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Aluminium content of selected foods and food products

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Abstract

For many years aluminium was not considered harmful to human health because of its relatively low bioavailability. In 1965, however, animal experiments suggested a possible connection between aluminium and Alzheimer's disease.

Oral intake of foodstuffs would appear to be the most important source of aluminium. Consequently, the joint FAO/WHO Expert Committee on Food Additives reduced the provisional tolerable weekly intake value for aluminium from 7 mg kg^{-1} body weight/week to 1 mg kg^{-1} body weight/week. Analysis of aluminium content of a number of foods and food products was therefore undertaken in order to evaluate the nutritional intake of aluminium.

A total of 1,431 samples were analysed within the scope of this study. The data obtained allow a preliminary but current depiction of the aluminium content of selected non-animal foods, food products and beverages.

Keywords: aluminium, contamination, food additives, PTWI, TDI

Introduction and objective

Aluminium is the third most abundant element in the earth's crust and is therefore a natural component of drinking water and foodstuffs and is a component of many manufactured materials. Exposure of the human body to aluminium may be by food [1-8] including drinking water, fruit juices wine and beer [9-11], articles of daily use that are made of aluminium, cosmetics and pharmaceuticals such as local therapeutic agents, anti-diarrhoeal drugs or antacids. Increased aluminium exposure can be compensated for by excretion via intestines and normal, healthy kidneys. Kidney insufficiency was shown to result in increased aluminium concentrations in the kidneys of dialysis patients, possibly resulting from dialysis fluids that may contain substantial concentrations of aluminium [12].

For many years, aluminium was not considered a health threat because of its relatively low bioavailability. In 1965, however, animal experiments suggested a possible connection between aluminium and Alzheimer's disease, whereby aluminium salts were injected directly into rabbit brain where they caused tissue alterations (for a review see [13]). Increased aluminium concentrations were found in the brains of deceased Alzheimer's patients. Other studies, however, have been unable to find definite indications supporting the hypothesis that aluminium plays a causative role in Alzheimer's disease or causes pathological alterations *in vivo* in the species studied [14-20].

In the 1970s, the issue of toxicity of aluminium gained importance after Berlyne et al. (1970) reported on increased aluminium concentrations in the serum of nephropathic persons [21]. The findings of Alfrey et al. [22] increased concerns about an increased oral intake of aluminium since these findings were the first to establish a connection between neurologic diseases of dialysis patients and an increased intake of aluminium in the organism. In a more recent study aluminium is also discussed as as an endocrine disruptor in female Nile Tilapia (*Oreochromis niloticus*) [23].

Food is unquestionably the main source of aluminium intake, whereby the source is considered either primary or secondary. The primary content is the natural content of food caused by uptake from the geologic surrounding during growth and is for all practical purposes unavoidable. The secondary content is the primary content plus any possible contamination from aluminium

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articles that come into contact with food and additives as well as veterinary drugs, fertilisers and the air. Table 1 shows the main, permissible secondary aluminium sources that may lead to an aluminium accumulation in food.

The health safety of aluminium intake has long been a subject of controversial debate. Table 2 depicts some of the related evaluations published by International Scientific Committees. In the European Community, the Scientific Committee for Food of the European

Table 1 Permissible secondary aluminium sources

Regulation	Designation	Permitted concentration	Admitted for
Feedstuffs Regulation	E 561 Vermiculi Mg Al Fe silicate		All feedstuffs
	E 554 Na Al silicate		All feedstuffs
	E 559 Kaolinite clay containing Al silicate		All feedstuffs
	E 599 Perlite Na Al silicate	50,000 mg/kg[a]	In mineral feedstuffs only
Drug colouring regulation	E 173 Aluminium		
	Aluminium salts of all other colours		
Animal drug residues regulation	Aluminium distereate		All animals for food production
	Aluminium hydroxy acetate		
	Aluminium phosphate		
	Aluminium tristereate		
	Aluminium trisalicylate		
	Magnesium aluminium silicate		
	Aluminium hydroxide		
Cosmetics regulation	Some particular aluminium compounds only are forbidden or subject to limited admission		
Ecology regulation	Aluminium calcium phosphate		As fertiliser/soil improvement agent
Tobacco regulation	Aluminium hydroxide		White burning agent
	Aluminium oxide		Material for filters
	Aluminium sulphate		Material for filters
	Aluminium oxide		Material for filters
	Aluminium		Material for filters
	Aluminium potassium sulphate		Chewing tobacco
Drinking water regulation.	Aluminium	0.2 mg/l[b]	
Wine regulation.	Aluminium	8 mg/l[c]	Wine and other products
Food additives admission	E 173 Aluminium	qs*	Coverings of sugar confectionery for cake decoration, pastries
	E 520 Aluminium sulphate	30 mg/kg[d]	Egg white
	E 521 Aluminium sodium sulphate		
	E 523 Aluminium ammonium sulphate	200 mg/kg[d] expressed as aluminium	Candied, crystallized crystallised and glazed fruit and vegetables
	E 541 sodium aluminium phosphate, acidic	1,000 mg/kg[d] expressed as aluminium	Fine bakery wares (scones and sponge wares only)
	E 554 Sodium aluminium silicate	10 g/kg[d]	Sliced or grated hard, semi-hard and processed cheese
	E 556 Calcium aluminium silicate	30 g/kg[d]	Spices
	E 556 Calcium aluminium silicate	30 g/kg[d]	Products for the greasing of baking trays
	E 559 Aluminium silicate	As required	Confectionery excluding chocolate (surface treatment only)
	E 555 Aluminium potassium silicate	As required	Dye for colouring of Easter eggs

[a]Feedstuffs Regulation (FuttMV 1981) Full citation: "Feedstuffs regulation in the version introduced on 24 May, 2007 (BGBl. I S. 770), that was last amended by Article 1 of the regulation from December 20th, 2010 (eBAnz 2010 AT135 V1)", Germany. [b]Regulation concerning the quality of water for human usage (Drinking Water Regulation - TrinkwV 2001), Germany. [c]Wine Regulation (WeinV 1995), Full citation:"Wine regulation in the version introduced on 21 April, 2009 (BGBl. I S. 827), that was last amended by article 4 of the regulation from March 28th, 2011 (BGBl. I S. 530) ", Germany. [d]Additive Regulation (ZVerkV 1998), Full citation: "Additive regulation from 29 January, 1998 (BGBl. I S. 230, 269), that was last amended by article 2 of the regulation from March 28th, 2011 (BGBl. I S. 530)", Germany.

Table 2 Evaluation of aluminium and aluminium compound intake from foodstuffs

International Body			Evaluation
JECFA 1970	Aluminium silicate Sodium aluminium silicate		No limit
JECFA 1978	Aluminium (metal) for surface colouring		Not considered to present a hazard
JECFA 1982	Sodium aluminium phosphate (acidic and basic)	Preliminary ADI value	0 to 0.6 mg Al/kg/bw/day
JECFA 1986/87	Aluminium salts (additives)	Preliminary ADI value	0 to 0.6 mg Al/kg/bw/day
JECFA 1986/87	Calcium aluminium silicate	ADI (group ADI for SiO_2 and silicates)	Not specified
JECFA 1989	Aluminium (foodstuffs incl. additives)		PTWI 0 to 7 mg Al/kg/bw/week
SCF 1990	Aluminium (all sources)		PTWI 0 to 7 mg Al/kg/bw/week
JECFA 2007	Aluminium (all sources)		PTWI 1 mg Al/kg/bw/week
SCF 2008	Aluminium (all sources)		PTWI 1 mg Al/kg/bw/week

JECFA, Joint FAO/WHO Expert Committee on Food Additives [25]; ADI, acceptable daily intake, PTWI, provisional tolerable weekly intake; SCF, Scientific Committee for Food of the European Communities [24].

Communities (SCF) [24] set the provisional tolerable weekly intake (PTWI) used by Joint FAO/WHO Expert Committee on Food Additives (JECFA) for substances that may accumulate in the body) value at 0 to 7 mg kg^{-1} body weight per week (Table 2). This occurred in 1990, 1 year after the Joint FAO/WHO Expert Committee on Food Additives [25] presented their evaluation. Among various topics considered during its 77th meeting in 1995, JECFA [25] dealt with the toxicological evaluation of aluminium. The committee reduced the PTWI value for aluminium from 7 mg kg^{-1} body weight per week to 1 mg kg^{-1} body weight per week. Additionally, the European Food Safety Authority EFSA set the weekly aluminium intake to 1 mg kg^{-1} body weight in 2008 (Table 2).

On the basis of values obtained from the literature and data from the 1984 German Society of Nutrition Report, Treptow and Askar tried to establish the daily aluminium intake in the Federal Republic of Germany. The authors estimated the aluminium intake to be less than 5 mg per day [26]. Treier and Kluthe [27] analysed 446 foodstuff samples as well as 94 daily meals and determined that food of plant origin (n = 283) had significantly higher aluminium content than food of animal origin (n = 163). In a study of Treier [28], an aluminium intake of less than 5 mg per day and person was found.

Schlegel and Richter [29] reported on the analysis of 451 foodstuffs of different categories. These authors also determined that the aluminium contents of plant derived foods were more significant and that processed foods contained more aluminium than the primary products [29]. Consequently, food was considered to be increasingly contaminated by aluminium packaging [30-33].

In general, data on the nutritional intake of aluminium in Germany still seems to be incomplete and is, for the most part, not up to date. In particular, data on the amounts of foodstuffs that must be ingested to reach the toxicologically tolerable intake of aluminium is lacking. It was therefore of interest to analyse an extensive number of foodstuffs for their aluminium content in order to develop a current basis for the evaluation of the nutritional intake of aluminium.

Materials and methods

A total of 1,431 samples [65 flours, 37 baking premixes, 107 breads, 60 loaf-shaped yeast fruit cakes, 38 pastries in aluminium trays, 185 salted pretzels and similar savoury biscuits, 24 pastas, 12 herb-teas, 37 cocoa powders, 84 chocolates, 115 confectioneries, 50 malts, 237 beers (177 bottled beers, 40 draught beers and 20 canned beers), 59 fruit juices and fruit juice beverages, 65 wines and fruit wines, 31 ready-cooked meals in aluminium trays, 16 soups from the catering trade, 171 mineral waters and spring waters and 38 diverse products] were analysed.

The samples which were for the most part from Hessian enterprises (mills, bakeries) and from the retail market were collected for analysis within the scope of governmental control and inspection of foodstuffs.

For quantitative determination of aluminium concentration, samples were pre-treated in accordance with DIN EN 13804 [34] depending upon the physical properties of the sample. Liquid and powder samples, the homogeneity of which was considered sufficient according to manufacturing procedures, were shaken in order to avoid segregation effects and were then dissolved in solvent. Lumpy food samples were homogenised by means of a Retsch Grindomix GM 200 knife mill (Retsch, Haan, Germany).

Homogenised compact samples, soups, fruit juices as well as red wines and fruit wines were solubilised by means of heated-under-pressure microwave dissolution in accordance with DIN EN 13805 [35]. Samples of one 1 mg were added to 2 ml of deionised water (18.2 MΩ cm), 3 ml of nitric acid (Merck, 65% Suprapur®) and 2 ml of hydrogen peroxide (Merck, 30% Suprapur®) as oxidiser. Dissolution for three parallel batches each per sample was made in an Anton Paar Multiwave 3000 microwave digestion system (Anton Paar, Graz-Straßgang, Austria), whereby the power was increased to 1,000 W after 15 min and maintained for 30 min.

The aluminium concentrations in the dissolution solutions as well as in mineral waters and white wines that had been acidified with nitric acid were determined by means of inductively coupled plasma mass spectrometry (ICP-MS), in accordance with DIN EN ISO 17294 [36]. ICP-MS metering was performed using a Perkin-Elmer SCIEX ELAN 5000 (PerkinElmer, Waltham, MA, USA) with cross-flow nebulizer and Scott type spray chamber. For calibration, commercially obtainable ICP standards by Merck (CertiPUR®, Whitehouse Station, NJ, USA) were used. Standards and samples were diluted with deionised water (18.2 MΩ cm) and high-purity acids. Rhenium was used as internal standard. Method validation for the aluminium parameter demonstrated a limit of detection of 0.4 mg kg^{-1} (mineral water 0.1 mg l^{-1}). Quality assurance comprised the use of standard reference materials and comparative inter-laboratory studies.

Quality assurance

Since adequate round-robin testing has not been performed on aluminium in foodstuffs the validation of the methods was carried out in the following manner. Validation was performed on material from pretzels. In eleven samples of water extracted pretzel material (0.5 g/10 ml each) an arithmetic mean of 3.6 mg aluminium per kilogramme material was determined. TM-28.2 Lake Ontario water from Environment Canada, Quebec, (47 μg/l) was used a standard reference additive. A solution containing 0.1 mg/l was added to four of the pretzel extractions and a solution containing 0.2 mg/l was added to another five extractions. The following statistical characteristics were determined: limit of detection 0.8 mg/kg, limit of quantification 2.6 mg/kg, standard deviation of repeatability = 0.4 mg/kg, relative standard deviation of repeatability (coefficient of variation) 15%, correlation coeffizient 0.97, recovery 103%.

Using the t-factor method the standard deviation of repeatability was calculated to represent an uncertainty of measurement of 0.2 mg/kg (95% level) or 0.04 mg/kg (99% level). These statistical characteristics show that the methods for the samples to be tested provide excellent accuracy, precision and sensitivity.

Results and discussion

Aluminium compounds may be used as additives in foodstuffs. Additional contamination can occur through articles that come into contact with food and that contain aluminium since aluminium is unstable in the presence of acids and bases if the protective transparent oxide film is damaged, e.g. by fine fissures. Aluminium concentrations of the samples analysed in this study were between 0.4 and 737 mg kg^{-1} or mg l^{-1} (Table 3).

This wide range of concentrations may be due to the effect of important secondary factors. The relative frequency distribution of all analysed samples is shown in Figure 1.

Of all the samples, 77.8% had an aluminium concentration of less than 10 mg kg^{-1}. Of the samples, 17.5% had aluminium concentrations between 10 and 100 mg kg^{-1}. In only 4.6% of the samples, aluminium concentrations greater than 100 mg kg^{-1} were found.

Cereal products

The aluminium concentrations of cereal products (flour, baking premix, bread and pastries in aluminium trays, salted pretzels and similar savoury biscuits) were between 1 and 737 mg kg^{-1} (Table 3). Figure 2 provides a summary of the relative frequency distribution of the aluminium contents of all cereal products.

Of these samples, 82% (n = 425) had an aluminium concentration below 10 mg kg^{-1}. In 81 samples (20%), an aluminium concentration of 10 to 100 mg kg^{-1} was found. Only 2% of the samples (n = 10) had aluminium concentrations of more than 100 mg kg^{-1}. The average aluminium concentration of the diverse flour products was 4 mg kg^{-1} (Table 3).

The baking premixes are ready-to-bake mixtures that are used to make breads or various pastries after adding liquid and are mainly produced for home use. The aluminium concentrations of the baking premixes analysed in this study (mainly ready-to-bake bread flours) were between 1.3 and 737 mg kg^{-1}. The median value, however, was only 6 mg kg^{-1}. In three samples, aluminium concentrations greater than 100 mg kg^{-1} were detected. The sample containing 113 mg kg^{-1} of aluminium was a ready-to-bake premix containing nuts. The other two samples (ready-to-bake pastry flours) with a relatively high aluminium concentration (566 and 737 mg kg^{-1}) were baking premixes to which sodium aluminium sulphate had been added and that were, according to the manufacturer, originally produced for export but were mistakenly put into circulation in Germany.

The aluminium concentration of the analysed 107 breads was between 1 and 14 mg kg^{-1}; 105 samples had aluminium concentrations of less than 10 mg kg^{-1}; only two samples only had concentrations of more than 10

Table 3 Aluminium in foodstuffs (milligrammes per kilogramme or milligrammes per litre)

Product	Number	Minimum	Maximum	Mean value[a]	Median value
Flour	65	1	19	4	3
Baking premix	37	1	737	51	6
Bread	107	1	14	3	2
Loaf-shaped yeast fruit cakes	60	3	22	10	9
Fine bakery wares in aluminium trays	38	1	537	19	3
Salt pretzels and similar savoury biscuits	185	2	218	13	4
Pasta	24	1	76	10	4
Herb-teas	12	14	67	40	45
Cocoa powder	37	80	312	165	160
Chocolate	84	6	150	48	39
Confectionery	115	1	184	17	8
Malt	50	1	12	7	7
Beer and mixed drinks containing beer, draught beer	237	0.4	4.2	0.5	0.4
Fruit juice and fruit juice drinks	59	0.4	47	3	1
Wine and fruit wine	65	0.4	15	2	1
Mineral water, spring water and table water	171	0.1	0.07	0.01	0.006
Ready-cooked meals in aluminium trays	31	1	13	3	1
Soups	16	1	15	5	3
Diverse products	38	1	138	16	7
Total	1,431	n.n.	737	19	2

[a]Arithmetic mean

mg kg^{-1} (Table 3). As shown in Table 3, the aluminium concentrations of the pastas analysed in this study were between 1.4 and 76 mg kg^{-1}. Two samples with aluminium concentrations of 71 and 76 mg kg^{-1} were imported products.

The investigation results (1999 to 2006) of the Land of Saxony Control and Inspection Agency show that 89% (n = 227) of the baking premixes analysed there had aluminium concentrations of less than 5 mg kg^{-1}; the aluminium concentration of the remaining samples was below 20 mg kg^{-1}. Treier [28] determined a mean aluminium concentration of 1.9 to 2.6 mg kg^{-1} for bread and pastry. Thus, the results obtained in the present study correspond well with those of this previous study.

The aluminium contents of "pastries in aluminium trays" and "salted pretzels and similar savoury biscuits" are described under "Articles intended to come into contact with foodstuffs" subsection due to a possible migration of aluminium from the trays or the baking trays into these foods.

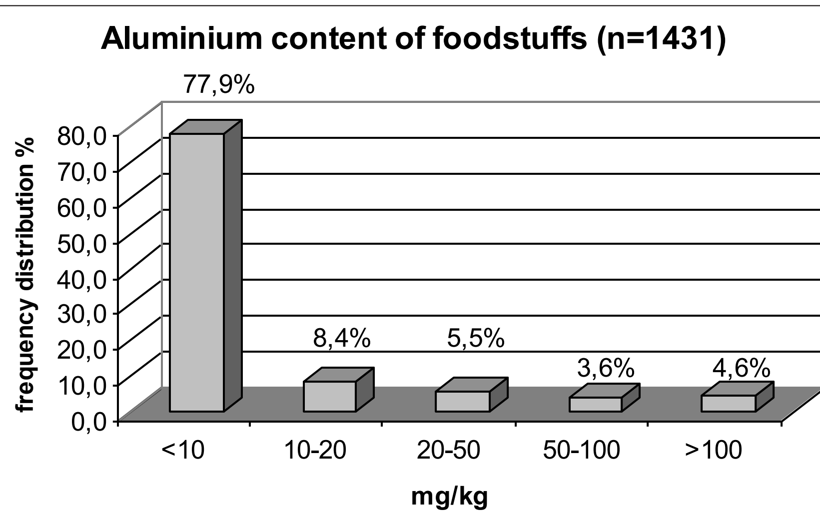

Figure 1 Relative frequency distribution of the aluminium content of foodstuffs (***n*** **= 1,431)**.

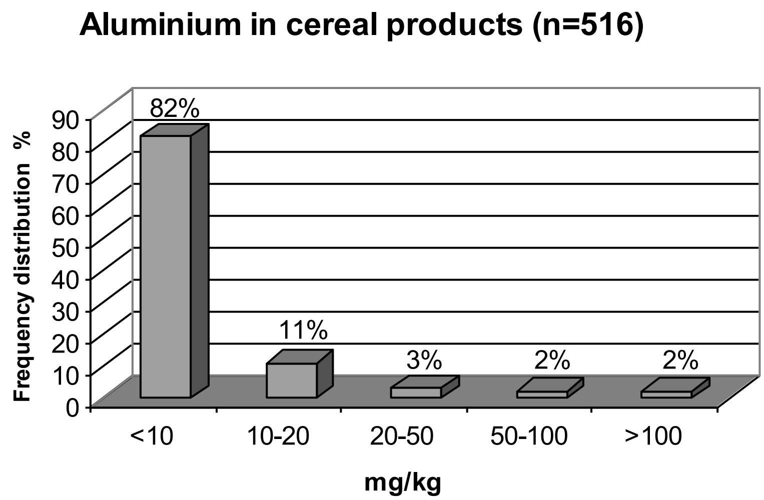

Figure 2 Relative frequency distribution of the aluminium content in cereal products. Baking premixes, breads, pastries, salt pretzels and similar savoury biscuits (*n* = 516).

Herb-tea

The aluminium concentrations of the analysed herb-teas were between 14 and 67 mg kg^{-1} (Table 3). The mean value of the concentrations was 40 mg kg^{-1}; the median value was 45 mg kg^{-1}.

Confectioneries

The comprehensive term confectionery applies to a heterogeneous group of products mainly containing sugar. They are grouped into hard and soft caramels, jelly products, chocolates, chocolate articles and others.

Figure 3 summarises the relative frequency distribution of the aluminium contents of 236 confectioneries analysed in this study (cocoa powders, chocolates and products containing sugar). The mean value of the concentrations of these products was 51 mg kg^{-1}, the relevant median value was 26 mg kg^{-1} (not shown in Table 3). More than one third of these samples contained aluminium concentrations greater than 50 mg kg^{-1} (Figure 3).

The aluminium contents of the products containing sugar and/or chocolate (bear-shaped confectionery made of "gummi" substance, "gummi" sticks, soft gums, fruit gums, sour apple candies, foamed sugar confectionery, etc.) were between 1 and 184 mg kg^{-1} (Table 3). Fifty five percent of the samples (*n* = 62) had less than 10 mg kg^{-1}. Thirty six percent had aluminium concentrations between 10 and 50 mg kg^{-1}.

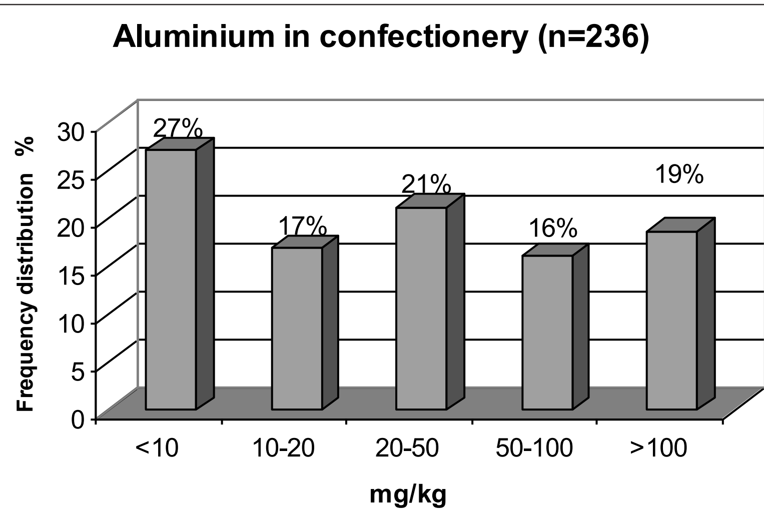

Figure 3 Relative frequency distribution of the aluminium content in confectionery. Cocoa powder, chocolate and confectionery (*n* = 236).

In accordance with the German "Additives Admission Regulation" [37] additives containing aluminium (releasing agents and colour lakes) may be added to such products. According to the content declarations, additives containing aluminium were, however, not used in any of the products tested here.

The term cocoa is as a rule used synonymously for cocoa powder and for products made from the powder. According to the German Cocoa Regulation, cocoa powder is a product made of roasted cocoa beans ground to powder, purified and pared containing at least 20% cocoa butter in the dry matter and not more than 9% water. Cocoa powder can be low in fat or fatless. Cocoa powders were found to have higher aluminium concentrations than almost all of the other foodstuffs analysed in this study (Table 3). The mean aluminium concentration of the products was 165 mg kg^{-1}, the median value was 160 mg kg^{-1}. Since aluminium-containing additives are not allowed to be added to cocoa powder, these values are assumed to be the result of the natural substance in the growing soil. A certain accumulation during production through aluminium utensils and processing equipment cannot, however, be excluded.

Chocolate is a product made of cocoa products and sugars that contains a minimum of 35% total cocoa dry matter of which at least 18% is made up of cocoa butter and at least 14% non-fat cocoa solids. The chocolates analysed in this study are products with varying proportions of cocoa. Considering the aluminium levels found in cocoa, the determined aluminium values can be attributed to the cocoa portion. Contamination caused by aluminium utensils and production equipment cannot, however, be excluded. In summary, cocoa powder and chocolate count amongst those foods that showed the highest aluminium concentrations found in this study (Table 3). Schlegel and Richter [29] analysed ten cocoa and chocolate samples and found between 50 and 150 mg kg^{-1} aluminium concentrations [29] which is within the same range as the values found in the present study.

Malt, beer, mixed beverages containing beer

As shown in Table 3, aluminium concentrations of the malt samples analysed in this study were between 0.4 and 12 mg^{-1} kg. The bottled, canned and draught beers examined here revealed aluminium concentrations between 0.4 and 4.2 mg l^{-1}, with a mean value of 0.5 mg l^{-1} and a median value was 0.4 mg l^{-1}. Aluminium concentrations of the bottled, canned and draught beers varied so slightly that an examination of statistically significant differences of the aluminium contents was dispensed with.

Schlegel and Richter [29] analysed 30 beers and determined aluminium concentrations of less than 1 mg l^{-1}. Jorhem et al. [38] and Müller et al. [39] demonstrated that there was virtually no difference in aluminium content between beverages in cans and beverages in glass bottles. These authors also reported aluminium values of below 1 mg l^{-1}.

Seruga et al. [32] reported that the aluminium contents of seven analysed soft drinks (two cola, two orange, two lemons, one tonic) in aluminium cans increased over a duration of 12-months storage time. The authors attribute this to the low pH value (pH = 2.80 to 3.20) of these beverages. The values determined every month by these authors show, however, comparably low aluminium concentrations between 0.04 mg l^{-1} and 0.8 mg l^{-1} after 12 months at the maximum. Seruga et al. [33] established that after expiration date canned beers contained more aluminium than bottled beers. Aluminium concentrations of canned beers and of bottled beers were, however, determined to be less than 1 mg l^{-1}. Modern aluminium cans are stove-enamelled inside and seams are provided with an additional layer of protecting lacquer. The minimal increase of aluminium content of beverages in cans must be attributed to the high quality of the cans used. This may be assumed to be the reason for the low contents of aluminium in beverages in aluminium cans. Modern beer kegs and draught installations are made of stainless steel with plastic pipes. Accordingly, aluminium concentrations of less than 0.5 mg l^{-1} were determined for draught beers in the present study.

Fruit juice, fruit wine and wine

The aluminium content found was below 5 mg l^{-1} for most fruit juices and fruit juice beverages, but three samples (one sour cherry nectar and two apple juices) showed aluminium concentrations of 19 to 47 mg l^{-1}. According to generally accepted standards in Germany [40], the aluminium concentration of fruit juices should not be greater than 8 mg l^{-1}.

Section 13, subsection 1 number 2 of the German Wine Regulation [41], states that wines may have a maximum aluminium concentration of 8 mg l^{-1}. Only one of the wines analysed in this study had an aluminium concentration that exceeded that limit (14.7 mg l^{-1}). The aluminium concentrations of the remaining samples were below 6 mg l^{-1} (Table 3). The arithmetic mean value of the concentrations was 2 mg l^{-1}. The median value was 1 mg l^{-1}.

Mineral water

A total of 171 samples were analysed for their aluminium content. As shown in Table 3, aluminium concentrations of the mineral and spring waters were far below the permissible limit for drinking water of 0.2 mg l^{-1} (Table 3).

Diverse foodstuffs

These are samples of various foodstuffs such as soy products, food supplements, baby food, tomato pulp, dietary foods, etc. The aluminium concentration of baby food (dry powder, not prepared for immediate consumption) was below 5 mg kg^{-1}.

The "tomato pulp" samples had an aluminium concentration of 1.5 to 15 mg kg^{-1}. The aluminium concentration of dietary foodstuff products was between 1 and 53 mg kg^{-1}. One "soy product" sample had an aluminium concentration of 138 mg kg^{-1}.

Articles intended to come into contact with foodstuffs

Aluminium trays, aluminium baking trays and aluminium cans for beverages fall under this category. These articles, as defined by the Food and Feedstuffs Law as well as by Regulation (EC) No. 1935/2004 are materials and objects destined to come in contact with foodstuffs or already in contact with foodstuffs [41].

As shown in Figure 4, 62 food samples in aluminium trays (91%; cakes to be consumed without further preparation and prepared ready-to-serve meals with various ingredients) had an aluminium concentration below 10 mg kg^{-1}. Six products (9%) contained aluminium in concentrations between 10 and 15 mg kg^{-1}. The mean value of the aluminium concentrations in these foodstuffs was 4 mg kg^{-1}. The relevant median value was 2 mg kg^{-1}.

According to the results of in this study, the aluminium contents of foods in aluminium packaging (beverages, cakes and various ready-to-serve meals) are so small that one must draw the conclusion that migration of aluminium from the packaging into the food can be ignored.

Based on the results of this study, it might even be worth considering the possibility of coating certain packaging (e.g. for milk and milk products, juices and other beverages) with aluminium in order to avoid the migration of heavy metals, additives from packaging materials and printing colours into the foodstuffs. By using aluminium trays or foils, the development of unwanted substances, such as 3-monochlorpropandiol or polycyclic aromatic hydrocarbons, e.g. during barbecuing, might be avoidable.

Aluminium baking trays, like aluminium cans and trays, also belong to the class of materials referred to as articles intended to come into contact with foodstuffs. According to section 31 of the German Food and Feedstuffs Law (2005) it is prohibited to use such objects as intended to come into contact with foodstuffs that do not meet the requirements of Regulation (EC) No. 1935/2004. The use of aluminium baking trays would therefore be inadmissible if aluminium can migrate to brined dough and/or salted pretzels and similar savoury biscuits.

In the present study, 185 salted pretzel samples from Hessian bakeries were analysed for their aluminium contents (Table 3) and were found to be between 1 and 218 mg kg^{-1}. A total of 148 samples (81%) had aluminium concentrations below 10 mg kg^{-1}. In 37 samples (19%), aluminium concentrations between 10 and 218 mg kg^{-1} were found (Table 3). According to generally accepted standards, aluminium concentration of such products as a rule should be below 10 mg kg^{-1}. This can be readily achieves since migration is technically avoidable by Teflon coating or the use of baking paper.

Sixteen soup products (ten ready-for-consumption soups from the Asian catering trade and six dry soups) were analysed. The aluminium concentrations of the ready-for-consumption soups were between 1 and 4 mg

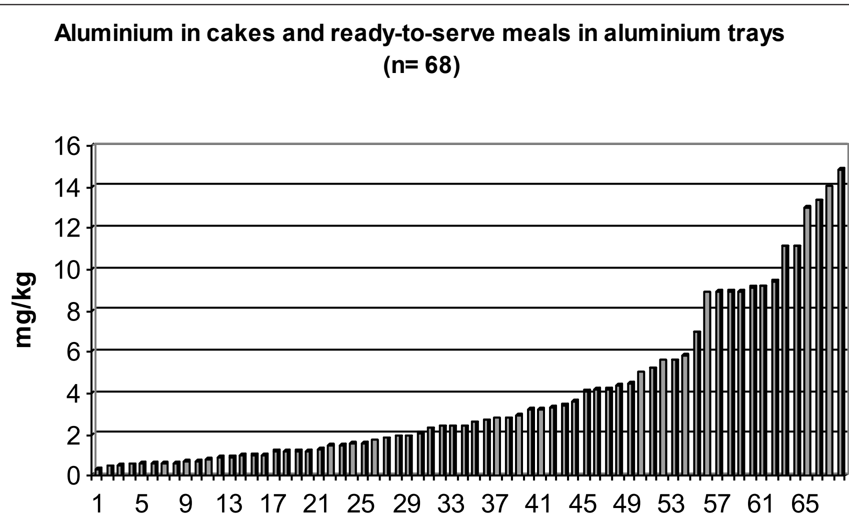

Figure 4 Aluminium in cakes and ready-to-serve meals in aluminium trays (*n* = 68).

kg^{-1}; the dry soups had aluminium concentrations between 5 and 15 mg kg^{-1}. These results suggest that migration of aluminium from cookware as well as from packing into the food may be ignored.

Evaluation of a tolerable weekly intake

The results obtained by the present study do not allow a final evaluation of the PTWI of 1 mg Al kg^{-1} body weight (Table 2) since only foodstuffs of plant origin and beverages were analysed within the scope of this study. Table 4 shows the percentage of the PTWI acquired for adults (70 kg) and children (30 kg) in Germany. These values are based on the weekly uptake in foods, wherever useable data or studies were available, as well as the median and arithmetic mean of aluminium content. Due to the comparatively low aluminium content of beverages, these were disregarded. Table 4 shows that the PTWI for aluminium can be reached only by consumption of large amounts of chocolate [42-44].

In Table 5, the amounts of various foods that would need to be consumed to reach the PTWI values of 70 mg per week (adults) and of 30 mg per week (children) are summarised on the basis of the median, lowest and maximum aluminium concentration (see also Table 4). For adults, the PTWI value would thus be reached in the event of an average weekly consumption (median value) of, e.g. 1,795 g of chocolate and for children 772 g of chocolate. On the basis of the maximum aluminium content of chocolate, the PTWI for adults would be reached after a weekly consumption of 467 g and for children of only 201 g. To reach the PTWI value of 70 mg per week (adults) and of 30 mg per week (children), the weekly consumption of bread would have to be on an average of 5 kg (adults) and 2.15 kg (children).

The data obtained in the present study allow a preliminary but current description of contamination by aluminium through selected foods of plant origin as well as beverages. For these foods, the amount consumed to reach the PTWI could be calculated and possible consumer risks could be shown (Table 4). Other groups of foodstuffs such as tea, coffee, meat, meat products and fish as well as milk and milk products which may contribute to an aluminium intake will be analysed for their aluminium contents by our laboratory. This will allow a more accurate description of the intake of aluminium

Table 4 Percent of the PTWI through uptake of aluminium for adults (70 kg) and children (30 kg)

Product	Average daily intake [kg]	Average weekly intake [kg][a]	Average weekly Al uptake [mg] [b]	Percent PTWI acquired adult [%][b]	Percent PTWI acquired child [%][b]	Average weekly Al uptake [mg][c]	Percent PTWI acquired adult [%][c]	Percent PTWI child [%][c]
Bread[d]	0.178	1.246	3.74	5.34	12.46	2.49	3.56	8.31
Bread[e]	0.133	0.931	2.79	3.99	9.31	1.86	2.66	6.21
Fine bakery wares in aluminium trays[f]	0.058	0.406	7.71	11.02	25.71	1.22	1.74	4.06
Fine bakery wares in aluminium trays[g]	0.045	0.315	5.99	8.55	19.95	0.95	1.35	3.15
Salt pretzels and similar savoury biscuits[h]	0.110	0.767	9.97	14.25	33.24	3.07	4.38	10.23
Pastas[i]	0.067	0.469	4.69	6.70	15.63	1.88	2.68	6.25
Pastas[j]	0.052	0.364	3.64	5.20	12.13	1.46	2.08	4.85
Cocoa powder[k]	0.002	0.015	2.53	3.62	8.44	2.45	3.51	8.18
Chocolate[l]	0.019	0.133	6.38	9.12	21.28	5.19	7.41	17.29
Chocolate[m]	0.065	0.455	21.84	31.20	72.80	17.75	25.35	59.15
Confectionery[n]	0.021	0.147	2.50	3.57	8.33	1.18	1.68	3.92
Confectionery[o]	0.026	0.182	3.09	4.42	10.31	1.46	2.08	4.85
Ready-cooked meals in aluminium trays[p]	0.028	0.193	0.58	0.83	1.93	0.19	0.28	0.64

Percent of the PTWI through uptake of aluminium for adults (70 kg) and children (30 kg) based on the median and arithmetic mean. [a]Calculated by multiplying the average daily uptake (column 2) by a factor of 7. [b]Calculated on the basis of the arithmetic mean - see Table 3. [c]Calculated on the basis of the Median - see Table 3. dMen [42]. [e]Women [42]. [f]Based on an average consumption of males 13 to 64 years in age per year in Germany under the assumption that all fine baking goods were packaged in aluminium containers [43]. [g]Based on an average consumption of females 13 to 64 years in age per year in Germany under the assumption that all fine baking goods were packaged in aluminium containers [43]. [h]Based on the USA with an average yearly consumption of 40 kg. [i]Based on an average consumption of males 13 to 64 years in age per year in Germany [43]. [j]Based on an average consumption of females 13 to 64 years in age per year in Germany [43]. [k] Based on Germany with an average yearly comsumption of 0.8 kg Verzehr. http://www.theobroma-cacao.de/wissen/wirtschaft/deutschland/konsum/. [l]Based on normal consumption [44]. [m]Based on high consumption [44]. [n]Based on an average consumption of males 13 to 64 years in age per year in Germany [43]. [o]Based on an average consumption of females 13 to 64 years in age per year in Germany [43]. [p]Assuming a per-person consumption in Germany in 2010 of 40.2 kg frozen food 25% of which was frozen in aluminium trays. http://www.tiefkuehlkost.de/tiefkuehlmarkt/statistiken/.

Table 5 Consumption of various foods required to achieve the PTWI values adults and children

	Adults Weekly consumption (g)			Children Weekly consumption (g)		
Product	**Minimum[a]**	**Maximum[b]**	**Median value[c]**	**Minimum[a]**	**Maximum[b]**	**Median value[c]**
Bread	70,000	5,000	35,000	30,100	2,150	15,050
Fine bakery wares in aluminium trays	70,000	130	23,333	30,100	56	10,033
Salt pretzels and similar savoury biscuits	35,000	321	17,500	15,050	138	7,525
Pastas	70,000	921	17,500	30,100	396	7,525
Cocoa powder	875	224	438	376	96	188
Chocolate	11,667	467	1,795	5,017	201	772
Confectionery	70,000	380	8,750	30,100	164	3,763
Ready-cooked meals in aluminium trays	70,000	5,385	70,000	30,100	2,315	30,100

Consumption of various foods required to achieve the PTWI values of 70 mg/week (adults) and of 30 mg/week (children) on the basis of the median, the lowest and the maximum aluminium concentration. aWould have to be consumed weekly on the basis of the lowest aluminium concentration, see Table 3. bWould have to be consumed weekly on the basis of the highest aluminium concentration, see Table 3. cWould have to be consumed weekly on the basis of the median value, see Table 3.

through nourishment based on the per capita consumption and will make possible a calculation of the amounts of various foods that must be consumed to reach the PTWI.

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Authors' contributions

TS performed the data calculations, participated in its coordination and drafted the manuscript, HT participated in the study design and coordination, HB conceived of the study, and participated in its design and coordination. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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