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The Effect of Using Scratched Stewpots on Food Contamination with Heavy Metals

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تأثير استخدام أواني الطهي المخدوشة على تلوث الطعام بالعناصر الثقيلة

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Abstract:
Heavy metals can be released from cooking stewpots into food, causing food contamination and posing a risk to human health. This research study investigated the release of heavy metals into food from stewpot cookers used in Gharyan, Libya. As well as, investigating the effect of scratches, acidic food, and storage on the concentrations of those metals. Eight new and used cooking stewpots (with scratches) made of: (Tefal, stainless steel, copper and aluminum) were collected, and then fresh tomato juice was cooked in these stewpots and left in the refrigerator for six days, the samples were digested and analyzed by atomic absorption spectrophotometer (AAS) to measure the concentration of the heavy metals: (cadmium, lead, iron, copper and zinc). Results showed that cadmium and lead concentrations were less than the detection limit of the AAS instrument, except for one sample (old Tefal stewpot with scratches), in which the lead concentration was (0.046ppm). The concentration of iron, copper and zinc in the samples was less than the maximum limit allowed by WHO/FAO; except for one sample in which the iron concentration exceeded that value (old copper pot with scratches) and its value was (2.892ppm). The results of the descriptive statistical analysis showed that the average concentration of iron (0.493 ppm) was higher than that of zinc and copper (0.054 ppm), (0.016 ppm), respectively. In addition, the rate of leakage of those elements from old (scratched) cookware was higher than that from the new ones made from the same material, and the concentration of these elements increases significantly when these stewpots contain scratches, since that increases the rate of leakage of those metals, especially in acidic food such as tomatoes.

Keywords: Stewpots, Heavy Metals, Leaching, Atomic Absorption.

الملخص:
يمكن أن تتحرر المعادن ثقيلة من أواني الطهي إلى الطعام، مما يتسبب في تلوث الطعام ويشكل خطرًا على صحة الإنسان. أجريت هذه الدراسة البحثية للتحقيق في إطلاق المعادن الثقيلة في الأغذية من أوعية الطهي المستخدمة في غريان، ليبيا. وكذلك دراسة تأثير الخدوش والأغذية الحمضية والتخزين على تركيزات تلك المعادن. تم جمع ثمانية قدور طهي جديدة ومستعملة (بها خدوش) مصنوعة من: (تيفال والستانلس ستيل والنحاس والألمنيوم)، ثم تم طهي عصير الطماطم الطازج في هذه القدور وتركت في الثلاجة لمدة ستة أيام، ثم تم هضم العينات وتحليلها بواسطة جهاز مطياف الامتصاص الذري (AAS) لقياس تركيز المعادن الثقيلة: (الكادميوم، الرصاص، الحديد، النحاس، الزنك). أظهرت النتائج أن تركيزات الكادميوم والرصاص كانت أقل من حد الكشف بجهاز AAS، باستثناء عينة واحدة (قدر تيفال القديم به خدوش) حيث بلغ تركيز الرصاص (0.046 جزء في المليون). وكان تركيز كل من: الحديد والنحاس والزنك في العينات أقل من الحد الأقصى المسموح به من قبل منظمة الصحة العالمية/منظمة الأغذية والزراعة؛ باستثناء عينة واحدة تجاوز فيها تركيز الحديد تلك القيمة (وعاء نحاس قديم به خدوش) وبلغت قيمته (2.892 جزء في المليون). أظهرت نتائج التحليل الإحصائي الوصفي أن متوسط تركيز الحديد (0.493 جزء في المليون) أعلى منه في الزنك والنحاس (0.054 جزء في المليون)، (0.016 جزء في المليون) على التوالي. إضافة إلى ذلك، فإن معدل تسرب تلك العناصر من

الأواني القديمة (بها خدوش) كان أعلى من تلك الموجودة في أواني الطبخ الجديدة المصنوعة من نفس المادة، كما أن تركيز هذه العناصر يزداد بشكل ملحوظ عند احتواء هذه القدور على خدوش، حيث أن ذلك يزيد من معدل تسرب تلك المعادن، وخاصة في الأغذية الحمضية مثل الطماطم.

الكلمات المفتاحية: أواني الطهي، عناصر ثقيلة، تسرب، امتصاص ذري.

Introduction

Foodstuff safety is an important food quality attribute, and a major issue of public concern [1]. In recent years, some heavy metals have been reported to affect food security and human health [2]. These metals, which have a specific density > 5 g/cm^3, include cadmium (Cd), copper (Cu), iron (Fe), lead (Pb), zinc (Zn), chromium (Cr), nickel (Ni) and manganese (Mn).

Some of these metals, such as Fe and Cu, are necessary nutrients for various biochemical and physiological functions; however, excessive amounts of such necessary metals cause a variety of adverse effects and human illnesses. Other heavy metals, such as Pb and Cd, are toxic, and their toxicity is affected by several factors, including the dose, exposure route, and chemical species, as well as the age, gender, and dietary intake and nutrition status of those who are exposed [3]. Severe and prolonged toxic effects of heavy metals affect different body organs leading to complications that include gastrointestinal and kidney dysfunction, nervous system disorders, immune system dysfunction, and cancer. In addition, cumulative effects occur with exposure to two or more metals simultaneously [4-6].

Heavy metals can enter our bodies through the skin, inhalation, drinking contaminated water, or consumption of contaminated food [3-7], and any heavy metal that presents in food and reduces the safety or quality of food is considered a food contaminant [8]. Food contamination can occur at different stages. It can occur when raw materials are being transported, or when food is being conditioned, which involves the storage of raw materials, preheating, disinfection, and cleaning. It can also occur during heating steps either by boiling, baking or frying. In addition to that food contamination can occur when food is being packaged, as well as at the transport, storage and distribution of packaged food [9].

Metals and alloys are commonly used for the production of cookware, including cooking stewpots, due to their high thermal conductivity and thermostability. The most commonly used metals include Aluminum (Al), Cu, Fe, Cr, and Ni; however, toxic heavy metals can be included in the manufacturing process as contaminants or impurities. This indicates that metal exposure may be a concern for people who use metallic cookware in their daily lives, particularly while boiling water and cooking food [10]. Several studies have demonstrated that heavy metals can leach from cooking utensils into food [6, 11-13] and that the release of these metals from cookware into food has a positive relationship with cooking time and container age due to frequent use and damage or abrasion [14].

People in Gharyan, Libya, where this study was conducted, use a variety of stewpots, which could be potential sources of metal release into the prepared food, posing a health risk to the population. To the best of our knowledge, no data from Libyan researchers on the release of heavy metals into food from cookware has been reported. Therefore, this study aimed to investigate the release of heavy metals into food from stewpots used in Gharyan, Libya.

Materials

Concentrated nitric acid (HNO_3), (E-MERCH DARMSTAD), concentrated hydrochloric acid (HCl), (WIN LAB). Triple distilled water (3D-H_2O), provided by the Libyan Oil Institute (Gargarash, Tripoli). Standard solutions for the elements lead, cadmium, iron, copper and zinc at a concentration of 1000 ppm, (VWR PROLABO, UK).

Methods

Eight cooking stewpots were collected: two of them were made of Tefal (new and old (with scratches), two of them were made of stainless steel (new and used), two of them were made of copper (new and used) and the last two were made of aluminum (new and used), some of them are shown in figure 1. Then, a quantity of fresh tomatoes was collected, chopped and divided to prepare eight samples in equal quantities and each sample was cooked in one of the collected cooking stewpots for equal periods of time, then each of the samples was digested and placed in polyethylene containers. All these steps took place at the laboratory of the Faculty of Science at Gharyan University/ Gharyan. The analysis process was conducted at the laboratory of the Libyan Oil Institute located in the Gargarish/Tripoli. The concentration of heavy metals was measured, namely: lead, cadmium, iron,

copper and zinc, in order to determine the extent of heavy metals leakage into food during the cooking process and the effect of scratches in cooking stewpots on the rate of this leakage.

A B C

Figure 1 Some kinds of the cooking stewpots used in this research.

A. Tefal, B. Stainless steel, C. Aluminum

Procedure:

Samples of fresh tomatoes was chopped using electric blender, then 10 grams of each tomato sample was weighed and cooked in one of the above-mentioned types of stewpots. After that, 30 ml of distilled water was added three times in three stages (10 ml each time) for two hours at temperature of 40º C, until water evaporated. Then, the samples were covered and placed in the refrigerator for six days.

Digestion stage:

1 g of each of the prepared tomato samples was weighed and placed in a clean beaker and 1ml of concentrated nitric acid and 3 ml of concentrated hydrochloric acid were added to it, (ratio of 1:3). The acids were added in three stages in the hood until the digestion process was complete. If the reaction was slow, the beaker was placed on the heater at a temperature of 40ºC, stirred with glass rod, and then covered for an hour until the reaction ended. Then the sample was left to cool. It was filtered into a glass beaker, then the filtrate was transferred to 50ml standard flask and the volume was completed with distilled water up to the mark. 25 ml of the solution was measured and stored in 25 ml polyethylene container and placed in the refrigerator until it was transported to the laboratory for measurement.

Instrumentation and measurement conditions:

Fast Sequential Atomic Absorption Spectrophotometer AA240 FS (VARIAN), error was corrected by Reference Materials, flame temperature was 3000ºC, sample flow rate inside the flame was 3-5 ml/min, gas used was acetylene with air, radiation source (hollow cathode lamp) for each of the analyzed elements. Table1 shows the wavelengths of each analyzed element [15], detector used double photocell.

Table 1 The wavelength of the analyzed elements.

Element	Fe	Cd	Pb	Zn	Cu
Wavelength (nm)	248	288.8	213.3	213.9	324.8

Results and discussion

Some heavy metals are necessary for healthy and normally growing for the human body, but the increase in their concentration and accumulation in the body in the long term may cause several health problems [16, 17]. The results of this research showed variation in the concentration of heavy metals in different cooking stewpots. Table 2 shows the results of heavy metal concentrations that were measured in tomato samples that were cooked in the cooking stewpots under study. As shown in the table, the cadmium concentration in all samples was less than the detection limit of the instrument and was excluded from the discussion of the results. This is also the case for lead, except

for one sample, which is the sample of the old Tefal utensil (with scratches), and its value was 0.046 ppm, this value did not exceed the maximum limit allowed by the WHO/FAO organizations, which is (0.05mg/kg) [18-20]. This result indicates that scratches on the surface of utensils made of Tefal may lead to the leakage of heavy elements such as lead into the food, especially in acidic environments (tomatoes), and its long-term accumulation may lead to health problems for the consumer [21].

Table 2 Concentration of lead, iron, copper, zinc and cadmium in ppm in tomato samples that were cooked in the cooking stewpots under study.

Stewpot Type	Concentration/ppm				
	Cd	Zn	Cu	Fe	Pb
Old Tefal(scratched)	ND	0.058	0.016	0.171	0.046
New Tefal	ND	0.036	0.024	0.074	ND
Old Copper(scratched)	ND	0.050	0.024	2.892	ND
New Copper	ND	0.034	0.032	0.378	ND
Old Stainless steel(scrached)	ND	0.061	0.001	0.038	ND
New Stainless steel	ND	0.064	0.002	0.064	ND
Old Aluminum(scratched)	ND	0.083	0.023	0.251	ND
New Aluminum	ND	0.043	0.003	0.076	ND

ND = not detected

Results of statistical analyzes and their interpretation:

The results of the descriptive statistical analysis of the concentration of iron, copper, and zinc for the samples that were cooked in the cooking stewpots under study (Table 3) showed that the average concentration of iron (0.493 ppm) in these samples is higher than the average concentrations of zinc and copper (0.054 ppm, 0.016 ppm), respectively, for the same samples; This may be due to the fact that most traditional cooking stewpots contain high percentage of iron in their manufacturing materials and the scratches and damage in these pots increase the possibility of iron transferring to the food [22]. As shown in Table 2, the highest value recorded for iron in the sample of the copper pot that contains scratches is (2.892 ppm), followed by the sample of the new copper pot (0.378 ppm), which indicates that the cooking pot made of copper contains high percentage of iron.

Table 3 Results of descriptive statistical analysis of the samples.

Metal	mean/ppm	Standard deviation/ppm	Maximum Value/ppm	Minimum Value/ppm
Iron Fe	0.493	0.976	2.892	0.038
Copper Cu	0.016	0.012	0.032	0.001
Zink Zn	0.054	0.016	0.083	0.034

It was also found that the concentration of iron in the tomato sample that was cooked in the old aluminum pot was 0.251 ppm, which is much higher than its concentration in the sample that was cooked in a new pot of the same type (0.076 ppm), which confirms that scratches in old pots may cause more iron to leak into the food.

This can also be observed when comparing the iron concentration in old pots made of Tefal and stainless steel (with scratches) and new ones, (Table 2). These results are similar to the results

obtained from previous studies [6, 23], as these studies showed that the amount of iron that leaks from cookware that has scratches into the food is more than its amount from unused cookware of the same type. This leakage increases in the presence of acidic substances [24], such as tomato sauce.

By comparing the iron concentration results obtained in this research with the maximum allowed by WHO\FAO, which is (0.8mg/kg) [18, 20], it was found that only one sample (the old copper pot) of these samples exceeded its value (2.892ppm) maximum as shown in Figure 2; Although iron is considered essential element in cellular metabolism and oxygen transport, and its deficiency leads to anemia, exceeding the permissible limit may be harmful to health [6, 24].

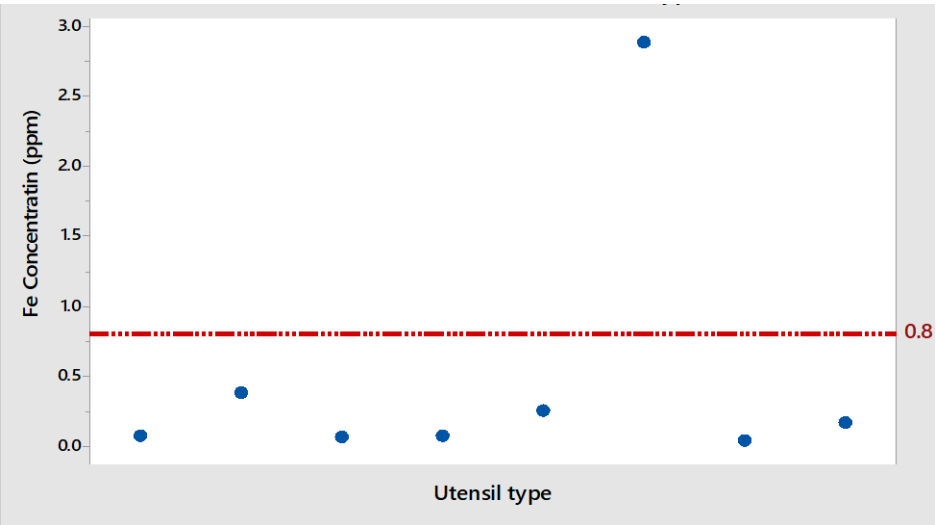

Figure 2 Concentration of iron in tomato samples vs. maximum concentration allowed by WHO\FAO.

As for the copper element, the results of this research showed that the copper concentration in the studied samples ranged between 0.001ppm and 0.032 ppm, with a mean value of 0.016 ppm and a standard deviation of 0.012, as shown in Table 3, and that the highest value was for the sample that was prepared in copper stewpots. These values were within the maximum allowed by WHO\FAO, which is (0.1mg/kg) [18-20], also shown in Figure 3; the concentration of copper in all samples did not exceed this value. Copper is considered an essential mineral for the human body, but exceeding the permissible limit may cause several health problems [25].

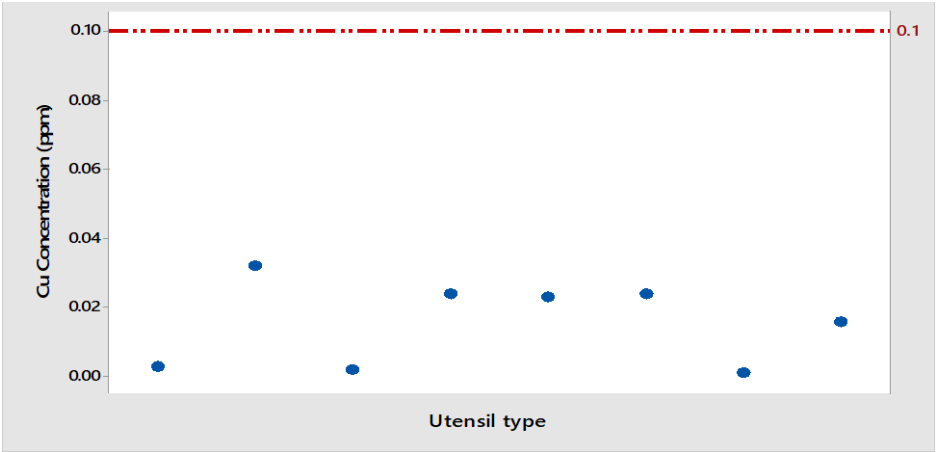

Figure 3 Concentration of copper in tomato samples vs. maximum concentration allowed by WHO\FAO.

The concentration of zinc in the samples ranged between 0.034 ppm and 0.083 ppm, with average of 0.054 ppm, and standard deviation of 0.016 ppm (Table 3). The highest value of zinc was in the sample that was cooked in a stainless-steel pot, followed by the sample that was prepared in an aluminum utensil. The reason for that could be that stainless steel utensils contain 5.59% zinc, while aluminum pots contain only 0.05% [26].

According to WHO/FAO, the maximum permissible concentration of zinc is (10mg/kg) [18- 20], and therefore none of the samples exceeded this limit (Figure 4); however, zinc leakage from cooking utensils into food leads to its accumulation in the body in the long term, which may cause damage to the liver, anemia, and other health problems [27].

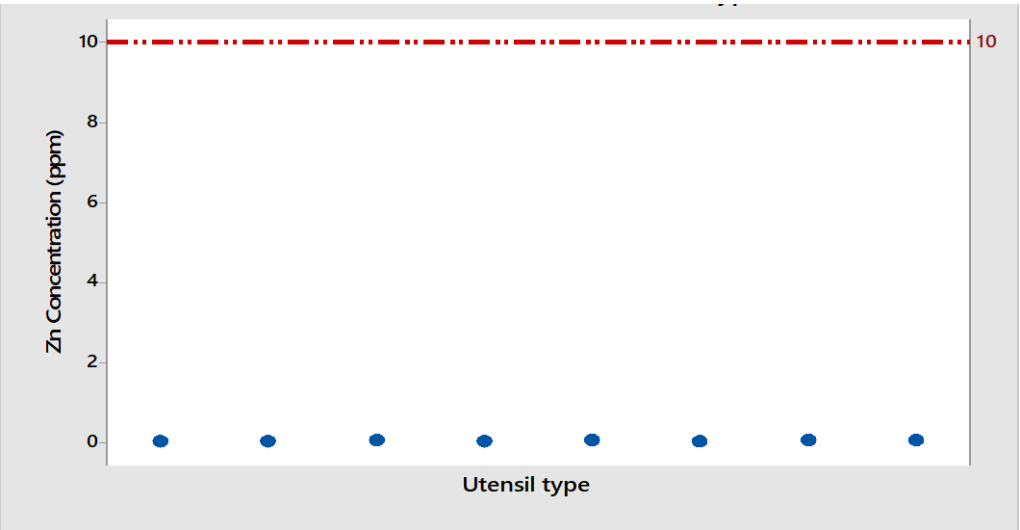

Figure 4 Concentration of zinc in tomato samples vs. maximum concentration allowed by WHO\FAO.

When performing ratio test by finding the ratio between the concentration of iron to copper (Fe/Cu), iron to zinc (Fe/Zn) and zinc to copper (Zn/Cu) in all samples; to compare these elements in terms of their leakage into food from different cooking stewpots. The results of this test (Table 4) showed that the concentration of iron in all samples is higher than the concentrations of zinc and copper, but the percentages vary due to the difference in the percentage of iron in the material in which the pot is made, as well as its condition (new or damaged). As shown in Table 4, these percentages were higher in old pots compared with new ones of the same type, which confirms that the leakage of the iron increases with scratches in the cooking utensils. For example, in the stainless steel pot (with scratches), the ratio of iron to copper was 120.5, while their ratio in the new stainless steel pot was only 10.9, which indicates that the leakage of iron into food is higher than copper leakage. That could be due the fact that stainless steel vessels contain 70.59% iron in their composition, and only 1.92% copper [28]; the presence of scratches in the stewpots leads to a greater amount of iron leaking into the food.

Table 4 Results of ratio test.

Stewpot Type	**Ratio**		
	Zn/Cu	**Fe/ Zn**	**Fe/Cu**
Old Tefal(scratched)	3.6	2.9	10.6
New Tefal	1.5	2.1	3.1
Old Copper(scratched)	32.0	1.0	32.0
New Copper	1.1	11.1	11.8
Old Stainless steel(scrached)	2.1	57.8	120.5
New Stainless steel	3.6	3.0	10.9
Old Aluminum(scratched)	50.5	5.1	38.0
New Aluminum	14.3	1.7	25.3

The values of iron and copper were close in most samples, although there was a variation in the values, due to the difference in the concentration of the two elements in the material used to make the cooking stewpots studied.
This is what was shown by the results of the cluster analysis of the elements iron, zinc, and copper, shown in Figure 5. A similarity ratio of 68.51% was observed between iron and copper, and the similarity ratio between these two elements and zinc is only 35.99%. This similarity could be due to the fact that most types of cooking stewpots contain iron and copper and other elements in their composition, and these pots must be treated and covered with protection layer that prevents these and other elements from leaking into the food. When scratches occur in their surfaces, leakage increases especially in the presence of acidic food, heavy metals transfer to the food, and their long-term accumulation in the body leads to several diseases and health problems [29].

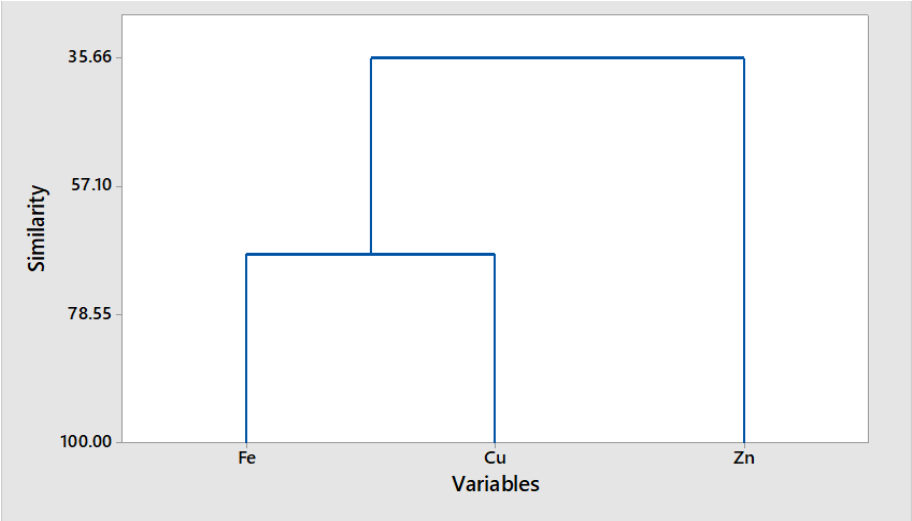

Figure 5 Results of cluster analysis of the concentration of iron, copper, and zinc metals in the samples.

When comparing the average concentrations of the three elements (Fe, Cu, Zn) in old stewpots (with scratches) to their counterparts in new ones, it was noted that the concentration of heavy metals in samples cooked in old stewpots is higher than in new ones, and this can be observed in Figures (6-9) for Tefal, copper, stainless steel and aluminum pots, respectively.
From the results, it can be seen that the various cooking stewpots used for cooking differ in the number of heavy elements that leak into the food during the cooking process. The concentration of these elements increased significantly when these pots contain scratches resulting from the use of sharp tools or as a result of damage to their inner surface in contact with food, especially acidic foods. Therefore, it is recommended to avoid using cheap, untreated cooking stewpots, as well as getting rid of old pots that have scratches because of their risks to human health.

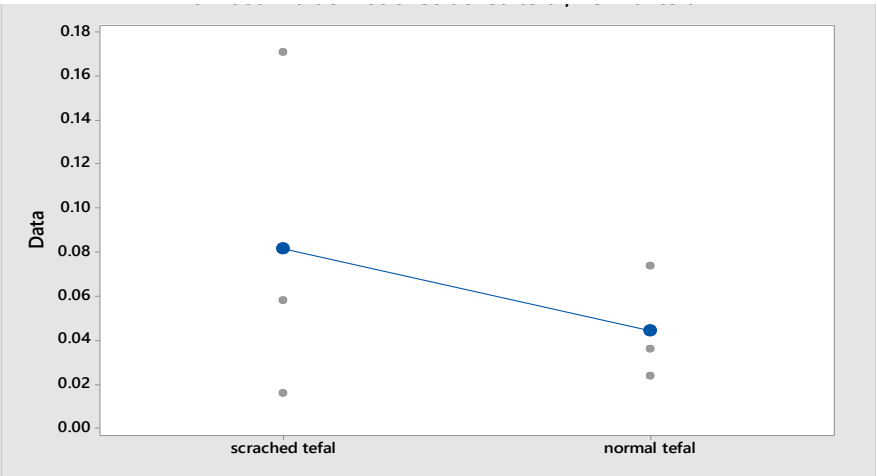

Figure 6 Results of comparing the average concentrations of elements (Zn, Cu, Fe) in old and new Tefal stewpots.

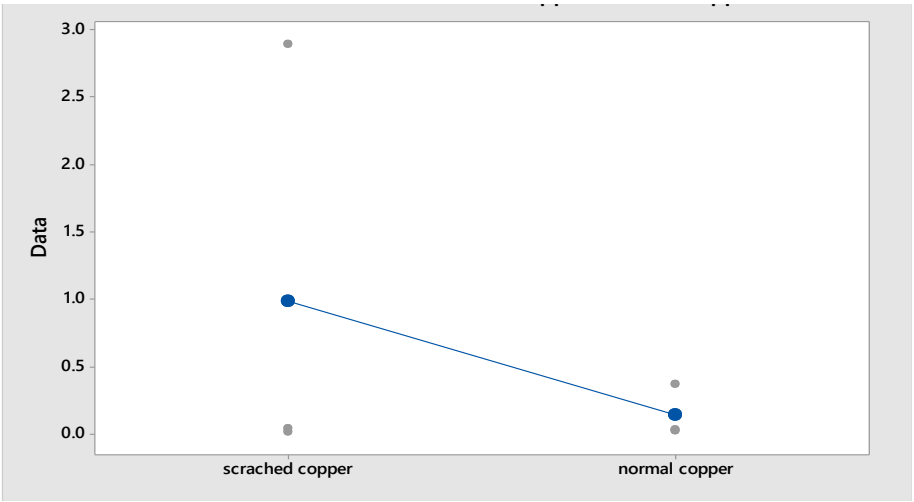

Figure 7 Results of comparing the average concentrations of elements (Zn, Cu, Fe) in old and new copper stewpots.

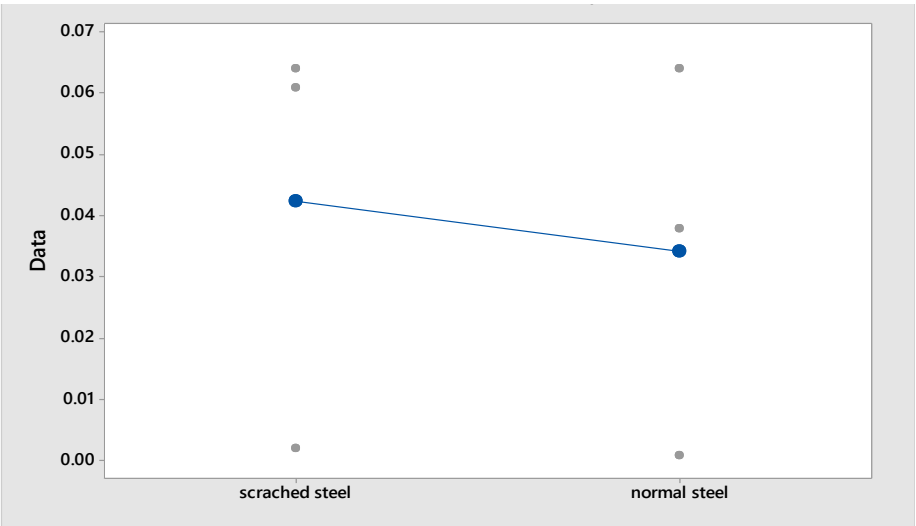

Figure 8 Results of comparing the average concentrations of elements (Zn, Cu, Fe) in old and new stainless steel stewpots

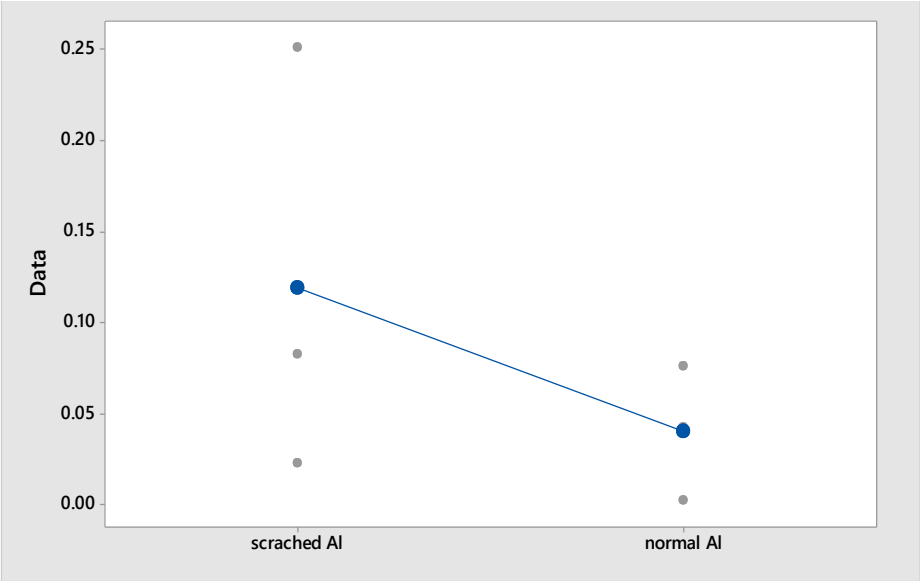

Figure 9 Results of comparing the average concentrations of elements (Zn, Cu, Fe) in old and new aluminum stewpots

Conclusion

Through this study, it can be concluded that cooking stewpots have a considerable effect on the leakage of heavy metals into food and that the repeated use of these pots could damage their inner surface when in contact with food. The presence of scratches in these pots also increases the rate of leakage of those metals, especially in acidic food such as tomatoes, and thus increases their accumulation in the human body, which in the long term may cause much harm.

According to the results of this research, the concentration of heavy metals in 99% of the samples studied did not exceed the maximum permitted by the WHO and FAO organizations. Only one sample exceeded this limit; despite that, repeated and daily use of cooking stewpots, especially those with scratches, could lead to the accumulation of heavy metals in the body, which may harm the body's cells in the long term and cause several diseases.

It is recommended to conduct more extensive research that includes other heavy metals found in cooking stewpots, such as aluminum, arsenic, and others. Also, conducting more extensive research that includes a larger number of samples and other types of cookware, and applying other statistical tests to provide more information. In addition, using more sensitive instruments for measuring the concentration of toxic heavy metals such as lead, cadmium, etc. As well as, ensuring that the stewpots used for cooking are made of high-quality, rust-resistant materials that can withstand high heat and are not easily scratched and avoiding the use of sharp tools when stirring food inside cooking pots and cleaning them.

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