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ПРОБЛЕМНАЯ СТАТЬЯ

THE BIOLOGICAL IMPORTANCE OF ALUMINIUM IN THE FOOD CHAIN OF ANIMALS AND MAN: INTAKE, APPARENT ABSORPTION RATE, BALANCE AND LIMITING CONCENTRATIONS

БИОЛОГИЧЕСКОЕ ЗНАЧЕНИЕ АЛЮМИНИЯ В ПИЩЕВЫХ ЦЕПЯХ ЖИВОТНЫХ И ЧЕЛОВЕКА: ПОТРЕБЛЕНИЕ, УСВОЕНИЕ, БАЛАНС И ПРЕДЕЛЬНЫЕ КОНЦЕНТРАЦИИ

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КЛЮЧЕВЫЕ СЛОВА: алюминий, пищевая цепь, взаимодействие, животные, человек

ABSTRACT: The geological origin of the soil and its pH value influence the aluminium content of the flora. The weathered soils of the Rotliegende, phyllite, granite and gneiss produce vegetation rich in aluminium. A low pH value of the soil (e.g. acid rain) increases the aluminium solubility in the soil, the aluminium content of the plants and decreases their calcium, magnesium, iron and zinc uptake and content. In areas with acid rain, the aluminium concentration in the organs of domestic ruminants is increased, whereas calcium, magnesium, iron and zinc content are decreased. Target organs of the aluminium intake are skeleton and brain, which accumulate high amounts of this ultratrace element (microcytic anaemia, osteomalacia, and encephalopathy). Desferrioxamine supplementation removed the aluminium from the tissue by renal excretion. Aluminium may be essential for life (experiments with goats and hens). It is necessary for normal reproduction performance and life time. The proof of aluminium in an essential organic compound (hormones, enzymes, proteins e.g.) of the body is pending. The basal requirement of herbivores is ~10 mg/kg food dry matter (DM). Highest amounts of aluminium are found in black tea (900 mg/kg dry matter — dm), in spices (170 mg/kg dm), herbs (155 mg/kg dm) and vegetables (81 mg/kg dm) medium aluminium concentration were stored in fruits (20 mg/kg dm), white meat, fish, dairy products, bread, pastries. Sugar-rich food is poor in aluminium (ca. 10 mg Al/ kg dm). On average, peoples with mixed diet get 70% of their daily aluminium-intake from vegetable food, 21% from animal food and 9% from drinks. Ovo-lacto vegetarians consume about 30% more aluminium than persons living on a mixed diet. The tolerable daily intake may be 1 mg/kg body weight. Woman and men in Germany eat 3.1 and 3.2 mg Al/day on a weekly average. Currently, the German population with mixed diet consumed 40 to 50 μg/kg body weight. It is only 5% of the tolerated daily intake. Thus, the nutritional intake of aluminium possesses no toxical or nutritional risk to humans.

РЕЗЮМЕ: Содержание алюминия в растениях зависит от геологического происхождения почв и их pH. На почвах, образованных в результате выветривания нижнепермских песчаников, филлитов, гранитов и гнейсов, растения содержат много алюминия. Низкие величины pH почвы (например, в зонах кислотных дождей) повышают растворимость алюминия, его содержание в растениях, и снижают ассимиляцию растениями кальция, магния, железа и цинка. В таких зонах также увеличено содержание алюминия и

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снижено содержание кальция, магния, железа и цинка в органах сельскохозяйственных животных. Органы-мишени для алюминия — скелет и головной мозг (микроцитарная анемия, остеомаляция, энцефалопатия), в них этот ультрамикроэлемент накапливается в больших количествах. Алюминий выводится из организма через почки, его удалению из тканей способствует десферриоксамин. Эксперименты на козах и курах показали, что алюминий необходим для нормальной жизнедеятельности и размножения. Базовая потребность в алюминии составляет для травоядных животных ~10 мг/кг сухого веса пищи. Наибольшее содержание алюминия обнаружено в черном чае (900 мг/кг сухой массы), пряностях (170 мг/кг), зелени (155 мг/кг) и овощах (81 мг/кг); относительно много его во фруктах (20 мг/кг сухой массы), белом мясе, рыбе, молочных продуктах, хлебе, выпечке. Пища, богатая сахаром, содержит мало алюминия (около 10 мг/кг). В среднем люди со смешанным питанием получают 70% алюминия с растительной пищей, 21% — с животной и 9% — с напитками. Оволактовегетарианцы потребляют примерно на 30% больше алюминия, чем люди со смешанным питанием. Переносимый уровень потребления алюминия предположительно равен 1 мг/кг массы тела в сутки. Женщины и мужчины в Германии потребляют в среднем 3,1 и 3,2 мг Al в день, соответственно, это ~5% от допустимого уровня. Таким образом, поступление алюминия с пищей не связано с риском для здоровья.

INTRODUCTION

Aluminium is the third most abundant element in the Earth's crust after oxygen and silicon. Although it has been produced in commercial quantities for just over 100 years, it is used more today than any other metal, except iron. Aluminium makes up 8.8% of the Earth's crust. Only the ^{27}Al isotope is natural stable. Nearly all aluminium is obtained from bauxite, which contains 40—60% aluminium. In the year 2000 worldwide aluminium production was ~ 24×10^6 metric tons (Yokel, 2004).

The major uses of aluminium: Transportation 35%, packaging 25%, building 15%, consumer durables 8% electrical 7%, and other 10% (Plunkert, 2002).

Environmental exposure of aluminium is from natural sources and is increased by acidification. Animal and human exposure is primarily from the diet.

Aim of these experiments, which started 1986, was the identification of the aluminium in the food chain of animals and man, the toxicity and essentiality of this ultratrace element for animal and man and their intake, excretion, apparent absorption, balance and normative requirement in case of its essential or limiting concentrations in case of toxicity (Anke et al., 1990, 1991).

Table 1. ***Experiments, number of the animals and origin of the goats of aluminium-deficiency experiments***

Year	Control goats	Al-deficient goats	Remarks
1	6	5	Bought
2	6	5	1 Al-deficient kid
3	4	5	Bought
4	4	1	1 Al-deficient kid
5	5	5	Bought
6	4	1	1 Al-deficient kid
7	4	1	1 Al-deficient kid
8	2	—	—
9	1	—	—
Total	36	23	—

MATERIAL AND METHODS

All samples were collected in plastic vessels and stored at -18°C until use. The samples were dried at 105°C, ashed in a muffle furnace at 450°C, dissolved in 25% HCL and diluted to make up a 2.5% solution.

Aluminium determination was carried out by graphite furnace atomic absorption spectrometry using a combination of AAS 3030, HGA 400 and AS (Perkin Elmer) and the magnesium nitrate matrix modifier (Merck). Reference material JAEAH-9 (mixed human diet) was used checking the method. A good accordance between reference results (16.6 µg_Al/g) and own analysis (17.4 µg_Al/g) could be observed. Relative and absolute detection limits of the method are at 1.3 mg/l and 26 µg (pipette volume 20 ml) (Muller et al., 1995d, 1998)

All foodstuffs were prepared ready for cooking, but raw. All inedible parts, for instance the cores of apples and pears were removed from the samples.

The experiments with goats and a semi-synthetic aluminium poor ration began on July 1st 1986 with female kids of goats (White German goats, Thuringian Forest goats, Dappled German goats of the dark breed).

The stock was replenished by the purchase of animals or by animals from our own breeding. All animals remained in the experiment till their natural death. Deceased goats delivered different organs for analytical purposes. The animals were kept in individual or group bases with cellulose as litter. Their drinking water was distilled. The control- and aluminium goats were fed a semi-synthetic ration which contained per 100 kg: 48.3 kg potato starch, 32 kg beet sugar, 10 kg casein, 3 kg urea and 3 kg sunflower oil. The ration was fortified with macro, trace and ultratrace elements and the vitamins E, A and D_3 (Anke, Groppel, 1989).

Per 100 kg control ration 200 g $Al_2(SO_4)_3$ were added, which was not done in the group with aluminium poor rations. The aluminium content in the deficiency ration amounted to 6.5 mg/kg dry matter. The animals were fed ad libitum on the clean trough. The cellulose intake of control and aluminium-deficient

Table 2. ***Composition of the semisynthetic diet for goats (100 kg)***

Components	Amount	Components	Amount
Potato starch	48.29 kg	KI	100 mg
Turnip sugar	32.00 kg	$SnCl_2$ x 2 H_2O	95 mg
Casein	10.00 kg	$(NH_4)_6Mo_7O_{24}$ x 4 H_2O	92 mg
Urea	3.00 kg	SeO_2	80 mg
Sunflower oil	3.00 kg	$CoSO_4$ x 7 H_2O	80 mg
KH_2PO_4	1.340 kg	$La_2(CO_3)_3$ x 3 H_2O	80 mg
$CaCO_3$	1000 g	CH_3COOAg	76 mg
K_2SO_4	400 g	$BiO(NO_3)$ x H_2O	73 mg
NaCl	350 g	BeO	70 mg
MgO	200 g	RbCl	70 mg
$Al_2(SO_4)_3$, 60%ig	200 g	PbC_{12}	67 mg
$ZnSO_4$ x 7 H_2O	50 g	Sb_2O_5	66 mg
$FeSO_4$ x 7 H_2O	50 g	$Tl(NO_3)$	65 mg
$MnSO_4$ x H_2O	30 g	Nb (metal)	50 mg
Na_2SiO_3 x 8 H_2O	21 g	As_2O_3	40 mg
H_3BO_3	14 g	$CdCl_2$ x H_2O	36 mg
$BaCl_2$ x 2 H_2O	9 g	$Cs(NO_3)$	29 mg
Li_2CO_3	5 g	$2(NH_4)_2SO_4$ x $Ce(SO_4)_2$ x 2 H_2O	23 mg
$CuSO_4$ x 5 H_2O	4 g		
KBr	3 g	$K_2(PtCl_4)$	11 mg
Ni_2SO_4 x 7 H_2O	2 g	$(CH_3COO)_2UO_2$	8.9 mg
$Sr(NO_3)_2$	1 g	$PdCl_2$	8.4 mg
Vitamin E	20 g	$Na_2(IrCl_2)$ x H_2O	7.8 mg
Vitamin A	2 g	$HgSO_4$	7.4 mg
Vitamin D_3	400 mg	GeO_2	7.2 mg
TiO_2	835 mg	Ga_2O_3	6.7 mg
TeO_2	625 mg	OsO	6.7 mg
$Cr_2(SO_4)_3$ x 18 H_2O	600 mg	Ta_2O_5	6.1 mg
NaF	220 mg	AuCl	5.9 mg
NH_4VO_3	200 mg	HfO_2	5.9 mg
$NaWO_4$ x 2 H_2O	180 mg	ThO_2	5.7 mg
$ZrOCl_2$ x 6 H_2O	159 mg	Sc_2O_3	0.8 mg

goats were calculated to 100 g/day and animal. The mean aluminium content of the used cellulose was about 180 mg/kg dry matter. The live weight of the animals was registered every fortnight.

The aluminium intake of adults in Germany was investigated be means of the so called double portion technique.

The experiments were carried out with 14 test-populations of East Germany about seven subsequent days. Al least seven men and seven women took part in each trial. They collected visually estimated duplicates of their daily meals including all beverages and snacks and they wrote a protocol from their intake. A total of 1456 duplicates were available for analysis. Table 3 informs about the analysed samples.

Table 3. ***Kind of samples, table numbers and quantity of samples analysed***

Kinds of samples	Table	Number of samples
Indicator plants	4	5
Plants, species, age, part	5; 6; 7	296
Fissures of animal species	8; 9	1351
Influence of acid rain	10; 11	1363
Essentiality of aluminium	13	102
Indicator organs of aluminium states	14	1188
Aluminium in food stuffs and beverages	15 to 19	1456
Aluminium intake of humans	21	1456
Sum		7996

RESULTS

1. ALUMINIUM IN THE VEGETATION

1.1. INFLUENCE OF THE GEOLOGICAL ORIGIN

On average, the 16 km thick Earth crust contains about 81.3 g_Al/kg (Falbe, Regitz, 1989). Aluminium, the third most abundant terrestrial element, is ubiquitously distributed throughout the environment. Aluminium in the rocks commonly ranged from 4.3 to 100 g/kg. Granites and gneisses store 72 to 88 g_Al/kg, sandstone 25 to 43; Muschelkalk and Keuper layers — 4.3 to 13 g_Al/kg respectively.

The total aluminium content of soils is inherited from parent rocks. In acid soils with pH below 5.5, the mobility of aluminium increase sharply and very actively competes with other cations for exchange site. A sudden increase of the aluminium solubility is observed mainly at the narrow range of the pH from 4.5 to 4.0. Soil acidification due to the atmospheric deposition of sulphur dioxide increases aluminium solubility in soils. The mobile aluminium in soils can be taken up rapidly by plants and it creates a problem of chemical stress (Kabata-Pendias, Pendias, 2001). The soils of weathered Rotliegende, phyllite and granite produce vegetation, which is rich in aluminium (Table 4).

Table 4. ***Influence of the geological origin of the relative aluminium content the flora (n = 784)***

Geological origin of the site	Relative index
Rotliegende weathering soils	100
Phyllite weathering soils	93
Granite weathering soils	90
Gneiss weathering soils	83
Loess	78
Diluvial sands	78
Boulder clay	73
Bog, peat	73
Muschelkalk weathering soils	71
Bunter weathering soils	68
Keuper weathering soils	54

Medium aluminium contents are found in the flora on Pleistocene formations (loess, diluvial sands, boulders clay) and Holocene formations (peat, moor, riverside plains). The vegetations are poorest on weathering soils of the Triassic (Keuper, Bunter, Muschelkalk) (Anke et al., 2001).

1.2. INFLUENCE OF THE PLANT SPECIES

The aluminium content of the vegetation is species-specific (Table 5). Besides the geological origin of the soil, the species of the plant influence their aluminium content significantly.

Table 5. ***Species-specific aluminium content of the plants (mg/kg DM) (n = 942)***

Usual soil Species (n)	x, s	Muschelkalk Species (n)	x, s
Meadow red clover	70 ± 28	Wheat (6)	121 ± 54
Field red clover	54 ± 22	Couch grass (6)	115 ± 64
Meadow grass (104)	52 ± 30	Field red clover (6)	65 ± 55
Rye (231)	43 ± 20	Lucerne (6)	65 ± 37
Wheat (369)	36 ± 20	Rape (6)	58 ± 21

Note: x, s = arithmetic mean, standard deviation, n = number of samples.

Meadow red clover in blossom, field red clover in the bud, rye in blossom and shooting wheat accumulate in Germany between 36 mg_Al/kg DM in wheat and 70 mg_Al/kg DM in meadow red clover. On the other hand plant species grown at the upper Muschelkalk and harvested at the end of May store between 58 and 121 mg_Al/kg DM (Anke et al, 2001).

1.3. INFLUENCE OF THE PLANT PART

The aluminium content of the plants is highest in leaves. Ears and flowers accumulate only ~ 40% of the aluminium content found in the stalks of wheat and 14 to 28% in the stalks of field red clover. The differences are significant ($p < 0.05$). The leaf to stalk ratio of the species influences aluminium content of the flora (Muller, 2007)

Table 6. ***Aluminium content of several plant parts (mg/kg DM) (n = 42)***

Wheat (n)	x, s	Field red clover (n)	x, s
Leaves (12)	128 ± 50	Leaves (6)	144 ± 19
Ears (6)	52 ± 8	Flowers (6)	64 ± 13
Stalks (6)	15 ± 12	Stalks (6)	41 ± 22

1.4. INFLUENCE OF THE PLANT AGE

With increasing age, the aluminium content decreases (Table 7). The influence of age on the aluminium concentration of the vegetation is very strong and highly significant.

In early spring the aluminium intake of wild grazing ruminants and plant eaters is very high (Muller et al., 1995a, Anke, 2004a).

2. ALUMINIUM IN ANIMALS

The aluminium concentration of several tissue were analysed from wild and domestic animal species (Table 8). Animals, irrespective of species and feeding habits, incorporate a lot of aluminium in their tissues. Highest amounts of aluminium are accumulated in skeleton of all tested species (59 to 87 mg/kg DM). The cerebrum stores also relatively high amounts of aluminium (25 to 61 mg_Al/kg DM).

Hair, bristles and feathers contains like kidneys 38 and 35 mg_Al/kg DM. Liver and muscles are relatively aluminium poor (28 and 14 mg/kg DM).

Sheep subjected to prolonged aluminium loads (16 g_Al/kg_feed DM) have shown increased aluminium content in all tissue (Table 9).

The main target organ of aluminium is the nervous system. In the cerebrum aluminium concentration was increased six-fold, in ribs threefold, and in liver and kidneys relatively threefold to twofold. Brain and bone accumulate highest amounts of aluminium. In case of the aluminium intoxication both organs show the symptoms of aluminosis.

3. ECOTOXICOLOGY AND TOXICITY

3.1. PLANTS

The interest in aluminium as a toxic ultratrace element has increased during last 30 years. Acid rain (by sulphuric acid and/or NO_x), which decreased pH of soil (< 4.2), and increased solubility of the aluminium compounds, was a reason for the present forest decline. At pH values < 4, the acid stress in the soils is due to protons. On the other hand, aluminium ions are accidentally transported through the ATP-dependent calcium channels. Increased ratios of aluminium to calcium and magnesium ions in the soil solutions decrease or even prevent the uptake of these essential cations by the root and their transport to the shoots. Even aluminium-tolerant species may be severely stressed by continuous damage to the root system and insufficient nutrient uptake. A continuous damage to the root system can be overcome by root regeneration, but it consumes photosynthesis products, which are missing for production in the shoot.

Table 7. ***Effect of the stage of vegetation (age) on the aluminium content in various species (mg/kg DM) (n = 120)***

Species (n)	04.05	17.05	31.05	14.06	%*	LSD**
Wheat (6)	194 ± 58	191 ± 131	121 ± 54	57 ± 55	29	102
Field red clover (6)	88 ± 34	92 ± 29	65 ± 55	36 ± 20	41	44
Lucerne (6)	112 ± 29	102 ± 18	65 ± 37	54 ± 23	48	37
Couch grass (6)	138 ± 72	142 ± 84	115 ± 64	87 ± 34	63	82
Rape (6)	71 ± 29	72 ± 27	58 ± 21	52 ± 27	73	35

Note: n = number of samples;* 04.05 = 100%, 14.06 = x%; **LSD = Least significant difference.

Table 8. ***Species-specific aluminium content of various indicator organs (mg/kg DM) (n = 1351)***

Species	Tissue					
	Rib	Cerebrum	Hair*	Kidneys	Liver	Muscle
Sheep	59 ± 15	61 ± 18	—	43 ± 13	37 ± 13	—
Pig	63 ± 20	36 ± 10	39 ± 25	42 ± 24	18 ± 10	12 ± 47
Cattle	68 ± 15	48 ± 20	19 ± 62	32 ± 13	32 ± 13	20 ± 8,6
Hen	69 ± 14	50 ± 12	38 ± 14	38 ± 13	42 ± 22	6,4 ± 25
Fallow deer	73 ± 34	56 ± 20	—	34 ± 20	30 ± 18	—
Hare	86 ± 30	56 ± 20	35 ± 10	33 ± 13	16 ± 30	16 ± 6
Cat	87 ± 33	25 ± 13	44 ± 22	24 ± 16	21 ± 16	—
Mean	72.1	47.4	35.0	35.1	28.0	13.6
Fp Species	< 0,001	< 0,001	< 0,001	< 0,001	< 0,001	< 0,001

* Hair, bristles, feathers; Fp = signifinace level in one- or multifactorial variance level analysis.

Table 9. ***Influence of aluminium exposure on the aluminium content of various tissues of adult sheep (mg/kg DM) (n = 48)***

Tissue	Control	Al exposure	p*	%
Kidneys	43 ± 13	89 ± 31	< 0,001	207
Liver	37 ± 13	96 ± 31	< 0,001	260
Rib	58 ± 15	195 ± 54	< 0,001	330
Cerebrum	61 ± 18	397 ± 114	< 0,001	651

* 04.05 = 100%, 14.06 = x%; **p = significance level in Student's t-test.

3.2 ANIMALS

Supplementation of 150 mg_Al/day (and 50 mg) about ~40 months increased histological lesions in CNS and phosphorylated neurofilaments in monkeys (Garruto et al., 1989).

In rabbits long time supplementation (12 months) of the drinking water with 5 mg_Al/l depressed growth and caused accumulation of aluminium in the tissue. It induced no histological changes (Wills et al., 1993). Aluminium in the water reduced fluorine retention (Ahn et al., 1995).

Chicken with 0,5% Al in the diet in 3 weeks developed a high mortality (> 80%) and decreased weight gain and bone ash content (Storer, Nelson, 1968). 1500 and 3000 mg_Al/kg_diet in form of aluminium sulphate in 22 weeks depressed growth and bone ash, too, and reduced egg production (3000 mg_Al/kg_diet) (Wisser et al., 1990). On the other hand, aluminium (40, 80, 520, 1040 mg_Al/kg_diet) in 112 days reduces toxic effect of fluorine on feed intake, egg production and tissue fluorine retention (Hahn, Guenther, 1986). In turkeys, aluminium decreased the toxic effects of fluorine, too (Cakir et al., 1977).

In sheep, 3.3 g_$AlCl_3$/day in 25 months increased faecal fluorine excretion and decreased mottling of the teeth (Said et al., 1977), 2000 mg_Al/kg_diet reduced (56 days) weight gain and phosphorus, calcium absorption (Valdivia et al., 1982), 1450 mg_Al/kg_diet (76 days) had the same effect (Rosa et al., 1982). Similar effects of aluminium intoxication were seen in rats, mice and fish (Anonymous, 2005).

Little is known about ecotoxicological effects of aluminium in mammals. The effect was analysed in tissues of cattle, sheep, fallow deer, hare and roe deer living in regions with acid rain and without. In areas with acid rain, they incorporate 10—60% more aluminium in cerebrum, rib, kidney and liver (Table 10). The differences in aluminium content in the organs of the species are partly significant.

Table 10. ***Influence of acid rain on the aluminium status of cows and wild deer (mg/kg DM)Tissues***

Tissues	Cow				Wild deer			
	Acid rain		p	%	Acid rain		p	%
	without	with			without	with		
Rib	68 ± 15	73 ± 22	> 0.05	107	73 ± 34	79 ± 20	> 0.05	108
Kidney	32 ± 13	39 ± 24	> 0.05	132	34 ± 20	50 ± 12	< 0.01	147
Liver	32 ± 13	44 ± 16	< 0.05	138	30 ± 18	49 ± 7.1	< 0.01	163
Cerebrum	48 ± 20	69 ± 15	< 0.05	144	56 ± 20	68 ± 16	> 0.05	121

Table 11. ***Influence of acid rain on the calcium, magnesium, iron and zinc content of several tissues of roe deer and sheep (mg/kg DM)***

Tissues		Roe deer				Sheep			
		Acid rain		p	%	Acid rain		p	%
		without	with			without	with		
Ca g/kg	Kidney	0.82	0.49	< 0.01	60	0.80	0.62	> 0.05	78
	Rib	259	215	< 0.01	83	212	193	> 0.05	91
Mg mg/kg	Kidney	1255	984	< 0.05	78	1097	871	< 0.01	79
	Rib	4155	3513	< 0.05	85	5147	3275	0.01	64
Fe mg/kg	Kidney	298	162	< 0.05	54	—	—	—	—
	Rib	303	217	> 0.05	72	—	—	—	—
Zn mg/kg	Kidney	210	140	< 0.01	67	—	—	—	—
	Rib	111	79	< 0.01	11	—	—	—	—

The analysis of the macro and trace content of the tissues of roe deer and domestic sheep with high aluminium intake from regions with and without acid rain shows the interaction of these feeding regimes on the calcium, magnesium iron and zinc metabolism and tissue content.

This result demonstrates the ecotoxicological influence of acid rain and the increased aluminium absorption. The interactions of aluminium with the four nutritionally important elements calcium, magnesium, iron and zinc in wild and domestic species documents a relevant influence of emission of sulphuric acid and NO_x on the aluminium status and a secondary deficiency of calcium, magnesium, iron and zinc (Muller et al., 1995a; Muller, 2007; Anonymous, 2005).

3.3. HUMANS

One of the first cases of poisoning of man attributed to aluminium was reported in 1921. The patient, a metal worker, suffered memory loss, tremor, jerking movement, and incoordination. Five years later, attention was drawn to the potential health hazards of aluminium present in municipal drinking water supplies, various medicine and possible industrial exposures. During the 1930s and 1940s, cases of lung fibrosis were discovered in Germany, Sweden and Great Britain due to an occupational exposure to elemental aluminium dust. The new disease was called aluminosis. These cases of disease were only observed in workers exposed to high concentration of stamped aluminium powder, which had not been treated with stearine. Later, in 1947, lung fibrosis was observed in occupationally aluminium-exposed employees (production of corundum from bauxite). These are cases of «shaver's disease» and pulmonary aluminosis in particular.

In the 1970s, an increased aluminium level in dialysis patients with renal insufficiency was observed. Alfrey and co-authors (1972) and Alfrey (1978) were the first to describe the neurological syndromes of this «dialysis encephalopathy» or dialysis dementia. The excess of aluminium in serum and tissue resulted from therapeutically administered aluminium (e.g., aluminium containing phosphate-binding gels and aluminium contaminated dialysis solutions). After it could be proved that dialysis patients, compared to the general population, showed among other things, a higher aluminium level in the brain, a neurotoxic effect of aluminium was assumed. The targets of aluminium toxicity in humans are the central nervous system (encephalopathy), the bone tissue (osteomalacia), and the erythropoietic system (microcytic anaemia). The typical symptoms of the aluminium-induced dialysis encephalopathy developed in the following succession:

— Anaemia, normochromic normocytic

— Osteomalacia

— Soft tissue or metastatic extraskeletal calcification

— Speech and motor disturbances

— Increased predisposition to spasms

— In the last stage, rapidly progressing dementia

The only method available for removing substantial quantities of aluminium is chelation with desferrioxamine.

In several brain regions of patients with Alzheimer's disease aluminium concentrations was deducted. At present, there is no evidence to substantiate that Alzheimer's disease is induced by aluminium toxicity. The toxicological relevance of Aluminium in connection with the Parkinson dementia complex is the indigenous (Chamarro) population of Guam is under discussion.

The administration of aluminium-containing drugs is disputed. Aluminium hydroxide is contained in antacids, in substances applied in the therapy of hyperphosphatemia and ulcers, and — as an auxiliary — in antidiarrhetics, buffered analgetics, and various vaccines. In this way, the daily intake with, e. g. antacids, may be 1 g of aluminium and more. For a healthy organism with normal kidney formation, these quantities are safe, as a rule, whereas the drugs mentioned must not be administered to patients with chronic renal insufficiency.

Attentions should be paid also to oral aluminium intake via food additives, migration from packing materials, and drinking water. Insogna and co-authors (1980) registered after long-term intake of 0.5—9.6 g aluminium hydroxide (antacid) /day osteomalacia, bone pain, bone loss, hyperphosphatemia and elevated alkaline phosphatase activity.

Greger and Baier (1983a,b) observed after intake of 125 mg_Al/day in form of aluminium lactate about 20 days a transient decrease of phosphorus absorption, decreased urine fluoride, and increased serum content.

A mutagenic and carcinogenic potential of aluminium has not been reported (Spofforth, 1921; Betts, 1926; Persson, 1980; Leonard, 1988; Dietrich et al., 1988; Schenkel et al., 1989; Savory, Wills, 1991; Marquardt, Schafer, 1994; Schaller et al., 1994).

4. ESSENTIALITY OF ALUMINIUM

Irrespective of these toxic effects aluminium may be an element necessary for fauna life. The aluminium-content of the earth crust, the soils and food is so high, that the requirement of animal and man is covered and no deficiency develops. Semisynthetic rations with low aluminium content and intrauterine aluminium depletion induce in the second and third generation thoroughly deficiency symptoms. Goats with an aluminium intake of < 6.5 mg_Al/kg_feed dry matter developed after intrauterine aluminium depletion an increased mortality, impaired reproduction performance and diminished milk yield in comparison to goats with 38 mg_Al/kg_feed dry matter (Table 12) (Anke, 2004b; Anke et al., 1990, 2001,

Table 12. ***Effects of aluminium-poor nutrition of goats***

Parameter	Control goats	Deficiency Goats	p	%
Feed intake (g/day), adult goats	627	668	< 0.001	107
Live weight, 91th day of life (kg)	17.7	15.8	> 0.05	90
Weight gain, 100th—268th day of life (g/day)	98	101	> 0.05	103
Success of first insemination (%)	74	57	< 0.05	—
Conception rate (%)	90	86	> 0.05	—
Abortion rate (%)	1	14	< 0.001	—
Services per gravidity	1.3	1.8	< 0.001	—
Male/female ratio	1.8	2.2	> 0.05	—
Mortality, kids (%)	8.0	42	< 0.001	—
Mortality, first year of life (%)	0	35	< 0.001	—

2005, 1991; Anonymous, 1996; Angelow et al., 1993; Muller et al., 1995d, 1996).

Feed intake and growth were not significantly influenced in goats, but they were in hens (Carlisle, Curran, 1993).

The difference in aluminium supply had an insignificant effect on different components of blood plasma and the activity of selected enzymes (Table 13), except that the urea content in the blood plasma of goats with aluminium-poor nutrition was significantly higher than that in the control group. This change was the only indication of metabolic changes due to aluminium-poor nutrition. So far, no analysed essential body substance has been found to depend on aluminium (Anke et al., 2001).

The aluminium poor nutrition altered the aluminium content of aorta, spleen, rib and carpal bones significantly (Table 14). The aluminium content in the aorta of aluminium-deficient goats decreased to 44% of the normal aluminium concentration in this tissue.

Pathological changes in goats with aluminium poor nutrition manifested themselves as weakness of the hind legs, and disturbed coordination resulted therefore. The basal aluminium requirement of animals is satisfied under practical conditions. The basal requirement of herbivores is less than 10 mg_Al/kg_feed dry matter. This aluminium amount we can find in all practical feed rations of domestic or wild animal species. Aluminium deficiency can be excluded under practical conditions.

To sum up, changes in the goats and hen's chicken as a model animal, observed during experiments with 6.5 mg_Al/kg_feed dry matter, suggested the potential essentiality of this element. However, the essentiality of aluminium for animal and man must still be characterised as a component or activator of an enzyme, hormone or essential protein in the body. Older experiments (Horecker et al., 1939) suggested that aluminium promoted the reaction between cytochrome C and succinate dehydrogenase in vitro. The activation of a purified guanine nu-

Table 13. ***The influence of aluminium-poor nutrition on several biochemical parameters of blood plasma***

Parameter	Control goats		Deficiency Goats		p	%
	x	s	x	s		
Protein (g/100ml)	7.2	1.0	7.4	0.7	> 0.05	103
Cholesterol (mmol/l)	1.5	0.6	1.1	0.5	> 0.05	73
Glucose (mmol/l)	3.0	0.4	2.6	0.7	> 0.05	87
Kreatinine (mmol/l)	89	14	95	16	> 0.05	107
Total bilirubin (µmol/l)	2.3	0.8	2.4	0.7	> 0.05	104
Glycerine (mmol/l)	52	16	84	45	> 0.05	162
Phospholipids (mmol/l)	1.2	0.3	1.0	0.4	> 0.05	83
Free fatty acids (µmol/l)	192	90	224	67	> 0.05	117
Triglyceride (mmol/l)	0.2	0.1	0.2	0.1	> 0.05	100
Urea (mmol/l)	4.6	1.8	7.2	2.0	< 0.01	156
Sodium (mmol/l)	146	4.7	150	2.4	> 0.05	103
Potassium (mmol/l)	4.3	0.5	4.2	0.6	> 0.05	98
Phosphorus (mmol/l)	1.8	0.5	1.5	0.5	> 0.05	83
Chloride (mmol/l)	111	2.4	113	3.0	> 0.05	102
AP (U/l)	519	572	717	556	> 0.05	138
GGT (U/l)	48	18	49	10	> 0.05	102
ASAT (U/l)	68	26	66	20	> 0.05	97

Table 14. ***The influence of aluminium-poor nutrition on the aluminium content of various organs and tissues of goats (mg_Al/kg DM)***

Parameter	Control goats		Deficiency Goats		p	%
	x	s	x	s		
Aorta	46	26	20	10	< 0.001	44
Spleen	23	14	13	7.3	< 0.01	56
Muscle	20	15	12	5.0	> 0.05	60
Carpal bone	100	40	72	13	< 0.001	72
Heart	15	7.4	12	7.8	> 0.05	80
Rib	97	33	78	20	< 0.05	80
Kidneys	15	8.2	13	9.4	> 0.05	87
Lungs	14	8.2	13	8.5	> 0.05	93
Ovary	78	38	74	31	> 0.05	95
Hair	16	10	17	8.4	< 0.01	106
Uterus	21	12	23	14	> 0.05	110
Brain	17	5.5	19	14	> 0.05	112
Liver	20	12	27	16	> 0.05	135
Pancreas	26	13	37	24	> 0.05	142

cleotide binding protein, the regulatory component of adenylate cyclase by fluoride was shown to require the presence of Al^{3+} in vitro (Sternweis, Gilman, 1982; Kahn, 1991). The significance of these observations in vitro is not known. However, aluminium may be a cariostatic agent by itself and in combination with fluoride (Kleber, Putt, 1984). Also, the investigations suggest possibly essential character of aluminium for the fauna.

5. ALUMINIUM IN FOODSTUFFS AND BEVERAGES

5.1. FOODSTUFFS

The analysis of 128 food and drink samples in 6—15 repetitions before and after reunification (n = 1821) showed that a major part of men prefer plant foodstuff. If the aluminium content of the single foodstuff before and after reunification is significantly different (e.g. table salt: 1,4—15 mg/kg DM).

In case of no relevant differences, the aluminium concentration is given in form of the average and standard deviation. Poor in aluminium (1 to 10 mg_Al/kg DM) are table salt, sugar- and starch rich foodstuff like sugar, cereals, flour products, pulses, bread, cake and pastries.

On the average mushrooms, fruits, cabbage, kohlrabi, potatoes, cocoa, carrots and cauliflowers accumulate10 to 75 mg_Al/kg dry matter.

Their aluminium content is middle and can fluctuate considerably (e.g. cocoa 37 to 111 mg_Al/kg DM). Asparagus and chive deliver a very high aluminium amount in the food chain of men and animals.

Extremely high amounts of aluminium (110 to 900 mg_Al/kg dry matter) accumulate spices and herbs, cinnamon, parsley, paprika, dill and marjoram, but also cucumber, mixed mushrooms, lettuce and several kinds of tea deliver very high amounts in the food chain of humans. Black tea accumulates 900 mg_Al/kg dry matter, the highest amount of this ultratrace element.

The animal foodstuff stores only 4—9 mg_Al/kg dry matter in milk, diary products, fishes and hens eggs (Table 17) with the exception of mutton, chicken, beef and pork, which contains on the average 12 to 14 mg_Al/kg DM. Sausages have 11 to 14 mg_Al/kg DM, with highest amounts of aluminium in blood pudding. Limburg, Camembert and Tilsit cheese accumulate on average 11 mg_Al/kg DM, whereas curd stores 16 mg_Al/kg DM. Soft cheese, which is produced from badly turned-out cheese being smelted after addition of sodium phosphate, delivers, beside sodium and phosphate also aluminium and other elements.

Liver and kidneys of pig and cattle accumulate 28 and ~40 mg_Al/kg DM, the highest amount of this ultratrace element in the food chain of animal food (Muller et al., 1998, 1997, 1997b; Anke et al., 2001).

Table 15. ***Aluminium contents of plant food, poor in aluminium (mg/kg DM)***

Food	Al	Food	Al
Table salt	1.4—15	Oat pulp	7.6 ± 6.1
Candies	3.6—9.8	Bee honey	7.9 ± 2.1
White beans	3.6—12	Vanilla pudding powder	8.0 ± 4.0
Banana	3.7 ± 11	Sponge cake	8.0 ± 4.9
Wheat flour	4.1—19	Jam	8.3—21
Semolina	4.2—18	White and rye bread	8.3 ± 4.0
Sugar	4.4 ± 1.8	Peas. green	8.7 ± 3.8
Crisp bread	5.2 ± 2.2	Rolls	9.0 ± 4.1
Thin sponge cake with crumble tapping	5.4 ± 1.7	Rusk	9.0 ± 3.4
Pancake wheat	5.4 ± 1.0	Whole wheat flour	9.1 ± 4.1
Corn flour	5.0 ± 1.4	Rice	9.7 ± 4.8
Pasta	6.2 ± 2.4	Toasted bread	9.8 ± 4.2
Corn-grained whole meal rye bread	6.3 ± 2.2	Peas dried	9.9 ± 4.8
Oat flakes	7.1 ± 3.1	Milk. chocolate	9.9 ± 3.5
Pearl barley	7.3 ± 2.3	White bread	10 ± 7.2
Mustard seeds	7.4—12	Cornflakes	10 ± 3.7

Note: data are presented as mean ± SD, or as range.

Table 16. ***Aluminium contents of plant food, rich in aluminium (mg/kg DM)***

Food	Al	Food	Al
Cake with egg topping	11 ± 6.7	Ready-to-serve-soup	38 ± 35
Synthetic honey	11 ± 6.6	Tomatoes	39 ± 18
Chocolate spread for bread	13 ± 4.6	Dwarf beans. tinned	40 ± 18
Mushrooms	14 ± 6.8	Carrots	46—65
Pineapples	14 ± 3.5	Cauliflower	46 ± 25
Maize flour	15 ± 9.2	Kiwi	50 ± 16
White cabbage	15 ± 6.4	Pepper	51 ± 317
Pears	15 ± 3.7	Asparagus	66 ± 33
Lemons	16 ± 8.5	Chive	77 ± 50
Chocolate cream	17 ± 11	Cinnamon	111 ± 48
Lentils	17 ± 10	Parsley	133 ± 38
Oranges	17 ± 10	Paprika. hot	140—281
Apples	18 ± 9.4	Paprika. sweet	174 ± 74
Coffee	19 ± 10	Cucumber	161 ± 100
Sauerkraut	19—68	Dill	255 ± 45
Red cabbage	21 ± 12	Mixed mushrooms	284 ± 18
Biscuits	22 ± 17	Fruit tea	292 ± 157
Kohlrabi	23 ± 16	Mountain-herb tea	419 ± 159
Apple puree	24 ± 11	Lettuce	453 ± 269
Caraway	25 ± 6.5	Peppermint tea	477 ± 52
Mustard	27—46	Aromatised black tea	763 ± 195
Potatoes	30 ± 11	Majoran	797 ± 176
Chocolate pudding	32 ± 15	Black tea	899 ± 292
Cocoa	37—111		

Note: data are presented as mean ± SD, or as range.

Table 17. ***Aluminium content of animal food (mg/kg DM)***

Food	Al	Food	Al
Margarine	3.5 ± 1.1	Salami	11 ± 4.0
Milk	3.8 ± 1.7	Thick Frankfurter	11 ± 4.8
Butter	4.2 ± 2.6	Mackerel filet	11 ± 4.1
Rosefish, filet	4.9 ± 2.1	Limburger cheese	11 ± 6.0
Goat cheese	5.0 ± 2.1	Camembert cheese	11 ± 8.6
Tollense cheese	5.4 ± 2.5	Tilsit cheese	11 ± 5.8
Emmental cheese	5.4 ± 2.3	Chicken	12 ± 6.0
Bismarck herring	5.9 ± 1.7	Mortadella	12 ± 4.0
Yoghurt	6.6 ± 2.9	Liver	12 ± 6.6
Herring fillet	7.1—18	Trout. fresh	12 ± 5.5
Mutton	7.6 ± 2.8	Sardines	13 ± 4.2
Edam cheese	7.9 ± 3.3	Beef	13 ± 7.3
Herring in tomatoes	8.0 ± 4.0	Pork	14 ± 5.6
Salted herring	8.1 ± 4.0	Blood pudding	14 ± 6.0
Condensed milk	8.2 ± 4.5	Trout. smoked	14 ± 8.1
Fried herring	8.3 ± 4.4	Curd	16 ± 8.0
Egg	8.4 ± 1.6	Soft cheese	18—39
Liver sausage	8.5 ± 4.0	Liver	28 ± 15
Gouda cheese	10 ± 5.6	Kidneys	43 ± 25

Note: data are presented as mean ± SD, or as range.

5.2. BEVERAGES

The aluminium content of drinking water in Eastern Germany was not significantly influenced by geological origin of the earth crust (Table 18). Regardless of this fact, their aluminium content varied between 200 µg/l in Keuper regions and 450 µg/l in gneiss rocks. On an average its aluminium concentration amounted 284 µg/l, its median aluminium content arrived 215 µg/l, with a minimal value of 35 µg/l and maximal value of 1249 µg/l. The limit of 250 µg_Al/l was exceeded in 55% of the samples. Well water delivered on an average 196 µg_Al/l, drinking water from the water pipe 292 µg/l. After reunification of Germany the aluminium content of the drinking water decreased in East Germany to ~50 µg/l. The German «drinking-water order» allow 200 µg/l (Höll, 2002).

Lemonade, schnapps and brandy store ~500 µg_Al/l. Beer and coke accumulate a little more aluminium. Juice, champagne, red and white wine are with 2500 µg_Al/l richer in aluminium.

The transfer of aluminium from coffee and tea in the beverage is quite different (Table 20). Coffee delivers only 19 mg_Al/kg on the beverage, where only ~ 3% of this amount is transferred in the beverage (77 µg/100 ml). Peppermint tea and fruit tea contain much more aluminium, but only 1 to 6% of this aluminium is transported into the tea. Black tea is with 900 mg_Al/kg DM much richer in aluminium and its transfer rate is with ~ 27% much higher. Black tea delivers an average 4190 µg_Al/l to the consumer (Anke et al., 2001; Höll, 2002; Muller, 2007).

Table 18. ***The aluminium content of drinking water in East Germany and Germany (µg/l) (n = 151)***

Geological origin	µg/l	%
Keuper (6)	447 ± 306	100
Phyllite (7)	350 ± 399	78
Bunter (6)	317 ± 223	71
Granite (7)	315 ± 317	71
Boulder clay (31)	307 ± 226	69
Loess (20)	288 ± 160	64
Muschelkalk (8)	258 ± 159	58
Pleistocene sand (56)	245 ± 139	55
Gneiss (10)	204 ± 59	46
Fp*	> 0.05	
Average	284 ± 200	
Median	215	
Minimum	35	
Maximum	1249	
Limit exceeding %	55	
Well	196 ± 69	
Water pipe	292 ± 207	
Water pipe after reunification	48 ± 14	

*Fp = significance level in one- or multifactorial variance level analysis.

On an average, people with a mixed diet get 70% of their daily aluminium intake from vegetable food, 21% from animal food and 9% from drinks. Among vegetable foods, vegetables contribute about 27%, bread, cakes and pastries 20%, and fruit 10% of the aluminium consumption (Anke, 2004 c).

6. ALUMINIUM INTAKE OF HUMAN ADULTS

The aluminium intake of adults in Germany was investigated by means of the double portion technique. The experiments were carried out with 13 test teams about seven subsequent days at least with 7 women and 7 men of omnivores in an age of 20 to 69 years. The 14th test team of 10 women and 10 men consist of ovo-lacto vegetarians. All of them collected a visually estimated duplicate of their daily meals including all beverages and snacks and they wrote a record of their consumption. A total of 1456 duplicates were available for analysis. Medications were recorded but not added to the duplicates.

The systematic analysis of the aluminium intake revealed that the aluminium intake after reunification dropped to about one half of that before reunification (Table 21).

Table 19. ***The aluminium contents of beverages***

Beverage	Al (mg/l)		
	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	2.4	2.2	0.5–8.2
Champagne	2.5	1.2	1.0–5.1
Red wine	2.6	0.8	1.5–3.8
White wine	2.6	1.5	0.5–4.6

Table 20. ***Transfer of aluminium from coffee and several kinds of tea in the beverages***

Kind of beverage	µg Al per tea bag	µg Al per 100 ml	Transfer rate%	Multiplication of Al in 100 ml
Coffee	105	7.7	2.8	1.0
Peppermint tea	506	13.5	1.3	1.8
Fruit tea	603	42.1	6.2	5.5
Black tea	1543	419.0	27.0	54

Table 21. ***Aluminium intake of German adults depending on type of diet and sex (mg/day) (n = 1456)***

Type of diet, reunification (n)		Women		Men		Fp P	%
		s	x	x	s		
Mixed diet (md)	before reunification	3.5	5.4	6.5	3.6	< 0.01	120
	after (1) reunification	3.1	4.6	4.9	3.5		107
	after (2) reunification	1.9	3.1	3.2	2.2		103
Vegetarian (v)	after reunification (2)	3.2	4.1	4.1	1.9	> 0.05	100
Fp	md before: after (2) **	< 0.001				—	
p	v: after (2) ***	< 0.001					
%	md before: after (2)	57		49			
	v: md after (2)	132		128			

* Women = 100%, men = x%; ** Before = 100%, after (2), 1996 = x%; *** Mixed diet after (2) = 100%, vegetarians = x%.

The significant decrease in aluminium intake seems to be due to the reduction of sulphur emission, which lowers soil pH value, as well as to the more thorough cleaning of vegetables, fruits, spices and the more aluminium poor drinking water. As expected, ovo-lacto vegetarians consumed about 30% more aluminium than persons living on a mixed died.

Apart from time (before and after reunification) and the type of diet, aluminium consumption was shown to vary also with sex, though less than expected. On average, males with mixed diet consume much more dry matters than their female counterparts. This considerable difference between the sexes did not alter the aluminium intake figures, as most women consume more vegetables and with them more aluminium than do most men. The geological origin of living area of the subjects influences the aluminium intake. Persons living in Thuringia consumed highest aluminium amounts. The reason for that could be the higher aluminium content of their local drinking water.

The men with mixed and vegetarian diet consumed dry matter, really poorer in aluminium than the women (Table 22). They prefer aluminium-poor diet. The difference is significant. Usually, the difference between women and men in the intake of macro, trace and ultratrace elements disappears after relation of the intake to the consumed dry matter.

The aluminium intake possesses no risk to humans with healthy kidneys. It has been provisionally suggested that tolerable daily intake on the average of a week may be approximately 1 mg/kg body weight.

Table 23 shows that the daily intake of both sexes varied between 42 and 89 µg_Al/kg body weight. Thus, the nutritional intake of aluminium possesses no toxicological risk to healthy subjects (Muller, 1995a, Anke et al., 2001).

The calculation of aluminium intake in humans by the basket method overestimates the aluminium intake on average about 10 to 20% (Table 24). This Method should not longer been used (Anke, 2004).

CONCLUSION

The provisional tolerable weekly intake (PTWI) of aluminium, according to the FAO/WHO, is 7 mg/kg body weight per day. In Germany, on average of a week, the daily aluminium intake is only about 5% of the PTWI value (Anonymous, 1989, 1996). In the number States of European Union, the limit value for aluminium in drinking water has been set at 200 µg/l. In Germany, the use of aluminium-containing food additives is permitted for definite and specific purposes only, mostly to a limited extent.

The amount of aluminium taken by individuals as aluminium containing pharmaceutical preparations (antacid, analgesics, anti-ulcerative) reported to be 126-5000 mg/day (Schafer, Anke, 2006; Schafer et al, 2006).

Table 22. ***The aluminium concentration of the dry matter consumed by adult Germans with mixed and ovo-lacto vegetarian diet (mg/kg DM)***

Type of diet, year (n)		Women		Men		Fp P	%
		s	x	x	s		
Mixed diet (md)	before reunification	12	18	17	9.5	< 0.01	94
	after reunification (1)	10	15	13	8.5		87
	after reunification (2)	5.6	10	8.3	5.2		83
Vegetarian (v)	after reunification (2)	8.3	11	9.1	4.3	< 0.01	83
Fp	md before: after (2)	< 0.001				—	
p	v: after (2)	> 0.05					
%	md before: after (2)	56		49			
	v: md after (2)	110		110			

Table 23. ***Aluminium intake per kg body weight depending on sex, time und type of diet (µg/day/kg body weight)***

Type of diet (n)		Women		Men		Fp P	%
		s	x	x	s		
Mixed diet (md)	before reunification	54	81	89	51	> 0.05	110
	after reunification (1)	57	74	62	42		84
	after reunification (2)	32	47	42	30		89
Vegetarian (v)	after reunification	47	70	60	27	> 0.05	88
Fp	md before: after (2)	< 0.001				—	
p	v: after (2)	< 0.001					
%	md before: after (2)	58		47			
	v: md after (2)	149		143			

Table 24. ***The aluminium intake measured with the duplicate method and calculated with the basket method (mg/day)***

Method	Before reunification		After reunification		Al mg/day		%	
	Women	Men	Women	Men	Women	Men	Women	Men
Duplicate	5.4	6.5	4.6	4.9	5.00	5.70	100	114
Basket	5.8	7.4	5.2	6.3	5.50	6.85	100	125
Overestimation,%	107	114	113	129	110	120	—	

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