

Research Article

Aluminium Exposure Through the Diet

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Abstract

Aluminium is one of the most common metals found in the environment and consequently, in food. However, Al levels have been increasing over time due to acidification of the soils and anthropogenic activities. Al is a known neurotoxic agent because this metal tends to accumulate in the brain. Several studies have reported the correlation between Al levels and different diseases such as Alzheimer's disease. In addition, aluminium can interfere with some essential metals. In order to study the toxic risk of Al intake, data on Al levels in several types of food have been compiled and compared with the aim of estimating the total dietary intake of the metal. The most widely used analytical techniques for Al determination were Inductively Coupled Plasma mass Atomic Spectroscopy and Atomic Emission Spectroscopy (ICP-OES and ICP-AES). The highest mean Al content was found in vegetables (16.8 mg/kg), fish and seafood (11.9 mg/kg) and roots and tubers (9.60 mg/kg). The food group with the most notable contribution to tolerable weekly intake were fruits (18.2% adults, 29.4% children) and vegetables (32.5% for adults and children). Al dietary intake can pose a health risk resulting from Al accumulation in the brain caused by long-term intake.

Keywords: Aluminium, Aluminium toxicity, Analytical methods, Dietary intake, Exposure, Food

Introduction

Aluminium (Al) is the third most common element in the Earth's crust [1] and is naturally present in the environment. Al is a versatile metal with several properties and has a wide range of uses. Al is used in different alloys with other metals such as copper, zinc or magnesium [2].

The concentrations of this metal in food and drink have been increasing over time due to the acidification of the soils that transfers the aluminium from the soil to the aquatic environment [3] and anthropogenic activities such as mining (bauxite extraction), aluminium industries, and others [2].

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Diet is the main exposure route to metals [4]. Aluminium is found in different drinks and foods such as drinking water, where it is added as a flocculant (aluminium sulphate, $Al_2(SO_4)_3$), processed foods, where it is used as an additive and as a result of the packaging used and even in fresh foods such as vegetables and fruit because of the Al contained in the soils [5].

In addition, aluminium utensils and tea consumption are also responsible for the increase of the Al in the diet [2]. Tea leaves have been reported to have higher Al levels because tea plants are grown in acidic soils [5].

Al is present in an ionic form as Al^{3+} . The absorption of Al depends on several factors such as the pH level, the presence of organic acids (citrate, lactate), etc [2,6].

The oral bioavailability of Al, which is the amount that can be absorbed, in drinking water is around 0.3%, while this is approximately 0.1% in food and drinks [7]. Al absorption increases as the pH level decreases.

Aluminium is a toxic metal that does not have a function in the human and animal organism [8]. The toxicity of Al depends on the exposure route and the solubility of the aluminium compounds [9]. Aluminium tends to accumulate in the body, in tissues such as the brain, bones, kidney and liver. Long-term exposure to low Al levels leads to toxic effects [10].

Based on information provided by the European Food Safety Authority (EFSA), the exposure of the European population to Al is 28.6-214 $\mu g/kg$ body weight per day [11].

Because of the abovementioned reasons, different institutions have established maximum limit intake levels for aluminium. The EFSA has established a Tolerable Weekly Intake (TWI) of 1 mg Al per kg of bodyweight [7]. The FAO/WHO Expert Committee on Food Additives, has evaluated the Al bioavailability and concluded by setting a Provisional Tolerable Weekly Intake (PTWI) of 2 mg/kg of bodyweight/week [12], and this value is twice that established by EFSA.

Since Al is a toxic metal found in foods and beverages, the data previously obtained by our research group and others have been compiled to compare the levels of aluminium content in different types of food and drinks to study the toxic risk from the diet, and to compare the levels found over time to estimate the general variations in Al content.

Data Collection Method

The present study was conducted from March 2017 to May 2017. The review of the scientific papers was performed with online databases such as the Web of Sciences (WOS), MEDLINE (PubMed), Google Scholar, Research Gate and the library of the Universidad de La Laguna, where versions of most important scientific journals are found. The aim of the review was to compile the aluminium levels in different food groups and the relevant information about the methodology, toxic effects, limits, etc.

Search terms used were "aluminium", "aluminium levels", "aluminium neurological damages", "inductively coupled plasma", "ICP-OES",

“aluminium toxicity”, “toxic metals”, “Alzheimer”, “food additives”, “provisional tolerable weekly intake aluminium”, “tolerable weekly intake aluminium”, “aluminium sources”, “aluminium exposure”, “aluminium metabolism”, “spectrophotometry”, “food composition”, “food analysis”, “contaminants”, “ICP-AES”, and “atomic emission spectroscopy”.

The papers reporting Al levels in the most commonly consumed foods using the analytical techniques with the highest levels accuracy and precision for Al determination were selected. The studies where a quality control using reference materials was not included were excluded to ensure the quality of the selected studies. In addition, the included papers were classified into two periods: “before 2000” and “after 2000”.

Sources of aluminium in the environment

Aluminium is found in the environment released by natural and anthropogenic processes (Figure 1).

Al is present in the Earth’s crust as a part of silicates (mica or feldspar), hydroxo oxides like bauxite, which are used to extract aluminium and cryolite (Na_3AlF_6) [13,14].

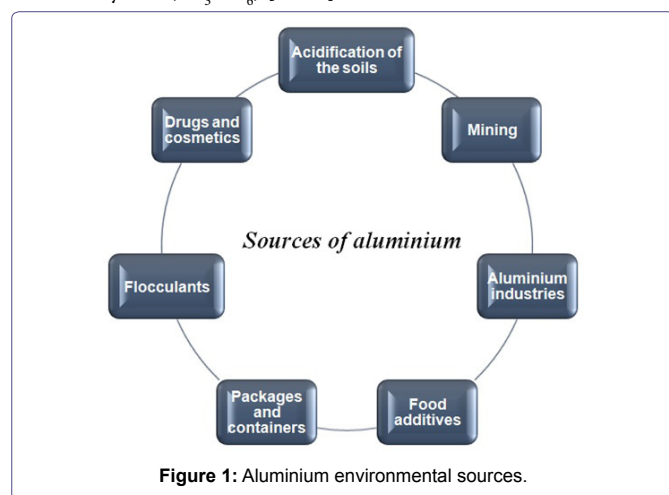


Figure 1: Aluminium environmental sources.

Environmental transport and distribution of aluminium depends on its coordination chemistry [15]. Acidification of soils is a cause of aluminium release in an ionic form (Al^{3+}) to the aquatic systems. Al^{3+} is highly soluble in water.

Regarding anthropogenic sources, industrial processes and mining are the main routes of aluminium release into the environment, mainly into the atmosphere. Al content in food packaging and in cooking utensils is an important route of Al release into the environment [15]. Al is also found in drugs, cosmetics and food additives, whose residues can release Al into the environment.

Metabolism of Aluminium

Absorption

Aluminium is absorbed in a proportion of 0.1-0.3% by the gastrointestinal tract, and occurs in the upper intestine [2] where absorption is higher due to lower pH levels [16].

Absorption of aluminium from foods and drinks depends on numerous factors. Several studies have reported the higher aluminium absorption when citrate and fluoride are present [2,14,16,17]. Meanwhile, the presence of silicon and calcium decrease aluminium

absorption [16] due to the formation of insoluble products with aluminium, the above mentioned compounds are frequently found in processed food as additives.

Transport and distribution

When Al is absorbed it reaches the blood bound to transferrin molecules and circulates across the blood-brain barrier. Al^{3+} can bind with the transferrin molecule thanks to its oxidative state which is the same as the serum iron (Fe^{3+}) [18].

Figure 2 shows the transferrin-transferrin pathway. Aluminium bound to the transferrin molecule enters the cell, where there are transferrin molecule receptors [1,2]. The Al-transferrin complex, binding to the transferrin receptors, is inside the cytosol, where it experiences a decrease of pH level at 5.5 and Al^{3+} is released from the complex [19].

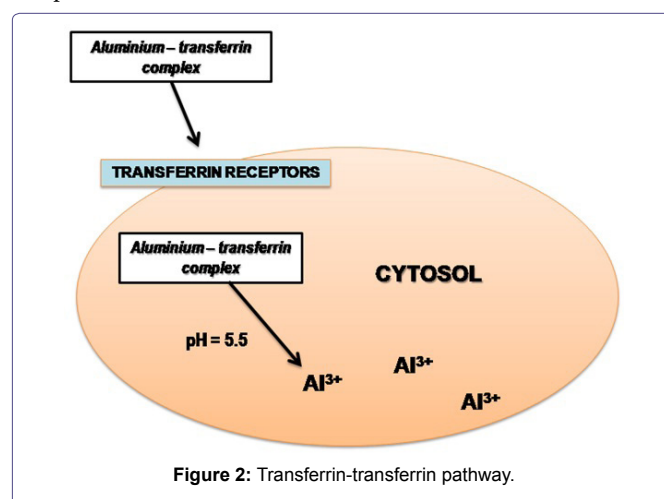


Figure 2: Transferrin-transferrin pathway.

Elimination

The kidneys eliminate the absorbed aluminium in amounts of 15-55 $\mu\text{g}/\text{day}$ through urine and faeces [2,16]. Al excretion is lower in people with reduced renal function and this can lead to toxic effects because of the nephrotoxicity of Al [14,16].

Mechanism of aluminium toxicity

Aluminium can interfere with certain essential elements such calcium and calcium metabolism is one of the most important processes in the human organism. Aluminium has been shown to be able to replace Ca disrupting the mineralization and bone cell growth [9,20]. However, other studies have reported that Al can increase the stability of IRP2 (Iron Regulatory Protein-2), which is a protein whose function is the regulation of the iron levels [9].

Aluminium is considered to be a neurotoxic agent that could increase the probability of developing Alzheimer’s disease [1,21-25]. In addition, Al can lead to cognitive damage and neurological diseases. The probability of having any of these diseases is higher in children and people with kidney problems because the kidney plays a key role in the excretion of Al [25].

Analytical methods in food samples

The analytical methods used for the determination of aluminium in food samples, consists of a first step of solubilisation of the samples. The samples are previously treated with microwave-assisted acid digestion or with an incineration process to obtain ashes [25,26]. The

consulted authors used with a quality control process using certified reference materials for Al, which was treated under the same conditions as the samples.

The most commonly used methods of aluminium measurement are Flame Atomic Absorption Spectrometry (FAAS), Graphite-Furnace Atomic Absorption Spectrometry (GFAAS), Inductively Coupled Plasma-Optical/atomic Emission Spectrometry (ICP-OES/AES) or inductively coupled plasma mass spectrometry (ICP-MS). These methods provide a high selectivity, with low detection limits (0.1 mg/kg or lower) [26,27]. Table 1 shows the comparison between the Limit of Detection (LOD) and the necessary sample volume of the different techniques for aluminium determination.

Technique	Sample volume required	LOD (ng/mL)	Reference
FAAS	1 mL	30	Skoog et al. [28]
GFAAS	0.5-10 µL	0.2	Skoog et al. [28]
ICP-AES	20-100 µL	0.2	Skoog et al. [28]
ICP-OES	20-100 µL	4	Luis et al. [4]
ICP-MS	20-100 µL	0.003	Lloyd et al. [29]

Table 1: Comparison of the LOD (ng/mL) and the volume of sample required between the different analytical techniques used in Al determination.

Some of the above techniques are based on Atomic Absorption Spectrometry (AAS). AAS is a modern technique with the following advantages: non-spectral interferences and low levels of chemical interference, high-accuracy and sensitivity, and chemical separations are not normally required [28].

However, techniques based on inductively coupled plasma are used the most because of their high stability and lack of interferences. In addition, this technique offers a simultaneous multi-element determination. Cost and maintenance are the main problems with this technique [4,28,30,31].

Flame Atomic Absorption Spectrometry (FAAS)

This technique is based on the flame as the atomization system. The sensitivity, precision and accuracy of FAAS are of a high quality in wavelengths between 200-800 nm. However, the atomization through flame needs a high-volume of sample (1 mL approximately) and a high-flow of flame gases (10 mL per minute) that produce a short-residence time of the analyte [28].

Graphite-Furnace Atomic Absorption Spectrometry (GFAAS)

This analytical technique has certain advantages and is based on an electrochemical atomization system [28]. Strict temperature control is required to assure a high reproducibility. The atomization of the analyte is quick and practically quantitative. GFAAS provides high sensitivity with a small sample volume (0.5-10 µL).

Inductively Coupled Plasma-Optical/atomic Emission Spectrometry (ICP-OES/AES)

Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) is an optical method whose measurement is based on energy exchange between electromagnetic radiation and the matter. The main advantages are the quickness, relatively low detection limits, wide concentration ranges, automation and the wide instrumentation available [4,28].

Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) is a technique based on Atomic Emission Spectrometry (AES). Certain instrumental conditions restrict the use of this technique to the analysis of certain elements because: the detectors provide an acceptable sensitivity only in a specific wavelength range, the flame has a strong absorption at wavelengths < 200 nm, and oxygen absorbs with UV radiation. However, AES equipment can provide good results in the wavelength range between 190-850 nm [28,30,31].

Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

ICP-MS is an analytical technique able to develop multi-elemental analysis with a high sensitivity [32]. The mass spectrometer increases sensitivity [2]. The mass spectrometer splits the ionic species according to their mass/charge proportionality [28,33]. The main disadvantage concerns matrix effects (non-spectroscopic interferences) which are produced by electrostatic effects in the generation stage of the aerosol.

Aluminium levels in food and drinks

Aluminium concentrations in foods depend on several factors. Aluminium found in foods is from different origins. Aluminium levels can be found naturally in foods, but these levels can be higher due to the use of food additives (sulphates, phosphates, silicates, etc.) and the use of cooking utensils made of aluminium [34].

Water and drinks

Table 2 shows the average Al levels (mg/kg) in water and selected drinks reported by different authors.

Aluminium is present in drinking water, where it is added as a flocculant or coagulant like sulphate ($Al_2(SO_4)_3$). The lowest Al level found in drinking water and bottled drinking water is 0.12 mg/L from Spain [25]. On the other hand, the highest Al level was found in water from Germany with an average content of 2 mg/L [35].

Al levels found in several types of non-alcoholic drinks were higher than those found in water. The highest levels were found in fruit juice and fruit juice drinks from Germany, with a mean content of 3 mg/L [35]. In general terms, the beverages which were stored in aluminium cans contained higher Al levels, but the consulted studies did not confirm the statistical differences between aluminium cans, glass bottles and plastic containers [35].

Regarding alcoholic drinks, wine is the beverage showing the highest Al levels at 2.42 mg/L in Spanish wines [25], and 2 mg/L in German wines [35]. The high acidity of the soils where the grapes were grown may explain these high Al contents.

With respect to tea, tea (*Camellia sinensis*) leaves are known to make a notable contribution to the dietary intake of Al, due to the acidity of the soil where the plant is grown, because these plants are acidophilic [39]. A study conducted by Sweileh et al., [36] reported an average Al concentration of 312 ± 18 µg/g in tea infusions, however, the brewed tea infusions were found to have lower concentrations (2.1 ± 0.1 mg/L). Other studies reported Al concentrations in brewed tea ranging between 3-4 mg/L, these concentrations are 10 times higher than those found in coffee [40].

Tea infusions can contribute 50% of the total daily intake of Al in cases of high consumption.

Fruit and vegetables

Table 3 shows the mean Al content (mg/kg) in selected fruit and vegetables according to country of origin.

Product	Number of samples	Al mean content (mg/L) ± SD	Origin	Reference
Water				
Water	20	0.12 ± 0.06	Spain	González-Weller et al. [25] ¹
Mineral water, spring water and table water	171	2	Germany	Stahl et al. [35] ²
Drinking water	3	0.016 ± 0.0004	Jordan	Sweileh et al. [36] ³
Tap water	3	0.026 ± 0.0006	Jordan	Sweileh et al. [36] ³
Non-alcoholic drinks				
Soft drinks	20	1.24 ± 0.70	Spain	González-Weller et al. [25] ¹
Soft drinks (Non-cola, cans)	106	0.90	Australia	Duggan et al. [37]
Soft drinks (Cola, cans)		0.66	Australia	Duggan et al. [37]
Soft drinks (Non-cola, glass bottle)		0.15	Australia	Duggan et al. [37]
Soft drinks (Cola, glass bottle)		0.24	Australia	Duggan et al. [37]
Fruit juice and fruit juice drinks	59	3	Germany	Stahl et al. [35] ²
Fruit juices	-	0.043-4.130*	USA	Schenk et al. [38]
Alcoholic drinks				
Wine and fruit drinks	65	2	Germany	Stahl et al. [35] ²
Wine	20	2.42 ± 2.03	Spain	González-Weller et al. [25] ¹
Beer and draught beer	237	0.50	Germany	Stahl et al. [35] ²
Alcoholic beverages (gin, whiskey, rum, beer)	20	0.50 ± 0.32	Spain, England, Scotland, The Netherlands	González-Weller et al. [25] ¹
Tea, herbal infusions and coffee				
Sweetened tea infusions	3	2.2 ± 0.1	Jordan	Sweileh et al. [36] ³
Tea infusions	3	2.1 ± 0.1	Jordan	Sweileh et al. [36] ³
Arabian coffee	3	0.63 ± 0.03	Jordan	Sweileh et al. [36] ³
Herbal teas	-	0.14-1.065*	USA	Schenk et al. [38]
Instant coffee	-	0.02-0.581*	USA	Schenk et al. [38]
Whole coffee	-	0.235-1.163*	USA	Schenk et al. [38]

Table 2: Mean aluminium content in drinking water and other drinks.

*Maximum and minimum value, respectively

-Unknown number of samples

¹Study conducted using ICP-OES

²Study conducted using ICP-MS

³Study conducted using FAAS/GFAAS

The aluminium in vegetables and fruits depends on the irrigation water, the soils where they grow, the plant variety, etc.

During the growth of the plant, Al is transferred from the soil to the different parts of the plant. Depending on the species, some of which tend to accumulate higher Al concentrations in the roots, while others such as the tea plant accumulate higher Al levels in the leaves [44-46].

In regard to vegetables, the highest Al levels were reported by González-Weller et al., [25] in the vegetable group of squashes, carrots, marrow, cabbage, watercress and spinach from Spain, with an average Al concentration of 27.5 mg/kg. Baked potatoes from USA had a high Al level, with an average level of 26 mg/kg [15]. The lowest Al concentrations were reported by Soni et al., [15] in cooked green beans (3.40 mg/kg).

The highest Al level found in fruit was that reported by González-Weller et al. [25] in bananas from Spain, with a mean content of 32.8 mg/kg. This concentration is much higher than that found by

Soni et al., [15] in bananas from USA, with a mean concentration of 0.40 mg/kg.

Animal products

Table 4 shows the mean Al concentration (mg/kg) in different animal products according to origin.

Aluminium, as in humans, is accumulated in animals from the water, the diet and environmental pollution [47]. In addition, in the case of processed products such as cheese, the use of Al-based additives in cheese production processes is a source of Al in these products.

Regarding the Al levels in fresh meat, the highest level was found in viscera from Spain, with an average concentration of 11.2 mg/kg [25], whereas the lowest level was reported in porcine meat (muscle) from France, with a mean Al level of 0.21 mg/kg [48].

The hamburger from the USA was the processed meat product with the highest Al level with a mean concentration of 20 mg/kg. This could be explained by the use of Al-additives.

Product	Number of samples	Al mean content (mg/kg) ± SD	Origin	Reference
Vegetables				
Green beans, cooked	-	3.4	USA	Soni et al. [15]
Potato, baked	-	26	USA	Soni et al. [15]
Potato, cooked	-	10.8	India	Soni et al. [15]
Potatoes	20	5.88 ± 3.29	Spain	González-Weller et al. [25] ¹
Potatoes	-	0.90	USA	MAFF [41]
Squash, carrots, marrow, cabbage, watercress, spinach	20	27.47 ± 38.47	Spain	González-Weller et al. [25] ¹
Tomatoes and onions	20	5.41 ± 2.16	Spain	González-Weller et al. [25] ¹
Asparagus	-	4.4	USA	Greger et al. [42]
Green vegetables	-	3.1	USA	MAFF [41]
Fruits				
Apple sauce	-	0.10	USA	Soni et al. [15]
Apple, fresh	-	0.14	USA	Pennington [43]
Apples and citrus	20	4.73 ± 3.33	Spain	González-Weller et al. [25] ¹
Banana, fresh	-	0.05	USA	Pennington [43]
Banana	20	32.80 ± 33.05	Spain	González-Weller et al. [25] ¹
Peaches, pears, plums	20	9.68 ± 6.88	Spain	González-Weller et al. [25] ¹

Table 3: Mean aluminium content in selected fruit and vegetables.

-Unknown number of samples

¹Study conducted using ICP-OES

²Study conducted using ICP-MS

Product	Number of samples	Al mean content (mg/kg) ± SD	Origin	Reference
Fresh meat				
Poultry, rabbit	20	6.35 ± 2.83	Spain	González-Weller et al. [25] ¹
Viscera	20	11.19 ± 6.42	Spain	González-Weller et al. [25] ¹
Red meat	40	9.31 ± 4.85	Spain	González-Weller et al. [25] ¹
Porcine meat (muscle)	-	0.21	France	Leblanc et al. [48] ²
Porcine meat (kidney)	-	0.52	France	Leblanc et al. [48] ²
Processed meat products				
Ham	20	1.99 ± 0.44	Spain, Italy	González-Weller et al. [25] ¹
Ham, cooked	-	1.2	USA	Greger et al. [42]
Sausage, bologna, salami	20	3.06 ± 1.09	Spain, Italy	González-Weller et al. [25] ¹
Hamburger	-	2.00	USA	Soni et al. [15]
Milk and its derivatives				
Cheddar cheese	-	3.9 ± 3.9	USA	Saiyed and Yokel [40] ³
Processed American cheese slices	-	470 ± 200	USA	Saiyed and Yokel [40] ³
American pasteurized prepared cheese	-	6.6 ± 4.4	USA	Saiyed and Yokel [40] ³
Natural cheese	-	1.57	USA	Soni et al. [15]
Processed cheese	-	29.7	USA	Soni et al. [15]
Soft cheese	-	0.4	Greece, Turkey	Elbarbary and Hamouda [49] ³
Milk, whole	20	0.37 ± 0.09	Spain	González-Weller et al. [25] ¹
Milk, skimmed and semi skimmed	20	0.82 ± 1.59	Spain	González-Weller et al. [25] ¹
Milk	-	0.70	USA	Soni et al. [15]
Yogurt	40	0.82 ± 0.50	Spain	González-Weller et al. [25] ¹
Plain yogurt	36	0.72 ± 0.57	Spain	Luis et al. [4] ¹

Flavored yogurt	36	0.45 ± 0.27	Spain	Luis et al. [4] ¹
Eggs				
Hens' eggs (homogenized)	40	2.93 ± 2.95	Spain	González-Weller et al. [25] ¹
Hens' eggs	-	0.107	USA	Schenk et al. [38]

Table 4: Mean aluminium content in animal products.

-Unknown number of samples

¹Study conducted using ICP-OES

²Study conducted using ICP-MS

³Study conducted using FAAS/GFAAS

Product	Number of samples	Al mean content (mg/kg) ± SD	Origin	Reference
Fresh fish				
<i>Mullus surmuletus</i> (Red rock mullet)	16	3.48 ± 3.96	Spain	Dorta et al. [50] ¹
<i>Pseudupeneus prayensis</i> (African mullet)	12	0.92 ± 0.71	Spain	Dorta et al. [50] ¹
Sarpa salpa	40	2.86 ± 2.23	Spain	Afonso et al. [51] ¹
Chelon labrosus	40	2.84 ± 2.69	Spain	Afonso et al. [51] ¹
White fish	20	3.57 ± 3.23	Spain, Morocco, South Africa, Mauritania	González-Weller et al. [25] ¹
Oily fish	20	3.90 ± 1.97	Spain, Morocco, South Africa, Mauritania	González-Weller et al. [25] ¹
Fish	-	0.40	USA	Soni et al. [15]
<i>Trachurus</i> genera (blue jack mackerel)	60	1.343-49.24*	Turkey	Özden [52] ²
<i>Trachurus</i> genera	24	10	Turkey	Küpeli et al. [53] ¹
<i>Trachurus</i> genera	142	2.408-5.857*	Spain	Rivas et al. [54] ³
<i>Saurida undosquamis</i>	45	0.831	Turkey	Türkmen et al. [55] ³
<i>Sparus aurata</i>	45	0.919	Turkey	Türkmen et al. [55] ³
<i>Mullus barbatus</i>	45	2.228	Turkey	Türkmen et al. [55] ³
Seafood				
<i>Sepia officinalis</i> (cuttlefish)	-	10.2	Spain	Villanueva and Bustamante [56] ²
<i>A crassispina</i> (sea urchin)	26	26.9 ± 30.1	South Korea	Choi et al. [57] ¹
<i>S. japonicus</i> (sea cucumber)	63	30.3 ± 25.1	South Korea	Choi et al. [57] ¹
<i>H. roretzi</i> (sea squirts)	67	38.8 ± 22.6	South Korea	Choi et al. [57] ¹
<i>S. plicata</i> (warty sea squirts)	66	204.6 ± 166.4	South Korea	Choi et al. [57] ¹
Seaweed				
Red seaweed	18	27.1 ± 22.6	Asian and European Union	Rubio et al. [29] ¹
Brown seaweed	22	7.43 ± 5.26	Asian and European Union	Rubio et al. [29] ¹
<i>Laminaria</i> (brown seaweed) and <i>Porphira</i> (red seaweed)	-	8.78 ± 18.6	Spain	Larrea-Marín et al. [58] ¹
Laver	53	15.5 ± 9.36	South Korea	Khan et al. [59] ²
Sea tangle	45	4.89 ± 4.15	South Korea	Khan et al. [56] ²
Sea mustard	58	4.14 ± 3.36	South Korea	Khan et al. [56] ²
Hijiki	27	6.56 ± 4.47	South Korea	Khan et al. [56] ²
Gulf weed	15	52.1 ± 7.34	South Korea	Khan et al. [56] ²

Table 5: Mean aluminium content in different fish and seafood according to country of origin.

*Maximum and minimum value, respectively

-Unknown number of samples

¹Study conducted using ICP-OES

²Study conducted using ICP-MS

³Study conducted using FAAS/GFAAS

Finally, the highest concentration of Al found in milk and its derivatives was that found in processed American cheese slices from the USA, with an average content of 470 mg/kg, this is due to the use of sodium aluminium phosphate as the emulsifying agent in processed cheeses [15,40].

Fish and other seafood

Table 5 shows the Al content (mg/kg) in different fish and seafood depending on the origin. As a result of the acidification of the soils, soluble aluminium (Al³⁺) can reach the aquatic environment easily. In addition, pollution from anthropogenic activities is an important means of releasing Al into the marine environment. However, Al accumulation in fish, marine plants and other organisms depends on several factors such as the species, age, sex, etc [60,61]. Several studies have reported that the aluminium polymerization from aqueous aluminium, which takes place in a pH of between 5.0 and 6.0, which leads to acute toxicity in fish because it causes damage to the functioning of fish gills resulting in respiratory and ion regulatory dysfunctions [62,63].

As for fresh fish, the highest Al levels were reported in samples of oily fish (3.90 mg/kg) from Spain, Morocco, South Africa and Mauritania [25].

Seafood is the group with the highest reported Al levels, with an average Al concentration of 204.6 mg/kg in warty sea squirts from South Korea [57]. This fact could be explained by the characteristics of the organisms, e.g., cephalopods which filter the water and thereby accumulate higher metal contents [64,65].

Al content in edible seaweeds is higher than in fish, reaching an Al level of 52.1 mg/kg in gulfweed (microalgae) from South Korea [59]. Seaweeds can accumulate metals present in the aquatic environment and in several cases, seaweeds can act as bio indicator of marine contamination as a method of monitoring the pollution [66].

Estimation of the spatiotemporal aluminium levels along the time

Figures 3, 4, 5 and 6 show the general evolution of the Al content in the different types of food analysed by the consulted authors.

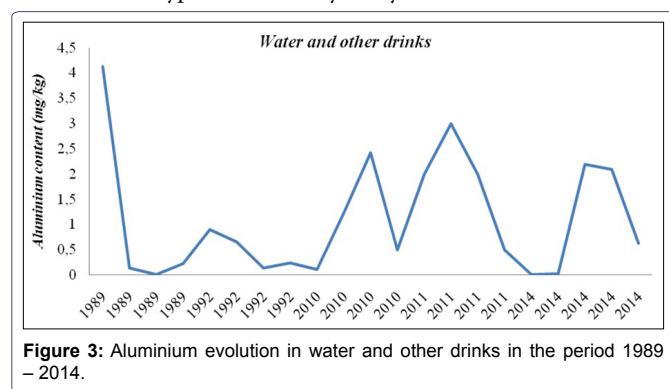


Figure 3: Aluminium evolution in water and other drinks in the period 1989 – 2014.

Al levels in drinking water and other beverages have increased over time and this fact can be explained by the use of Al-based compounds in the treatment of drinking water [21].

In the case of fruit and vegetables, the Al concentration was highest in 2010, and it should be mentioned that the highest concentrations were found in selected fruit and vegetables from the Canary Islands (Spain), where the crop soils are acidic as a result of the volcanic nature of the islands [25].

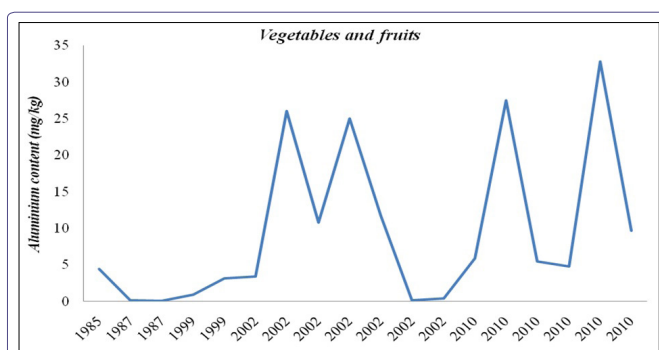


Figure 4: Aluminium evolution in fruit and vegetables in the period 1985 – 2010.

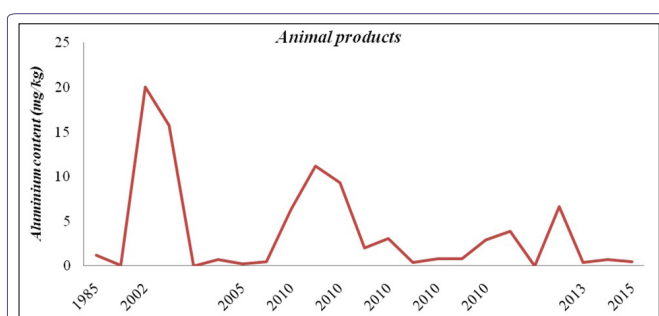


Figure 5: Aluminium evolution in animal products in the period 1985 – 2015.

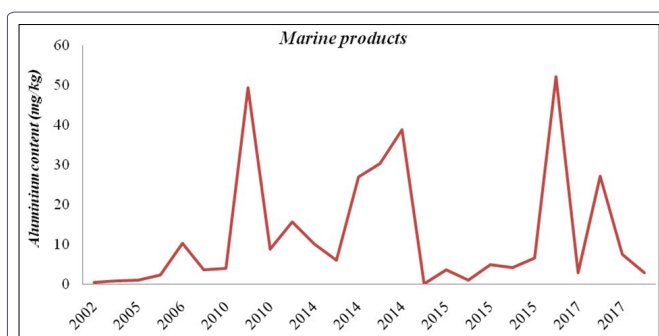


Figure 6: Aluminium evolution in marine products in the period 2002 – 2017.

On the other hand, Al levels in different animal products have decreased over time.

Al concentrations in different marine products peaked in some cases in 2010, 2014 and 2015. However, Al levels between 2015 and 2017 were slightly lower than in the first period (2002-2014). Nevertheless, there are wide differences in marine products between the areas where the products were collected.

The differences of Al levels over time may be due to the development of analytical techniques, which are currently able to detect lower levels. However, the wide use of Al-packaging to preserve food and acidification of soils can lead to higher Al levels in food.

Estimation of the dietary intake

The Tolerable Weekly Intake (TWI) set by the EFSA at 1 mg/kg body weight/week [7] and, the average consumption data of each food group for children and adults given by the AESAN (Spanish Agency of Food Safety and Nutrition) have been used to estimate the Al dietary intake [67]. The average consumption data by the Spanish population are: roots and tubers (67.6 g/day children, 71.71 g/day adults),

Food group	Al mean content (mg/kg) ^a	Children (7-12 years old, 34.48 kg ^b)		Adults (≥17 years old, 68.48 kg ^b)	
		EDI (mg/day)	% TWI	EDI (mg/day)	% TWI
Beverages	1.11	0.05	1.02	0.14	1.43
Eggs	1.52	0.04	0.81	0.05	0.51
Fish and seafood	11.9	0.75	15.2	1.12	11.4
Fruits	6.84	1.45	29.4	1.78	18.2
Meat and its derivatives	5.98	0.92	18.7	0.99	10.1
Milk and its derivatives	3.05	1.31	26.6	1.07	10.9
Roots and tubers	9.60	0.65	13.2	0.69	7.05
Vegetables	16.8	1.60	32.5	3.18	32.5

Table 6: Estimated intake of aluminium through the diet.

^aUsing the average aluminium content but removing the deviated results

^bAverage weight established by the AESAN [64]

EDI: Estimated Daily Intake

TWI: Tolerable Weekly Intake

vegetables (95.35 g/day children, 189.12 g/day adults), fish and seafood (63.3 g/day children, 94.41 g/day adults), eggs (24.34 g/day children, 31.21 g/day adults), fruits (211.45 g/day children, 259.56 g/day adults), milk and its derivatives (428.37 g/day children, 350.52 g/day adults), meat and its derivatives (153.8 g/day children, 167.15 g/day adults), beverages (42.61 g/day children, 128.55 g/day adults).

Table 6 shows the Estimated Daily Intake (EDI), which is the approximated intake of Al per day, and the percentages of contribution to the TWI of aluminium from the diet. The results of EDIs in mg/day are obtained multiplying the average consumption data by the Al concentration found in each food group. Otherwise, the percentages of contribution to the TWI are obtained by dividing the EDIs values by the TWI and by the average body weight, which is given by the AESAN [67], and multiplying the result by 7 to express the result per week as the TWI, and by 100 to express the result as a percentage.

The EDIs for children follow the sequence: vegetables > fruits > milk and its derivatives > meat and its derivatives > fish and seafood > roots and tubers > beverages > eggs. In the case of adults, the EDIs values are sequenced as follows: vegetables > fruits > fish and seafood > milk and its derivatives > meat and its derivatives > roots and tubers > beverages > eggs.

The food groups with the most notable contribution to the TWI of Al are fruits (18.2% adults, 29.4% children) and vegetables (32.5% for adults and children).

Depending on the average consumption of each food group, the total dietary intake of Al can be above the TWI established by the EFSA.

Conclusion

Aluminium is a toxic metal known as a neurotoxic agent. The accumulation of Al in the brain can lead to diseases such as the impairment of memory and neurodegenerative diseases like Alzheimer's disease, Parkinson's disease, etc.

Aluminium is widely present in the diet. Al levels in vegetable, fruit or seafood groups are higher than in other groups. The gastrointestinal absorption of Al is low, but there are many dietary sources and therefore, in some cases, the Al level may pose a health risk. It is therefore necessary to control the level of aluminium in certain types

of foods, because of the toxic effects of aluminium in the human organism.

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