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Aluminum Leaching during Food Preparation and Storage

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ABSTRACT

Aluminum (Al) is extensively used in many industrial sectors, including those dealing with transportation, electricity, machinery, equipment, food processing, packaging, storage, and a wide range of home appliances. Fruits and vegetables, which might have varied quantities depending on the plant's capacity to absorb Al, are usually the main dietary exposure sources. Depending on a variety of variables, such as cooking temperature, technique of cooking, and type of additives used, using Al utensils leads to a significant addition of the metal to the diet.This research focuses on studying the effect of food pH, cooking time and storage period on the leaching of Al from three cooking utensils (high quality aluminum pot, low quality aluminum pot and clay pot) into cooked organic tomato and spinach. The results demonstrated that the leaching of Al from pots of low-quality aluminum was greater than that of the other pots recording 5.94 and 13.09 mg Al/kg for tomato and spinach samples cooked in low quality aluminum pots, respectively, followed by high quality aluminum pots cooked tomato and spinach samples reaching 4.59 and 8.94 mg/kg, respectively after 30min of cooking. On the other hand, the clay cooked tomato and spinach samples recorded the lowest (Al) content reaching 1.33 and 5.44 mg/kg, respectively after 30min of cooking.

Keywords: Aluminum, Clay pot, Tomato, Spinach, Storage

INTRODUCTION

Aluminum (Al) is the third most prevalent element in the Earth's crust by weight, after oxygen and silicon, making up around 8.1% of the crust overall (Sjögren *et al.,* 2015). Alum, also known as aluminum potassium sulphate, was first used by the Egyptians 5,000 years ago as a color fixative, and the Greeks used anhydrous aluminum sulphate as an astringent to cure wounds (Yokel, 2016). Due to its qualities including low weight and density, ductility, high electrical and thermal conductivity, resistance to corrosion, and outstanding formability, aluminum is a particularly useful metal. As a result, it is frequently utilized in a variety of industries, among which are those that involve with transportation, electricity, building and construction, machinery, and equipment, food processing, packaging, and storage, as well as a wide range of home appliances. It is used in the manufacturing of personal hygiene and health care items such as toothpaste, antiperspirants, acne treatments, dermal care, antacids/antiulcerative medications, vaccines, and antidiarrheal medications (Gándara, 2013; Ng *et al.,* 2017; and Rahimzadeh *et al.,* 2022). Moreover, it is frequently used as a food ingredient as a buffer, firming agent, dyeing agent, anticaking agent, neutralizing agent, dough stabilizer, thickener and curing agent (Yeh *et al.,* 2016 and Yokel, 2016).

The main source of dietary exposure is often found in fruits and vegetables, which can have different levels depending on the plants' capacity to store aluminum, such as tea plants, which can contain deposits of the metal in their leaves (Yeh *et al.,* 2016). Aluminum cookware, such as drink containers, coffee pots, grill pans, pressure cookers, skillets, and saucepans, as well as foil used as wrappers, is one of the potential sources of extra dietary (Al). Depending on an array of variables, such as cooking temperature, technique of cooking, and type of additives used, using aluminum utensils leads in a significant addition of the metal to the diet (Semwal *et al.,* 2006). Aluminum utensils are manufactured from waste metals and recycled aluminum-containing alloys in underdeveloped nations. A number of toxic compounds, including heavy metals like cadmium, lead, nickel, arsenic, copper, and aluminum, have been documented to leach into cooked food as a result of such impurities present in the utensils, which could accelerate the dissolving of metal. According to studies, a number of criteria influence how rapidly metals leach from aluminum cookware (Alabi and Adeoluwa, 2020). Aluminum's relatively low bioavailability made it generally recognized as safe (GRAS) for human health for a very long time. Nonetheless, it has been established that they pose health risks once consumed (Verssimo and Gomes, 2008). Aluminum's total weight in a healthy person is between 30 and 50 mg. 25% of the weight is in the lungs, where the amount of aluminum rises with ageing, and around 50% of the weight is in the skeleton. Aluminum concentrations in bone tissue range between 5 and 10 mg/kg. Healthy individuals have serum levels around 1-3 µg/L. (Larsen, 2008). Between 75% and 95% of the aluminum we eat or drink is absorbed by our bodies; the remainder is absorbed and may accumulate in several organs, including the brain, liver, kidney, thyroid, and lungs (Al Juhaiman, 2012). According to studies, youngsters have a low tolerance for this non-essential element and are particularly susceptible to (Al) exposure because of their developing renal systems (De Paiva *et al.,* 2020). Furthermore, a higher plasma aluminum concentration has also been linked to increased DNA damage, oxidative stress, and

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neurodegenerative/neurodevelopmental illnesses such multiple sclerosis, Parkinson's disease, and Alzheimer's disease (Celik *et al.,* 2012). Alzheimer's disease, autism, and epilepsy patients' brain tissue has recently been found to contain increased levels of aluminum (Mold *et al.,* 2019). In comparison to people without dementia, Alzheimer's patients accumulate 2-3 times more aluminum in their cortices and hippocampus (Samir and Rashed, 2018, Yang *et al.,* 2019). According to data from a different study, people who ate meals with high aluminum content had a twofold increased probability of developing Alzheimer's disease (Rogers and Simon, 1999).

Based on toxicological research and exposure, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a Provisional Tolerable Weekly Intake (PTWI) for (Al) of 7 mg/kg body weight (bw) in 1990. In 2008, the JECFA reexamined the safety of aluminum and reduced the PTWI to 1 mg/kg bw due to the possibility that aluminum could damage the nervous and reproductive systems of experimental animals. But in 2011, the Committee added a new modification that included a suggested PTWI of 2 mg/kg bw in response to additional bioavailability and toxicological information (Sato *et al.,* 2014 and EL Daouk *et al.,* 2022).

For centuries, we have been cooking in clay or earthen pots (Tajen) for ages. Meals cooked in clay pots will stay hot and moist for a longer period of time, allowing for cooking with less liquid and fat than usual (Paranjape and Kulkarni, 2018). It is among the most widely available and economical refractory raw materials. Compared to other cookware materials like steel, iron, or aluminum, clay has some advantages. It takes a while for the heat to be absorbed, after which it distributes the heat evenly throughout the clay pot's body before slowly releasing it into the food that is cooking within. Clay's partially porous nature allows heat and moisture to circulate through the pot, promoting gradual and even cooking. By slow cooking, flavors can develop gradually, spices can permeate a dish more thoroughly, and in the case of meat, tough slices can become succulent. From a health perspective, clay is a natural substance free of potentially harmful substances that could react with food, cooking in it helps in the ingredients' nutritional and vitamin retention. The meal keeps its authentic flavors. Moreover, clay is eco-friendly and biodegradable (Fwalo *et al.,* 2017). Thus, the aim of this study is to investigate the effect of food pH, cooking time and storage period on the leaching of aluminum from three different cooking utensils (high quality aluminum pot, low quality aluminum pot and clay pot) into cooked organic tomato and spinach.

MATERIALS AND METHODS

Materials:

Three types of pots were utilized in this study, high quality aluminum pot (HqAl), low quality aluminum pot (LqAl) and clay pot (C). Aluminum pots were purchased from local market, while clay pot was purchased from the manufacturer in Naj Hammadi, Qena governorate. Organic tomato (*Solanum lycopersicum L.*) and spinach (*Spinacia oleracea L.*) were obtained from a local market in Cairo, Egypt.

Methods:

Sample preparation:

Fresh organic tomatoes and spinach were cleaned with tap water and homogenized. In each of the three pots, two kilograms of tomato and two kilograms of spinach were cooked. To facilitate the cooking process, 100 ml of tap water per kg of spinach was added. Samples were withdrawn from the pots after 10, 20 and 30 min of cooking and after1, 12, 24 and 48 hours of storage at 4°C. Samples were stored in polyethylene bags and frozen until analysis.

pH measurement:

The pH values of fresh tomato and spinach samples were measured using Jenway 3510 pH Meter.

Aluminum content determination:

The aluminum content of water, clay pot, fresh, cooked and stored tomato and spinach samples were quantified using triple quadrupole Inductively Coupled Plasma Mass Spectrometry (Agilent 8800 ICP-QQQ, Agilent technologies, Perkin Elmer Inc., USA) on a single run using appropriate cell gases and internal standards according to Geiger and Soffey (2020). Results were expressed as mg/kg fresh weight (Fw).

Statistical analysis

The obtained results were statistically analyzed using the SPSS program version 20. *P*-value of less than 0.05 was considered significant.

RESULTS AND DISCUSSION

(Al) is present in the majority of natural water sources, including rivers, lakes, groundwater, coastal sea water, and open ocean water, with trace concentrations of about 0.15, 0.4, 0.1, 0.001-0.007, and 0.001 mg/L, respectively. As a result, it can be found as a trace element in numerous foods and drinking water (Yokel, 2016). The World Health Organization, however, advised that the concentration of aluminum in drinking water should not exceed 200 µg/L. (World Health Organization, 2003). The Al content of tap water used with spinach samples was 0.004 mg/L which is negligible.

Effect of cooking time on aluminum content in tomato and spinach cooked in different pots:

According to results shown in Table (1) aluminum concentration in fresh spinach sample recorded higher (Al) content than fresh tomato sample reaching 4.44 and 0.82mg/kg, respectively. In accordance with our results, leafy vegetables including spinach, lettuce, water spinach and Chinese cabbage (baby bok choy) were reported to be high in aluminum (Liang *et al.,* 2019). It has been demonstrated that leafy vegetables have a significant capacity to accumulate aluminum and other metals from soil throughout the growth phase (Stahl *et al.,* 2017, Xu *et al.,* 2013 and Yokel, 2016).

Table 1. Effect of cooking time on aluminum content mg/kg (Fw) in tomato and spinach cooked in high quality aluminum pot (HqAl), low quality aluminum pot (LqAl) and clay pot (C).

	HqAl		LqAl		C	
Cooking time	**Tomato**	**Spinach**	**Tomato**	**Spinach**	**Tomato**	**Spinach**
0 min	0.82^{n}	4.44^{j}	0.82^{n}	4.44^{j}	0.82^{n}	4.44^{j}
10 min	1.99^{l}	5.02^{h}	2.47^{k}	8.49^{d}	1.13^{m}	4.89^{i}
20 min	2.81^{k}	7.18^{e}	4.59^{i}	11.01^{b}	1.26^{m}	5.20^{g}
30 min	4.59^{i}	8.94^{c}	5.94^{f}	13.09^{a}	1.33^{m}	5.44^{g}

Different letters indicate significant differences at (*P*<0.05). LSD 0.353
HqAl:high quality aluminum pot, LqAl: low quality aluminum pot, C:clay pot

Data in Table (1) revealed that (Al) content in tomato and spinach samples cooked in the three different pots significantly increased during cooking. There were numerous factors that contributed to the elevated level of (Al) including the type of utensil (steel, aluminum, earthen), temperature, the duration that the foodstuff was in contact with the pot, as well as the nature of the food (Jabeen *et al.*, 2016). Mohammad et al. (2012) and Al Juhaiman (2012) reported that longer cooking time elevates the leaching of aluminum in all food solutions used in their study. Heating of aluminum cookware increased the rate of Al migration to cooked food (Alasfar and Isaifan, 2021 ; Ertl and Goessler, 2018).

It could be observed that LqAl cooked tomato and spinach samples recorded the highest (Al) content reaching 5.94 and 13.09 mg/kg, respectively, followed by HqAl cooked tomato and spinach samples reaching 4.59 and 8.94 mg/kg, respectively after 30min of cooking. On the other hand, the C cooked tomato and spinach samples recorded the lowest (Al) content reaching 1.33 and 5.44 mg/kg, respectively after 30min of cooking. Clay pots were tested for aluminum content, which was quantified and found to be 6.97%. Al Juhaiman (2010) noted that the differences in (Al) leaching were affected by the composition of the alloyed components. Moreover, the concentration of (Al) in food samples differed between two different types of (Al) pans, ranging from 0.009 to 2.48 mg/L. (Stahl *et al.*, 2017). However, when clay and metal kitchenware were evaluated as potential sources of heavy metal contamination, it was discovered that metal pots contributed the most heavy metals. Significant sources of these heavy metals include the composition of rocks used to create the raw materials for clay pots and the composition of the scrap metal used to create metal pots (Uriah *et al.* 2014). Meanwhile, Arunal *et al.* (2020) observed that cooking food in earthenware may help us avoid the harmful effects of toxic elements in our daily diet. The most effective way to prevent the negative health impacts might be to use clay cookware. The same table's data revealed that (Al) increment rate in cooked tomato samples (pH 3.5) was significantly higher than in spinach (pH 5.7).These findings concur with those provided by Szefer and Nriagu (2006), who found that the high acidity of tomato puree can account for its high (Al) content, which is released from the (Al) container that it is packaged in, compared to non-acidic meals. In addition, it was observed that an acidic additive could raise the quantity of (Al) in cooked food by 46.85 mg/kg, compared to a non-acidic additive's rise of 20.38 mg/kg (Osman *et al.*, 2017). Moreover, samples with high acid concentrations showed greater increases in total (Al) content than the other samples under study, according to Ertl and Goessler (2018) and Duru and Duru (2020). When oxygen is in contact with aluminum, a very thin layer of aluminum oxide (Al_2O_3) is created; this layer is tough, non-flaking, colourless, and can be dissolved by only a few chemicals. It quickly changes into a layer of corrosion on the (Al) surface, especially when the pH is below 4.5, increasing the amount of (Al) in the aqueous medium and simultaneously exposing a new, active (Al) surface to more action. Food acidity or high salt concentration can encourage migration (Chappard, 2016 and Fermo *et al.*, 2020). On the other hand, Fwalo *et al.* (2017) illustrated that because clay is naturally alkaline; it helps to neutralize the pH of sour or bitter acids in food with ingredients like tomatoes, mellows the dish and imparts a natural sweetness to it as a result when clay comes into contact with the food while it cooked.

Effect of storage time at 4°C on aluminum content in tomato and spinach:

The results presented in Table (2) elucidated that during storage (1, 12, 24 and 48hr) at 4°C of cooked tomato and spinach (Al) content significantly increased in all cooked tomato and spinach samples recording 23.63, 22.18, 49.39, 44.74, 9.61 and 11.64 for HqAl, LqAl and C, respectively after storage for 48hr. However, it could be observed that cooked tomato samples recorded higher increment rate than cooked spinach samples. It is well known that using Al utensils for cooking and storing food increases its (Al) content. Temperature, pH, the duration of contact, and the presence of several food additives including sugar, salt, and organic acids are factors that affect the amount of Al leaching from (Al) utensils (Semwal *et al.,* 2006). Cooking and storing food in (Al) cookware after cooking will accelerate (Al) leaching even more (Al Zubaidy *et al.,* 2011). Yokel (2016) found that 72 hours of tomato puree storage in an (Al) pan increased the amount of (Al) in the mixture. Meanwhile, Stahl *et al.* (2017) investigated how drinking bottles and pots made of (Al) affected the amount of (Al) present in water, apple juice, tea, and coffee and came to the conclusion that acidic drinks shouldn't be stored in (Al) drinking bottles. According to Onyeka *et al.* (2020), increasing the contact duration between food and pots composed of recycled metal and alloy increased the migration of (Al) and Pb ions. Due to the chemical structure of the clay used to make the pots, clay pots have the notably lowest rate of increase during storage. (Al) is bound by strong covalent bonds in clay, making it challenging to remove (Al) from the molecular structure of clay (Nyangababo and Droti, 2006).

Table 2. Effect of storage time at 4°C on aluminum content mg/kg (Fw) in tomato and spinach cooked in high quality aluminum pot (HqAl), low quality aluminum pot (LqAl) and clay pot (C).

	HqAl		LqAl		C	
Storage time	**Tomato**	**Spinach**	**Tomato**	**Spinach**	**Tomato**	**Spinach**
1hr	5.49^{n}	10.80^{i}	9.26^{k}	17.24^{g}	1.49^{p}	5.70^{n}
12hr	7.83^{l}	13.69^{h}	18.19^{g}	24.43^{e}	2.63^{p}	5.91^{n}
24hr	10.11^{k}	18.28^{g}	28.19^{d}	30.14^{c}	4.59^{o}	6.93^{l}
48hr	23.63^{e}	22.18^{f}	49.39^{a}	44.74^{b}	9.61^{k}	11.64^{i}

Different letters indicate significant differences at (*P*<0.05). LSD 1.26
HqAl:high quality aluminum pot, LqAl: low quality aluminum pot, C:clay pot

CONCLUSION

There is a growing concern about the dietary exposure of aluminum and other metals as a result of leaching from cookware into food due to factors such as temperature, pH, duration of cooking, storage period and quality of cookware. Aluminum leaching into cooked food can be considerably reduced by using clay pots.

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انتقال الألومينيوم أثناء تحضير الطعام و تخزينه

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الملخص

يستخدم الألمنيوم على نطاق واسع في مختلف مجالات الصناعة مثل صناعات النقل والصناعات الكهربائية والآلات والمعدات وتجهيز الأغذية وعمليات التعبئة والتغليف بالاضافة الى مجموعة واسعة من الأجهزة المنزلية. وجد أن المصدر الرئيسي للتعرض الغذائي للألومنيوم عادةً من الفواكه والخضروات والتي يمكن أن تحتوي على مستويات مختلفة منه اعتمادًا على قدرة النباتات على تخزينه . تؤدي أواني الألومنيوم إلى زيادة نسبة المعدن بشكل كبير فى النظام الغذائي اعتمادًا على عدد من العوامل ، بما في ذلك درجة حرارة الطهي وطريقة الطهي وطبيعة الإضافات المستخدمة. يركز هذا البحث على دراسة تأثير درجة حموضة الطعام ووقت الطهي وفترة التخزين على انتقال الألومنيوم من ثلاثة أواني للطهي (وعاء ألومنيوم عالي الجودة ، وعاء ألومنيوم منخفض الجودة ، وعاء فخاري) إلى الطماطم والسبانخ العضوية المطهية. أوضحت النتائج أن انتقال الالومنيوم من أوعية الألومنيوم منخفضة الجودة كان أعلى من انتقاله فى الأوعية الأخرى.