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Determination of aluminium in Slovenian foodstuffs and its leachability from aluminium-cookware

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Abstract

Al toxicity is well known, particularly in patients with chronic renal failure. To estimate the dietary intake of Al in man it is necessary to determine the total Al concentrations in different foodstuffs. For this purpose twenty one different vegetables growing on peat and clay soils were analysed. Water-soluble Al was determined in soil samples to estimate the easily soluble Al fraction available to plants and to find whether plants may also accumulate higher Al concentrations from soils low in Al. Al was further determined in wheat, eggs, baby foods, fish, mussels and twenty samples of total diet. In addition, beverages and herbal and black tea infusions were analysed for Al content. Total Al in foodstuffs was determined by ICP-AES after microwave digestion. The accuracy of the procedure was checked by the analysis of CRM 1570a (Spinach leaves). Good agreement between measured and certified values was obtained. In order to estimate the extent of leaching from Al utensils, sauerkraut and sour turnip were cooked in Al-cookware. The results indicate that concentrations of Al in foodstuffs (dry weight) were in general below 30 mg kg^{-1}, while in total diet they ranged from 3 to 6 mg kg^{-1} Al. High Al concentrations were found in mussels (about 300 mg kg^{-1}) parsley and lamb's lettuce. Lamb's lettuce contained about 400 mg kg^{-1} of Al, irrespective of whether it was growing on peat soil low in total and water-soluble Al or in clay soil rich in Al. Black tea infusions contained up to 300 mg kg^{-1} of Al, which is 30 times more than herbal tea infusions. Concentrations of Al in beverages were below 0.4 mg dm^{-3}. The results indicated that sauerkraut and sour turnip leach appreciably high concentrations of Al during cooking in Al utensils (313 and 260 mg kg^{-1}, respectively), so the use of such cookware is not recommended for acidic foodstuffs.

1. Introduction

Aluminium (Al) comprises 8% of the Earth's crust and is bound predominantly in oxides and alumosilicates. In contrast to its abundance, it is very insoluble in the neutral pH range. However, its solubility may significantly increase under acidic ($pH < 6$) and alkaline ($pH > 8$) conditions and/or in the presence of inorganic and organic complexing ligands (Mulder & Stein, 1994). Al is not considered to be an essential element in humans, but its toxicity is known, particularly in patients with chronic renal failure (Alfrey, 1994; D'Haese & De Broe, 1994). Concern over a possible relation between Al exposure and Alzheimer's disease initiated investigations on the potential intake of Al into the human body, including foods and drinking water (Gauthier, Fortier, Courchesre, Pepin, Mortimer, & Gaoureau, 2000). Al concentrations in foodstuffs expressed on a dry weight basis range in meat from 0.5 to 30 mg kg^{-1} (Müller, Anke & Illing Günther, 1998; Oniawa, Ikadeh, & Nwese, 1997; Gramiccioni, Ingrao, Milana, Santaroni, & Tomassi, 1996) in flour, bread and rice from 2 to 22 mg kg^{-1} (Müller et al., 1998; Oniawa et al., 1997) and in milk from 2 to 6 mg kg^{-1} Al (Müller et al., 1998). In beverages and wine Al concentrations range in general between 0.2 and 8 mg dm^{-3} (Gramiccioni et al. 1996; Müller et al., 1998; Oniawa et al., 1997). Since Al-tolerant plants can accumulate Al from soil solution, higher concentrations of Al may be found in vegetables (up to 80 mg kg^{-1}) (Müller et al., 1998; Oniawa et al., 1997), in different kinds of lettuce (up to 1000 mg kg^{-1}) (Müller et al., 1998), in herbs and spices (up to 300 and 1000 mg kg^{-1} Al, respectively) (Müller et al., 1998) and in black tea leaves (from 600 to 1200 mg

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kg^{-1} of Al) (Müller et al., 1998). Al enters drinking waters mainly due to the use of $Al_2(SO_4)_3$ that is applied as a coagulant to clarify turbidity (Martin, 1986). Another possible source of the entry of Al into foodstuffs is the use of Al containing food additives, which in cheese processing may increase Al concentrations up to 70 mg kg^{-1} (Müller et al., 1998). Al may also be leached from Al-cookware in the presence of organic acids in foods. Gramiccioni et al. (1996), reported that tomato juice leached moderate Al concentrations from Al-cookware, which increased after cooking from 3 to 5.5 mg kg^{-1}. A potential pathway of entry of Al into foodstuffs is also the use of Al foils and Al foil containers. However, it was demonstrated that foodstuffs with neutral pH and low salt content like milk and diary products, edible fats and oil do not significantly leach Al from Al food packing (Severus, 1989). In order to know the contribution of particular food products to the daily intake of Al, numerous investigations have been carried out to determine the total Al in different foodstuffs (Nuurtamo, Varu, Saari, & Koivistoinen, 1980a, 1980b; Pennington & Jones, 1989; Sherlock, 1989; Varo, Nuurtamo, Saari, & Koivistoinen, 1980a, 1980b, 1980c; Varo, Lahelma, Nuurtamo, Saari, & Koivistonen, 1980; Wang et al. 1994; Ward & Savage, 1994). Studies of the average Al concentration in 24 h diets indicated that under normal circumstances the average dietary intake of Al is about 6 mg per day (Gramiccioni et al., 1996; Sherlock, 1989). Although the provisional weekly intake of Al established by the FAO/WHO Expert Committee on Food Additives is quite high (60 mg per day for an adult man) (World Health Organization, 1989), it is important to check the Al concentration in various foods, since it may sometimes depend on the region of food production.

The aim of our work was therefore to determine Al concentrations in various Slovenian foodstuffs. The leachability of Al from Al-cookware into foods was also investigated and the accumulation of Al in lamb's lettuce grown in soil with high and low Al concentrations was investigated.

2. Materials and methods

2.1. Instrumentation and reagents

Al was determined on a Perkin-Elmer (Norwalk, CT, USA) Plasma 40 emission spectrometer, adjusted to a wavelength of 396.152 nm. The plasma was operated at a power of 1.0 kW and a frequency of 40 MHz, with argon as the coolant gas at a flow rate of 15 dm^3 min^{-1}, and a nebulisation flow rate of 1 dm^3 min^{-1}. Microwave digestion of samples was performed on a CEM Corporation (Matthews, NC, USA) CEM MARS 5 Microwave Acceleration Reaction System. An Iskra (Kranj, Slovenia) UZ 2R ultrasonic bath was used for the degassing of beverages. A WTW (Weilheim, Germany) pH 330 pH meter was employed to determine pH of samples.

Suprapur acids (Merck, Darmstadt, Germany) and water doubly distilled in quartz were used for the preparation of samples and standard solutions. An Al standard stock solution (1000±2 mg dm^{-3}, Al in 5% HCl) was purchased from Merck (Darmstadt, Germany). All other chemicals were of analytical-reagent grade and were also purchased from Merck (Darmstadt, Germany). Sartorius (Göttingen, Germany) 0.2 μm cellulose nitrate membrane filters of 25 mm diameter were used in the filtration procedure. The quality of data was checked by the analysis of standard reference material CRM 1570a Trace and minor elements in spinach leaves (NIST, Gaithersburg MD, USA).

2.2. Sample preparation and determination of Al in foodstuffs

Cabbage, wheat, potato, eggs, instant milk, mussels, marine fish (gilthead bream, golden grey mullet, anchovy) and river fish (trout) were selected for analysis. Baby milk formulae and baby foods were analysed as well. Cabbage, wheat and potato were obtained from Slovenian agricultural farms. Fish was bought in fish market while other food items were bought in grocer's shop. Twenty-one vegetables grown on peat and clay soils were also investigated. Crops were obtained from the local gardeners. Samples were first lyophilised and homogenised (particles less than 0.2 mm). Approximately 0.35 g of lyophilised sample was weighed into a Teflon beaker. Afterwards 4 cm^3 of HNO_3 (supra pure) and 0.1 cm^3 of HF (supra pure) were added and the samples subjected to closed vessel microwave digestion at a maximal power of 1200 W: ramp to temperature 20 min, 180 °C, pressure 10 bar, hold 20 min, cooling 20 min. HF was added to vegetable samples in order to dissolve silica. The clear solutions obtained were quantitatively transferred to 25 cm^3 polyethylene graduated tubes and filled to the mark with water doubly distilled in quartz. The same procedure (acids without food samples) was applied for the blank analysis.

Al was determined by ICP-AES using yttrium (Y) (10.0 μg cm^{-3}) as internal standard. The analyses were performed in two parallels. Al in each parallel sample was determined at least five times. Sample aliquots were taken for the determination of the moisture content. Approximately 1 g of sample was kept at 80 °C until constant weight and the moisture content determined on the basis of the mass loss. If not stated otherwise, the concentrations of Al in foodstuffs were calculated on a dry weight basis.

In addition the aim of our work was to determine Al content in total diets of Slovenian army. Slovenian army nutrition is based on its own nutritional standards, which determine macro and micro nutrients in daily diet (Republic of Slovenia, MORS, 1994). Total diet samples (including beverages and drinking water) were taken five times in four barracks in a time span of two months (May, June). Each menu was repeated twice in different barracks. The average energy value of daily menu was 3770 ± 537 kcal. Samples were collected in polyethylene bags and the contents weighed. The mass of the total diet samples ranged from 2900 to 4600 g, the average was 3956 ± 400g. Samples were collected in polyethylene dish, mixed and homogenised with a mixer with titanium knife and frozen at −24 °C. After that samples were lyophilised and milled in an agate Fritch mill, Pulversiette 7, (particles less than 0.2 mm) and stored in plastic bottles, which had been washed before use with dilute nitric acid (pro analysis) (1:1). Al was then determined under the recommended procedure described above.

2.3. Sample preparation and determination of Al in beverages

Beverages containing CO_2 were degassed before analysis in an ultrasonic bath for 30 min. Al in beverages was determined without sample pre-treatment by ICP-AES.

2.4. Sample preparation and determination of total and water-soluble Al in soils

Total Al in soil samples was determined by ICP-AES after HNO_3–$HClO_4$–HF digestion (Ščančar, Milačič, & Horvat, 2000). For the determination of total water-soluble Al 2.00 g of moist soil sample was shaken for 16 h with 20 cm^3 of water, centrifuged (10 000 rev min^{-1}, 20 min), decanted, filtered through a 0.2 μm membrane filter and Al determined by ICP-AES (Mitrović, Milačič, & Pihlar, 1996).

2.5. Sample preparation and determination of Al in tea infusions and in leachates from Al-cookware

20 cm^3 of a boiling water was poured over 2.00 g of black or herbal tea, left for 10 min, decanted, filtered (0.2 μm) and after equilibration to room temperature Al in the tea infusions analysed by ICP-AES. In order to study the leachability from Al-cookware espresso coffee was cooked in a standard stainless steel and in Al-containers (25 g of coffee, 250 cm^3 of water, boiling 2 min). After equilibration to room temperature Al was determined by ICP-AES. In another experiment to 0.5 kg of sauerkraut or sour turnip 0.5 dm^{-3} of water was added and contents were cooked for 2 h in Al-cookware. After equilibration to room temperature the liquid from the sauerkraut and sour turnip was filtered (0.2 μm) and Al determined by ICP-AES.

3. Results and discussion

3.1. Repeatability of measurement, linearity, limit of detection

The repeatability of measurement of Al was tested on six parallel samples of lamb's lettuce and six parallel samples of chicory. The average Al concentration for lamb's lettuce was 413 mg kg^{-1} with a relative standard deviation ±5%, while the average Al concentration in chicory was 130 mg kg^{-1} with a relative standard deviation ±4%, indicating good repeatability of measurement of Al. Linearity of measurement was obtained over a concentration range from 0.1 to 10 μg cm^{-3} Al. The LOD calculated on a 3s basis (a value of three standard deviations of the blank) was found to be 1.5 mg kg^{-1} for foodstuffs and 0.02 mg dm^{-3} of Al for beverages. The accuracy of the procedure was tested by the analysis of the standard reference material CRM 1570a 'Trace and minor elements in spinach leaves''. Good agreement between the value of Al determined (301 ± 5 mg kg^{-1}) and the reported certified value for Al (310 ± 11 mg kg^{-1}) was obtained.

3.2. Determination of Al in vegetables

Various vegetable samples grown on clay soil high in Al and on peat soil low in Al were analysed. The concentration of Al in vegetable was determined using the recommended analytical procedure. The results, calculated on a dry weight basis are presented in Table 1. These data indicate that the concentration of Al in 50% of the samples analysed is less than 25 mg kg^{-1}. In the other vegetable samples the concentration of Al is much higher, ranging from 70 to 400 mg kg^{-1}. The highest concentration of about 400 mg kg^{-1} of Al was found in lamb's lettuce and in garden lettuce. Similar observations for Al concentrations in various vegetables, ranging from 2 to 80 mg kg^{-1}, and from 18 to 80 mg kg^{-1} of Al, respectively, were reported in the literature (Müller et al., 1998; Oniawa et al., 1997). Müller et al., (1998) also found higher Al concentrations in various sorts of lettuce (200–1000 mg kg^{-1}Al).

3.3. Determination of total Al and water-soluble Al in soil samples

In order to find whether some sorts of lettuce may accumulate Al from soils containing different Al concentrations, clay and peat soil were analysed for total and water-soluble Al content. The pH of the aqueous soil extract was also determined. The results are presented in Table 2. It is evident that both soil extracts have a slightly acidic pH. Clay soil contains appreciably higher water-soluble and total Al concentrations than peat soil. Nevertheless, lamb's lettuce grown on peat and clay soils contains similar Al concentrations, about 400 mg kg^{-1} (Table 1). This indicates that the concentration of Al in lamb's lettuce is not influenced significantly by the total water-soluble Al content in soil. On the basis of these observations it can be concluded that lamb's lettuce may accumulate Al even from soils low in Al.

3.4. Determination of Al in foodstuffs

In addition to vegetables some other Slovenian foodstuffs were also analysed. The results are presented in Table 3. These data indicate that concentrations of Al are generally low. An exception was mussels containing 330 mg kg^{-1} of Al. In wheat the Al content was found to be about 6 mg kg^{-1}, in "Čokolešnik" baby food 4 mg kg^{-1}, in anchovy and trout 2 mg kg^{-1}, while in yolk and white of egg, instant milk, "Aptamil" baby milk formulae, "Milubrei" baby food, gilthead bream and golden grey mullet the Al concentration was less than 1.5 mg kg^{-1}. Low Al concentration in fish, ranging from 0.5 to 4 mg kg^{-1} Al was also reported by Oniawa et al. (1997).

3.5. Determination of Al in beverages

Twenty different beverages were also analysed for Al content. The results are presented in Table 4. It is evi-

Table 1
Analysis of Al in some Slovenian vegetables after microwave digestion of samples and determination of Al by ICP-AES (the results are calculated on a dry weight basis)

Sample	Soil type	Concentration of Al[a] (mg kg^{-1})
Cabbage	Clay	11.7±0.2
Cauliflower-head	Clay	17.9±0.3
Cauliflower-leaves	Clay	21.4±0.3
Onion	Clay	6.7±0.1
Chives	Clay	76±1
Horse radish	Clay	10.7±0.2
Potato	Clay	51.8±0.1
Paprika	Clay	11.5±0.2
Tomato	Clay	6.0±0.1
Red beet	Clay	14.9±0.2
Parsley leaves	Clay	282±5
Leek	Clay	22.1±0.4
Cabbage Chicory	Clay	11.3±0.2
Cabbage lettuce	Clay	27.0±0.4
Garden lettuce	Clay	379±5
Lamb's lettuce	Clay	332±4
Lamb's lettuce	Peat	413±6
Kale	Peat	22.8±0.3
Mangold	Peat	92±2
Rhubarb	Peat	117±2
Chicory	Peat	130±2

[a] Mean of two parallel determinations±standard deviation of measurements.

Table 2
Determination of total Al, water-soluble Al and pH of aqueous soil extracts of clay and peat soils by ICP-AES

Soil sample	Concentration of total Al[a] (mg kg^{-1})	Concentration of total water-soluble Al[a] (mg kg^{-1})	pH of soil extract[a]
Clay	41000±700	7.7±0.1	6.5±0.1
Peat	6800±100	0.62±0.01	6.2±0.1

[a] Mean of two parallel determinations±standard deviation of measurements.

Table 3
Analysis of Al in some Slovenian foodstuffs after microwave digestion of samples and determination of Al by ICP-AES (the results are calculated on a dry weight basis)

Sample	Concentration of Al[a] (mg kg^{-1})
Wheat	5.7±0.1
Yolk of egg	<1.5
White of egg	<1.5
Baby milk formulae "Aptamil"	<1.5
Baby food "Čokolešnik"	3.95±0.08
Instant milk	<1.5
Baby food "Milubrei"	<1.5
Mussels	330±5
Gilthead bream	<1.5
Golden grey mullet	<1.5
Anchovy	2.00±0.06
Trout	1.70±0.05

[a] mean of two parallel determinations±standard deviation of measurements.

Table 4
Analysis of Al in various beverages by ICP-AES

Sample	Concentration of Al (mg dm^{-3})[a]
Mineral water "Zala"	<0.02
Mineral water "Miral"	<0.02
Mineral water "Donat"	<0.02
Coca Cola	<0.02
Coca Cola canned	<0.02
"Fanta" orange beverage	<0.02
"Fanta" orange beverage canned	0.023±0.002
Orange nectar	<0.02
"ACE" multivitamine beverage	0.035±0.002
"FRUC" fruit beverage	0.031±0.002
Apple juice	0.437±0.009
Red wine "Teran"	0.125±0.007
Red wine "Merlot"	0.080±0.005
Red wine "Cabernet"	0.120±0.007
Red wine "Refošk" Koper	0.118±0.007
Red table wine of local grower	0.059±0.003
White table wine "Janžek"	0.283±0.009
Light beer "Laško"	0.039±0.003
Laško beer "Zlatorog"	0.027±0.002
Laško beer "Zlatorog" canned	0.035±0.003

[a] mean of two parallel determinations±standard deviation of measurements.

dent that the Al concentration in mineral waters, refreshing beverages, orange nectar and beer is very low, less than 0.04 mg dm^{-3} of Al. It can be also seen that there is no leaching of Al in beverages stored in Al containers, since the cans are protected by a plastic foil. A slightly higher concentration of about 0.44 mg dm^{-3} of Al was found in apple juice. The concentration of Al in wines ranged between 0.06 and 0.3 mg dm^{-3}. On the basis of these observations it can be concluded that the beverages analysed do not contribute appreciably to the daily intake of Al. Al concentrations reported in different beverages ranged from 0.2 to 7 mg dm^{-3} (Gramiccioni et al., 1996; Müller et al., 1998; Oniawa et al., 1997).

3.6. Determination of Al in total diet samples

Twenty lyophilised total diet samples obtained from the Slovenian Army were analysed for Al content. In order to estimate the daily intake of Al, the mass of total diet was account. Al was determined in lyophilised diet samples and calculated on a fresh weight basis. To compare these data with other foodstuffs, Al concentration was also expressed on a dry weight basis. The results are presented in Fig. 1. It is evident that the concentration of Al expressed on a dry weight basis ranged from 2.9 to 6.9 mg kg^{-1}. Taking into consideration that the moisture content in total diet samples is about 80%, the concentration of Al expressed on a fresh weight basis is appreciably lower. Data from Fig. 1 also indicate that the total daily intake of Al ranged from 2 to 6 mg per day. Our observations are somewhat lower than the average dietary intake of Al reported by Gramiccioni et al. (1996) and Sherlock (1989) who found about 6 mg Al per day in total diet samples. These concentrations are far below the maximal permitted intake of 60 mg Al per day for an adult man, established by the FAO/WHO Expert Committee on Food Additives (World Health Organization, 1989).

3.7. Determination of total and extractable Al in black and herbal tea samples

For the analysis of the total concentration of Al in black and herbal teas, samples were digested and Al determined as described in Section 2.2. Extractable Al was determined in infusions left to stand for 10 min, as described in Section 2.5. The results calculated on a dry weight basis are presented in Table 5. It can be seen that total Al in black tea ranged from 685 to 1200 mg kg^{-1}. Similar high concentrations of Al in black tea leaves from 600 to 1200 mg kg^{-1} were reported in the literature (Müller et al., 1998). Data from Table 5 also indicate that the concentrations of total Al in herbal teas are appreciably lower, between 30 and 200 mg kg^{-1}. Extractable Al in black tea is also high, between 170 and 280 mg kg^{-1}, which corresponds to 17 and 28 mg dm^{-3} of Al in a black tea infusion, while extractable Al in herbal teas ranged from 5 to 20 mg kg^{-1}, corresponding to 0.5 to 2 mg dm^{-3} of Al in a herbal tea infusion.

3.8. Leaching of Al from Al-cookware

In order to find whether cooking may leach higher concentrations of Al, espresso coffee was prepared in stainless steel and in Al containers. Coffee prepared

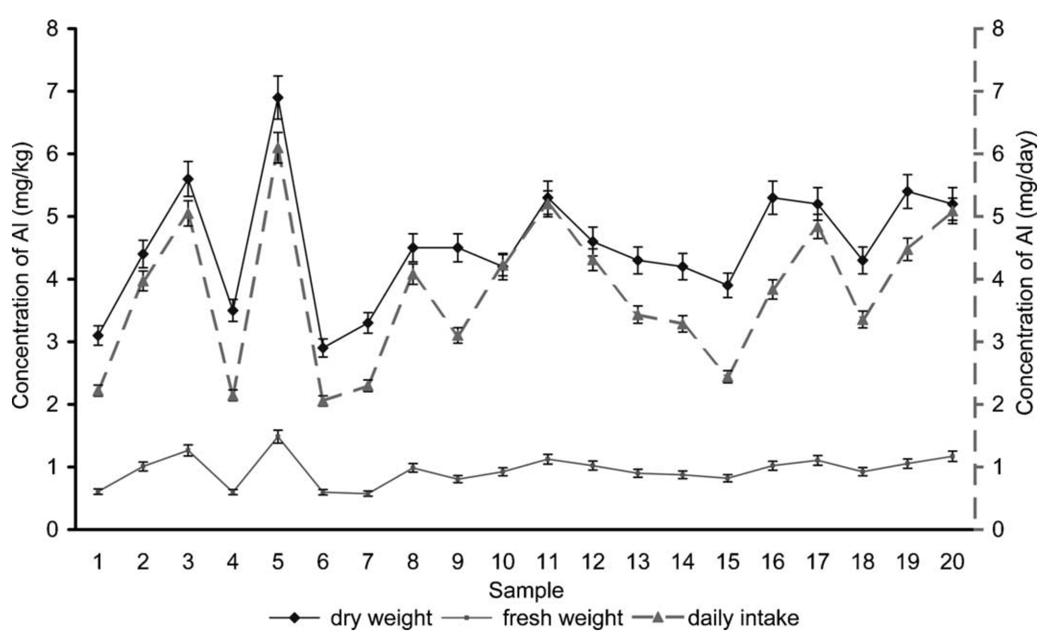

Fig. 1. Determination of Al in twenty total diet samples from the Slovenian Army after microwave digestion and determination of Al by ICP-AES. The results, expressed on a fresh weight basis, a dry weight basis and as daily intake of Al, represent the mean of two parallel samples.

Table 5
Analysis of total and extractable Al in black and herbal teas by ICP-AES (the results are calculated on a dry weight basis)[a]

Sample		Concentration of extractable Al (mg kg^{-1})	Concentration of total Al (mg kg^{-1})
Black tea	Tung Ting Oolantez	226±5	1190±25
	Williamson and Magor	194±5	725±20
	Twinings English Breakfast tea	280±6	800±20
	Fomosa Finest Oolong	226±5	1180±25
	Twinings Orange Pekoe tea	247±6	685±15
	Twinings Darjeeling tea	167±5	1185±25
Herbal tea	Bilberry tea	8.3±0.1	32.5±0.5
	Rose hip tea	11.1±0.2	105±2
	Camomile tea	12.1±0.2	184±3
	Peppermint tea	5.2±0.1	88±2
	Lime-blossom tea	13.6±0.2	46.5±0.8
	Hibiscus tea	21.3±0.4	210±4

[a] Mean of two parallel determinations±standard deviation of measurements.

in stainless steel contained 10.5 mg kg^{-1} of Al (dry weight basis), which corresponded to 0.35 mg dm^{-3} of Al in a coffee infusion. Coffee made in an Al container leached moderate concentrations of Al during preparation, and the concentration of Al increased to 33 mg kg^{-1} (dry weight basis). This concentration corresponded to 1.1 mg dm^{-3} of Al in a coffee infusion.

In the second experiment sauerkraut or sour turnip were cooked in Al-cookware and the concentrations of Al in the liquid of sauerkraut and sour turnip were determined as described in section *2.5*. Before cooking the sauerkraut liquid contained 2.2 mg kg^{-1} of Al, calculated on a dry weight basis of sauerkraut, and the liquid of sour turnip 1.5 mg kg^{-1} of Al (dry weight basis of sour turnip). The corresponding concentrations in the liquid of sauerkraut and sour turnip, expressed in mg dm^{-3}, were 0.22 and 0.15 mg dm^{-3} of Al, respectively. After cooking in Al-cookware the concentration of Al in sauerkraut increased to 313 mg kg^{-1} (dry weight basis) and in sour turnip to 260 mg kg^{-1} (dry weight basis). The concentrations after cooking, expressed in mg dm^{-3} corresponded to 31.3 and 26.0 mg dm^{-3} of Al, respectively. Therefore, it is evident that during cooking in Al-cookware sauerkraut and sour turnip leach substantial concentrations of Al. These concentrations are much higher than those reported for tomato juice. In tomato juice the concentration of Al increased after cooking in Al-cookware from 3 to 5.5 mg kg^{-1} (Gramiccioni et al., 1996).

3.9. Concentrations of Al in tap water

The regular monitoring of the Al concentration in tap water was performed in our group by electrothermal atomic absorption spectrometry on a Hitachi (Tokyo, Japan) Z-8270 polarized Zeeman atomic absorption spectrometer, in twelve dialysis centres in Slovenia (number of observation = 55) (Benedik & Milačič, 1997). The data indicated that the Al concentration in various municipal water supplies ranged from 0.001 to 0.2 mg dm^{-3}, with an average concentration of 0.037 mg dm^{-3} of Al. It was observed that Al concentrations in municipal water supplies that did not use Al-based flocculating agents were in general below 0.001 mg dm^{-3}, while in those that used Al-containing flocculants the concentrations varied from 0.03 up to 0.2 mg dm^{-3}. Variations in Al concentration in tap water were also observed in monitoring performed for 54 municipal water supplies in Quebec, Canada (Gauthier et al. 2000), indicating that Al concentrations ranged from 0.005 to 0.26 mg dm^{-3} of Al with an average Al concentration of 0.04 mg dm^{-3}.

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