cast iron, mainly acidic foods, can cause iron to leak into the cooked food leading to its storage in the body's organs. The build-up of iron in the body's organs may increase the risk of different diseases such as diabetes[22]. In addition, results showed that those who use stainless steel had a higher incidence of arthritis, diabetes, and liver cancer. Stainless steel cookware is made of a combination of nickel, chromium, and iron[4]. Leaked nickel and chromium in the food have been linked to the development of arthritis[23], whereas iron was linked to the development of diabetes and liver cancer[24].

Table 5. The types of cookware used by the Saudi sample and their health status.

Cookware Type	Health Status	Degree	*p*-value
Aluminum	Alzheimer	5.053	0.025
	Hair loss	81.57	0.000
	Distraction of focus	36.89	0.000
	No symptoms	81.031	0.000
Cast Iron	Diabetes millets	306.052	0.000
	Arthritis	40.040	0.000
	Heart disease	8.38	0.004
	Liver cancer	0.52	0.471
	Cirrhosis	0.87	0.351
	No symptoms	127.81	0.000
Stainless Steel	Diabetes millets	46.29	0.000
	Arthritis	16.50	0.000
	Heart disease	0.91	0.340
	Liver cancer	11.14	0.001
	Cirrhosis	1.22	0.268
	No symptoms	66.62	0.000

***p*-values ≤0.05 indicates a significant correlation**

Conclusions

In this study, it has been shown that choosing the preferable cookware can be based on the pH of the food. Acidic food is recommended to be cooked in cast iron due to the inability of cast iron to interact with the acidic media. However, for the alkalized foods like Meat, Eggplants and Carrots, it is preferable to use aluminum cookware due to its lower heat conduction ability, as alkalized food can interact with the cookware's metals under higher temperatures. In general, it is preferable to use stainless steel for cooking all types of foods with no matter of the pH value. However, for individuals who suffer from rheumatoid arthritis, it is recommended to avoid using stainless steel due to its possibility of leaking nickel and chromium into the food leading to more health complications regarding arthritis. Moreover, it is preferable to avoid using aluminum cookware due to its potential negative impact on health generally.

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