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Aluminum accumulation in the wheat production chain: a review

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Abstract

The consumption of food containing significant amounts of aluminum has been the focus of discussions related to health. Aluminum is distributed in nature and may be naturally present in raw materials or added to food as a result of activities or modification processes along the food production chain. This exploratory and descriptive research identified possible sources of aluminum in food, in general, and particularly in the wheat chain and its derivatives, which comprise an important class in the food chain and ordinary consumption in many cultures. Even though total aluminum values have been found in food, in general, and particularly in wheat derivatives, information about their origins is still incipient. The total content of this metal in the food may be the result of a sum of sources. For wheat and its derivatives, the potential origins are among environmental factors, such as soil and water, and even operational procedures, such as the use of food additives and chemical compounds for the control of stored grain pests. Despite these considerations, the actual contributions of each source are still purely speculative, since although aluminum contents have been quantified in most studies, their sources were impartially explored and clarified.

Keywords: aluminum; food safety; wheat; degenerative desease; risk assessmen.

Practical Application: This text proposes to investigate the sources of aluminum in food, with a focus on the wheat chain and its derivatives, which are important food sources. The ingestion of aluminum through food and drink has been associated with health problems like Alzheimer's disease. The authors aim to conduct a critical analysis of technical scientific articles, theses, legal requirements, and recommendations to determine the possible sources of aluminum in the production of wheat flour and its derivatives. The possible primary and secondary sources of aluminum were explored, including cross-contamination and additions of ingredients during the manufacturing process.

1 Introduction

The presence of aluminum in food, its possible sources, and impacts on health have been tirelessly explored over the years (Centre for Food Safety, 2009; Exley, 2001; International Programme on Chemical Safety, 1997; Joint FAO/WHO Expert Committee on Food Additives, 2016; Rondeau, 2002). Even so, its sources and mechanisms of absorption by the organism and impacts on health are still not clearly known (Rondeau, 2002).

Official bodies so far have only managed to establish recommended intake doses and scientific-technical papers show exposure risk assessments, which is calculated based on the total aluminum content in the food, intake rate, and body weight (Food and Agriculture Organization of the United Nations, 2009; International Programme on Chemical Safety, 1997; Joint FAO/WHO Expert Committee on Food Additives, 2016).

On the other hand, there are no limits established for the different classes of food and there is also a certain fear of doing so. The lack of accurate information makes it even more difficult to establish acceptable aluminum concentrations.

Given the above, this study aimed to explore and understand the impacts of aluminum on health and identify possible sources of this metal in foods, in general, and particularly in the wheat chain and its derivatives.

For this purpose, a bibliographic review was carried out through technical-scientific articles, theses, and legal requirements and recommendations made by reference bodies focused on the study topic. Then, a critical analysis of the obtained data was performed in order to clarify how possibly the aluminum appears in wheat flour and in its derivatives at each stage of production of these food products. Possible primary sources, naturally derived from the food itself, were explored, as well as secondary sources, which result from the addition of ingredients in food manufacturing processes or from cross-contamination.

Aluminum is a grayish, ductile, malleable metal and is naturally present in the earth's crust. Due to its good physicochemical characteristics, it is commonly used combined with other metals to form alloys (Agency for Toxic Substances and Disease Registry, 2008; International Programme on Chemical Safety, 1997).

The first mechanisms for the manipulation and industrial application of this metal began over a hundred years ago and since then, various advantageous industrial uses and their health and environmental implications have been questioned (Hachez-Leroy, 2013).

Even so, due to its desirable characteristics, aluminum is unquestionably widely used in a variety of household utensils,

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as well as in the production of packaging materials, including bottles, cans, aluminum foil, among others, in addition to being used as a micro ingredient in food additives (Centre for Food Safety, 2009, 2016).

Aluminum is ubiquitous in the environment and in our bodies; thus, studies on this metal are important because they make it possible to identify where it becomes, in fact, a precursor to health damage (Rondeau, 2002)..

2 Aluminum and health

The relationship between aluminum and health risk has been investigated for at least fifty years, and this metal is certainly not inert in the human body (Rondeau, 2002). Human exposure to this metal through food, environmental sources, bioavailability, organism absorption, and toxicological mechanisms has been studied in recent years, as observed in the reports presented by the Agency for Toxic Substances and Disease Registry, Center For Food Safety, and Joint FAO/WHO Expert Committee on Food Additives, in 2008, 2009 and 2016, respectively (Agency for Toxic Substances and Disease Registry, 2008; Centre for Food Safety, 2009; Joint FAO/WHO Expert Committee on Food Additives, 2016).

This exposure from the environment primarily occurs through food and water, being the former the main contributor. Exposure can also occur through air and drugs but measuring this exposure is still difficult, considering sampling, analytical methods, bioavailability, and absorption by the human body (Agency for Toxic Substances and Disease Registry, 2008; International Programme on Chemical Safety, 1997; Shaw & Tomljenovic, 2013).

The aluminum is a neurotoxicant element and its toxicity in human body lead to an oxidative stress, immunologic alterations, inflammatory effects, besides genotoxicity and a plenty of other cell disorders (Flaten, 2001; Shaw & Tomljenovic, 2013).

Aluminum absorption by the body is apparently low in humans, although our gastrointestinal absorption mechanisms are not yet fully understood (Agency for Toxic Substances and Disease Registry, 2008; International Programme on Chemical Safety, 1997). Despite this, its effects on health have been related to the development of degenerative diseases, such as Alzheimer's (Exley, 2001; Ferreira et al., 2008), multiple sclerosis (Mold et al., 2018), in addition to genetic disorders tested in laboratory conditions (Agency for Toxic Substances and Disease Registry, 2008).

Autistic children had higher amounts of aluminum in their hair than those of the control group, indicating a possible relationship between exposure to this metal in important periods of their development and the occurrence of autism (Mohamed et al., 2015). Aluminum may accumulate in bones throughout life. Its levels depend on the type of medication administered by the patient, exposure to chemicals, differences in body anatomy, and gender. This information is particularly important for further research on the role of aluminum in bone diseases (Zioła-Frankowska et al., 2015).

Considering the strong interest and concern of the scientific community for this relationship, complete studies gathering information on the effects of aluminum on health have been published in recent years (Crisponi et al., 2013; Klotz et al., 2017). Therefore, it is possible to find clarifications about aluminum absorption through oral ingestion, skin contact, or even via the respiratory tract (Agency for Toxic Substances and Disease Registry, 2008).

In addition to the medical diagnosis indicating the presence of aluminum in the patient's body (Mohamed et al., 2015; Mold et al., 2018), researchers can also use animals to explore the effects of aluminum toxicity (Geyikoglu et al., 2013; Kuznetsova et al., 2017; Martinez et al., 2017).

In rats, for example, the administration of an acceptable daily dose of aluminum showed rapid onset of deterioration in liver and kidney function, in addition to affecting kidneys, liver, and blood tissues even at low doses (Geyikoglu et al., 2013). Furthermore, it was also observed damage to cardiovascular health (Martinez et al., 2017).

The administration of aluminum citrate and aluminum chloride in mice evidenced that both compounds have a neurotoxic effect, and at the same dose and time of ingestion, the former had a more significant neurotoxic effect (Kuznetsova et al., 2017).

3 Aluminum and food

Reference bodies for contaminants in food have published relevant scientific technical data on decisions for limiting recommendations for the consumption of different food products. Aluminum is a metal that can accumulate in the organism for a period of time and has a potential risk for affecting the reproductive and nervous systems; therefore, it has a Provisional Tolerable Weekly Intake (PTWI) of 2 $mg.kg^{-1}$ body mass (Joint FAO/WHO Expert Committee on Food Additives, 2011).

The PTWI corresponds to the weekly acceptable exposure to contaminants inevitably associated with food consumption, although factors such as aluminum bioavailability and bioaccumulation in the body need to be clarified (Joint FAO/WHO Expert Committee on Food Additives, 2016). Due to the natural occurrence of aluminum in food, its complete elimination from food is practically impossible. Even when the maximum recommended doses are not exceeded, there is a notable concern of the scientific community for reducing the average daily consumption of aluminum in food, especially by children (Bagryantseva et al., 2016; Crisponi et al., 2013; Guo et al., 2015; Hartwig & Jahnke, 2017; Joint FAO/WHO Expert Committee on Food Additives, 2016; Ma et al., 2019; Yeh et al., 2016).

The risk of exposure to aluminum can be calculated, and it usually varies between adults and children. In the case of sweets and snacks, e.g., exposure to the contaminant is generally higher in children than in adults, since food consumption habit is considered in this assessment (Yeh et al., 2016).

Important data can also be presented as Estimated Daily Intake (EDI), which is determined based on the average aluminum content and estimated daily intake, for the same food (Antoine et al., 2017; Filippini et al., 2019); or even considering the estimated weekly intake per kilogram of body mass, usually expressed for a mass of 60 kg (Liang et al., 2019).

Given this standardized information, it is possible to compare some regions, food categories, or even reference values. The average weekly intake of aluminum per kilogram of body mass is shown in Table 1. These values differ in each region. The highest intake was observed in Tianjin, China, where high levels were recorded even for different age groups; higher than the recommended PTWI (Joint FAO/WHO Expert Committee on Food Additives, 2011; Ma et al., 2019).

The intake of aluminum from food, daily or weekly, has been calculated and different values are typically found in the literature. For this calculation, eating habits of each culture or region, age, gender, body weight, and quantity of aluminum specific for each food should be considered, and there may be variations for the same type of food from different locations (Hayashi et al., 2019; Joint FAO/WHO Expert Committee on Food Additives, 2011). The same variation behavior was observed in the wheat food category and its derivatives (Table 2).

In wheat flour, there was a significant difference between the obtained values. For example, in China (Ma et al., 2019), aluminum concentration is about twenty times higher than the values found in Germany (Stahl; Taschan; Brunn, 2011).

The total aluminum content in cereal products depends largely on the production process; therefore, some products have low amounts of aluminum and others will have significant quantities (Bratakos et al., 2012). In analyses performed by Ma et al. (2019) in a total of 69 samples, 62.32% were above the recommended value in Chine (100 $mg.kg^{-1}$), which indicates a possible increase due to the use of other ingredients containing aluminum.

The aluminum additives established by FAO/WHO (Food and Agriculture Organization of the United Nations, 2015) regulations to be used in wheat derivatives are shown in Table 3. A confectionery product such as a cake may contain multiple contributors to the total aluminum content. Both in basic ingredients, such as eggs, milk, and flour, as well as in toppings, fillings, and yeasts, in which additives are found (Bratakos et al., 2012).

It is also noteworthy that there are different laws in each region of the world. In Brazil, the use of the additive INS 541i (aluminum and sodium acid phosphate or acid sodium aluminum phosphate), until then authorized by ANVISA (Brasil, 1999) as a chemical yeast in bread production (recommended maximum limit of 1000 $mg.kg^{-1}$) was banned by ANVISA itself (Brasil, 2019) In China, this additive is prohibited (China, 2012).

Table 1. Weekly Average Intake of Aluminum from Food in the World.

Country	Weekly average intake of aluminum per kilogram of body mass ($mg.kg^{-1}$)	Reference
Italy	0.41*	Filippini et al. (2019)
China	0.60*	Liang et al. (2019)
China (Tianjin)	8.4*	Ma et al. (2019)
China (Shanghai)	0.51*	Guo et al. (2015)
South China	1.50*	Jiang et al. (2013)

*For 60 kg of body mass. Source: By the author (2019).

Stahl et al. (2011), analyzing cereal products (425 samples including flours, pre-prepared mixtures for baking, bread, pretzels, and savory cookies), found that 82% of the samples had values lower than 10 $mg.kg^{-1}$, 20% between 10 and 100 $mg.kg^{-1}$ and only 2% had values higher than 100 $mg.kg^{-1}$ of total aluminum.

The general average found for cereal products was 4 $mg.kg^{-1}$. However, aluminum contents varied from 1 and 737 $mg.kg^{-1}$ and the highest concentrations were found in samples of pre-prepared mixture for baking, containing nuts or aluminum and sodium sulfate additive. Bread and flours had the lowest levels of aluminum (1-14 $mg.kg^{-1}$ and 1-19 $mg.kg^{-1}$, respectively) of all cereal products analyzed (Stahl et al., 2011).

Despite the use of additives in these products, a significant reduction was observed in 2009 and between 2011 and 2014, which may be associated with public policies, even though the presence of additives in some specific foods may exceed the level recommended for children (Ogimoto et al., 2016).

Table 2. Aluminum Content in Foods Derived from Wheat Cereal.

Type of food	Total aluminum ($mg.kg^{-1}$)	Country	Reference
Bread	3.55	Italy	Filippini et al. (2019)
Bread	1.0-14.0	Germany	Stahl et al. (2011)
Bread	1.0-28.0	China (Hong Kong)	Centre for Food Safety (2009)
Bread	<0.01	Japan	Ogimoto et al. (2016)
Wheat flour	370.91 ± 370.22	China (Tianjin)	Ma et al. (2019)
Wheat flour	1.0-19.0	Germany	Stahl et al. (2011)
Pre-prepared mixture for bread	1.0-737.0	Germany	Stahl et al. (2011)
Pre-prepared mixture for confectionery	<0.01-1.06	Japan	Ogimoto et al. (2016)
Crackers, crispbread, salty snacks	3.51	Italy	Filippini et al. (2019)
Cookies	4.44	Italy	Filippini et al. (2019)
Cookies	0.01-0.05	Japan	Ogimoto et al. (2016)
Crackers and cookies	1-88	China (Hong Kong)	Centre for Food Safety (2009)
Cakes and pies	2.34	Italy	Filippini et al. (2019)
Cake	1-220	Chine (Hong Kong)	Centre for Food Safety (2009)
Fried bread	4.5-852.7	China	Li et al. (2017)
Flour and pasta (noodles)	14.0 ± 11.7	South China	Jiang et al. (2013)
Baked flour-based products	187.1 ± 189.4	South China	Jiang et al. (2013)
Fried flour-based products	282.7 ± 249.0	South China	Jiang et al. (2013)

Source: By the author (2019).

Table 3. Aluminum Additives Allowed by FAO/WHO in Wheat Derivatives.

Additive/Technological function	Food category	Maximum limit (mg.kg^{-1})
INS 523 - Aluminum and ammonium sulfate / acidity regulator, color stabilizer, firmness agent, body and stabilizing agents	Crackers, except for sweet cookies, other common bakery products such as bagels, Arabic bread, *muffins*	100
	Steamed bread and bread; mix for bread and common bakery products	40
	Fresh pasta and noodles and similar products	300
INS 541 i (aluminum and sodium acid phosphate) and INS 541 ii (basic aluminum and sodium phosphate) / Acidity regulator, emulsifier, stabilizer, growth agent, thickener	Flours	1600
	Crackers, except for sweet cookies, other common bakery products such as bagels, Arabic bread, muffins	100
	Steamed bread and bread; mix for bread and common bakery products	40

Source: Adapted from Food and Agriculture Organization of the United Nations (2015).

In Shanghai, exposure to aluminum from wheat flour and its derivatives ranged from 0.42 to 1.88 mg.kg^{-1} body weight per week. These values correspond to 97% and 77% of the maximum recommended intake (2 mg.kg^{-1}) for adults and children, respectively. Even so, results have shown that the exposure to aluminum through wheat flour and its derivatives appear to be no critical in the development of adverse health effects on the local population (Guo et al., 2015).

The comparison between aluminum concentrations in different food products is almost impractical because of the wide variation between foods from the same group or between those from different sources (Table 4). In the Italian population, e.g., aluminum was found in higher levels in drinks, cereals, and leafy vegetables (Filippini et al., 2019). Depending on the diet, cereals and vegetables tend to be the major contributors to aluminum intake in adults (Bratakos et al., 2012; Ma et al., 2019).

Nevertheless, vegetables with measured aluminum content may cause no health risk, as in the case of vegetables from Jamaica including bananas, potatoes, coconut, pumpkin, carrots, among others (Antoine et al., 2017). Significant levels of aluminum may be found in processed or in natura products. However, the presence of too much aluminum is notable due to the use of food additives in some preparations (Bagryantseva et al., 2016; Joint FAO/WHO Expert Committee on Food Additives, 2016).

Infant food formulations with levels above 5 μg.kg^{-1} of body mass, recommended by the FDA (Food and Drug Administration), are of concern because toxicity is more harmful in developing babies and children, in addition to significant differences between analytically measured values and calculated values, causing insecurity in consumption (Poole et al., 2010).

In infant formulations, aluminum occurs as a contaminant in the ingredients used in their composition (Poole et al., 2010;

Table 4. Food Aluminum Content in General.

Type of food	Total aluminum (mg.kg^{-1})	Country	Reference
Vegetables	7.37	Italy	Filippini et al. (2019)
Banana	93.12	Jamaica	Antoine et al. (2017)
Fermented tea	590-980	China	Cao et al. (2010)
Tea in general	487.57±234.46	China	Li et al. (2015)
Herbs tea	14.0-67.0	Germany	Stahl et al. (2011)
Raw rice	76.49±19.72	Thailand	Rittirong & Saenboonruang (2018)
Honey	15.299±6.75	Turkey	Altunatmaz et al. (2018)
Infant formulation	10.10±9.40	China (Tianjin)	Ma et al. (2019)

Source: By the author (2019).

Redgrove et al., 2019). However, this is not necessarily inevitable, as there are some measures to ensure the quality control of raw materials and a better selection of ingredients (Redgrove et al., 2019).

4 Origin of aluminum in food

The diet is unquestionably the major responsible for aluminum intake, and food sources can be primary or secondary. The primary source corresponds to the natural aluminum content of foods due to the absorption of metal from the environment, and it is inevitable. The secondary source corresponds to the primary one plus the addition of aluminum due to contamination through food contact with other sources, use of food additives containing aluminum, or even from veterinary drugs, fertilizers and air (Stahl et al., 2011).

In cereals, aluminum levels can already be found in plants and grains, which are the base of the production chain (Liang et al., 2019; Nanda et al., 2016; Szabó et al., 2015; Liang et al., 2019), analyzing 109 samples of unprocessed wheat grains in China, found values between 2.4-31.6 mg.kg^{-1}, a mean of 11 ± 6 mg.kg^{-1}, and approximately 80% of the samples presented values ranging from 5 to 20 mg.kg^{-1}. The results found by these authors are similar to those from previous studies on wheat grown in the same region.

Additionally, a study on aluminum quantification in different rice cultivars, collected from the same agricultural field under the same growing conditions, recorded values between 5.0-80.0 mg.kg^{-1}, indicating these cultivars have special characteristics that explain such differences (Nanda et al., 2016).

4.1 Soil

Aluminum is naturally present in the soil as silicate, oxides, and hydroxides, as well as associated with other elements. It is not found isolated due to its reactivity, but it can be found in the form of Al^{+3} ion; its mobility and transportation in the environment are determined by several factors, such as the presence of other components, organic matter, and water (Agency for Toxic Substances and Disease Registry, 2008; International Programme on Chemical Safety, 1997; Water Quality Association, 2013).

The presentation forms of aluminum also depend on pH. When pH increases, the aluminum ion is hydrolyzed, which may occur successively if pH continues increasing, consequently, changing the major aluminum presentation form (Dalović et al., 2012; Salet, 1998). Each aluminum species has a characteristic potential for toxicity to the plant, and the Al^{3+} ion has been identified as toxic to most plants (Salet, 1998). This effect was observed in specific studies on wheat (Del Guercio & Camargo, 2011; Delhaize et al., 2012; Iqbal, 2014; Li et al., 2022; Silva et al., 2010), and also in sorghoum growth (Miller et al., 2009).

The effect of aluminum toxicity on plants interferes with root growth and may occur from multiple mechanisms (Del Guercio & Camargo, 2011; Li et al., 2022; Liu et al., 2018; Silva et al., 2010; Szabó et al., 2015; Zhou et al., 2007).

In roots, the effect of aluminum toxicity is identified when their growth is inhibited (Del Guercio & Camargo, 2011; Liu et al., 2018; Silva et al., 2010; Szabó et al., 2015; Zhou et al., 2007), also followed by an increase in the content of aluminum and phosphorus, which may be a response of the plant's defense mechanism (Szabó et al., 2015). However, Al^{3+} detoxification by phosphorus occurs mainly in the soil and not in the plant tissue (Iqbal, 2014). Moreover, in a wheat cultivar considered to be tolerant, it was observed an increase in the antioxidant system activity and a reduction in H_2O_2 root accumulation, leading to less oxidative damage and more intense growth (Liu et al., 2018).

This same behavior was observed when comparing two wheat cultivars, resulting in a stimulus for root hair growth in the most tolerant cultivar, which reinforces the presence of physiological mechanisms in response to aluminum toxicity (Garcia-Oliveira et al., 2016). Plant's tolerance to Al^{3+} in acidic soil is related to genetic factors (Del Guercio & Camargo, 2011; Han et al., 2016). Different tolerances may be found between cultivars of the same species, which is observed, e.g., in some studies on wheat (Camargo et al., 2006; Del Guercio & Camargo, 2011; Delhaize et al., 2012; Garcia-Oliveira et al., 2016; Liu et al., 2018; Silva et al., 2010; Zhou et al., 2007) and oats (Crestani et al., 2011; Silveira et al., 2013). These results are especially important for genetic improvements to obtain more resistant cultivars (Del Guercio & Camargo, 2011; Han et al., 2016; Zhou et al., 2007).

In this context, special genes that promote better tolerance to aluminum toxicity present in bread-type wheat (*Triticum aestivum*) were used to improve this characteristic in durum wheat (*Triticum turgidum*) (Han et al., 2016).

Although plants have different tolerances to aluminum in the soil, these defense mechanisms are apparently common. Despite this subject has been addressed for at least fifty years, there is still a need for a better understanding of resistance mechanisms, including intracellular processes and biochemical mechanisms involved in signaling the stress caused by aluminum (Li et al., 2022; Singh et al., 2017).

In addition to toxicity, aluminum may migrate from soil to plant and thus contribute to increasing the total aluminum content, as evidenced by Cao et al. (2010) in tea leaves grown in China, and into vegetables cultivated in Central Africa as show in the research.(Ondo et al., 2013) Regarding wheat, there are no studies associating soil aluminum content with aluminum found in wheat grains, suggesting migration to this cereal.

However, aluminum is found in higher concentrations in roots than in the aerial part of wheat, suggesting there was a limited aluminum translocation (Szabó et al., 2015). Aluminum toxicity is more pronounced in the lower portion of the root (tip of the root), between 0-5 mm, where the highest aluminum concentration is found (Liu et al., 2018).

4.2 Pollution

In addition to soil composition natural factors, the area where food is produced may contribute to increasing the concentration of metals due to pollution generated by industries. In a study on three different species of mushrooms cultivated at different distances from the contaminating source, it was found that as the distance increased, the total aluminum content decreased differently between the species (Wesołowska et al., 2016). Thus, in addition to toxicity resistance, there is also a migration of the metal.

In honey, industrial applications and the use of chemicals, such as pesticides that pollute soil, water, and air, are the main factors associated with differences in residual aluminum content, according to the collection area. These chemicals may contain metallic elements in their composition, and bees and honey are exposed to them from pollen and contaminated water and air (Altunatmaz et al., 2018).

Water from extraction mines may also contribute to contaminating the environment. According to Lu et al. (2011), aluminum contained in mine water may be transported by watercourses, especially in flood periods.

4.3 Water

Water may contain aluminum from primary sources, due to leaching of rocks and soil (Water Quality Association, 2013). In a research performed by Akbari et al., (2018), Iranian cities were mapping and mean, minimum and maximum aluminum concentrations of 0.015, 0.0004 and 0.059 $mg.L^{-1}$, respectively, were found in the water resources of the studied municipalities. However, none of these values was a problem.

In a similar study, analyzing water from lakes and rivers in Switzerland, (Peydayesh et al., 2019) found values between 10.4 and 100 ppb, or approximately 0.01 and 0.10 $mg.L^{-1}$. Although no sample has exceeded the recommended values, the mapping made it possible to identify the most critical areas relating to the contaminant potential, in addition to showing that the differences are possibly due to the natural characteristics of each region (Akbari et al., 2018; Peydayesh et al., 2019).

Differences in aluminum content were also found in water available for consumption. Purified bottled water had the lowest values, followed by bottled spring water and tap water; the latter one had the highest variation according to the collection area (Peydayesh et al., 2019), suggesting a possible interference by the treatment.

A research on the identification of water aluminum levels, in Malaysia, recorded no values (0.11 and 0.12) above the recommended concentrations and no significant risk was found by the risk index calculation (Dzulfakar et al., 2011).

The increased aluminum concentration in water is usually due to the use of coagulating agents in its treatment, which may generate residual aluminum in the treated water (Jiao et al., 2015; Rosalino, 2011; WHO, 2008). The residual aluminum in water may cause changes in its color and deposition of sediments throughout the processes (WHO, 2008). Among the water treatment processes, coagulation, flocculation, decantation, and filtration are the most critical steps with regard to residual aluminum (Rosalino, 2011).

Aluminum and iron salts and organic polymers are some of the most common coagulating agents used in water treatment (Lombi et al., 2010). Those based on aluminum are usually used at doses between 2 and 5 mg.L^{-1}. The precipitated flake removes contaminants dissolved or suspended by neutralization, adsorption, and trapping mechanisms (WHO, 2008).

Aluminum polychloride is an example of an aluminum-based coagulant for water treatment and is available in different commercial types (Kimura et al., 2013). Aluminum chloride ($AlCl_3$), polymer $Al_{13}O_4(OH)_{24}^{7+}$ (Al_{13}), and polymer $(AlO_4)_2Al_{28}(OH)_{56}^{18+}$ (Al_{30}) are other examples (Shu-xuan et al., 2014).

Each coagulant has a particular efficiency and can result in different levels of residual aluminum in the treated water. For example, by comparing aluminum chloride ($AlCl_3$), polymer Al_{13}, and polymer Al_{30}, the former had the highest residual aluminum value (Shu-xuan et al., 2014).

Among several factors, pH strongly influences the residual aluminum content dissolved in treated water (WHO, 2008). The pH values close to neutrality indicate the lowest metal concentrations (Jiao et al., 2015; Kimura et al., 2013; Shu-xuan et al., 2014; Wang et al., 2010). Low temperatures also contribute to minimizing residual aluminum content (Kimura et al., 2013; Wang et al., 2010). These data are of fundamental importance in the choice of water treatment process parameters that provide lower levels of contaminant residues in treated water, which will later be directly consumed or used in industrial processes.

There are several studies aimed to know the characteristics of each coagulant; however, there is a desire for the development of new products that provide aluminum residues around 0.05 mg.L^{-1}, still considering pH variables (Kimura et al., 2013).

The use of other technological alternatives, such as that proposed by Peydayesh et al. (2019), also results in significant decreases in residual aluminum in water for consumption or beverages. These researchers evaluated the use of hybrid membranes, of which composition has a strong interaction with metals, resulting in an aluminum recovery above 98%.

The limit of aluminum in water for human consumption in Brazil and in the United States is 0.2 mg.L^{-1} (Brasil, 2021; United States Environmental Protection Agency, 2017). In a water treatment plant, operating under good conditions, residual aluminum values below 0.1 mg.L^{-1} are possible to be found, even using aluminum-based coagulants (World Health Organization, 2008). However, there is also a need for flexibility considering the usefulness of aluminum salts in the coagulation process for water treatment (United States Environmental Protection Agency, 2017).

The presence of residual aluminum in treated water is generally due to problems in the water treatment plant, which basically result from a deficiency in controlling and monitoring doses of chemical agents, poor maintenance conditions, among other factors. The lack of aluminum monitoring in the water treatment steps, as well as the non-elaboration of molecules profile in each step, lead to a non-identification and, consequently, non-resolution of failures. If analysis indicates a predominance of soluble aluminum species, it means there may be coagulation problems. If aluminum particulate species predominate, there is probably a problem in the filtration step (Rosalino, 2011).

A second problem regarding aluminum, in this same context, is related to water treatment residual by-products, of which destination has been the focus of some studies, since they may contain contaminants, in addition to the high cost of their disposal in landfills (Lombi et al., 2010).

The data obtained by Lombi et al. (2010) suggest that the use of water treatment residues, with high levels of aluminum, in lettuce cultivation affects plant growth due to the decreased availability of phosphorus in the soil, but not directly because of aluminum toxicity.

Some studies such as those by Ooi et al. (2018), who analyzed aluminum recovery in sludge resulting from water treatment by acid leaching, have contributed positively to minimizing these effects. As a main result, they found that the coagulant used in water treatment determines the quantity of total residual aluminum and factors such as acid concentration, solid/liquid ratio, temperature, and heating time in acid leaching are determinant for the sludge aluminum recovery. Under the best conditions, the experimental result was 68.8 ± 0.3% against 70.3% calculated, and the most impactful parameter for this recovery was the solid/liquid ratio.

All technical scientific information on water discussed here is essentially important since medical studies show a relationship between the consumption of water containing aluminum and the risk of developing dementia in men and women (Russ et al., 2020), especially in the elderly (Ferreira et al., 2009).

4.4 Chemicals in the treatment of stored grains

Stored grains need to be protected from pest attack. For this, chemical control can be preventively or curatively used (Lorini et al., 2015). The phosphine is one of the insecticides most used for this purpose; it is considered a common fumigant that can be obtained from aluminum or magnesium (Reed, 2013; Thabit & Elgeddawy, 2018).

The phosphine fumigant, formulated as an aluminum phosphate solid for the treatment of grain, has been used for at least 60 years. On the other hand, magnesium phosphate-based products became known in the market in the middle of 1975 (Reed, 2013). These fumigants are obtained by combining metal phosphates with a mixture of other solid ingredients, which are compressed into tablets or pellets (Reed, 2013). Tablets, sachets or plates are commercially available forms. The decision of which product should be used must consider the reaction speed, in order to protect the manipulator when the application is performed in a large area (Reed, 2013).

The use of phosphine, by purging or fumigation, must occur in a closed environment. The insecticide resulting from the vaporization of chemical compounds applied in solid form produces a lethal concentration to target pests (Lorini et al., 2002). Ensuring the sealing of the area, lethal concentration and homogeneous distribution of gas, there will be a desired death of the insects (Lorini et al., 2015; Thabit & Elgeddawy, 2018). The release of toxic fumigant gas (PH_3) is slow and gradual. From aluminum or magnesium phosphate, the release occurs according to the reactions $AlP + 3H_2O \rightarrow Al(OH)_3 + \uparrow PH_3$ and $Mg_3P_2 + 6H_2O \rightarrow 3Mg(OH)_2 + \uparrow 2PH_3$, respectively (Reed, 2013).

Considering the problem of aluminum, in the first reaction, in addition to gas, there is also the formation of residue (aluminum hydroxide), which is obtained as a gray powder (Khanchi et al., 2010; Reed, 2013). When the reaction finishes, the residue is no longer considered a pesticide, or a residual pesticide, because it has no insecticidal properties. Metal hydroxides are common in nature, and for this reason, regulatory bodies consider phosphine fumigants to be environmentally friendly (Reed, 2013).

In this way, doses lethal to pests, insect resistance to pesticides, and efficiency of different application procedures have been frequently investigated (Chen et al., 2015; Isa et al., 2016; Nguyen et al., 2015; Sağlam et al., 2015). However, there is no concern related to food contaminants intrinsic to the use of these chemicals, if management conditions make the metallic powder separation impossible.

Scientific references on this management bring important information about gas poisoning in humans, which in many cases may be lethal (Meena et al., 2015; Sinha, 2018; Yan et al., 2018). In addition to gas, via inhalation exposure to a metallic powder containing aluminum may increase the risk of cardiovascular diseases and degenerative diseases such as Alzheimer's (Peters et al., 2013). Nevertheless, there are controversies on this topic, since oral bioavailability is apparently low and considered insufficient to induce clear adverse effects that justify a characterization of this risk (Dekant, 2019).

The use of phosphine was first evaluated in 1965, from a toxicological, residual, and analytical point of view. Some revisions were performed in 1966, 1967, 1969 and 1971; however, the residual limit allowed in post-harvest grains had no change in this period and is still 0.1 $mg.kg^{-1}$ (Food and Agriculture Organization of the United Nations, 2015).

From the application of analytical methods, for measuring residual phosphine in treated cereal matrices, no values exceeding the recommended limits were found (Khanchi et al., 2010; Thabit & Elgeddawy, 2018). Analytical methods have been also used to confer legitimacy on the chemical product composition declared by its manufacturer (Santos et al., 2018).

As for the powdered residue (aluminum hydroxide), resulting from a fumigation reaction, there is neither research addressing its quantification in cereals or cereal products nor information in package inserts on its removal from silos. However, although there are more appropriate product management technologies, the application in the form of phosphine tablets in silos shows that the residue is inevitable, occurring its total incorporation because these tablets are directly distributed on the grain mass. The removal of this residual powder is very difficult and unlikely to be carried out.

In this way, the treatment of wheat grains stored with purging pellets may contribute to the increased concentration of residual aluminum in wheat flour, making it necessary to review good operational practices, so that the levels of purge residues in the treated grains are minimal. There are no other implications that interfere with the use of phosphine in the control of stored grain pests, until the present moment. There is no change in the technological quality of wheat flour obtained from purged grains (Faroni et al., 2002).

The recent studies (in process of publication, 2021) of our research group (Cooperativa Agraria Agroindustrial/Universidade Tecnológica Federal do Paraná-Francisco Beltrão) demonstrated that the total aluminum content in wheat farinaceous products can be the result of an accumulation along the production chain. For this, our research group conducted two experiments were carried out. The first evaluated the effect of soil correction with liming on the total aluminum content of wheat flour products. The second evaluated the post-harvest treatment of stored wheat grains as potential that contributes to the increase of aluminum in their derivatives. Chemical treatments used to control stored grain pests, can contribute significantly to the increase of aluminum in farinaceous products due to the incorporation of residues (aluminum hydroxide). Aluminum contamination is greater in bran than in flour. This fact, combined with the stimulus for the consumption of integral products due to health benefits, reinforces an alert about legal limits.

4.5 Additives

The secondary source of aluminum is given by the addition of ingredients containing this metal to food formulation in order to impart color, flavor, texture, aroma, or other quality or technological characteristics to the final product (Centre for Food Safety, 2016).

The use of food additives is justified when it has some technological advantages, such as (1) preservation of nutritional characteristics, (2) use as an ingredient or component for specific products for consumers who have special dietary needs, (3) for obtaining or maintaining food quality or stability, or (4) for improving its organoleptic properties, without changing food nature or quality (Food and Agriculture Organization of the United Nations, 2015).

The use of additives in food production is a common practice in the market, and some of them contain aluminum. In Brazil, this use was a legal practice but it was modified by Resolution RDC No. 285, May 21, 2019, of the National Health Surveillance Agency (ANVISA), which prohibits the use of food additives containing aluminum in several food categories, including anti-humectants, such as aluminum sodium silicate, aluminum silicate, aluminum salts, and chemical yeasts, such as sodium and aluminum acid phosphate (Brasil, 2019).

On the other hand, in China, some typical foods, such as Fried bread Youtiao, contain high concentrations of total aluminum from a secondary source, i.e., from the use of food additives to obtain the desired crispness (Li et al., 2017). Guo et al. (2015) and Jiang et al. (2013) found higher levels of aluminum in fried food made of flour in comparison with other food products; these studies recorded average total aluminum concentrations of 225.67 and 282.7 $mg.kg^{-1}$, respectively.

Furthermore, Chinese people are exposed to aluminum levels much higher than the recommended concentration (Jiang et al., 2013; Ma et al., 2019), even after the exposure limits being revised in the country, in 2014. Thus, public awareness and more vigorous supervision are recommended (Ma et al., 2019).

Conversely, there are conflicting conclusions in terms of exposure risk. For Guo et al. (2015), exposure risk from the consumption of wheat flour and its derivatives is relatively low, since they recorded an exposure of only 4.2% of the population, resulting from the consumption of these products.

The use of aluminum as a dye (food coloring) is regulated by EU No. 231/2012. It is a metallic pigment in the form of finely divided powder. However, aluminum may also be found in other types of dyes (European Union, 2012).

To reduce food aluminum content as much as possible, food companies should follow at least some basic principles, such as the maximum reduction of additive containing this metal, adhere to the policy of replacing it by other acceptable ingredients as much as possible, as well as finding technical alternatives for food processing that help to reduce this risk of contamination. Moreover, accurately informing the consumer is essential, i.e., all additives used must be described on food packaging (Centre for Food Safety, 2016).

As an additional alternative, it is essential to know the origin of food additives. Their quality, purity, composition, and obtainment method are essential to assess the risk associated with their use (Centre for Food Safety, 2016).

In pet food, which also have cereals in their formulations, some reports indicate total aluminum concentrations between 21-11900 $mg.kg^{-1}$ and 49-8500 $mg.kg^{-1}$ in adult and baby diets, respectively. These high values may also be due to the use of ingredients containing aluminum in order to confer some specific technological characteristics to the final product (De Nadai Fernandes et al., 2018).

4.6 Technological processes

Some technological processes may also contribute to the quantity of total aluminum in food. Tea fermentation, a process usually performed in China, increases the aluminum concentration in the plant leaves; however, this is not yet fully understood (Cao et al., 2010). Additionally, Li et al. (2015) found that the process of infusing tea leaves into the water also causes the migration of metals from the leaves to the liquid, with a decreasing aluminum concentration after each sequential infusion using the same leaves. Thus, it is recommended to discard the first infusion to decrease metal intake.

Conversely, cooking in aluminum pans, which are very common in the market, caused no significant increase in rice grains, or at least, no value that puts health at risk (Odularu et al., 2013; Rittirong & Saenboonruang, 2018). However, in acid food such as tomatoes, among others, cooking may favor the metal migration from the pan to the food, and this migration is directly proportional to the cooking temperature (Dantas et al., 2007; Sander et al., 2018).

Although this metal migration has been recorded, aluminum levels in food subjected to this process are lower than the internationally recommended limits and therefore, there were no health risks in the experimental conditions studied (Dantas et al., 2007). As for the process of boiling milk in aluminum utensils, there was no significant difference between the control sample and those boiled in stainless steel utensils, suggesting that metal migration by this process in milk is negligible. However, an increase of approximately 1%, during milk cooling storage in aluminum containers, was recorded (AI-Ashmawy, 2011). On the other hand, handmade pans based on lead and aluminum showed a significant risk of migration from the utensils to the food (Weidenhamer et al., 2014).

5 Conclusion

To control food aluminum concentrations, it is essential to know its source or sources; however, there is still a lack of scientific information in this context. The total metal content in food may be the result of a sum of amounts from several origins. Thus, there is a need for future studies aimed to elucidate these issues.

This fact, in addition to the absorption of aluminum from the diet, has been widely discussed because it is associated with the development of diseases. There is no doubt that aluminum is toxic, but there is still a need to clarify its mechanisms of toxicity, bioavailability, and bioaccumulation in the body.

Although there is a recommended maximum intake, no maximum aluminum content has been established for each class of food, preferably from a numerical and aluminum-based scale. Scientific technical studies have identified aluminum concentrations in a variety of foods, as well as calculated their respective percentage, based on maximum weekly recommendation and eating habits. However, from these studies, it is categorically impossible to state that a certain food is in accordance (or not) with the current regulations.

For this reason, current standards and recommendations will probably be revised so that it is even possible to perform inspections to ensure compliance with the established standards. Until then, according to the guideline, the use of aluminum in food production processes must be reduced as much as possible.

Author contributions

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