Minerals and trace elements in total diets in The Netherlands

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1. During a period of 2·5 years, every 3 months 221 different food items forming a 'market basket' were purchased, prepared and divided into twenty-three food-commodity groups. The 'market basket' was based on a study of the dietary intake of 18-year-old male, Dutch adolescents. In the (homogenized) food groups various minerals and trace elements were determined.

2. The mean daily amounts of cadmium (21 μg), mercury (0·7 μg), lead (32 μg), tin (0·65 mg), arsenic (38 μg) and bromine (8 mg) in the diet of adolescents, as calculated from the concentrations analysed in the food groups, were all (well) below the Food and Agriculture Organization/World Health Organization acceptable daily intake (ADI) value (Codex Alimentarius Commission, 1984).

3. The mean daily amounts of zinc (14 mg) and selenium (72 μg) seemed to be adequate compared with the Dutch recommendations, (Voedingsraad (Dutch Nutrition Council) 1986). The amounts of copper (mean value 1·5 mg/d) and iron (mean value 14 mg/d) in the total diet were marginal. Contents of calcium (1340 mg/d), magnesium (433 mg/d) and iodine (402 μg/d) were all well above the Dutch recommendations for male adolescents. Sodium intake, corresponding to 11 g sodium chloride/d, was higher than advised.

4. It is concluded that the amounts of As, Br and toxic heavy metals in Dutch total-diet samples of male adolescents are of little concern as regards health aspects. Among the essential trace elements, Fe and Cu contents seem to be marginal. Some concern regarding the salt content is indicated.

In the period 1976–8 the first total-diet study in The Netherlands with the 'market basket' approach was carried out (van Dokkum *et al.* 1982; de Vos *et al.* 1984). The aim of total-diet studies is to monitor the exposure to additives and contaminants through habitual diets and to estimate the health risk for the consumer, by comparing the actual, analysed contents with the acceptable daily intake (ADI) as established by the Food and Agriculture Organization/World Health Organization (FAO/WHO). Total-diet studies as defined and recommended by the FAO/WHO (WHO, 1976, 1985) have been carried out for many years in the USA (Johnson *et al.* 1984), the UK (Peattie *et al.* 1983) and other countries (Gorchev & Jelinek, 1985). Total-diet studies are also suitable for evaluating the nutritional quality of 'national' diets.

Results of the first study indicated that for most chemicals the concentrations found were below the Dutch tolerance limits. For practically all chemicals examined the mean and the maximum contents were well below the ADI. In view of the consumer's concern about food products and possible health risks, it is of importance to (national) governments to be able to guarantee and visualize the safety of the diet. Through a regular monitoring system it is possible to detect trends in the concentrations of significant food components in total diets.

In the present paper we report results of the second total-diet study in The Netherlands carried out in 1984–6, regarding heavy metals, minerals and trace elements.

METHODS

In the present study the approach of a 'market basket' survey was followed, based on the dietary intake by a defined population group. The composition of the diet analysed comprised the average total diet of 18-year-old males. This group probably has the highest food consumption compared with other age-categories and consequently is likely to have the highest intake of contaminants such as heavy metals.

Table 1. *Composition (g/d) of the 'market basket' constituting the total diet of 18-year-old Dutch males*

Group no. *	Item	1984–6 Study Main group	1984–6 Study Subgroup	1976–8 Study Main group
1	Cereal products	329	—	331
1A	Bread	—	249	—
1B	Biscuits	—	48	—
1C	Rice, macaroni, etc	—	32	—
2	Potatoes and potato products	260	—	229
2A	Potatoes	—	230	—
2B	Potato products	—	30	—
3	Vegetables	210	—	159
3A	Leafy vegetables	—	61	—
3B	Other vegetables	—	33	—
3C	Soups	—	116	—
4	Root vegetables	16	—	28
5	Legumes	18	—	22
6	Fruits	256	—	195
6A	Fresh fruits	—	130	—
6B	Canned fruits and fruit juices	—	126	—
7	Meat, poultry, eggs	141	—	159
7A	Meat and meat products	—	108	—
7B	Poultry and eggs	—	33	—
8	Fish	9	—	10
9	Milk and dairy products	602	—	636
9A	Milk	—	316	—
9B	Milk products and dairy products	—	286	—
10	Oils and fats	75	—	51
10A	Butter, margarine, oils	—	64	—
10B	Nuts	—	11	—
11	Sugar and sweets	78	—	81
12	Drinks and drinking water	1617	—	1642
12A	Drinks	—	1257	—
12B	Drinking water	—	360	—
Miscellaneous group (noodles, pizza, etc.)		20	—	—

* The 1976–8 'market basket' consisted of twelve food groups (no subdivision was made).

In September–October 1982 and January–February 1983 the food consumption of 18-year-old males was surveyed. Thus, 187 males were randomly selected from a group of about 2000 who underwent a medical examination for entering military service. Of these 187 males, 72% were students, 14% were employed and 14% were looking for a job.

The survey was carried out using a diet-history method with emphasis on the food consumption over the 'previous' 14 d. In total, 426 food products were mentioned by the 187 males. For each product the average daily consumption was calculated (g/d or ml/d) as well as the relative contribution to the total weight and energy content of the diet. Only food items that contributed by more than 0·2 g/kg to the total diet were incorporated in the final 'market basket'. Thus, various products were aggregated into groups of similar food items on the basis of food type and nutrient content.

Ultimately, 221 food products formed the 'market basket' of the present study. These products contributed approximately 98% of the weight and energy contents of the average total diet of male adolescents.

During a period of 2·5 years (1984–6), every 3 months the complete set of (221) foods was

Table 2. *Gross composition of the market basket of the 1984–6 total-diet study of 18-year-old Dutch males*

Energy (MJ/d)	13·2
Percentage of total energy:	
Protein	13·7
Fat	35·7
Total available carbohydrates	46·2
Alcohol	4·4
Dietary fibre (g/d)	24·5

purchased in Zeist, a town of 60000 inhabitants in the centre of The Netherlands. The sites of purchase were varied, a wide variety of product brands was chosen and seasonal variation in the consumption of fruits and vegetables was taken into account. In this way ten samplings ('market baskets') were collected in the period mentioned.

The various food items were prepared in the way they would normally be served and eaten. Cleaning and cooking were carried out according to standard procedures and supervised by a dietician. After preparation the foods were combined into twenty-three commodity classes, representing the basic 2-week diet of 18-year-old males. Foods in each of these twenty-three groups were homogenized and frozen at −20 °. Samples from each mixture were analysed for the presence of additives, contaminants and some food components of nutritional importance. Table 1 shows the composition of the 'market basket' divided into twenty-three commodity groups.

The food consumption survey of the 1976–8 study was carried out in a similar way, except that the foods were classified into twelve commodity groups which were not subdivided (see Table 1).

The main difference in food consumption between the 1976–8 males and by those from the 1984–6 study pertain to the increased amounts of potato (products), fruits and vegetables. Table 2 gives the gross (analysed) composition of the total diet in terms of energy percentage of protein, fat and carbohydrates. Details will be published elsewhere.

Analytical procedures

In all twenty-three food groups the following analyses were performed in duplicate.

Sodium and potassium. Samples were dry ashed at 500° and the ash was dissolved in hydrochloric acid. Both minerals were determined by atomic absorption spectrophotometry (AAS). Recovery tests yielded averages of 99% for Na and 98% for K. The detection limit for Na was 0·5 mg/kg and for K 0·1 mg/kg.

Calcium and magnesium. Samples were dry ashed according to Isaac & Johnson (1975). The ash was dissolved in HCl and both minerals were analysed by AAS. Recoveries were Ca 101%, Mg 100%; detection limits for Ca were 1 mg/kg and for Mg 0·1 mg/kg.

Zinc, iron and copper. The ashing method of Isaac & Johnson (1975) was used. After dissolving the ash in HCl, Zn and Fe were determined by AAS and Cu by flameless AAS. Recoveries were Zn 99%, Fe 101%, Cu 101%; detection limits: Zn and Cu 0·05 mg/kg, Fe 0·5 mg/kg.

Selenium. Se was determined by digesting the sample with nitric acid and perchloric acid. Se was reduced to selenite in HCl and coupled to 2,3-diaminonaphthalene at pH 1·5. The product formed was extracted with cyclohexane and the fluorescence of the complex was measured at 520 nm. Recovery was 99·5%; detection limit 10 μg/kg.

Tin. Samples were digested with sulphuric acid and HNO_3. After removal of nitrogen oxides, Sn was precipitated with an aluminium salt in an alkaline medium. The precipitate was centrifuged, washed and then dissolved in HCl. Sn was determined by alternating current polarography (Lingane, 1945). Recovery was 92%; detection limit 0·5 mg/kg.

Cadmium and lead. Cd and Pb were analysed according to Muys (1984), in which method a programmed ashing of the samples is applied. After complexing with sodium diethyl dithiocarbamate and extraction with methyl isobutyl ketone, both minerals were determined by flameless AAS. Recoveries were Cd 101%, Pb 100%. Detection limits Cd 1 μg/kg, Pb 5 μg/kg.

Mercury. Samples were digested with HNO_3 and analysed by the 'cold vapour' technique (Hatch & Ott, 1968). Recovery was 95%; detection limit 5 μg/kg.

Arsenic was analysed after wet digestion of the samples with sulphuric acid and hydrogen peroxide. Arsenic was reduced to AsH_3 and determined by AAS after passing of AsH_3 through a heated quartz cell (Fiorino *et al.* 1976). Recovery was 100%; detection limit 5 μg/kg.

Phosphate. After dry ashing of the sample and dissolving the ash in HNO_3, phosphorus was precipitated with a sulphate molybdenum reagent. The precipitated ammonium phosphomolybdate was determined gravimetrically (Schormüller, 1967). No recovery tests were carried out for P; the detection limit for P was 10 mg/kg.

Bromine and iodine. Br and I were determined by neutron-activation analysis (Tjioe *et al.* 1977; Cornelis & Hoste, 1973). Detection limits were Br 0·1 mg/kg, I 0·5 μg/kg.

RESULTS

Table 3 shows the contents of As, Br and the heavy metals (Cd, Hg, Pb and Sn) in the total diets of 18-year-old males. Both mean and median values are given, as well as the range. In calculating intake values, a content was assumed to be zero when a compound was reported as not detectable. The values are compared with the 1976–8 total-diet study (van Dokkum *et al.* 1982; de Vos *et al.* 1984) and the ADI for the various chemicals as calculated from the provisional tolerable weekly intake (Codex Alimentarius Commission, 1984) is provided as well. The contents of the essential elements are presented in a similar way in Table 4; the provisional recommendations for male adolescents are given instead of the ADI.

The highest contents of As were found in the group 'fish' (mean value 2050 μg/kg) and very low contents were detected in all other food groups. Br and Pb were more evenly distributed over the various food groups; the group 'nuts' showed the highest content of Br (about 8 mg/kg), but for Pb such a dominant group could not be indicated. Tin could almost only be detected in the group 'canned fruits' (mean content 4 mg/kg). Cd levels of 40 μg/kg were found in the groups 'bread', 'potato products' and 'nuts'. Hg could only be detected in the group 'fish' (mean content 80 μg/kg).

The highest contents of Na were found in the groups 'bread' (5·6 g/kg), 'meat and meat products' (7·8 g/kg) and 'fish' (5·0 g/kg). High contents of K were observed in the groups 'potato products' (7·4 g/kg) and 'nuts' (5·6 g/kg). High values for Ca in 'milk' (mean content 1180 mg/kg) and in 'milk products and dairy products' (2010 mg/kg) were not surprising. The content of Mg in 'nuts' (mean value 1500 mg/kg) was much higher than that in other food groups. Also for P, 'nuts' showed the highest values (mean 2·9 g/kg). The highest content of I was found in the group 'fish' (2110 μg/kg); high contents of I were also found in the groups 'sugar and sweets' (mean value 1400 μg/kg) and 'bread' (mean 650 μg/kg). It is possible that the high amount of I found in the group 'sugar and sweets' can be accounted for by the use in certain confectioneries of erythrosine, a colouring agent containing 57% iodine. High contents of Fe (15–20 mg/kg) were found in various food groups. 'Nuts' contained more Cu (4·5 mg/kg) than other groups. The groups 'nuts' and 'meat and meat products' showed the highest values for Zn (25 mg/kg). High Se values

Table 3. *Heavy metals, arsenic and bromine (μg/d) in the total diets of 18-year-old Dutch males*

Element	1984–6 Mean	Median	Range	1976–8 Mean	ADI*
Arsenic	37·8	37·5	20·6–70·1	15	120
Bromine	7800	8000	6000–8710	9400	60000
Cadmium	20·8	19·5	16·5–32·6	20	60–70
Mercury	0·72	0·60	0·43–1·44	5	43
Lead	31·8	30·7	25·7–40·9	ND	430
Tin	650	530	0–1840	1700	120000

ADI, acceptable daily intake; ND, not determined.
* For a body-weight of 60 kg (calculated from the FAO/WHO provisional tolerable weekly intake) (Codex Alimentarius Commission, 1984).

Table 4. *Minerals and essential trace elements (mg/d) in the total diets of 18-year-old Dutch males*

Element	1984–6 Mean	Median	Range	1976–8 Mean	RDA*
Calcium	1340	1320	1260–1490	ND	900–1200
Copper	1·5	1·5	1·3–1·7	ND	1·5–3·5
Iron	14·2	14·1	12·6–15·8	ND	15
Iodine	0·402	0·318	0·253–0·875	0·210	0·150
Potassium	4370	4340	4030–4640	ND	–
Magnesium	433	435	397–452	ND	300–350
Sodium	4420	4390	4310–4600	4200	–
Phosphorus	2010	2030	1870–2090	1860	900–1800
Selenium	0·072	0·069	0·047–0·109	0·078	0·050–0·140
Zinc	13·7	13·5	12·9–14·8	ND	7–10

RDA, recommended dietary allowance; ND, not determined. * Provisional Dutch recommendations for male adolescents (Voedingsraad (Dutch Nutrition Council), 1986).

were observed in the groups 'nuts' (370 μg/kg), 'fish' (320 μg/kg), 'meat and meat products' (135 μg/kg) and 'poultry and eggs' (190 μg/kg).

High concentrations, however, do not necessarily result in high daily intake values as can be concluded from the values observed in the food groups 'nuts' and 'fish': for both food groups the average daily consumption by male adolescents was low (about 10 g/d, see Table 1), so that these foods contributed only weakly to the mean daily intake of most of the metals and trace elements analysed.

In Table 5, for each element the three food groups are listed that contribute most to the average daily amounts in the total diet of our population group. Values higher than 50% were found for Sn ('canned fruits' 82%), for As (52% by 'fish') and Hg (100% by 'fish').

When the daily amounts of various minerals and trace elements consumed are compared with either the ADI or the recommended daily allowances (RDA), it can be concluded from Table 3 that all analysed toxic elements are (well) below the ADI values. Table 4 shows that the recommendations for most essential minerals and trace elements are met, but that the calculated daily values of Cu and Fe are on the lower side of the (Dutch) RDA for male adolescents.

Table 5. *Food groups contributing most to the total daily amounts of toxic and essential metals and trace elements in total diets of 18-year-old Dutch males*

Element	Group	%	Group	%	Group	%
Arsenic	Fish	52	Drinks	12	Bread	8
Bromine	Milk	26	Milk and dairy products	19	Bread	13
Calcium	Milk and dairy products	43	Milk	28	Bread	6
Cadmium	Bread	42	Potatoes	20	Leafy vegetables	6
Copper	Bread	24	Potatoes	13	Drinks	11
Iron	Bread	28	Meat and meat products	16	Potatoes	9
Mercury	Fish	100	–		–	
Iodine	Bread	41	Sugar and sweets	27	Milk and dairy products	5
Potassium	Potatoes	17	Bread	12	Milk	11
Magnesium	Bread	24	Drinks	13	Milk and dairy products	11
Sodium	Bread	32	Meat and meat products	22	Milk and dairy products	10
Phosphorus	Milk and dairy products	22	Bread	20	Milk	15
Lead	Bread	18	Milk and dairy products	11	Canned fruits	11
Selenium	Bread	30	Meat and meat products	23	Poultry and eggs	8
Tin	Canned fruits	82	Milk and dairy products	9	Other vegetables	4
Zinc	Meat and meat products	24	Bread	19	Milk and dairy products	18

DISCUSSION

The total daily amounts of the toxic elements analysed were found to be well below the ADI. Even the maximum value was not alarming. Compared with the first total-diet study, the daily amounts of Cd and Br had remained approximately constant, the decrease of the 'intake' of Sn and Hg was apparent and the increase of the daily amount of As resulted in values that were still (well) below the ADI. Interestingly the Br content in the group 'cereal products' (4·2 mg/kg) was much lower than the Br content in the same group analysed in the first total-diet study (12·7 mg/kg). This difference can be ascribed to the prohibition of potassium bromate for bread baking, before the 1984–6 total-diet study was carried out. In Table 6 the mean daily intake of some toxic metals in various countries is summarized. It should be mentioned that these values concern total diets of diverging age-categories and that the samples were collected by different methods ('market basket' *v.* duplicate portion). It is, however, striking that for As, Hg, Pb and Cd the conclusions were similar in all studies in that total daily exposure was well below the ADI.

From the results obtained it can be concluded that for male adolescents the amounts of Ca, Zn, P, Mg, I and Se in the total diet can be considered acceptable from a nutritional point of view. The amounts of Cu and Fe found were on the low side of the recommendations. The relatively low intake of Cu has also been observed in other countries (Table 6). The minimum values in our study indicate that the intake of both Cu and Fe is a source of concern. It should be kept in mind that the recommendations for certain nutrients may differ from country to country. This is well illustrated for Zn: the amount

Table 6. *Mean daily intake of some toxic and essential metals and trace elements in various countries*

Element	The Netherlands*	UK†	USA‡	USA§	Finland‖	Sweden¶	Belgium**	Canada††
Arsenic (μg)	38	129	65	ND	ND	ND	45	17
Cadmium (μg)	21	20	33	ND	13	10	18	14
Copper (mg)	1·5	1·5	ND	1·2	1·7	1·5	1·4	ND
Mercury (μg)	0·7	ND	5	ND	ND	ND	13·5	ND
Iron (mg)	14	11	ND	17	ND	14	ND	ND
Lead (μg)	32	115	82	ND	66	27	179	54
Selenium (μg)	72	60	138	90	ND	ND	ND	ND
Zinc (mg)	14	9	18	16	16	9	15	ND

ND, not determined.
* Present study ('market basket' for male adolescents).
† Hazell, 1985 (adults).
‡ Gartrell *et al.* 1985 ('market basket' for 16 to 19-year-old males).
§ Pennington *et al.* 1986 ('market basket' for 14 to 16-year-old boys).
‖ Varo & Kovistoinen, 1980 (adults).
¶ Slorach *et al.* 1983 (duplicate diets, adults).
** Buchet *et al.* 1983 (duplicate diets, adults).
†† Dabeka *et al.* 1987 (duplicate diets, adults).

found in our study can be considered adequate when the Dutch recommendations are applied, but according to the USA recommended intake (National Academy of Sciences, 1980) the diet has to be judged marginal with respect to the Zn intake. The basis for an established RDA is not always certain and the consequences for nutritional health when the RDA is not met are not well known. Moreover, for a number of nutrients the assessment of the 'status' is hampered because no suitable indices exist for measuring body stores.

The bioavailability and speciation of minerals and trace elements are other factors to be taken into account in the evaluation of the results. Details on this matter, however, are beyond the scope of the present study.

The total amount of Na in the total diet of male adolescents was similar to the level found in our first study, corresponding to approximately 11 g NaCl. The Voedingsraad (Dutch Nutrition Council) (1986) recommends that individual NaCl consumption should not exceed the current (Dutch) average consumption level of 9 g/d. Discretionary salt was not included in our study, so that the level of salt 'intake' was underestimated. Some concern may be expressed regarding the amount of Na in the total diet of male adolescents. Reduction of salt intake is therefore indicated, since excessive Na intake is considered to be an aetiological factor in hypertension.

The amount of P in the analysed total diet was at least adequate; the amount found was well below the ADI of 4200 mg. A decrease of P intake may be advisable since a Ca:P value of 1 is recommended by the (US) National Academy of Sciences (1980).

Although the mean Se amount in the total diet meets the recommendation, an ample supply of this essential trace element may lower the risk of development of certain types of cancer (Vernie, 1984). A low Se intake is also associated with a higher risk of cardiovascular diseases (Robinson & Thomson, 1983).

In conclusion, the amounts of As, Br and toxic trace elements found in total diets of male adolescents are not a source of concern. Most essential minerals and trace elements in the analysed total diets were present in amounts exceeding the recommendations. Only for Cu and Fe can the situation be evaluated as less desirable. The daily amount of Na intake may be considered too high.

The food intake of the group of male adolescents forming the basis of our total-diet study was relatively high, so that the exposure to additives and contaminants was also relatively high. For people in other age- and sex-categories, who may consume less food, the risk of intake of toxic chemicals may be lower, but the intake of essential minerals and trace elements may be below recommended levels. It should also be mentioned that the conclusions of our study are not applicable to groups or individuals with a more extreme dietary pattern (e.g. a high intake of fish or fish products).

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