

Aluminium in foodstuffs

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The aluminium content of a comprehensive food assortment typical of German nutritional habits was determined within the framework of market basket studies. Carried out in 1988 and 1991, a total of 128 items out of 12 groups of foodstuffs were included in this investigation. Aluminium content of the food assortment was low and comparable with literature data. Most investigated foodstuffs contained $< 5 \mu g \, Al \, g^{-1} \, FM$. Highest concentrations were determined in cocoa/cocoa products $(33 \, \mu g \, g^{-1})$, spices $(145 \, \mu g \, g^{-1})$ and black tea leaves $(899 \, \mu g \, g^{-1})$. In general, aluminium content of frequently consumed food, increased in the following order: beverages, food of animal origin, food of plant origin. With this low level of aluminium concentration in food, there is no danger of aluminium exposure in healthy persons. © 1998 Elsevier Science Ltd. All rights reserved

INTRODUCTION

Environment represents the main source of aluminium burden in human populations with food being of much greater importance than exposure via air (Jones and Bennett, 1986). Environmental aluminium is generally assessed as non-toxic (Bünnig, 1984). Thus, aluminium has been regarded as harmless for healthy human beings until recently. In the literature, however, orally-consumed aluminium is increasingly considered as a contaminant of the food chain, playing a role in the aetiology of neurodegenerative diseases like Morbus Alzheimer and amyotrophic lateral sclerosis (Candy et al., 1986; Crapper McLachlan, 1986; Flaten, 1990; Kruck and McLachlan, 1988; Martyn et al., 1989; McLachlan and de Boni, 1980; McLachlan et al., 1989; Perl, 1988; Perl and Brody, 1980).

General possibilities of oral aluminium exposure of humans occur via foodstuffs, (extensive) use of aluminium-containing food additives, migration of aluminium from saucepan or food packing into foodstuffs, and also drinking water.

Aluminium-containing food additives may considerably alter the aluminium content of foodstuffs. Aluminium, sodium aluminium phosphate, aluminium sulphate, as well as several aluminium silicates are well-established food additives all over the world (Lione, 1983; Domingo, 1993). These food additives are the main source of aluminium intake in the USA (Greger, 1985, 1992; Pennington, 1987). In Europe,

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aluminium-containing additives are of much less importance (Rickenbacher, 1984).

Relevance of aluminium migration into food was recognised at the beginning of the 1980s (Levick, 1980). Nevertheless, this potential source of aluminium exposure is negligible (Trapp and Cannon, 1981; Lione, 1983; Greger et al., 1985; Domingo, 1993; Müller et al., 1993). Only when cooking acid food (for instance tomato purée, sauerkraut, etc.) in uncoated saucepans, does aluminium migration occur (Inoue et al., 1988; Liukkonen-Lilja and Piepponen, 1992; Domingo, 1993).

Owing to its ubiquitous distribution in the environment, all foodstuffs contain aluminium with concentrations, in general, higher in food of plant origin than of animal origin. Aluminium content of comprehensive food assortments has been investigated in the USA and several European countries (Koivistoinen, 1980; Greger, 1985, 1988, 1992; Pennington, 1987; Schamschula *et al.*, 1988; Pennington and Jones, 1989; Sherlock, 1989; Jorhem and Haegglund, 1992; MAFF, 1993). In contrast, recent data on aluminium in German food assortments were only available from Treier and Kluthe (1988).

Therefore, the aluminium content of a comprehensive, typical German food assortment was determined within the framework of market basket studies in 1988 and 1991.

MATERIALS AND METHODS

Food assortments were bought in 9-fold repetition in 1988 and in 6-fold repetition in 1991, within the entire

territory of the new Federal lands of Germany. Individual investigation sites were: 1988: Berlin (twice), Potsdam, Rostock, Stralsund, Greifswald, Leipzig, Weimar and Jena; 1991: Berlin, Schwerin, Magdeburg, Dresden, Potsdam and Erfurt (Fig. 1). Altogether, 954 individual samples of 102 foodstuffs were available in 1988; in 1991, 726 samples of 117 foodstuffs were analyzed. According to Haenel (1979) they belong to the following groups of food: bread, cake and pastries, vegetables, fruit, milk and dairy products, meat and sausage, fish, farinaceous products and flour, cocoa and cocoa products, sugar and sugar-rich products, spices and beverages. All samples were stored until analysis at -18° C in plastic bags or originally packed. Contamination during storage had to be excluded.

Concentration units related to both, fresh matter (FM), i.e. the edible form, and dry matter (DM). Some food were only available in 1988 or in 1991. In the case of significant differences of foodstuff's aluminium content between investigation years, this is indicated by the year of investigation in parentheses.

All foodstuffs were prepared ready for cooking, but raw. In general, fruit and vegetables were peeled and/or washed. All inedible parts, for instance the cores of apples and pears, were removed from the samples. Any exogenous contamination of food can be excluded. Canned products were analyzed without liquid. Preparing for analysis samples were dried to constant weight at 105°C, dry-ashed in a muffle furnace at 450°C and dissolved in diluted hydrochloric acid. Aluminium determination was carried out by graphite furnace atomic absorption spectroscopy with the device combination AAS 3030, HGA 400, AS 40 (Perkin Elmer) and a magnesium nitrate matrix modifier (Merck). Reference material IAEA H-9 (mixed human diet) was used for checking the method. A good accordance between reference results $(16.6 \,\mu g \,Al \,g^{-1})$ and own analysis $(17.4 \,\mu g \, Al \, g^{-1})$ could be observed. Relative and absolute detection limits of the method are at $1.3 \,\mu g \, litre^{-1}$ and 26 pg (pipette volume 20 μ l), resp. The analytical procedure has been described in detail elsewhere (Müller et al., 1995).

Data processing and statistical assessment were carried out using data bank FoxPro for Windows (version 2.6, Microsoft Corp.) and statistical package SPSS/PC+ for Windows (version 6.01, SPSS Inc.).

RESULTS

Bread, cake and pastries

Bread, cake and pastries are of special importance for both consumed food quantity and energy intake. The aluminium content of bread, cake and pastries covered a range from 3.4 up to $22 \mu g$ per g FM (Table 1). Wheat and rye bread, toasted bread, white bread and rolls, which are the most frequently consumed bread varieties

in Germany, contained about $6 \mu g$ Al per g FM. The average aluminium concentration of coarse-grained wholemeal rye bread was lower; it amounted to $3.4 \mu g$ per g FM. The investigated kinds of cake showed aluminium concentrations of $4.3 \mu g$ per g (thin sponge cake with crumble topping) to $6.3 \mu g$ per g (sponge cake). With (on average) $22 \mu g$ Al per g FM, biscuits proved to be the item richest in aluminium of this group. This relatively high aluminium content may result from the use of aluminium-containing food additives. Influences of the investigation year on bread, cake and pastry aluminium concentrations were not observed.

Vegetables, herbs and pulses

As expected, aluminium concentrations of vegetables showed a relatively large variation (Table 2). While the average aluminium content of cultivated Agaricus bisporus was $0.7 \,\mu \mathrm{g} \, \mathrm{g}^{-1}$ FM, a maximum aluminium level of $33 \,\mu \mathrm{g} \, \mathrm{g}^{-1}$ was found in lettuce. The majority of vegetables investigated contained < $10 \,\mu \mathrm{g} \, \mathrm{Al}$ per g FM. Potato, another basic foodstuff, showed aluminium concentrations of $5.4 \,\mu \mathrm{g} \, \mathrm{g}^{-1}$. Compared with cultivated Agaricus bisporus, the aluminium level of mixed mushrooms, a mixture of several Boletaceae species, was high with $17 \,\mu \mathrm{g}$ per g FM. Significant differences (P < 0.001) between the aluminium contents of sauerkraut bought in 1988 and 1991, were found. Causes remain unknown.

Herbs had much higher aluminium concentrations, being comparable with the quantities found in lettuce. With an aluminium content of $8.2\,\mu g$ per g FM, chive was below this order of magnitude. The aluminium content of pulses amounted to about $10\,\mu g$ per g FM. Lentils proved to be the most aluminium-rich pulse, with $16\,\mu g\,g^{-1}$. Investigation year had a highly significant effect on the aluminium concentrations of white beans.

Fruit

The aluminium content of fruit, including apple purée showed a large variation, too (Table 3). Banana contained the lowest amounts (0.7 μ g Al per g FM). An approximately 10-fold higher aluminium concentration was found in kiwi, with 7.9 μ g per g FM. Most investigated fruit contained about 2.0 μ g Al per g FM.

The increase of the aluminium content of apple purée—in comparison with its 'raw material' apple—resulted from food processing and the addition of further components such as sugar and food additives. No differences were detectable between the aluminium contents of domestic fruit, citrus and tropical fruit.

Farinaceous products and flours

Besides the above listed basic foodstuffs of plant origin, farinaceous products and flours, sugar and sugar-rich products, cocoa and cocoa products cannot be excluded from the total assortment of foodstuffs. The average

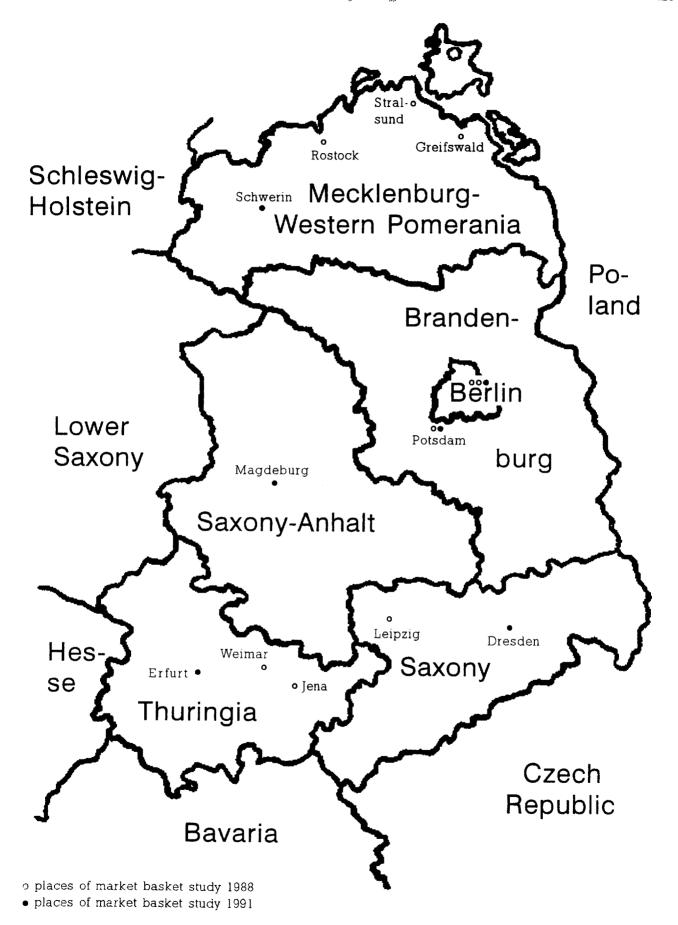


Fig. 1. Survey of investigation places.

aluminium amount of farinaceous products covered a wide concentration range, owing to the very different origin of individual items (Table 4). Thus, semolina bought in 1991 showed the lowest aluminium content with $3.8\,\mu\mathrm{g}$ per g FM, whereas ready-to-serve-soups contained up to 10-fold the aluminium quantities. However, the aluminium concentration of ready-to-serve-soups, flour for dumplings, pancake wheat and vanilla pudding powder is considerably diluted by the addition of water or milk during preparation.

Rice contained, on average, $8.5 \mu g$ Al per g FM. Aluminium concentrations of $5.5 \mu g$ per g FM were

Table 1. Aluminium contents of bread, cake and pastries

Food	μ g Al per g FM	μg	μg Al per g DM			
		\overline{x}	s	Range		
Coarse-grained whole meal rye bread ^a	3.4	6.3	2.2	4.3–10		
Thin sponge cake with crumble topping	4.3	5.4	1.7	3.4-8.6		
Cake with egg topping	4.3	11	6.7	4.5-29		
Crispbread	4.8	5.2	2.2	1.8 - 8.9		
Wheat and rye bread	5.3	8.3	4.0	2.4-18		
Sponge cake	6.3	8.0	4.9	2.2 - 21		
Toasted bread	6.6	9.8	4.2	2.4-18		
White bread ^a	6.7	10	7.2	4.4-22		
Rolls	6.8	9.0	4.1	3.6-19		
Rusk	8.4	9.0	3.4	3.2-17		
Cornflakes ^a	9.5	10	3.7	6.2 - 13		
Biscuits	22	22	17	7.9–60		

^aOnly available in 1991.

Table 2. Aluminium contents of vegetables, herbs and pulses

		,		•
Food	μg Al per g FM	με	r g DM	
		\overline{x}	s	Range
Agaricus bisporusa	0.7	14	6.8	6.5–23
White cabbage	1.3	15	6.4	6.0-28
Sauerkraut (1988)	1.8	19	9.7	8.4-35
Peas, green	1.9	8.7	7 3.8	2.7-15
Tomato	1.9	39	18	11-67
Red cabbage	2.0	21	12	2.8 - 39
Kohlrabi	2.4	23	16	8.1 - 64
Dwarf beans, tinned	2.5	40	18	2.3-60
Asparagus ^a	3.0	66	33	31-122
Carrots, fresh	3.2	46	34	10-91
Cauliflower ^a	3.7	46	25	23-85
Carrots, tinned	3.8	65	34	34-137
Sauerkraut, tinned ^b	4.0	55	28	21-100
Potato	5.4	30	11	12-52
Sauerkraut (1991)	5.8	68	13	47-81
Cucumber	8.4	161	100	29-358
Mixed mushrooms ^a	17	284	148	143-538
Lettuce	33	453	269	201-1011
Chive	8.2	77	50	29-169
Parsley	23	133	38	57-174
Dill	26	255	45	168-300
White beans (1988)	3.2	3.6		
Peas, dried	8.8	9.9	4.8	3.0-20
White beans (1991)	10	12		6.4-22
Lentils ^a	16	18	9.4	6.8 - 33

[&]quot;Only available in 1991.

measured in pasta. It is notable that, when statistical differences of the aluminium content between 1988 and 1991 were observed, items bought in 1991 generally contained lower amounts. Causes may be effects of harvesting site and/or changes in food processing.

Sugar and sugar-rich products

Sugar contained 4.4 μ g Al g⁻¹ (Table 5). The aluminium content of synthetic honey and bee honey, were 8.0 and 5.6 μ g g⁻¹, respectively. This difference remained insignificant. Time-dependent effects on aluminium concentration were observed in the case of candies and jam. Again, those products bought in 1991 were highly significantly (P<0.001) aluminium-poorer than those bought in 1988.

Cocoa and cocoa products

In general, cocoa contains high amounts of minerals and trace elements. Thus, high aluminium concentrations were expected in this foodstuff, as well as in the

Table 3. Aluminium contents of fruit

Food	μ g Al per g FM	μ g Al per g DM			
		\overline{x}	s	Range	
Banana ^a	0.7	3.7	1.1	2.1-5.1	
Lemon	1.4	16	8.5	7.9-38	
Pineapple ^a	1.8	14	3.5	9.4-18	
Pear ^a	1.8	15	3.7	8.9-20	
Apple	2.1	18	12	2.9-44	
Orange	2.2	17	10	6.5-40	
Apple purée	3.6	24	11	8.9-53	
Kiwi ^a	7.9	50	16	3068	

^aOnly available in 1991.

Table 4. Aluminium contents of farinaceous products and flours

Food	μ g Al per g FM		μ g Al per g DM			
		\overline{x}	S	Range		
Semolina (1991)	3.8	4.2	1.6	2.4-6.3		
Wheat flour (1991)	4.1	4.9	1.4	3.1 - 7.1		
Pancake wheat (1991)	4.8	5.4	1.2	3.7-6.8		
Corn flour ^a	5.2	5.9	1.4	3.6-7.8		
Pasta	5.5	6.2	2.4	2.0-11		
Oat flakes	6.3	7.1	3.1	3.8-14		
Pearl barley	6.4	7.3	2.3	3.6-11		
Oat pulp	6.7	7.6	6.1	2.5-27		
Vanilla pudding powder	6.9	8.0	4.0	3.0–17		
Flour for dumplings	7.7	8.8	3.6	2.8-14		
Whole wheat flour	8.1	9.1	4.1	4.1-16		
Rice	8.5	9.7	4.8	2.5–21		
Pancake wheat (1988)	8.9	10	1.9	7.6–13		
Maize flour	13	15	9.2			
Semolina (1988)	15	18	11	1.5–34		
Wheat flour (1988)	17	19	12	2.9–38		
Ready-to-serve-soups	34	38	35	6.0–137		

^aOnly available in 1991.

^bOnly available in 1988.

^bOnly available in 1988.

items made out of cocoa. Our results reflected this expectation (Table 6). Cocoa from 1988 contained, on average, $103 \,\mu g$ Al per g, whereas cocoa bought in 1991 showed an aluminium content of $33 \,\mu g \, g^{-1}$. Provenance of cocoa being unknown, may be the most important reason for this highly significant difference.

Cocoa products also contained relatively high aluminium quantities. They covered a range of 9.4 (milk chocolate) to $27 \,\mu g \, Al \, g^{-1}$ (chocolate pudding powder). Minimum and maximum values of aluminium content of cocoa products, which are related to DM basis, emphasize effects of provenance, as well as the quantity of cocoa added during production.

Spices and table salt

Spices are a further group of foods of plant origin. They are of no importance to food intake in man, but, generally contain high amounts of both minerals and trace elements. This could also be confirmed in the case of aluminium (Table 7). For practical reasons, table salt was included in this table.

As expected, a large range of aluminium concentrations was observed, from $6.5 \,\mu\text{g}\,\text{Al}\,\text{g}^{-1}$ (mustard, 1991) to $695\,\mu\text{g}\,\text{Al}\,\text{g}^{-1}$ (marjoram). In particular, in this group of foodstuffs, many time-dependent differences occurred with the aluminium content of items in 1988 being, in general, higher than 1991. Harvesting sites of spices may explain this finding. The fact

Table 5. Aluminium contents of sugar and sugar-rich products

	-	_		-		
Food	od μ g Al per g FM		μg Al per g DM			
		\overline{x}	s	Range		
Candies (1991)	3.4	3.6	0.4	3.0-4.1		
Jam (1991)	4.3	8.3	4.7	4.5-16		
Sugar	4.4	4.4	1.8	1.1 - 8.8		
Bee honey ^a	5.6	7.9	2.1	5.8 - 12		
Synthetic honey ^b	8.0	11	6.6	4.4-18		
Candies (1988)	9.5	9.8	5.6	4.3 - 23		
Jam (1988)	12	21	8.8	5.5-36		

^aOnly available in 1991.

Table 6. Aluminium contents of cocoa and cocoa products

Food	μg Al per g FM	μg Al per g DM			
		\overline{x}	s	Range	
Milk chocolate	9.4	9.9	3.5	4.5–18	
Chocolate spread	12	13	4.6	7.6-21	
for bread ^a					
Chocolate cream	16	17	11	3.9-45	
Chocolate pudding	27	32	15	16-73	
Powder cocoa	33	37	22	16-75	
(1991)					
Cocoa (1988)	103	111	48	21-161	

^aOnly available in 1991.

that table salt from 1991 contained up to 10-times the aluminium concentration of 1988 is also noteworthy.

Milk and dairy products

Aluminium concentrations of milk and dairy products reflected the influence of aluminium-containing food additives (Table 8).

Whereas nearly all dairy products contained $< 5 \,\mu g \,Al$ per g FM, soft cheese bought in 1991 and 1988, respectively, showed aluminium concentrations of 8.3 and $16 \,\mu g$ per g FM. The use of aluminium-containing emulsifiers (sodium aluminium phosphate) could explain this large concentration difference. Nevertheless,

Table 7. Aluminium contents of spices and table salt

Food	μ g Al per g FM	μ g	μ g Al per g DM		
		\overline{x}	s	Range	
'Sanisal'a	0.5	0.5	0.2	0.3-0.7	
Table salt (1988)	1.4	1.4	0.8	0.2 - 2.8	
Table salt (1991)	15	15	4.3	11-22	
Mustard (1991)	6.5	27	8.0	15-36	
Mustard seed (1991)	6.8	7.4	1.6	5.7-9.6	
Mustard (1988)	12	46	12	32-59	
Caraway	23	25	6.5	16-35	
Mustard seed (1988)	35	37	14	13-58	
Pepper (1991)	44	51	6.1	43-58	
Cinnamon	98	111	48	42-238	
Paprika, hot (1988)	122	140	74	57-285	
Paprika, sweet	153	174	74	62-349	
Paprika, hot (1991)	254	281	65	178-326	
Pepper (1988)	288	317	67	266-414	
Marjoram	695	797	176	531-1186	

^aOnly available in 1988.

Table 8. Aluminium contents of milk and dairy products

Food	μ g Al per g FM		μ g Al per g DM		
		\overline{x}	s	Range	
Yoghurt ^a	1.2	6.6	2.9	3.9-12	
Condensed milk	1.6	8.2	4.5	2.2 - 18	
Hen's egg	2.1	8.4	1.6	6.5 - 12	
"Altenburger goats cheese"	2.4	5.0	2.1	2.3-9.2	
Margarine	2.7	3.5	1.1	2.2-5.3	
Butter	2.8	4.2	2.6	1.2-11	
Curds	2.9	16	8.0	5.5-39	
Tollenser ^b	3.0	5.4	2.5	3.6 - 9.1	
Emmental cheese ^a	3.4	5.4	2.3	1.9-8.8	
Milk ^c	3.8	3.8	1.7	2.5-6.1	
Edam cheese ^b	4.3	7.9	3.3	4.3 - 13	
Limburg cheese ^b	5.0	11	6.0	4.9-25	
Camembert	5.0	11	8.6	3.2 - 40	
Gouda cheese	6.0	10	5.6	5.9-25	
Tilsit cheese ^a	6.4	11	5.8	3.3 - 21	
Soft cheese (1991)	8.3	18	10	9.8-38	
Soft cheese (1988)	16	39	20	11-70	

[&]quot;Only available in 1991

^bOnly available in 1988.

^bOnly available in 1988.

cRelated to ml milk.

the aluminium content of soft cheese between 1988 and 1991 also varied highly significantly (P < 0.001). The other kinds of cheese contained aluminium concentrations of 2.4 μ g per g FM ("Altenburger goat's cheese") to 6.4 μ g per g FM (Tilsit cheese). On average, the aluminium content of milk amounted to 3.8 μ g ml⁻¹. Hen's egg and margarine practically included in this table showed aluminium concentrations of 2.1 and 2.7 μ g per g FM, respectively.

Meat, sausage and offal

Average aluminium concentration of meat and sausage was $< 10 \,\mu g$ per g FM (Table 9). Highest concentrations within this group were found in both offal, liver and kidney containing 8.3 and $10 \,\mu g$ Al per g FM.

Beef and pork, which are consumed in large quantities in Germany, showed aluminium contents of about $4 \mu g g^{-1}$ FM. Aluminium concentrations of the different kinds of sausage investigated covered a relatively small range of 4.2 (liver sausage) to $7.4 \mu g$ per g FM (black pudding). Related to fresh matter, the aluminium content of meat is lower than that of sausage. However, related to dry matter basis, this tendency disappeared.

Table 9. Aluminium contents of meat, sausage and offal

Food	food μ g Al per g FM		μ g Al per g DM		
		\overline{x}	s	Range	
Mutton	2.5	7.6	2.8	4.2-14	
Chicken	3.5	12	6	4.1 - 21	
Beef	3.6	13	7.3	3.7-29	
Pork	3.8	14	5.6	5.0-26	
Liver sausage	4.2	8.5	4.0	3.1 - 18	
Thick Frankfurter	4.8	11	4.8	4.3-22	
Mortadella ^a	5.1	12	4.0	6.2-19	
Liver loaf ^a	5.4	12	6.6	6.0-25	
Salami	6.7	11	5.6	3.7-25	
Blackpudding	7.4	14	6.0	7.4-32	
Liver	8.3	28	15	9.4-59	
Kidneys	10	43	25	9.3-82	

^aOnly available in 1991

Table 10. Aluminium contents of fish and tinned fish

Food	μg Al per g FM	μ g A	Al pe	r g DM
		\overline{x}	s	Range
Rosefish fillet ^a	1.2	4.9	2.1	2.9-7.8
Bismarck herring ^a	1.7	5.9	2.3	2.6-8.7
Herring fillet (1991)	2.5	7.1	5.1	2.5-16
Herring in tomatoes ^a	2.5	8.0	4.0	6.0-17
Fried herring ^a	2.5	8.3	4.4	4.2 - 16
Salted herring ^b	3.1	8.1	4.0	5.7-14
Trout, fresh ^b	3.5	12	5.5	5.2 - 20
Trout, smoked ^b	4.0	14	8.1	6.5-31
Mackerel fillet	4.2	11	4.1	6.4-19
Herring fillet (1988)	4.9	18	6.4	7.7 - 28
Sardines ^a	5.5	13	4.2	5.2-17

^aOnly available in 1991.

Thus, addition of spices during sausage production had no effect on the aluminium content of sausages when compared with its raw material meat.

Fish and tinned fish

Fish and tinned fish had aluminium concentrations comparable with those found in meat (Table 10). The range was from 1.2 (rosefish fillet) to $5.5 \mu g$ per g FM (sardines).

The different items made out of herring and representing approx. half the investigated products within this food group contained $1.7 \,\mu\text{g}\,\text{Al}$ per g FM in the case of Bismarck herring up to $4.9 \,\mu\text{g}\,\text{Al}$ per g FM in the case of herring fillet (1988). Smoking of trout had only insignificant effects on its aluminium concentration.

Beverages, tea and coffee

Refreshing drinks and beer contained $< 0.7 \text{ mg Al litre}^{-1}$ (Table 11).

In contrast, aluminium content of juice was considerably higher and varied considerably, covering a range of 0.5–8.2 mg litre⁻¹. Fruit raw material and juice processing are of decisive influence. The alcoholic beverages vermouth, white, red and sparkling wine had aluminium concentrations of 2.0–3.0 mg litre⁻¹. In contrast, the spirits, schnapps and brandy, had relatively low aluminium concentrations (0.5 mg litre⁻¹).

Tea, especially black tea, can contain up to $20,000 \,\mu\text{g}$ Al per g DM (Matsumoto et al., 1976). In our investigations, black tea leaves of different provenance (Ceylon, India, Kenya) had aluminium concentrations of $899 \pm 292 \,\mu\text{g}$ per g DM. On the other hand, ground coffee (powder) contained only $19 \pm 10 \,\mu\text{g}$ Al per g DM. Black tea and coffee infusions (normally prepared) showed aluminium concentrations of 4.2 and 0.1 mg Al litre⁻¹. In the case of black tea, a transfer rate of aluminium into infusion of about 30% was calculated (Müller et al., 1997).

Table 11. Aluminium contents of beverages

Beverage		mg Al p	er litre
	\overline{x}	s	Range
Lemonade	0.5	0.1	0.2-0.5
Schnapps	0.5	0.2	0.3-0.8
Brandy	0.5	0.3	0.2 - 1.1
Beer	0.6	0.2	0.3-0.9
Coke	0.7	0.3	0.3-1.3
Advocaat	1.3	0.4	0.7-1.9
Vermouth	2.1	1.0	0.5-4.1
Juice ^a	2.4	2.2	0.5-8.2
Sparkling wine	2.5	1.2	1.0-5.1
Red wine	2.6	0.8	1.5-38
White wine	2.6	1.5	0.5-4.6

^aOnly available in 1991.

bOnly available in 1988.

DISCUSSION

Within the framework of two market basket studies, the aluminium content of a comprehensive food assortment typical of German nutritional habits was determined. Carried out in 1988 and 1991, a total of 128 items out of 12 groups of foodstuffs were included in our investigation.

Most investigated foodstuffs contained $< 5 \mu g$ Al per g FM. Individual bread, cakes and pastries, some vegetables, sausage, offal, sugar-rich food and a majority of farinaceous products and flours had aluminium concentrations within the range of 5 to $10 \mu g$ per g FM. Aluminium concentrations of several farinaceous products, jam (1988), table salt (1991), lentils, soft cheese, biscuits, mixed mushrooms, herbs, lettuce, cocoa and cocoa products, spices and tea leaves exceeded $10 \mu g$ per g FM.

Summarizing our findings within the individual groups of foodstuffs, beverages and fruit had the lowest aluminium contents (Table 12). Food of animal origin contained between 3.2 and 5.4 μ g Al per g FM, with offals and soft cheese showing the highest aluminium concentrations within this group. On average, the aluminium content of table salt amounted to 5.6 μ g g⁻¹.

Aluminium contents of other foods of plant origin including vegetables, bread, cake and pastries, sugar and sugar-rich food, pulses, farinaceous products and flours were within a range of 5.7 to 9.5 μ g per g FM. On average, vegetables, as well as bread, cake and pastries consumed in relatively large quantities contained up to 2 μ g Al per g FM more than products of animal origin. This difference was not so large as is often, a priori, assumed in the literature. Herbs had approx. 19 μ g Al per g FM. By far the highest aluminium concentrations were found within the groups cocoa/cocoa products (33 μ g g⁻¹), spices (145 μ g g⁻¹) and tea leaves (899 μ g g⁻¹).

In general, aluminium content of frequently consumed food increased in the following order: beverages, food of animal origin, food of plant origin. Comparison of our results with literature values is difficult since cited data show large variations (Bjorksten *et al.*, 1988; Sherlock, 1989; Reilly, 1991).

Table 12. Aluminium contents of different groups of foodstuffs $(\mu g Al \text{ per g FM or } \mu g Al/ml)$ —summary survey

Group of food	Mean	Range
Beverages	1.5	0.4-2.6
Fruit	2.7	0.7 - 7.9
Fish, tinned fish	3.2	1.2 - 5.5
Milk, dairy products	4.5	1.2 - 16
Meat, sausage, offal	5.4	2.5-10
Table salt	5.6	0.5 - 15
Vegetables	5.7	0.7 - 33
Sugar, sugar-rich products	6.7	3.4-12
Bread, cake and pastries	7.4	3.4-22
Pulses	9.3	3.2 - 16
Farinaceous products and flours	9.5	3.8 - 34
Herbs	19	8.2 - 26
Cocoa, cocoa products	33	9.4-103
Spices	145	6.5-695

Results of recent investigations emphasize that the average aluminium content of most food covers the range $0.1-1.0 \,\mu g$ per g FM with only few foods showing aluminium amounts > $10 \,\mu g$ per g FM (Greger, 1985, 1992; Pennington and Jones, 1989; Sherlock, 1989; Wen et al., 1993; Xu et al., 1993).

Corresponding or lower trending aluminium concentrations were found in investigations in Germany (Treier and Kluthe, 1988), Taiwan (Hsu and Wu, 1990), Hungary (Schamschula et al., 1988), China (Wang et al., 1994) and Finland (Nuurtamo et al., 1980a,b; Varo et al., 1980a,b,c,d). Compared with monitorings carried out in the USA, our results in bread, cake and pastries, several vegetables (including potatoes), beverages, chocolate and spices, corresponded with US values, whereas food of animal origin investigated in the USA had considerably lower aluminium concentrations (Greger, 1985, 1992; Greger et al., 1985; Sullivan et al., 1987; Pennington and Jones, 1989). Recent investigations in the UK indicate comparable aluminium concentrations in bread, cake and pastries, sausage, fish, several vegetables, refreshing drinks and dairy products. In contrast, meat, offal, potatoes, milk, as well as lettuce had much lower aluminium concentrations (MAFF, 1993; Ward and Savage, 1994).

Different kinds of bread investigated in China had comparable aluminium concentrations within the range $1.0-10 \,\mu g$ per g FM (Yang et al., 1994). On the other hand, Egyptian bread had between 18 and 200 µg Al per g FM (Iskander et al., 1990). Fish (including tinned fish) had aluminium concentrations of about 1.0 to 18µg per g FM (Yang et al., 1994; Tahan et al., 1995) which corresponded to our measurements. Depending on provenance, cocoa contained between 1.0 and 239 μ g Al g⁻¹ (Knezevic and Berghammer, 1994). The same authors measured approx. 8 µg Al per g FM milk chocolate and demonstrated an increase of aluminium content in different chocolate products with increasing cocoa proportions. There are some studies on the aluminium content of beverages, showing good agreement with our results. In individual items, the following aluminium concentrations have been reported: refreshing drinks: $0.01-0.25 \text{ mg litre}^{-1}$ (Müller *et al.*, 1993), 0.1- $2.1 \,\mathrm{mg \, litre^{-1}}$ (Schenk *et al.*, 1989); juice: 0.04– 4.1 mg litre⁻¹ (Schenk et al., 1989), about 0.5 mg litre⁻¹ (Nagy and Jobst, 1994); different kinds of wine and sparkling wine: 0.4–2.8 mg litre⁻¹ (Schenk et al., 1989); spirits: $0.15-0.6 \,\mathrm{mg\,litre^{-1}}$ (Schenk et al., 1989). On average, beer contained 0.1-0.4 mg Al litre⁻¹; however, this varied in a concentration range of 0.02 up to $3.0\,\mathrm{mg}\,\mathrm{Al\,litre^{-1}}$ (Schenk et al., 1989; Johansson, 1994; Sharpe and Williams, 1995).

Very large, almost unlikely differences were observed when comparing our results with recent investigations in Sweden. Jorhem and Haegglund (1992) determined concentrations of 0.02 up to 0.09 μ g Al per g FM in a multitude of foodstuffs including vegetables, fruit, meat and sausage, fish, milk, beverages, bread, cake and pastries.

Thus, a majority of foods had aluminium levels which were, partly, up to 100-fold lower than the results observed within our market basket. These differences cannot be explained.

Nevertheless, investigations of Bjorksten et al. (1988) emphasized that aluminium determination in food is beset with difficulties, as well as inadequacies. Within the framework of a comparative study, aluminium contents of selected foods were analyzed in several laboratories under relatively standard conditions. Even in this case, unimaginable ranges of aluminium concentration occurred. On average, maximum values were about 400-times higher than minimum with individual cases showing a deviation in the order of a factor of 1000. The following selected examples are illustrative (as μ g Al per g FM): potatoes: minimum 0.3/mean 4.4/maximum 50; beef: minimum 0.1/mean 5.6/maximum 20; cheese: minimum 0.1/mean 5.1/maximum 20 (Bjorksten et al., 1988).

Results obtained in black tea and black tea infusions were also comparable with the literature (Fairweather-Tait et al., 1987; Schenk et al., 1989; Baxter et al., 1990; Owuor et al., 1990; Müller et al., 1993; Wen et al., 1993; Wang et al., 1994). This special part of our study has been described in detail (Müller et al., 1997).

Finally, one 'food' not included in our investigation should be mentioned. Very high aluminium concentrations have repeatedly been reported in chewing gum. Kupchella and Syty (1980), as well as Varo et al. (1980d) measured (in a 5-g stick of chewing gum) aluminium amounts within the range of 3.0–4.5 mg! Nevertheless, further studies proved that this high concentration is only 'available' to a low degree. Chewing mobilized 0.01–0.46 mg per stick (Lione and Smith, 1982). Thus, moderate 'consumption' of chewing gum has only a slight effect on aluminium intake (Lione and Smith, 1982).

Overall, aluminium content of this comprehensive food assortment was low and comparable with the literature data in the majority of cases. High aluminium concentrations were found in foodstuffs which are consumed in low quantities and/or relatively seldom. With this level of aluminium there is no danger of aluminium exposure.

However, patients with chronic kidney insufficiency represent a real risk group for aluminium exposure. Systemic toxicity could occur due to continuous aluminium accumulation resulting from an increasingly impaired ability to excrete absorbed aluminium via the renal pathway. These number about 5% of the population. For the interest of concerned persons, the aluminium contents of selected foods (for instance beverages, bread, vegetables, soft cheese and others) will be checked at regular intervals (4–5 years) within the framework of food surveillance.

Monitoring would also seem necessary in connection with the aetiological function of aluminium in neurodegenerative diseases. So long as such effects of aluminium cannot unequivocally be excluded, aluminium content of food, bioavailability and intake deserve further consideration.

REFERENCES

- Baxter, M. J., Burrell, J. A. and Massey, R. C. (1990). The aluminium content of infant formula and tea. *Food Additives and Contaminants*, 7, 101-107.
- Bjorksten, J., Yaeger, L. L. and Wallace, T. (1988). Control of aluminium ingestion and its relation to longevity. *Interna*tional Journal of Vitamin and Nutrition Research, 58, 462-465.
- Bünnig, K. (1984). Bewertung des Risikos einer Aluminiumvergiftung. GIT-Fachzeitschrift für Labormedizin, 7, 24-25.
- Candy, J. M., Oakley, A. E., Watt, F., Grime, G. W., Klinowski, J., Perry, R. H. and Edwardson, J. A. (1986). A role for aluminium, silicon and iron in the genesis of senile plaques. *Modern Trends in Aging Research*, 147, 443-450.
- Crapper McLachlan, D. R. (1986). Aluminum and Alzheimer's disease. *Neurobiology of Aging*, 7, 525-532.
- Domingo, J. L. (1993). Aluminium. In Encyclopaedia of Food Science, Food Technology and Nutrition, Vol. 1, eds R. Macrae, R. K. Robinson and M. J. Sadler. Academic Press, New York, pp. 126-135.
- Fairweather-Tait, S., Faulks, R. M., Fatemi, S. J. A. and Moore, G. R. (1987). Aluminum in the diet. Human Nutrition Food Science and Nutrition, 41F, 183-192.
- Flaten, T. P. (1990). Geographical associations between aluminium in drinking water and death rates with dementia (including Alzheimer's disease), Parkinson's disease and amyotrophic lateral sclerosis in Norway. *Environmental Geochemistry and Health*, 12, 152–167.
- Greger, J. L. (1985). Aluminum content of the American diet. Food Technology, 9, 73-80.
- Greger, J. L. (1988). Aluminum in the diet and mineral metabolism. In *Aluminum and its Role in Biology*, eds H. Sigel and A. Sigel. Marcel Dekker, New York, pp. 199–216.
- Greger, J. L. (1992). Dietary and other sources of aluminium intake. Ciba Foundation Symposium, 169, 26-49.
- Greger, J. L., Goetz, W. and Sullivan, D. (1985). Aluminum levels in foods cooked and stored in aluminum pans, trays and foil. *Journal of Food Protection*, 48, 772-777.
- Haenel, H. (1979). Energie- und Nährstoffgehalt von Lebensmitteln, Lebensmitteltabellen. Volk und Gesundheit, Berlin.
- Hsu, G. S. W. and Wu, J. Y. (1990). Aluminum content of foods in taiwan: Report I. Fu Jen Studies, 24, 69-76.
- Inoue, T., Ishiwata, H. and Yoshihira, K. (1988). Aluminum levels in food-simulating solvents and various foods cooked in aluminum pans. *Journal of Agricultural and Food Chemistry*, 36, 599-601.
- Iskander, F. Y., Davis, K. R., Ashour, H. and Hassan, H. F. (1990). Aluminum content of Egyptian breads. *Food Chemistry*, **35**, 197–208.
- Johansson, C. G. (1994). Determination of aluminium in beer by GF-AAS. *Journal of the Institute of Brewing*, 100, 255.
- Jones, K. C. and Bennett, B. G. (1986). Exposure of man to environmental aluminium an exposure commitment assessment. Science of the Total Environment, 52, 65-82.
- Jorhem, L. and Haegglund, G. (1992). Aluminium in foodstuffs and diets in Sweden. Zeitschrift für Lebensmittel-Untersuchung und -Forschung, 194, 38-42.
- Knezevic, G. and Berghammer, A. (1994). Über den Gehalt von Aluminium in Rohkakao und Kakaofertigprodukten. Zucker- und Süβwarenwirtschaft, 47, 73.
- Koivistoinen, P. (1980). Mineral element composition of Finnish foods: N, K, Ca, Mg, P, S, Fe, Cu, Mn, Zn, Mo, Co, Ni, Cr, F, Se, Si, Rb, Al, B, Br, Hg, As, Cd, Pb and ash. Acta Agriculturica Scandinavica, suppl. 22.
- Kruck, T. P. A. and McLachlan, D. R. (1988). Mechanisms of aluminum neurotoxicity-relevance to human disease. In Aluminum and its Role in Biology, eds H. Sigel and A. Sigel. Marcel Dekker, New York, pp. 285-314.

- Kupchella, L. and Syty, A. (1980). Determination of nickel, manganese, copper, and aluminium in chewing gum by nonflame atomic absorption spectrometry. *Journal of Agri*cultural and Food Chemistry, 28, 1035-1037.
- Levick, S. E. (1980). Dementia from aluminum pots. New England Journal of Medicine, 303, 164.
- Lione, A. (1983). The prophylactic reduction of aluminium intake. Food and Chemical Toxicology, 21, 103-109.
- Lione, A. and Smith, J. C. (1982). The mobilization of aluminium from three brands of chewing gum. Food Chemistry and Toxicology, 20, 945.
- Liukkonen-Lilja, H. and Piepponen, S. (1992). Leaching of aluminium from aluminium dishes and packages. Food Additives and Contaminants, 9, 213-223.
- MAFF (Ministry of Agriculture, Fisheries and Food) (1993). Aluminium in food. Food Surveillance Paper No. 39, HMSO, London.
- Martyn, C. N., Osmond, C., Edwardson, J. A., Barker, D. J. P., Harris, E. C. and Lacey, R. F. (1989). Geographical relation between Alzheimer's disease and aluminium in drinking water. *Lancet*, **59**, 62.
- Matsumoto, H., Hirasawa, E., Morimura, S. and Takahashi, E. (1976). Localization of aluminium in tea leaves. *Plant Cell and Physiology*, 17, 627-631.
- McLachlan, D. R. C. and de Boni, U. (1980). Aluminum in human brain disease—an overview. *Neurotoxicology*, 1, 3-16.
- McLachlan, D. R. C., McLachlan, C. D., Krishnan, B., Krishnan, S. S., Dalton, A. J. and Steele, J. C. (1989). Aluminum and calcium in soil and food from Guam (Marianas Islands, Pacific Ocean), Palau (Western Pacific Ocean) and Jamaica (West Indies): implications for amyotrophic lateral sclerosis and parkinsonism-dementia syndromes of Guam. *Environmental Geochemistry and Health*, 11, 45-53.
- Müller, J. P., Steinegger, A. and Schlatter, C. (1993). Contribution of aluminium from packaging materials and cooking utensils to the daily aluminium intake. Zeitschrift für Lebensmittel-Untersuchung und -Forschung, 197, 332–341.
- Müller, M., Anke, M., Illing, H. and Hartmann, E. (1995). Aluminiumbestimmung in Lebensmitteln mittels Graphitrohratomabsorptionsspektrometrie. *GIT-Fachzeitschrift für das Laboratorium*, 39, 795–796.
- Müller, M., Anke, M. and Illing-Günther, H. (1997). Availability of aluminium from tea and coffee. Zeitschrift für Lebensmittel-Untersuchung und -Forschung (in press).
- Nagy, E. and Jobst, K. (1994). Aluminium dissolved from kitchen utensils. Bulletin of Environmental Contamination and Toxicology, 52, 396-399.
 Nuurtamo, M., Varo, P., Saari, E. and Koivistoinen, P.
- Nuurtamo, M., Varo, P., Saari, E. and Koivistoinen, P. (1980a). Mineral element composition of Finnish foods. V. Meat and meat products. *Acta Agriculturica Scandinavica*, suppl. 22, 57-76.
- Nuurtamo, M., Varo, P., Saari, E. and Koivistoinen, P. (1980b). Mineral element composition of Finnish foods. VI. Fish and fish products. *Acta Agriculturica Scandinavica*, suppl. 22, 77-88.
- Owuor, P. O., Gone, F. O., Onchiri, D. B. and Jumba, I. O. (1990). Levels of aluminium in green leaf of clonal teas, black tea and black tea liquors, and effects of rates of nitrogen fertilizers on the aluminium black tea contents. Food Chemistry, 35, 59-68.
- Pennington, J. A. T. (1987). Aluminum content of foods and diets. Food Additives and Contaminants, 5, 161-232.
- Pennington, J. A. T. and Jones, J. W. (1989). Dietary intake of aluminum. In *Aluminum and Health*, ed. H. J. Gitelman. Marcel Dekker, New York-Basel, pp. 67-100.
- Perl, D. P. (1988). Aluminum and Alzheimer's disease: methodological approaches. In *Aluminum and its Role in Biology*, ed H. Sigel and A. Sigel. Marcel Dekker, New York-Basel, pp. 259–284.

- Perl, D. P. and Brody, A. R. (1980). Alzheimer's disease: X-ray spectrometric evidence of aluminium accumulation in neurofibrillary tangle-bearing neurones. *Science*, 208, 297–299.
- Reilly, C. (1991). Metal Contamination of Food. Elsevier, London. Rickenbacher, U. (1984). Toxikologie von Aluminiumverbindungen. Mitteilungen aus dem Gebiete der Lebensmitteluntersuchung und Hygiene, 75, 69-76.
- Schamschula, R. G., Sugar, E., Un, P. S. H., Duppenthaler, J. L., Toth, K. and Barmes, D. E. (1988). Aluminum, calcium and magnesium content of Hungarian foods and dietary intakes by children aged 3.9 and 14 years. *Acta Physiologica Hungarica*, 72, 237-252.
- Schenk, R. U., Bjorksten, J. and Yeager, L. (1989). Composition and consequences of aluminum in water, beverages and other ingestibles. In *Environmental Chemistry and Toxicology of Aluminum*, ed. T. E. Lewis. Lewis, Chelsea, Michigan, pp. 247-271.
- Sharpe, F. R. and Williams, D. R. (1995). Content, chemical speciation, and significance of aluminum in beer. *Journal of* the American Society of Brewing Chemistry, 53, 85–92.
- Sherlock, J. C. (1989). Aluminium in foods and the diet. In *Aluminium in Food and the Environment*, eds R. C. Massey and D. Taylor. Royal Society of Chemistry, Cambridge, pp. 68–76.
- Sullivan, D. M., Kehoe, D. F. and Smith, R. L. (1987). Measurement of trace levels of total aluminum in foods by atomic absorption spectrophotometry. *Journal of the Association of Official Analytical Chemists International*, **70**, 118–120.
- Tahan, J. E., Sanchez, J. M., Granadillo, V. A., Cubillan, H. S. and Romero, R. A. (1995). Concentration of total Al, Cr, Cu, Fe, Hg, Na, Pb, and Zn in commercial canned seafood determined by atomic spectrometric means after mineralization by microwave heating. *Journal of Agricultural and Food Chemistry*, 43, 910–915.
- Trapp, G. A. and Cannon, J. B. (1981). Aluminum pots as a source of dietary aluminum. *New England Journal of Medicine*, **304**, 172–173.
- Treier, S. and Kluthe, R. (1988). Aluminiumgehalte in Lebensmitteln. *Ernährungs-Umschau*, 35, 307-312.
- Varo, P., Nuurtamo, M., Saari, E. and Koivistoinen, P. (1980a). Mineral element composition of Finnish foods. IV. Flours and bakery products. Acta Agriculturica Scandinavica, suppl. 22, 37-56.
- Varo, P., Lähelmä, O., Nuurtamo, M., Saari, E. and Koivistoinen, P. (1980b). Mineral element composition of Finnish foods. VII. Potato, vegetables, fruits, berries, nuts, and mushrooms. Acta Agriculturica Scandinavica, suppl. 22, 89–113.
- Varo, P., Nuurtamo, M., Saari, E. and Koivistoinen, P. (1980c). Mineral element composition of Finnish foods. VIII. Dairy products, eggs and margarine. Acta Agriculturica Scandinavica, suppl. 22, 115-126.
- Varo, P., Nuurtamo, M., Saari, E. and Koivistoinen, P. (1980d). Mineral element composition of Finnish foods. IX.
 Beverages, confectioneries, sugar and condiments. Acta Agriculturica Scandinavica, suppl. 22, 127-140.
- Wang, L., Su, D. Z. and Wang, Y. F. (1994). Studies on the aluminium content in Chinese foods and the maximum permitted levels of aluminum in wheat flour products. Biomedical and Environmental Sciences, 7, 91–99.
- Ward, N. I. and Savage, J. M. (1994). Metal dispersion and transportational activities using food crops as biomonitors. *Science of the Total Environment*, 146/147, 309-319.
- Wen, W., Chen, B. Q. and Li, D. (1993). Research on aluminium content of the diet in haerbin and daily aluminium intake of the local people. *Chinese Journal of Preventative Medicine*, 27, 32-35.

- Xu, G. S., Jin, R. P., Zhang, Z. W., Zhang, W. Q., Ren, D. L., Chen, J. and Huang, G. W. (1993). Preliminary study on aluminum content of foods and aluminum intake of residents in Tianjin. *Biomedical and Environmental Science*, 6, 319–325.
- Yang, Q., Penninckx, W. and Smeyers-Verbeke, J. (1994). Closed-vessel microwave acid digestion of foodstuffs and trace aluminum determination by graphite furnace atomic absorption spectrometry. *Journal of Agricultural and Food Chemistry*, **42**, 1948–1953.