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Dietary intake and risk assessment of elements for 1- and 2-year-old children in the Netherlands

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ABSTRACT

In 2017, a total diet study (TDS) was conducted in the Netherlands to determine the intake of elements by 1- and 2-year-old children. Concentrations of 47 elements were analysed and long-term dietary intake was calculated for 24 elements. The 95th percentile (P95) intake estimates were compared with a tolerable daily or weekly intake (TDI or TWI) or tolerable upper intake level (UL), or a margin of exposure (MOE) was calculated. The P95 intake of cadmium and zinc exceeded the TWI or UL, respectively, and the P95 intake of inorganic arsenic and lead resulted in low MOEs. Food subgroups contributing most to the intake were “potatoes” for cadmium, “milk and milk-based beverages” for zinc, “concentrated fruit juices” and “rice” for inorganic arsenic, and “candies” for lead. For inorganic mercury, it could not be established if the intake was (too) high. P95 intake estimates of the other elements for which a risk characterisation could be performed were below the health-based guidance values. It was noted that the P50 intake estimate of manganese was a factor of 3 higher than the adequate intake level. Due to the absence of a UL, it is not clear if this intake is of concern.

1. Introduction

Metals and other elements, hereafter referred to as “elements”, are present in foods and beverages, due to their occurrence in soil and water. Their occurrence can be due to natural occurrence and anthropogenic sources, such as contamination by industrial and agricultural practices. Furthermore, elements may be present in foods and beverages due to migration from food contact materials or contamination during processing. When ingested, elements may cause adverse health effects. For example, lead can affect neurodevelopment of young children, resulting in a decrease in Intelligence Quotient (EFSA, 2010). Cadmium is known to accumulate in the kidney and may cause kidney failure at the age of 50 years or older (EFSA, 2009b, 2011a). On the other hand, some elements are essential for normal growth, development and health. Examples of such elements are copper, iron, magnesium and zinc (EFSA, 2014b, 2015a; b; c).

To assess whether the dietary intake of elements may pose a health risk, intake levels are compared to intake levels below which it is unlikely that there is a risk of adverse health effects in humans. Examples of such intake levels are tolerable daily or weekly intakes (TDIs or TWIs), and tolerable upper intake levels (ULs). When no such intake levels can be defined, a margin of exposure approach based on the lower limit of a benchmark dose (BMDL) may be used to assess a possible health risk. BMDLs are doses in toxicity studies at which a percentage (e.g., 1%, 5% or 10%) increase in an adverse effect is observed. In the European Union, TDIs, TWIs, BMDLs and ULs are established by the European Food Safety Authority (EFSA). ULs are usually established for elements that are also nutrients, such as copper, molybdenum and zinc (EFSA, 2006, 2018b, 2021b).

In 2018, food consumption data of the Dutch National Food Consumption Survey (DNFCS) of 2012–2016 among persons 1–79 years of age were published (van Rossum et al., 2020). Halfway through this survey, consumption data became available (van Rossum et al., 2016), which were used to set up a total diet study (TDS) covering foods and beverages consumed by children 1 and 2 years of age. This age group was selected, because little is known about the dietary intake of chemicals in this age group. Furthermore, young children consume higher amounts of foods and beverages per kilogram (kg) body weight than adults and may thus ingest higher amounts of food chemicals per kg body weight.

The aim of the TDS was to estimate the long-term dietary intake of protein, fat, mycotoxins, and elements for 1- and 2-year-old children. In

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a TDS, chemicals are analysed in foods and beverages prepared for consumption. A TDS is, therefore, considered 'to provide a solid basis for calculating population dietary exposure and assessing potential impact on public health' (EFSA, 2011b). TDSs have been performed in different countries, resulting in long-term intake estimates of a diverse range of chemicals that can be present in foods and beverages, such as mycotoxins, elements, and environmental or processing contaminants (Arnich et al., 2012; FSAI, 2016; Gaoa et al., 2016; Jean et al., 2018; Kolbaum et al., 2019; Sirot et al., 2013, 2018; Sprong et al., 2016).

The long-term dietary intake of protein, fat and mycotoxins was published elsewhere (Pustjens et al., 2021a; b). This paper reports on the long-term dietary intake of elements of 1- and 2-year-olds in the Netherlands. The intake estimates of elements for which a TDI, TWI, BMDL or UL has been derived were compared to these reference values to assess potential health risks.

2. Materials and methods

2.1. Total diet study (TDS)

The design of the TDS is described in detail by Pustjens et al. (2021a). Briefly, the TDS was based on individual food consumption data of 232 children aged 1 and 2 years that were collected during the first two years of the DNFCS of 2012–2016 on two non-consecutive days (van Rossum et al., 2016). A total of 1942 foods and beverages were grouped in 164 composite samples for three age groups: 12–17 months, 18–23 months, and 24–35 months. Foods and beverages were grouped for each age group to consider differences in both the amounts of and actual foods and beverages consumed. For example, children aged 12–17 months eat more boiled potatoes, whereas older children eat more baked potatoes and French fries (Pustjens et al., 2021a). The foods and beverages covered 96–98% of the total consumed amounts of all foods and beverages in these three age groups. The composite samples were grouped in 18 food groups and 59 food subgroups (Table 1).

Foods and beverages were prepared based on the information available in the DNFCS regarding how foods were consumed, such as cooked, fried or peeled. For the preparation, instructions on the label were applied or a standard cookbook was used, if relevant. Prepared or raw/fresh foods and beverages were pooled into the composite samples for the three age groups according to the consumed amounts, based on weight. Liquid composite samples were frozen immediately after pooling. All composite samples were stored at −20 °C until analysis.

Table 1
Food groups and food subgroups included in the total diet study.

Food group	Food subgroup[a,b]
Cereals and cereal-based products	Breads, breakfast cereals, crackers, pasta, porridge, rice
Children's meals	Children's meals
Confectionery	Biscuits, cakes, candies, chocolates
Dairy products	Cheeses, creams and ice creams, milk and milk-based beverages, yoghurts and other dairy products
Eggs	Eggs
Fish and shellfish	Fish and shellfish
Fruit	Apple, apple sauce, banana, children's fruits, citrus fruits, dried fruits, other fruits-1[c], other fruits-2[d]
Follow-on formula	Follow-on formula
Legumes	Legumes
Meat	Beef, chicken, meat on bread, offal, pork, sausages, sausages on bread
Non-alcoholic beverages	Apple juice, concentrated fruit juices, other juices, soft drinks, syrups, tea, drinking water
Nuts and seeds	Nuts and seeds
Oils and fats	Deep-frying fat[e], margarines, oils
Potatoes	Potatoes
Sauces	Sauces
Savoury snacks	Savoury snacks
Soy products	Soy products
Vegetables	Brassica vegetables, onion and leek, fruiting vegetables, leafy vegetables, mixed vegetables, mushrooms, other vegetables, root vegetables, stem vegetables, tomatoes and tomato products

[a] For more details about the foods and beverages sampled per food subgroup, see Pustjens et al. (2021a).

[b] Per food subgroup, including the food groups without a subgroup, one composite sample was prepared for each of the three age groups, except for apple juice, banana, beef, concentrated fruit juice, deep-frying fat, and mushrooms. For these food subgroups, one combined composite sample was prepared.

[c] Grape, kiwi and pear.

[d] Black/blue berries, kaki, mango, peach, pineapple, strawberry and watermelon.

[e] This oil was used to deep-fry foods.

2.2. Element analysis

A total of 47 elements were analysed in all composite samples using inductively coupled plasma-mass spectrometry (ICP-MS). Using this multi-compound approach, total element concentrations were determined. Additionally, inorganic arsenic was analysed in the composite samples for "fish and shellfish" and "rice" using high performance liquid chromatography ICP-MS (HPLC-ICP-MS). Inorganic arsenic is the most relevant toxic form of arsenic (EFSA, 2009a). A detailed description of the analytical methods can be found in Supplementary Material Text S1 and the ICP-MS settings can be found in Table S1. The limits of detection (LODs) were calculated for each element and each sample separately based on the fresh weight of the sample and, where applicable, the drying factor. The LODs ranged from 0.05 to 120 μg/kg (Table 2).

2.3. Element concentrations used in dietary intake calculations

Table 2 provides an overview of the elements analysed in the composite samples. The elements were divided in two groups: elements with concentrations analysed above the LOD in at least six composite samples and elements with concentrations analysed above the LOD in five or less composite samples. The dietary intake was only calculated for the first group of elements. The number of six samples was arbitrarily chosen as the minimal number of samples that could still provide a meaningful dietary intake estimate.

Concentrations analysed above the LOD were used as such in the dietary intake calculations. These concentrations were also above the limit of quantification (LOQ), except for 27 composite samples analysed for arsenic (n=3), barium (n = 13), caesium (n = 1), iron (n = 3), molybdenum (n = 3), strontium (n = 3) and yttrium (n = 1). These samples had a concentration between the LOD and the LOQ. These concentrations and those reported at a value below the LOD are uncertain. This uncertainty was addressed by including these concentrations according to two scenarios in the dietary intake calculations: (1) concentrations below the LOD are equal to zero and those between the LOD and the LOQ are equal to the LOD (lower-bound (LB) scenario), and (2) concentrations below the LOD are equal to the LOD and those between the LOD and the LOQ are equal to the LOQ (upper-bound (UB) scenario). The resulting LB and UB intake estimates represent the lowest and highest estimates of intake, respectively, based on the analysed concentrations.

To estimate the intake of inorganic arsenic based on total arsenic concentrations in the composite samples, except for "fish and shellfish" and "rice" (see section 2.2), it was assumed that 70% of total arsenic was inorganic arsenic (EFSA, 2014a). For total mercury, the relevant forms for risk assessment are inorganic mercury and methylmercury. To estimate the intake of inorganic mercury, total mercury concentrations in all composite samples were considered to equal inorganic mercury concentrations, except for "fish and shellfish". Total mercury concentrations in this food subgroup were assumed to refer for 20% to inorganic mercury (EFSA, 2012d). Methylmercury was considered to be only present in "fish and shellfish". Therefore, total mercury analysed in this

Table 2
Limits of detection (LODs), and number and percentage of samples above the LOD for each element[a].

Element (abbreviation)	LOD in μg/kg fresh weight	No of samples above the LOD (%)	Element (abbreviation)	LOD (μg/kg)	Nr of samples above the LOD (%)
Elements with at least six composite samples that had a concentration above the LOD and for which dietary intake was calculated					
Arsenic (As)[b]	0.4–20	77 (50)	Lithium (Li)	10–50	47 (29)
Arsenic, inorganic[c] (iAs)	40–50	3 (50)	Magnesium (Mg)	120	163 (99)
Barium (Ba)	0.7–4	159 (97)	Manganese (Mn)	18–45	143 (89)
Bismuth (Bi)	0.1–2	10 (6)	Mercury (Hg)[b]	1–20	7 (4)
Cadmium (Cd)	0.3–2	77 (48)	Molybdenum (Mo)	3–4	142 (87)
Caesium (Cs)	0.08–1	75 (46)	Nickel (Ni)	5.5–41	76 (47)
Cerium (Ce)	0.2–4	23 (14)	Rubidium (Rb)	60–80	159 (97)
Cobalt (Co)	0.2–2	81 (49)	Strontium (Sr)	3	163 (99)
Copper (Cu)	20–24	149 (93)	Titanium (Ti)	4.8–32	124 (76)
Iron (Fe)	56–60	153 (95)	Vanadium (V)	0.2–3	76 (46)
Lanthanum (La)	0.26–5	20 (12)	Yttrium (Y)	0.1–1	15 (9)
Lead (Pb)	0.3–5	41 (25)	Zinc (Zn)	140	149 (91)
Elements with five or less composite samples that had a concentration above the LOD and for which dietary intake was not calculated					
Antimony (Sb)[d]	1.6–27	0 (0)	Neodymium (Nd)	0.1–2	0 (0)
Beryllium (Be)	10–50	1 (1)	Praseodymium (Pr)	0.1–2	0 (0)
Dysprosium (Dy)	0.05–1	1 (1)	Rhenium (Re)	0.15–3	1 (1)
Erbium (Er)	0.05–1	1 (1)	Samarium (Sm)	0.05–1	1 (1)
Europium (Eu)	0.05–1	1 (1)	Scandium (Sc)	0.6–11	0 (0)
Gadolinium (Gd)	0.05–1	1 (1)	Tellurium (Te)	0.36–7	3 (2)
Gallium (Ga)[6]	0.2–3	5 (3)	Terbium (Tb)	0.05–1	1 (1)
Germanium (Ge)	0.1–1	4 (2)	Thorium (Th)	1.7–34	0 (0)
Hafnium (Hf)	1.6–32	0 (0)	Thulium (Tm)	0.05–1	1 (1)
Holmium (Ho)	0.05–1	1 (1)	Uranium (U)	0.1–2	3 (2)
Indium (In)	0.05–1	2 (1)	Ytterbium (Yb)	0.05–1	1 (1)
Lutetium (Lu)	0.05–1	1 (1)	Zirconium (Zr)	1.6–32	5 (3)

LOD: limit of detection; No: number.

[a] A total of 164 composite samples were analysed. For eight elements, concentrations from the "water" composite samples were not included in the intake assessment (Table 3). For these elements, the percentage of composite samples with a concentration above the LOD was based on 161 samples.

[b] Refers to total arsenic and total mercury.

[c] Inorganic arsenic was analysed in the food subgroups "fish and shellfish" (n = 3) and "rice" (n = 3).

[d] Antimony could only be reliably analysed in 91 composite samples.

Table 3
LODs, total number of samples, number of samples above the LOD and mean concentrations as used in the intake calculation of eight elements analysed in drinking water[a].

Element	LOD in μg/L	No of samples	No samples > LOD (%)	Mean concentration in μg/L[b]	
				Lower bound	Upper bound
As	0.5–0.7	813	213 (26)	0.46	0.83
Cd	0.02–1.0	1314	30 (2)	0.001	0.06
Cu	0.05–1.0	1578	1572 (100)	172	173
Fe	5–15	1581	564 (36)	6.0	11
Hg	0.02–0.06	699	0 (0)	0	0.04
Mn	0.4–10	1578	27 (2)	0.04	4.5
Ni	0.5–17	1578	531 (34)	1.6	2.1
Pb	0.28–5.3	1578	591 (37)	0.55	0.86

As: arsenic (total); Cd: cadmium; Cu: copper; Fe: iron; Hg: mercury (total); LOD: limit of detection; Mn: manganese; Ni: nickel; No: number; Pb: lead.

[a] These data were obtained from the REWAB (Registration Tool Water Quality Data) database. For more details, see section 2.3.

[b] For an explanation of the calculation of mean lower-bound and upper-bound concentrations, see section 2.3.

food subgroup was equalled to methylmercury to assess the intake of this organic form of mercury (EFSA, 2012d).

Concentrations of eight elements in drinking water were taken from the REWAB (Registration Tool Water Quality Data) database covering measurements of 2017–2019 to estimate their dietary intake (Table 3). This database contains concentrations of numerous chemicals analysed in Dutch drinking water, generated as part of the Dutch Drinking Water Act and which are obtained using analytical methods that have lower LODs than those used in our study (Tables 2 and 3). For the other elements for which the dietary intake was estimated, no concentrations in drinking water were present in the REWAB database and the concentrations analysed in the TDS were used. Concentrations in the REWAB database refer to mean concentrations and are either reported as below the LOD or as positive concentrations based on the assumption that concentrations below the LOD were equal to ½ LOD (medium-bound scenario). For the dietary intake calculations, the mean concentrations in drinking water reported as below the LOD were equalled to zero in the LB scenario and to the LOD in the UB scenario. Total arsenic and total mercury concentrations in the REWAB database were considered to refer to inorganic arsenic and inorganic mercury, respectively, as both elements are mainly present in their inorganic form in drinking water (EFSA, 2012d, 2014a).

2.4. *Dietary intake calculations*

Long-term dietary intake was calculated using the Observed Individual Means (OIM) model as implemented in the calculation tool Monte Carlo Risk Assessment (MCRA) version 9.1 (https://mcra.rivm.nl/documentation). Using this model, a daily consumed amount of a food or beverage of each child was multiplied with the element concentration of the relevant composite sample resulting in an intake through each food or beverage. These intakes were summed to derive the total intake per day of each child and subsequently divided by the individual child's body weight, which was measured during a home visit. To obtain a measure for long-term intake, these daily intakes were averaged over the two days available in the DNFCS for each child, resulting in a distribution of two-day mean intake estimates expressed per kg body weight. A total of 100 distributions were generated in this way using a bootstrap approach (Efron, 1979; Efron and Tibshirani, 1993). The bootstrap is an accepted methodology to quantify sampling uncertainty in the intake distribution (EFSA, 2012b, 2018a). For this, 100 food consumption databases were generated by resampling of the original database and each was used to calculate a two-day mean intake distribution. For each distribution, the 50th (median; P50) and 95th (P95) percentiles of intake were calculated for all persons. Median of both intake percentiles and the 95th uncertainty interval around these percentiles are reported. This uncertainty interval quantifies the uncertainty in the intake percentiles due to the sample size of the food consumption database. Uncertainty due to the sample size of the concentration database could not be quantified, because only one concentration per composite sample was available.

Long-term dietary intake was calculated for the total group of 1- and 2-year-olds using the consumed amounts recorded across all four years in the DNFCS 2012–2016 (n = 440). Dietary intake was expressed in μg/kg body weight per day, except for copper, iron, magnesium, manganese, molybdenum and zinc. For these elements, the intake was expressed in mg per day as is customary for these elements (EFSA, 2013a; b, 2014b, 2015a; b; c; van Rossum et al., 2020).

Contribution of each food subgroup to the total two-day mean dietary intake distribution expressed as a percentage was also calculated for each of the 100 distributions. Mean contribution of these 100 distributions is reported.

2.5. Risk characterisation

A risk characterisation was performed for eight elements: inorganic arsenic, cadmium, copper, inorganic mercury, methylmercury, molybdenum, nickel and zinc. For these elements, a tolerable daily or weekly intake (TDI or TWI), tolerable upper intake level (UL) or lower limit of a benchmark dose (BMDL) have been established by EFSA or the FAO/WHO Joint Expert Committee on Food Additives (JECFA). The risk characterisation was performed by comparing P95 intake estimates with the TDI, TWI or UL, or by calculating a margin of exposure (MOE). MOE was calculated by dividing the BMDL of an element by its P95 intake estimate. For cadmium, inorganic mercury and methylmercury, a TWI has been derived. To compare the TWI with P95 intake estimates expressed per day, the TWI was divided by 7. For the risk characterisation of inorganic arsenic, the BMDL of 3 μg/kg body weight per day of JECFA (2011) was used instead of the BMDL of 0.3–8 μg/kg body weight per day as derived by EFSA (2009a): the BMDL of JECFA is based on studies with a relatively long follow-up and large study population.

3. Results

3.1. Concentrations of elements in composite samples

Concentrations of 24 elements were above the LOD in five or less composite samples (Table 2). Of the other elements, the percentage of composite samples with a reported concentration above LOD ranged from 4% (n = 7) for total mercury to 99% (n = 163) for magnesium and strontium. The composite sample with the highest number of concentrations above the LOD was “candies” for children 12–17 months of age. In this sample, 24 elements had a concentration above the LOD, including trace concentrations of dysprosium, gadolinium and samarium (at indicative concentrations below the LOQ of 1.1–1.4 μg/kg). This sample was followed by “leafy vegetables” for children 12–17 months and 24–35 months of age with 23 elements each, and seven composite samples, including “tomatoes and tomato products”, “stem vegetables” and “dried fruit” for the youngest age group, “leafy vegetables”, “root vegetables”, and “mixed vegetables” for the middle age group, and “mixed vegetables” for the oldest age group with 21 elements each. The percentage samples with a concentration above the LOD in drinking water ranged from 0% for total mercury to 100% for copper (Table 3).

Tables 4 and 5 list the range of mean LB and UB concentrations in the composite samples per food group and element as used in the dietary intake calculations. Table 4 lists the concentrations for the elements for which a risk characterisation was performed, and Table 5 for the elements for which this was not possible due to the absence of reference values (see section 2.5). For ten elements, the mean LB and UB concentrations did not differ within food groups (Tables 4 and 5). For these elements, the percentage of samples with a concentration above the LOD was 87% or more (Table 2). Table 3 lists the mean LB and UB concentrations of eight elements in drinking water as used in the dietary intake calculations.

Concentrations in the individual composite samples in which at least one element listed in Tables 4 and 5 was detected are presented in Table S2 of the Supplementary Material. Inorganic arsenic concentrations in the individual “fish and shellfish” and “rice” samples can be found in Table S3 of the Supplementary Material.

3.2. Dietary intake estimates and contributions of food subgroups to the intake

The LB and UB P50 and P95 intake estimates for the elements listed in Tables 4 and 5 are presented in Table 6. The LB and UB intake estimates were the same for the ten elements for which the LB and UB

Table 4

Range of mean lower-bound and upper-bound concentrations in the composite samples per food group for elements with at least six composite samples that had a concentration above the LOD and for which a risk characterisation was performed.

Food group	No of composite samples analysed	Concentration in μg per kg fresh weight for each element[a,b,c,d,e]								
		iAs	Cd	Cu	iHg	MeHg	Mo	Ni	Pb	Zn
Cereals and cereal-based products	18	16–17	19	1700	0–13	-	250	125–130	1–4	14,000
Children’s meals	3	11	7	550	0–3	-	110	67	2–2	4900
Confectionery	12	1–4	17	2100	0–18	-	100	328–351	16–18	7200
Dairy products	12	1–3	0–1	130	0–15	-	70	0–30	0–4	11,000
Eggs	3	0–1	0–1	640	0–6	-	50	0–12	0–1	16,000
Fish and shellfish	3	0–87	1–2	450	4	22	30	19–30	0–2	5000
Follow-on formula	3	0–3	0–2	480	0–20	-	20	0–41	0–5	9400
Fruits	22	5	0–1	1100	0–5	-	30	40–45	4	1400
Legumes	3	3–4	13	2100	0–6	-	910	269	3	8500
Meat	19	3–4	2–3	1300	0–9	-	80	5–23	1–3	23,000
Non-alcoholic beverages	17	1–3	0–2	170	0–16	-	10	0–34	0–4	320
Nuts	3	4–6	53	5600	0–19	-	1800	1627	0–5	31,000
Oils and fats	7	3–5	0–2	170	0–18	-	10	0–37	3–7	1300
Potatoes	3	1–2	31	1200	2–6	-	100	27–32	3–4	5200
Sauces	3	5	2	500	0–5	-	20	58	3–4	2000
Savoury snacks	3	5–10	23	1800	0–19	-	260	48–75	0–5	12,000
Soy products	3	0–3	0–2	890	0–20	-	170	237–250	0–5	2100
Vegetables	27	4	19	710	0–2	-	120	57	5	3400

iAs: inorganic arsenic; Cd: cadmium; Cu: copper; iHg: inorganic mercury; LOD: limit of detection; MeHg: methylmercury; Mo: molybdenum; Ni: nickel; No: number; Pb: lead; Zn: zinc.

[a] Ranges refer to the lowest mean lower-bound and the highest mean upper-bound concentration per food group. These concentrations were the same when only one concentration is reported. For an explanation of the calculation of mean lower-bound and upper-bound concentrations, see section 2.3.

[b] Fresh weight concentrations were recalculated from freeze-dried concentrations where applicable.

[c] Concentrations of iAs were based on the assumption that 70% of total arsenic analysed is iAs, except for “fish and shellfish”, “rice” (belonging to the food group “cereals and cereal-based products”) and “drinking water” (belonging to the food group “non-alcoholic beverages”). In these foods, iAs was measured as such or total arsenic was assumed to be 100% iAs.

[d] Concentrations of iHg were based on the assumption that 100% of total mercury analysed is iHg, except in “fish and shellfish”. In this food group, iHg concentrations were assumed to be 20% of total mercury.

[e] For MeHg, only lower-bound concentrations were calculated as it is very likely that MeHg is only present in “fish and shellfish”. Furthermore, ‘-’ means that no methylmercury was considered to be present in these samples (see section 2.3).

Table 5
Range of mean lower-bound and upper-bound concentrations in the composite samples per food group for elements with at least six composite samples that had a concentration above the LOD and for which no risk characterisation could be performed.

Food group	No of composite samples analysed	Concentration in μg per kg fresh weight for each element[a,b]														
		Ba	Bi	Cs	Ce	Co	Fe	La	Li	Mg	Mn	Rb	Sr	Ti	V	Y
Cereals and cereal-based products	18	990	0–1	1	3–5	8	21,000	3–6	6–33	427,000	9800	1800	1200	210	9–10	0–1
Children's meals	3	320	0–3	3	0–1	5	5100	0–1	11	128,000	860	870	810	80	3	0–2
Confectionery	12	790	0–2	4–5	8–10	44–45	23,000	2–6	0–44	297,000	4300	2900	1300	1200	26	0–1
Dairy products	12	290	7–8	0–1	3	0–1	750	0–4	7–37	142,000	90	1300	1000	160	1–3	0–1
Eggs	3	360	0–1	0–3	0–1	0–1	21,000	0–1	0–13	133,000	350	1800	590	170	0–1	0–3
Fish and shellfish	3	190	0–1	22	0–2	0–1	4300	0–2	16–23	270,000	1100	930	1300	400	11	0–4
Follow-on formula	3	4–5	0–2	0–1	0–4	0–2	12,000	0–5	0–50	81,000	90	570	370	0–30	0–3	0–1
Fruits	22	340	0–1	5	6–7	4	5000	4	24–30	132,000	1300	1500	1100	210	6	1
Legumes	3	400	0–1	2	0–1	15	14,000	0–2	11–24	344,000	3300	1300	2200	200	2–3	0–3
Meat	19	110	0–1	17	0–2	1–2	20,000	0–2	51–63	221,000	840	4000	340	250	6	0–5
Non-alcoholic beverages	17	130–140	0–2	2–3	0–4	4–5	1600	0–5	3–50	43,000	880	950–960	320	0–30	4–6	0–1
Nuts	3	2500	0–2	31	0–4	25	18,000	0–5	0–50	1,479,000	16,000	5800	3400	500	10–12	0–1
Oils and fats	7	9–30	0–2	0–1	0–4	0–2	330	0–5	0–46	4800	90	290–310	150–180	20–50	1–4	0–1
Potatoes	3	60	0–1	2	0–1	3	4500	0–2	0–13	231,000	1100	1000	320	60	1–2	0–3
Sauces	3	130	0–5	7	1–2	12	3100	0–1	4–14	64,000	1100	850	560	89	7	0–2
Savoury snacks	3	800	0–2	0–1	0–4	13	14,000	0–5	0–50	474,000	6700	1900	1600	250	6–8	0–1
Soy products	3	130	0–2	0–1	0–4	0–2	2800	0–5	0–50	125,000	1400	690	400	30	0–3	0–1
Vegetables	27	220	0–3	2	2	4–5	4600	1	10–14	148,000	1700	930	1400	80	3	1

Ba: barium; Bi: bismuth; Cs: caesium; Ce: cerium; Co: cobalt; Fe: iron; La: lanthanum; Li: lithium; LOD: limit of detection; Mg: magnesium; Mn: manganese; No: number; Rb: rubidium; Sr: strontium; Ti: titanium; V: vanadium; Y: yttrium.

[a] Ranges refer to the lowest mean lower-bound and the highest mean upper-bound concentration per food group. These concentrations are the same when only one concentration is reported. For an explanation of the calculation of mean lower-bound and upper-bound concentrations, see section 2.3.

[b] Fresh weight concentrations were recalculated from freeze-dried concentrations where applicable.

Table 6
Long-term lower-bound and upper-bound dietary intake of elements for children 1 and 2 years of age.

Element	Lower-bound[a,b,c]		Upper-bound[a,b]	
	P50	P95	P50	P95
Elements for which a risk characterisation was performed				
In μg/kg body weight per day				
iAs	0.10 (0.09–0.12)	0.44 (0.42–0.48)	0.31 (0.29–0.32)	0.74 (0.69–0.87)
Cd	0.31 (0.29–0.33)	0.64 (0.59–0.69)	0.43 (0.41–0.46)	0.79 (0.74–0.83)
iHg	0 (0–0)	0.05 (0.04–0.06)	1.3 (1.3–1.4)	2.2 (2.1–2.5)
MeHg	0 (0–0)	0.07 (0.05–0.09)	-	
Ni	2.4 (2.1–2.6)	6.5 (5.6–7.3)	5.0 (4.7–5.4)	9.2 (8.6–9.4)
Pb	0.09 (0.08–0.10)	0.23 (0.20–0.27)	0.41 (0.40–0.43)	0.70 (0.63–0.74)
In mg per day				
Cu	0.51 (0.49–0.53)	0.87 (0.81–0.90)	-	
Mo	0.060 (0.057–0.062)	0.12 (0.11–0.12)	-	
Zn	4.9 (4.8–5.1)	8.3 (7.9–8.7)	-	
Elements for which no risk characterisation could be performed				
In μg/kg body weight per day				
Ba	21 (19–22)	35 (33–37)	-	
Bi	0.02 (0.02–0.02)	0.09 (0.07–0.09)	0.16 (0.15–0.16)	0.27 (0.26–0.3)
Cs	0.15 (0.14–0.16)	0.64 (0.49–0.80)	0.22 (0.20–0.22)	0.68 (0.55–0.80)
Ce	0.03 (0.03–0.04)	0.11 (0.09–0.13)	0.30 (0.28–0.31)	0.51 (0.46–0.53)
Co	0.20 (0.18–0.21)	0.67 (0.61–0.84)	0.32 (0.30–0.34)	0.73 (0.67–0.90)
La	0.02 (0.02–0.02)	0.07 (0.07–0.08)	0.35 (0.34–0.37)	0.58 (0.55–0.63)
Li	0.35 (0.32–0.39)	1.4 (1.1–1.6)	3.5 (3.3–3.7)	6.0 (5.6–6.5)
Rb	117 (111–123)	218 (202–262)	-	
Sr	54 (52–56)	83 (78–87)	-	
Ti	5.6 (5.3–6.2)	14 (13–15)	7.6 (7.2–8.0)	16 (15–17)
V	0.19 (0.18–0.21)	0.49 (0.46–0.57)	0.37 (0.34–0.40)	0.69 (0.64–0.75)
Y	0.01 (0.00–0.01)	0.03 (0.02–0.02)	0.07 (0.07–0.08)	0.13 (0.12–0.14)
In mg per day				
Fe	5.6 (5.0–5.9)	10 (9.8–12)	-	
Mg	142 (140–151)	225 (212–239)	-	
Mn	1.4 (1.3–1.5)	2.4 (2.4–2.6)	-	

iAs: inorganic arsenic; Ba: barium; Bi: bismuth; Cd: cadmium; Cs: caesium; Ce: cerium; Co: cobalt; Fe: iron; iHg: inorganic mercury; La: lanthanum; Li: lithium; MeHg: methylmercury; Mg: magnesium; Mn: manganese; Mo: molybdenum; Ni: nickel; P50: median or 50th percentile; P95: 95th percentile; Rb: rubidium; Sr: strontium; Ti: titanium; V: vanadium; Y: yttrium.

[a] Lower-bound and upper-bound intake estimates were calculated as described in sections 2.3 and 2.4.

[b] Intake estimates in brackets are the lower and upper limit of the 95% confidence interval (see section 2.4).

[c] For MeHg, only a lower-bound intake estimate was calculated as this element is very likely only present in "fish and shellfish". For the other elements with no upper-bound intake estimate, this estimate was equal to the lower-bound intake estimate.

concentrations did not differ (see section 3.1). For methylmercury, only an LB intake estimate was calculated as this element is very likely only present in "fish and shellfish". The LB P50 intake estimates for inorganic mercury and methylmercury were zero: for inorganic mercury only seven samples had a concentration above the LOD (Table 2) and for methylmercury less than 50% of the children consumed fish on at least one of the reporting days. The lowest LB P95 intake estimate was 0.03 μg/kg body weight per day for yttrium. The lowest UB P50 and P95 intake estimates were also estimated for yttrium at 0.07 and 0.13 μg/kg body weight per day, respectively. The highest intake estimates were for rubidium, for which the LB estimates equalled the UB estimates: P50 and P95 intake of 117 and 218 μg/kg body weight per day (Table 6).

For the six elements for which the intake was expressed in mg per day, LB estimates equalled UB estimates. The lowest P50 intake was for molybdenum at 0.060 mg per day, and the highest for magnesium at 142 mg per day. The P95 intake estimates ranged from 0.12 mg per day to 225 mg per day for the same two elements (Table 6).

Table 7 lists the mean percentage contributions of the three food subgroups that contributed most to the LB intake distribution for each element. Contribution to the LB intake distribution was reported as this reflects the contribution based on concentrations above the LOD and does not depend on the concentrations assigned to concentrations below the LOD.

Table 7
Mean contributions (%) of the three food subgroups contributing most to the total lower-bound dietary intake distribution[a] of each element for children 1 and 2 years of age.

Element	Contribution (%)[b]		
Elements for which a risk characterisation was performed			
iAs	Concentrated fruit juices (21%)	Rice (19%)	Children's meals (11%)
Cd	Potatoes (28%)	Breads (11%)	Leafy vegetables (7.3%)
Cu	Banana (8.1%)	Potatoes (8.4%)	Breads (7.1%)
iHg	Potatoes (58%)	Fish and shellfish (23%)	Children's fruits (16%)
Mo	Milk and milk-based beverages (14%)	Nuts and seeds (10%)	Breads (7.4%)
Ni	Chocolates (24%)	Nuts and seeds (18%)	Breakfast cereals (7.3%)
Pb	Candies (27%)	Potatoes (14%)	Chocolates (8.5%)
Zn	Milk and milk-based beverages (16%)	Follow-on formula (9.4%)	Beef (8.3%)
Elements for which no risk characterisation could be performed			
Ba	Syrups (12%)	Concentrated fruit juices (8.9%)	Breads (8.8%)
Bi	Cheeses (91%)	Pork (4.2%)	Creams and ice creams (3.2%)
Cs	Concentrated fruit juices (32%)	Other fruits 1 (Grapes and pears) (8.4%)	Nuts and seeds (6.4%)
Ce	Chocolates (33%)	Dried fruits (29%)	Candies (9.4%)
Co	Chocolates (32%)	Concentrated fruit juices (18%)	Breads (7.1%)
Fe	Syrups (22%)	Follow-on formula (10%)	Breakfast cereals (8.4%)
La	Other fruits 1 (Grapes and pears) (34%)	Dried fruits (19%)	Candies (15%)
Li	Concentrated fruit juices (23%)	Pork (18%)	Sausages (10%)
Mg	Milk and milk-based beverages (14%)	Banana (8.1%)	Yoghurts and desserts (6.8%)
Mn	Breads (15%)	Breakfast cereals (11%)	Syrups (10%)
Rb	Milk and milk-based (19%)	Concentrated fruit juices (11%)	Yoghurts and desserts (8.4%)
Sr	Milk and milk-based (11%)	Syrups (11%)	Yoghurts and desserts (6.4%)
Ti	Chocolates (25%)	Milk and milk-based (8.2%)	Cheeses (7.3%)
V	Other juices (mixed juice, nectar and orange juice) (17%)	Chocolates (13%)	Breads (10%)
Y	Dried fruits (40%)	Other fruits 1 (Grapes and pears) (24%)	Leafy vegetables (14%)

iAs: inorganic arsenic; Ba: barium; Bi: bismuth; Cd: cadmium; Cs: caesium; Ce: cerium; Co: cobalt; Cu: copper; Fe: iron: iHg: inorganic mercury; La: lanthanum; Li: lithium; MeHg: methylmercury; Mg: magnesium; Mn: manganese; Mo: molybdenum; Ni: nickel; Pb: lead; Rb: rubidium; Sr: strontium; Ti: titanium; V: vanadium; Y: yttrium.

[a] Lower-bound dietary intake distributions were calculated as described in sections 2.3 and 2.4.

[b] Dietary intake of MeHg was for 100% due to consumption of "fish and shellfish".

3.3. Risk characterisation

To assess any potential risk, P95 dietary intake estimates of cadmium, copper, inorganic mercury, methylmercury, molybdenum, nickel and zinc were compared with a TDI, TWI or UL (see section 2.5). The intake estimates of copper, methylmercury, molybdenum and nickel did not exceed the relevant reference values, as well as the LB intake estimate of inorganic mercury (Table 8). The LB and UB intake estimates of cadmium exceeded the TWI by a factor of 1.8 and 2.2, respectively. The UB intake estimate of inorganic mercury exceeded the TWI by a factor of 3.7, and the intake estimate of zinc exceeded the UL by a factor of 1.1 (Table 8).

For inorganic arsenic and lead, an MOE was calculated based on the LB and UB P95 intake estimates and the relevant BMDLs. The MOE of inorganic arsenic was 6.8 for the LB estimate and 4.1 for the UB

Table 8
Factor of the tolerable daily or weekly intake (TDI or TWI), tolerable upper intake level (UL) and margins of exposure (MOE) for the long-term lower-bound and upper-bound P95 dietary intake estimates of elements for children 1 and 2 years of age.

Element	P95 intake[a]		TDI or TWI[b]		Factor of the TDI or TWI[c]
	Lower-bound	Upper-bound	Value	Reference	
	In μg/kg body weight per day				
Cd	0.64 (0.60–0.69)	0.79 (0.74–0.83)	0.36	(EFSA, 2009b, 2011a)	1.8–2.2
iHg	0.05 (0.04–0.06)	2.2 (2.1–2.5)	0.6	EFSA (2012d)	0.1–3.7
MeHg	0.07 (0.05–0.09)	-	0.19		0.4
Ni	6.5 (5.6–7.3)	9.2 (8.6–9.4)	13	EFSA (2020c)	0.6–0.7
	P95 intake In mg per day		UL	Reference	Factor of the UL[c]
Cu	0.87 (0.81–0.90)		1	(EFSA, 2006, 2018b)	1
Mo	0.12 (0.11–0.12)		0.1		1
Zn	8.3 (7.9–8.7)		7		1.1
	P95 intake In μg/kg body weight per day		BMDL	Reference	MOE[d,e]
iAs	0.44 (0.42–0.48)	0.74 (0.67–0.87)	3[f]	JECFA (2011)	4.1–6.8
Pb	0.23 (0.20–0.27)	0.70 (0.63–0.74)	0.5	EFSA (2010)	0.71–2.2

iAs: inorganic arsenic; BMDL: lower limit of the 95th confidence interval of the estimated benchmark dose with a 0.5% (iAs) or 5% (Pb) additional risk; Cd: cadmium; EFSA: European Food Safety Authority; iHg: inorganic mercury; JECFA: FAO/WHO Joint Expert Committee on Food Additives; MeHg: methylmercury; Mo: molybdenum; MOE: margin of exposure; Ni: nickel; P95: 95th percentile; Pb: lead; TDI: tolerable daily intake; TWI: tolerable weekly intake; UL: tolerable upper intake level; Zn: zinc.

[a] Lower-bound and upper-bound intake estimates were obtained from Table 6.

[b] TWI was expressed in μg/kg body weight per day by dividing the TWI of 2.5 μg/kg body weight per week for Cd, of 4 μg/kg body weight per week for iHg and of 1.3 μg/kg body weight per week for MeHg by 7.

[c] Factor of the TDI, TWI and UL was calculated by dividing the P95 best intake estimate by the TDI, TWI (expressed per day) or UL.

[d] MOE was calculated by dividing the BMDL by the P95 best intake estimate.

[e] Minimal value of the MOE for a negligible health risk is 10 for Pb. For iAs, EFSA (2009a) and JECFA (2011) have not indicated how large the MOE should be for a negligible health risk.

[f] EFSA (2009a) and JECFA (2011) derived BMDLs for iAs. JECFA's BMDL was used for the risk characterisation of iAs, because this BMDL is based on studies with a relatively long follow-up and large study population.

estimate. The corresponding MOEs of lead were 2.2 and 0.71, respectively. Minimal value of the MOE for a negligible health risk for lead is 10 (EFSA, 2010). For inorganic arsenic, EFSA (2009a) and JECFA (2011) have not indicated how large the MOE should be for a negligible health risk.

4. Discussion

4.1. Concentrations of elements

A multi-element inductively coupled plasma-mass spectrometry (ICP-MS) method was used to analyse the elements in the composite samples. Using this method, several elements were included in the TDS that have not been analysed in earlier TDSs in Europe that looked into the dietary intake of elements by young children from France (Chekri et al., 2019), Ireland (FSAI, 2016) and the UK (FSA, 2015). These elements are rare earth elements, such as indium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, lutetium and rhenium. On the other hand, some relevant elements were not included due to the use of this multi-element method, including aluminium, chromium, fluorine, iodine, potassium, sodium and selenium.

Using a multi-element ICP-MS method for the analysis implies that not all measurement parameters were optimised for each element. Consequently, the limits of detection (LODs) for the analysis of antimony and total mercury were relatively high compared to the LODs used in the

Table 9
Overview of dietary intake estimates of six elements of young children from total diet studies in Europe and as reported for Europe in most recent EFSA opinions.

Element	Country[a]	Year of sampling	Age (years)	Intake in µg/kg body weight per day		Main contributors (min 10%) to LB intake at food group level[e]	Reference
				Mean[b,c]	High (P90, P95 or P97.5)[b,d]		
iAs	Netherlands	2017	1–2	0.15–0.36	0.44–0.74[c]	Cereals and cereal-based products (33%), non-alcoholic beverages (21%), children's meals (11%), fruit (11%) and vegetables (10%)	Our study
	France	2011–2012	1–2	0.174–0.221	0.256–0.308[d]	Meat/fish based ready-to-eat meal (27%), rice and wheat products (19%)	Sirot et al. (2018)
	Ireland	2012	5–12	0.03–0.05	0.13–0.14[e]	Cereals (94%)	FSAI (2016)
	UK	2014	1.5–3	0.13–0.68	0.28–1.1[e]	Cereals	FSA (2015)
	Europe	2021	1–2	0.12–0.61	0.24–0.99[c]	Rice (9–36%), drinking water (6–39%), and cereals and cereal-based products (9–30%0)	EFSA (2021a)
Cd	Netherlands	2017	1–2	0.33–0.46	0.64–0.79[c]	Potatoes (28%), cereals and cereal-based products (24%), vegetables (21%) and confectionery (14%)	Our study
	France	2011–2012	1–2	2.04–2.18	3.42–3.61[d]	Potatoes (24%), and vegetables (18%)	Jean et al. (2018)
	Ireland	2012	5–12	0.24–0.32	0.47–0.59[e]	Cereals (48%), and vegetables (30%)	FSAI (2016)
	UK	2014	1.5–3	0.32–0.54	0.54–0.85[e]	Cereals	FSA (2015)
	Europe	2012	1–2	2.85–7.84	4.37–12.1[c]	Cereals and cereal-based products (15.7–34.5%), vegetables and vegetable products (9.02–20.5%), and starchy roots and tubers (6.49–26%)	EFSA (2012a)
iHg	Netherlands	2017	1–2	0.009–1.39	0.05–2.2[c]	Potatoes (58%), fish and shellfish (22%), and fruit (16%)	Our study
	France	2011–2012	1–2	0.003–0.045	0.01–0.06[d]	-	Sirot et al. (2018)
	Ireland	2012	5–12	0.02–0.17	0.11–0.31[e]	Fish and shellfish (99%)	FSAI (2016)
	Europe	2012	1–2	0.27–2.16	0.67–4.06[c]	Milk and dairy products (16–29%), fruit and vegetable juices (8.9–34%), and fish and other seafood (1.6–29%)	EFSA (2012d)
MeHg	Netherlands	2017	1–2	0.01	0.07[c]	Fish and shellfish (100%)	Our study
	France	2011–2012	1–2	0.006–0.006	0.009–0.009[d]	-	Sirot et al. (2018)
	Ireland	2012	5–12	0.02	0.11[e]	Fish and shellfish (100%)	FSAI (2016)
	Europe	2012	1–2	0.09–1.65	0.66–2.74[c]	Fish meat (59–100%), fish products (0–40%), and molluscs (0–5.3%)	EFSA (2012d)
Ni	Netherlands	2017	1–2	2.9–5.4	6.5–9.2[c]	Confectionery (24%), cereals and cereal-based products (20%), nuts and seeds (18%), and fruit (15%)	Our study
	France	2011–2012	1–2	2.68–4.39	4.57–6.51[d]	-	Sirot et al. (2018)
	UK	2014	1.5–3	4.4–5.2	7.1–8.1[e]	Cereals	FSA (2015)
	Europe	2020	1–2	6.23–14.6	10.7–24.8[c]	Cereals and cereal-based products (10–50%), sugar and confectionary (5–50%), and milk and dairy products (5–50%)	EFSA (2020c)
Pb	Netherlands	2017	1–2	0.10–0.43	0.23–0.70[c]	Confectionery (24%), Cereals and cereal-based products (20%), nuts and seeds (18%) and fruit (15%)	Our study
	France	2011–2012	1–2	0.199–0.209	0.287–0.295[d]	Vegetables (15%), and water (11%)	Sirot et al. (2018)
	Ireland	2012	5–12	0.04–0.17	0.09–0.27[e]	Cereals (27%), non-alcoholic beverages (19%), vegetables (12%), and fruit (10%)	
	UK	2014	1.5–3	0.15–0.28	0.25–0.46[e]	Milk group	FSA (2015)
	Europe	2012	1–2	0.81–1.77	1.18–3.27[c]	Milk and dairy products (13.7–29.1%), cereals and cereal-based products (9.1–19.4%), and drinking water (1.3–20.9%)[6]	EFSA (2012c)

iAs: inorganic arsenic; Cd: cadmium; EFSA: European Food Safety Authority; iHg: inorganic mercury; LOD: limit of detection; LOQ: limit of quantification; MeHg: methylmercury; Ni: nickel; Pb: lead; P50: median or 50th percentile: P90: 90th percentile; P95: 95th percentile; P97.5: 97.5th percentile.

[a] Dietary intake estimates for Europe are the range of lowest lower-bound and highest upper-bound estimates for children of 1 and 2 years of age across European dietary surveys.

[b] Dietary intake ranges refer to lower-bound and upper-bound intake estimates. The intake estimate of methylmercury is a lower-bound estimate (see section 2.3).

[c] For reasons of comparison, the mean intakes of the current study are reported.

[d] c = P95; d = P90; e = P97.5.

[e] For reasons of comparison, the main contributors to the lower-bound intake distribution for the current study are presented at food group level (Table 1).

other three TDSs in young children (Chekri et al., 2019; FSA, 2015; FSAI, 2016). LODs for the other elements were comparable. Antimony was not detected in any of the samples, whereas this element was detected in fruit juices, cereal-based foods and fruit purees in the French TDS (Chekri et al., 2019). However, the percentage of samples with a concentration above the LOD was also low in the French TDS, only 11.3%. Total mercury was detected in only 7 of the 161 composite samples included in the intake assessment, resulting in a large difference between the lower-bound (LB) and upper-bound (UB) intake estimates (Table 6). A targeted analytical method with a lower LOD may result in a more reliable intake estimate. Despite this, total mercury was detected in seven samples, including one sample of "potatoes" (12–17 months), "leafy vegetables" (12–17 months), "mushrooms" (all age groups) and "chicken" (12–17 months) (Table S2 of the Supplementary Material), and all three samples of "fish and shellfish", as expected.

For a substantial part of the elements, no intake calculations were performed because they were detected in none or only a limited number of the composite samples (Table 2). For example, hafnium, neodymium and thorium were not detected in any of the samples, and gallium and zirconium were detected in five samples. Gallium was detected in "leafy vegetables" ($n = 2$), "root vegetables" ($n = 1$), "dried fruits" ($n = 1$) and "candies" ($n = 1$) in concentrations ranging from 1 μg/kg to 1.6 μg/kg, and zirconium was detected in "cheeses" ($n = 3$) and "chicken" ($n = 2$) in concentrations ranging from 50 μg/kg to 460 μg/kg. For these elements, more sensitive methods are needed to reliably assess their intake. From a risk assessment point of view, this is relevant if a possible health risk cannot be excluded based on a UB intake estimate. For these elements, however, no tolerable daily or weekly intakes (TDIs or TWIs) or lower limits of a benchmark dose (BMDLs) are (yet) available to ascertain this.

4.2. Dietary intake of elements in young children

For the six elements with a TDI, TWI or BMDL, we compared the estimated intakes to the intakes reported in the TDSs in young children from France, Ireland and UK (Table 9). The ages of the children were 1 and 2 years for the French TDS, 5–12 years for the Irish TDS and 1.5–3 years for the TDS from the UK. Food consumption data were collected on 3, 7 or 4 consecutive days, respectively (Sirot et al., 2018; O'Connor et al., 2013; Roberts et al., 2018). In all these studies, the dietary intake was calculated according to an LB and UB scenario. The comparison showed that especially our UB dietary intake estimate of inorganic mercury exceeded the corresponding estimates in the other TDSs, due to a relatively high LOD in our study (see section 4.1). The UB high intake estimate of lead was also relatively high compared to the other TDSs. For this element, 75% of the composite samples in our TDS had a concentration below the LOD (Table 2). For the other elements, our intake estimates were within the range of estimates of the other TDSs. EFSA also calculated the dietary intake of these elements for Europe. In these calculations, individual national food consumption data of European countries covering 2–7 days were combined with a merged dataset of concentrations of elements in foods from European Member States. The LB and UB P50 and P95 intake estimates calculated by EFSA for children of 1 and 2 years across European dietary surveys were in general higher than our intake estimates (Table 9).

For most of the six elements, similar food groups were responsible for their dietary intake in the different studies, such as "cereals and cereal-based products" for inorganic arsenic, "potatoes" for cadmium, and "fish and shellfish" for inorganic mercury and methylmercury. A marked difference between our study and the other studies was a high contribution of "potatoes" to the LB intake distribution of inorganic mercury in our study (Table 9). This high contribution was due to a high consumption of potatoes and a positive total mercury concentration in one of the three composite samples for "potatoes" (12–17 months) of 10 μg/kg (Table S2 of the Supplementary Material). In addition, due to a relatively high LOD for this element, most food groups were not included in the LB estimate of inorganic mercury, which may have increased the contribution of "potatoes" to the intake of inorganic mercury. In 2012, EFSA (2012d) reported on total mercury levels in different food groups obtained from 20 European countries, covering the period of 2004–2011. Of the 421 levels reported for the food group "potatoes and potatoes products", 92% were reported below the LOD or LOQ, and the mean LB level was 0.1 μg/kg.

The dietary intake estimates of copper, iron, magnesium and zinc were compared to those reported by van Rossum et al. (2020). van Rossum et al. (2020) calculated the intake by combining the food consumption data of DNFCS 2012–2016, the same food consumption data source as used in our study, for 1-3-year-old children with concentrations from the Dutch food composition database NEVO. Only small differences in intake were observed (Table 10). The Dutch food composition database NEVO contains nutritional information of approximately 2100 foods, including foods for infants and young children. As shown for the dietary intake of protein and fat (Pustjens et al., 2021a), intake calculations of elements such as copper, iron, magnesium and zinc based on a TDS may be a good alternative for countries with no or a less complete food composition database.

EFSA also calculated the dietary intake of copper, iron, magnesium and zinc for children aged 1 and 2 years in Europe by combining individual national food consumption data of European countries with concentrations from the EFSA Nutrient Composition Database. The intake estimates calculated by EFSA were either comparable (iron, zinc) or higher (copper, magnesium) than our intake estimates (Table 10). Important contributors to the intake reported by EFSA were comparable to those found in our study, including "cereals and cereal-based products" (copper, iron, magnesium, zinc), "milk and milk-based beverages" (magnesium, zinc) and "meats and meat products" (zinc) (Table 10). van Rossum et al. (2020) did not report on the contribution of food groups to the intake of copper, iron, magnesium and zinc, and both van Rossum et al. (2020) and EFSA did not report on the dietary intake of manganese and molybdenum in young children.

4.3. Uncertainties in the dietary intake assessment

An important uncertainty in the dietary intake calculations of elements presented in this paper is due to the concentrations below the LOD, and to a lesser extent between the LOD and LOQ. If concentrations are below the LOD, it is not clear whether the chemical is present in the food at a concentration that cannot be detected, or whether it is not present at all. To address this uncertainty, the intake was calculated according to an LB and UB scenario (see section 2.3). The LB and UB intake estimates provide a lower and upper limit of the actual intake considering the analysed concentrations. For 10 elements, the number of samples with a concentration above the LOD was so high that the LB and UB intake estimates did not differ (Table 6). For other elements, this difference was small. The highest difference was observed for inorganic mercury, for which the UB intake estimate was a factor 44 higher than the LB estimate. The uncertainty related to samples with a concentration below the LOD is a problem if the LB intake estimate does not show a potential health risk but the UB estimate does. This situation occurred for inorganic mercury (see section 4.4).

To estimate the intake of inorganic arsenic, the total arsenic concentrations were converted to inorganic arsenic concentrations assuming that 70% of total arsenic is inorganic arsenic, except for "fish and shellfish" and "rice" in which inorganic arsenic was analysed as such (see sections 2.2 and 2.3). This assumption about the presence of inorganic arsenic based on total arsenic concentrations is a source of uncertainty in the dietary intake estimates of this element. Recently, EFSA (2021a) has calculated the percentage of inorganic arsenic to total arsenic for a selection of food groups. Excluding "fish and shellfish" and "rice", these ratios were 61%–91% in processed cereal products, including "biscuits, rusks and cookies for children", "cereal-based food for infants and young children" and "unleavened bread, crisp bread and rusk". Based on these percentages, the dietary intake of inorganic

Table 10
Overview of dietary intake estimates of elements for young children in the Netherlands based on our total diet study and food composition data[a], and as reported for Europe in most recent EFSA opinions.

Element	Country[b]	Age (years)	Intake in mg per day		Main contributors to the intake at food group level[d]	Reference
			P50 or mean[c]	High (P95)		
Cu	Netherlands	1–2	0.53[a]; 0.51[b]	0.87	Fruit (22%), grains and grain-based products (17%) and Confectionery (12%)	Our study
		1–3	0.7[a,b]	1.1	-	van Rossum et al. (2020)
	Europe	1–2	0.6–0.94[a]	0.94–1.2	Grains and grain-based products (24–37%), milk and dairy products (6–19%), and fruit and fruit products (5–17%)	EFSA (2015a)
Fe	Netherlands	1–2	6.0[a]; 5.6[b]	10	Non-alcoholic beverages (26%), grains and grain-based products (17%), confectionary (13%), meat (13%) and follow-on formula (10%)	Our study
		1–3	5.8[a]; 5.6[b]	8.6	-	van Rossum et al. (2020)
	Europe	1–2	5.0–7.0[a]	7.6–11.4	Grains and grain-based products (31–42%), food products for young population (4–22%), meat and meat products (5–14%)	EFSA (2015b)
Mg	Netherlands	1–2	150[a]; 142[b]	225	Dairy products (23%), Grains and grain-based products (15%), fruit (14%), non-alcoholic beverages (13%),	Our study
		1–3	182[a]; 177[b]	267	-	van Rossum et al. (2020)
	Europe	1–2	153–188[a]	228–278	Grains and grain-based products (23–34%), milk and dairy products (20–31%), and food products for young population (2–11%)	EFSA (2015c)
Zn	Netherlands	1–2	5.2[a]; 4.9[b]	8.3	Dairy products (32%), meat and meat products (197%), and grains and grain-based products (14%)	Our study
		1–3	5.7[a]; 5.6[b]	8.4	-	van Rossum et al. (2020)
	Europe	1–3	4.6–6.2[a]	6.7–9.0	Grains and grain-based products (19–32%), milk and dairy products (27–34%), meat and meat products (10–24%)	EFSA (2014b)

Cu: copper; Fe: iron; Food comp: Food composition data; Mg: magnesium; Zn: zinc.

[a] Intake estimates based on food consumption data for children aged 1–3 years from the same national food consumption survey as used in our study and concentrations of elements from the Dutch Food Composition Table (NEVO) (van Rossum et al., 2020).

[b] Dietary intake estimates for Europe are based on individual national food consumption data of European countries with concentrations from the EFSA Nutrient Composition Database. Range refers to the lowest and highest estimate across European countries and sex.

[c] a = mean; b = median (P50).

[d] For reasons of comparison, the main contributors for the current study are presented at food group level (Table 1). van Rossum et al. (2020) did not report the main contributors to the dietary intake.

arsenic could either have been overestimated or underestimated in our study. However, considering the low margin of exposure (MOE) for inorganic arsenic (Table 8), this uncertainty has very likely not affected the conclusion of the risk characterisation of inorganic arsenic (see section 4.4). This uncertainty in the intake assessment can be removed by analysing all foods for inorganic arsenic.

The intake estimates of inorganic mercury were subjected to the same uncertainty as for inorganic arsenic due to conversion of concentrations of total mercury to inorganic mercury (see section 2.3). These conversion factors, including the assumption that “fish and shellfish” contain inorganic mercury at a concentration equal to 20% of total mercury, were conservative and will have resulted in an overestimation of the dietary intake of inorganic mercury (EFSA, 2012d). However, this likely overestimation will not have affected the risk characterisation of inorganic mercury because the LB estimate was far below the TWI, and the UB estimate largely exceeded this limit value (Table 8).

Foods and beverages included in our study were sampled from August to November 2017 and may not be representative for all foods and beverages available on the market on an annual basis (Elegbede et al., 2017). However, the presence of elements in foods does not depend on, for example, weather or storage conditions and is considered not to vary considerably between seasons and years. Therefore, the intakes of elements presented in this paper are considered to be indicative for the intakes of the age group studied.

Dietary intake of elements was calculated using the OIM model. This model is commonly used by EFSA to calculate the long-term intake of mycotoxins (e.g. (EFSA, 2020b; d)), environmental contaminants (e.g. (EFSA, 2020f)) and food additives (e.g. (EFSA, 2020a; e)), and to calculate the chronic cumulative intake of groups of pesticides (EFSA, 2020g). Using this model, it is assumed that the mean intake over the available consumption days for each person, in our case two days, is a good estimate of the long-term intake. Given the limited number of person-days present in a food consumption database per person and the variation in daily food consumption by an individual, the distribution of mean intakes per individual obtained with the OIM model will often be too wide compared to distributions of “true” long-term intakes per person (Goedhart et al., 2012). Due to this, P95 intake estimates may be overestimated. However, we do not expect that this has affected the results of the risk characterisation considering the degree of exceedance of the TDI, TWI or tolerable upper intake level (UL), and the magnitude of the MOE (Table 8). The mean and P50 intake estimates are not affected by using this model.

Our TDS was based on food consumption data from the first two years of the DNFCS 2012–2016 and intake was estimated using the data for the period 2012–2016. The 2012–2016 period includes a larger group of children (n = 440 compared to n = 232) and is more representative due to the inclusion of food consumption data of two more recent years. A study into the intake of protein and fat and of mycotoxins, based on this TDS, showed that the intake for the whole period (2012–2016) did not differ significantly from that for the first two years of the DNFCS (Pustjens et al., 2021a; b). There is no reason to assume that this will be different for the estimated element intake.

4.4. Risk characterisation

For inorganic arsenic, cadmium, lead and inorganic mercury, the LB and/or UB P95 intake estimates for Dutch children aged 1 and 2 years were above the TWI or resulted in low MOEs (Table 8). The health effects on which these reference values are based occur after a “lifelong” period of intake. An intake exceeding the TWI or resulting in a low MOE during a limited time period, such as early childhood, may therefore not necessarily pose a health concern later in life. For example, a study into the dietary intake of cadmium in the Dutch population aged 2–69 years showed that the mean cadmium intake, according to a medium-bound scenario, dropped below the TWI around the age of 12 years and that the overall mean intake across all ages was below the TWI (Sprong and Boon, 2015). An exception is lead. For this element, a BMDL has been derived that is relevant for children 1–7 years of age as the adverse health effect concerns neurological development (EFSA, 2010).

For inorganic arsenic, it is not clear how large the MOE should be for a negligible health risk (EFSA, 2009a; JECFA, 2011). Based on intake estimates close to the BMDL, EFSA concluded 'Therefore, there is little or no MOE and the possibility of a risk to some consumers cannot be excluded.' (EFSA, 2009a). In our study, MOEs were 6.8 for the LB P95 intake estimate and 4.1 for the UB P95 intake estimate, which is low (Table 8).

The UB P95 intake estimate of inorganic mercury largely exceeded the TWI, whereas the LB P95 intake estimate was far below this TWI (Table 8). Concentrations obtained with a more sensitive analytical method are needed to establish if the intake of inorganic mercury is (too) high. For lead, the LB P95 intake estimate resulted in an MOE below 10 but above 1, which indicates that 'risk is likely to be low, but not such that it could be dismissed as of no potential concern" (EFSA, 2010). The UB P95 intake estimate of lead was below 1, which indicates that "possibility of an effect in some children cannot be excluded" (EFSA, 2010). This UB intake estimate was largely influenced by samples with a lead concentration below the LOD (75%; Table 2).

Comparing the P95 intake estimates of copper, molybdenum and zinc to the UL showed that the intake of zinc exceeded the UL (Table 8). van Rossum et al. (2020) reported also an exceedance of the UL for zinc for Dutch children aged 1–3 years; this group included the same individuals as included in our study plus 3-year-olds (Table 10). Also, the high intake of copper exceeded the UL in this study. Molybdenum was not included. In another Dutch study among 1526 children aged 10–48 months who attended 199 day-care centres, two-day food consumption records were collected for at least two days per week (Goldbohm et al., 2016). These authors concluded that 17% of the children exceeded the UL for zinc.

For several elements, including barium, rubidium, strontium, and titanium, concentrations were detected in all composite samples (Table 2), resulting in relatively high intake estimates (Table 6). For these elements, no health-based guidance values or BMDLs are available, and it was not possible to determine whether their intake was (too) high.

It was noted that for copper, iron, magnesium, manganese, molybdenum and zinc, also adequate intakes (AI) or average requirements (AR) have been set (EFSA, 2013a; b, 2014a, 2015a; b; c). These reference values indicate the average observed daily amount ingested by a population group (or groups) of apparently healthy people that is assumed to be adequate, and the estimated level of intake that is adequate for half of the people in a population group, respectively (EFSA, 2017). Comparing the relevant reference values with the P50 intake estimates of these six elements showed that the intake of manganese and molybdenum was a factor of 3–4 higher than the AIs (Table 11). For molybdenum, this was not accompanied by an exceedance of the UL by the P95 estimate (Table 8). However, no UL has been derived for manganese and it is therefore not clear if an intake of a factor of 3 higher than the AI may be of concern.

The risk characterisation was based on the best P95 dietary intake estimates (Table 8). Considering the uncertainty around these estimates, the upper limits of the confidence interval resulted in slightly higher factors of exceedance or lower MOEs. However, this did not affect the risk characterisation of these elements.

5. Conclusions

The P95 intake estimates of inorganic arsenic, cadmium, lead and zinc for Dutch children aged 1 and 2 years exceeded the tolerable daily or weekly intake or tolerable upper intake level or resulted in low margins of exposure. Food subgroups contributing most, i.e., more than 15%, to the dietary intake of these elements were "concentrated fruit juices" and "rice" for inorganic arsenic, "potatoes" for cadmium", "candies" for lead, and "milk and milk-based beverages" for zinc. For methylmercury and nickel, the P95 intake estimates did not exceed the tolerable daily or weekly intake. For inorganic mercury, it was not possible to ascertain if the intake was too high. Measurements with a more sensitive analytical method are needed to determine whether the intake of this element by young Dutch children is (too) high. For several elements, including barium, rubidium, strontium, and titanium, concentrations were analysed at levels above the limit of detection in all composite samples. For these elements, it was not possible to determine whether the intake was (too) high due to lack of reference values for risk characterisation. It was noted that the P50 intake estimate of manganese was a factor of 3 higher than the adequate intake level of this element. Due to the absence of a tolerable upper intake level, it is not clear if this intake is of concern.

Table 11
Long-term P50 intake of six elements of children 1 and 2 years of age and percentage of the AI or AR. The lower-bound and upper-bound intake estimates were identical.

Element	P50 intake[a,b]	AI or AR[c]	Factor of AI or AR[d]
	In mg per day		
Cu	0.51 (0.49–0.53)	0.7	0.7
Fe	5.6 (5.0–5.9)	5	1.2
Mg	142 (140–151)	170	0.9
Mn	1.4 (1.3–1.5)	0.5	3
Mo	0.060 (0.057–0.062)	0.015	4
Zn	4.9 (4.8–5.1)	3.6	1.4

AI: adequate intake; Cu: copper; AR: average requirement; Fe: iron; Mg: magnesium; Mn: manganese; Mo: molybdenum; P50: median or 50th percentile; Zn: zinc.

[a] Intake estimates were calculated as described in section 2.4.

[b] Intake estimates in brackets are the lower and upper limit of the 95% confidence interval (see section 2.4).

[c] AIs for Cu and Mg were derived for ages 1 and 2 years, and those for Mn en Mo for ages 1–3 years (EFSA, 2013a; b, 2015a; c). ARs were derived for Fe for children aged 1–6 years and for zinc for children aged 1–3 years (EFSA, 2014b, 2015b).

[d] Factor of AI or AR was calculated by dividing the best intake estimates of the P50 by the AI or AR. For this, the median intakes were rounded to the same number of decimals as the AI or AR, if needed.

CRediT authorship contribution statement

P.E. Boon: Conceptualization, Formal analysis, Writing – original draft, Revising the manuscript, critically for important intellectual content. **A.M. Pustjens:** Conceptualization, Funding acquisition, Writing – original draft. **J.D. te Biesebeek:** Conceptualization, Formal analysis, Writing – original draft. **G.M.H. Brust:** Funding acquisition, Formal analysis, Writing – original draft. **J.J.M. Castenmiller:** Conceptualization, Formal analysis, Writing – original draft, Approval of the version of the manuscript to be published (the names of all authors must be listed).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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