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Research article

Dietary heavy metal exposure of Finnish 1-year-olds

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Abstract: The exposure of Finnish 1-year-olds to cadmium, lead and inorganic arsenic via food and drinking water was determined. The food consumption data consisted of 3-day records from 1010 children aged 12 months, collected during 2002 to 2005 in Southwest Finland. One fifth of these children were still breastfed when the consumption data were collected and their exposure was assessed separately from the non-breastfed children. The heavy metal concentration data in foodstuffs were mainly analysis results from national authorities and they were mostly from the years 2005 to 2012. Dietary exposure assessment was performed probabilistically using the online program MCRA. With middle bound estimates, 89% of the non-breastfed and 56% of the breastfed children exceeded the tolerable weekly intake of cadmium. The benchmark dose ($BMDL_{01}$) for neurological damage caused by lead was exceeded by 60% of the non-breastfed and by 50% of the breastfed children, while the lowest $BMDL_{01}$ for cancer risk increase caused by inorganic arsenic was exceeded by 77% of the non-breastfed and by 61% of the breastfed children. The assessment did not include the unknown heavy metal exposure from breast milk. Heavy metal exposure differences between the boys and the girls were also assessed. Breastfed girls had significantly higher heavy metal exposure relative to their bodyweight than the breastfed boys, while in the non-breastfed group there were no differences by sex.

Keywords: dietary exposure; risk assessment; children; cadmium; lead; arsenic; Monte Carlo

Abbreviations: BMDL: Benchmark dose lower confidence limit; bw: Body weight; iAs: Inorganic arsenic; LB: Lower bound (<LOQ = 0); LOQ: Limit of quantification; MB: Middle bound (<LOQ = 50% of LOQ); TWI: Tolerable weekly intake

1. Introduction

Heavy metals are naturally present in the environment in varying amounts, depending on the soil composition of the area. In addition, humans have locally increased the environmental levels e.g., through pollution. The uptake of heavy metals by crops and animals from the environment varies with species, and the local environmental levels also play a role in the resulting heavy metal content in raw agricultural commodities. Legislative limits have been set to maximum permissible levels of heavy metals in many foods. In the EU, the legislation limiting heavy metal content of foods is the Commission Regulation (EC) No 1881/2006 setting maximum levels for certain contaminants in foodstuffs.

This study considers three heavy metals: cadmium, lead and arsenic, even though strictly speaking arsenic is a metalloid. The most toxic form of arsenic are its inorganic compounds, while some organic arsenic compounds are relatively nontoxic. Only inorganic arsenic is considered in this study.

Heavy metals are toxic to various parts of the human body. Many of their effects are caused at cell level by the inactivation of important enzymes or from oxidative stress, which may fatally damage the cells. Even though the heavy metals are only slightly genotoxic and mutagenic, they disrupt DNA repair mechanisms [1–3] and may therefore increase the damage caused by other mutagens such as UV radiation, smoking or oxidation.

Based on dose-response data, toxicological reference values for heavy metals have been determined. This study uses values determined in scientific studies by the European Food Safety Authority EFSA, and for inorganic arsenic comparison is also made with the value determined by the Joint FAO/WHO Expert Committee on Food Additives JECFA, which was determined later than the EFSA range of reference values.

The tolerable weekly intake (TWI) of cadmium is 2.5 μg/kg body weight (bw)/week based on kidney effects [4]. While JECFA has determined a tolerable monthly intake for cadmium, EFSA has since argued [5] that the lower intake limitation in their TWI is appropriate, which is why this study only compares the cadmium intake to the EFSA TWI. In the 2000s it was noticed that the critical effects caused by lead and inorganic arsenic do not appear to have a threshold, and a benchmark dose lower confidence limit (BMDL) is used as a reference value instead. The $BMDL_{01}$ for developmental neurotoxicity in children caused by lead was determined to be 0.50 μg/kg bw/day, corresponding to blood lead concentration of 12 μg/L [6]. The $BMDL_{01}$ based on the increase in the incidence of lung, bladder and skin cancers and skin lesions caused by inorganic arsenic (iAs) was determined to be in the range of 0.3–8 μg/kg bw/day [3], and the lower limit of this range was used as the reference value in this study. The corresponding $BMDL_{05}$ value determined for cancer caused by iAs has been determined by JECFA to be 3.0 μg/kg bw/day [7].

Children are a vulnerable group, because they have a higher relative energy need than adults and consequently they consume greater amounts of food in relation to the body weight than adults do. Therefore, the tolerable intake amounts are easily exceeded among children, especially among the youngest age groups. In addition, their developing organs are more prone to damage than those of adults. For these reasons, the legislative maximum limits for heavy metal content of (ingredients for)

industrially produced foods for children have been set very low in (EC) No 1881/2006. Ingredients of home-made food are subject to the general maximum permissible levels detailed in the same legislation.

In the current study, we investigated the dietary exposure of Finnish 1-year-olds to cadmium, lead and inorganic arsenic based on food consumption data collected nationally and on concentration data mainly consisting of foods and food ingredients analyzed in Finland. The main aim of this study was to determine whether the toxicological reference values for heavy metals are exceeded by the Finnish 1-year-olds and to what extent, as well as to find out the main dietary sources for the exposure.

Approximately one fifth of the studied population of 1-year-olds was still breastfed at the time of the collection of the consumption data, and these children are studied separately from the non-breastfed children. The dietary exposure of girls and boys in both groups was compared, and the results were compared with previously published results concerning heavy metal exposure of children aged 3 and 6 years in Finland [8]. Generally, the 1-year-olds in Finland consume commercial baby foods to a high extent, while the older age groups mainly eat "adult food", i.e., products not directed specially for children.

2. Materials and methods

2.1. Heavy metal concentrations in foodstuffs

The concentration data in this study covered the years 1995 to 2013, but most of the data were from the years 2005 to 2012. Table 1 shows the data divided by food group and heavy metal. The main part of the data consisted of analysis results of national authorities: the Finnish Food Safety Authority Evira (currently the Finnish Food Authority), the Finnish Customs Laboratory, the Finnish Environment Institute SYKE, the Natural Resources Institute Finland, as well as the Centres for Economic Development, Transport and the Environment (ELY Centres). These data have been partly published in [9,10]. Additional data from previous projects at the University of Helsinki [11,12], and self-monitoring data from the industry were also used.

A total of 4278 analysis results for cadmium, 3999 for lead and 1504 for arsenic were included in the study. The arsenic data were a mixture of inorganic arsenic results measured in rice and rice products, and total arsenic measured in other foods and calculated into inorganic arsenic as described later in this chapter. Lead concentrations in milk were included only for the last 7 years (2006 to 2012) since the levels have been decreasing. Additional literature data were used for tap water levels from waterworks providing water for ca. 90% of the Finnish population [13]. Further, average concentrations of European monitoring data, which is mainly Central European in origin, were taken from EFSA risk assessment reports [14–16] for foods with no national analysis results on a certain heavy metal. For some food groups there were many analysis results, in particular for meat (only cadmium and lead), fish, cereals, vegetables and dairy. For other food groups, the available results were mainly of one or two subgroups; e.g., the group fruit and berries mainly contained data on berries. The exposure assessment was performed at food subgroup level.

Most of the analyses had been performed with AAS or ICP-MS techniques, giving results for the total amount of different forms of the heavy metal. For rice, which is a known source of inorganic arsenic, data using an analytical speciation method were available. Data on arsenic in rice and baby foods were separated into iAs (inorganic As (III) and As(V) compounds) and organic

arsenic (MMA, DMA and arsenobetaine). For other foods, the iAs content was calculated from the measured total As. The relative proportion of iAs out of the total As was estimated to be 2% for fish, 3.5% for seafood [17], 100% for tap water (as well as bottled water, which the children drank only seldom) and 70% for other food groups [3].

Table 1. Years when the data were collected, at food group level. The food subgroups within the food group had sometimes data from only one year, but from the limitations of the available results, occurrence data were not always available for the same years even within a food group.

Food group	Cd and Pb data	As data
Cereals and cereal products	2000–2011	2000–2002, 2004, 2009, 2011
Vegetables including legumes, nuts and oilseeds	2002–2012	2004, 2006, 2008, 2009
Starchy roots	2003–2011	2009
Fruits and berries	2002–2011	2004, 2009, 2011
Meat and meat products	2003–2012	2009
Fish and seafood	1997–2000 (only Cd), 2001–2012	1998, 2001, 2002, 2004–2012
Milk and dairy products	1995–1998, 2002, 2004–2012 (Pb limited in the analysis to 2006–2012)	1996–2001, 2003, 2010–2012
Chocolate and cocoa	2002, 2003, 2010, Cd also 2011	2009, 2012
Juices and rice drinks	2004–2011, 2012	2009, 2011, 2012
Food for children	2003–2007, 2009–2013	2004, 2011–2013

2.2. Food consumption data

Food consumption data of 1-year-old children were collected during 2002–2005 in the Type 1 Diabetes Prediction and Prevention (DIPP) study [18] from children living in the Pirkanmaa area in Southwest Finland. These children have genetically increased risk of type 1 diabetes, but they were not diabetic at the time of the data collection for the current study. Therefore, it was assumed that their food consumption would be similar to 1-year-old Finnish children without the genetic type 1 diabetes risk. It is unknown how well the regional data describes the consumption by children in the whole country, but at the time the current study was done, the DIPP study data were the largest and most recent data on young children in Finland.

A total of 494 girls and 516 boys aged 1 year were studied. Of these children, 111 girls and 106 boys were still breastfed at the time when the food consumption data were collected and the rest (383 girls and 410 boys) were not. These two groups, differing by breastfeeding status, were studied separately. The mean weights of non-breastfed and breastfed children, respectively, were 9.76/9.54 kg for girls, 10.34/10.21 kg for boys and 10.06/9.87 kg for the whole studied population. The weights of the children ranged between 6.97 kg and 14.66 kg. Compared with WHO growth standards (weight for age), the mean weights in both groups were above the growth standard median, which is 8.9 kg for girls and 9.6 kg for boys.

Food records were completed for 3 consecutive days. The food consumption data were calculated using the Finnish Food Composition database (Fineli), which contains more than 7000 food items [19]. If a consumed food was not found in the Fineli database, it was saved as the sum of

its ingredients or as the nearest corresponding item. The detailed description of the food consumption data collection, entering, processing and calculation is published in [20].

In the current study, the heavy metal exposure of the breastfed children was determined separately from non-breastfed children because the diets of the two groups were different. The additional exposure of the breastfed children via breast milk was unknown because the amount of breast milk was not available. In Finland, only the cadmium content of human milk has been reported (most recently in [21]), and the cadmium levels were found to be lower than the values reported by WHO in [22]. In addition, the limited number of samples and the sensitivity of the analytical methods prevented the statistical comparison of heavy metal levels in follow-on formula, cow milk and breast milk in the current study. The infant formula, follow-on formula and cow milk samples were measured with a method where the limit of quantification for cadmium was the same as the cadmium concentration reported in [22] for human milk. Only one of the formula samples and 8% of the control programme cow milk samples exceeded this limit. Therefore, the average concentration in these foods is slightly lower than the concentration reported years earlier for breast milk, but the levels cannot be compared with accuracy.

In the current study, food consumption data were calculated to ingredient level for other foods than commercial baby foods. These were studied at food type level since the maximum allowed levels in legislation are lowest for baby foods and therefore the concentrations in baby foods were expected to be lower than in the other ingredients.

2.3. Exposure assessment

Exposure assessment for chronic (long-term) dietary exposure was performed with online program MCRA version 8 [23]. Concentration data and food consumption data for all three study days were downloaded to the program as individual results and combined probabilistically. For chronic exposure, which is the most relevant one for heavy metals, the MCRA program uses an average value for the concentrations in a single food item. It is assumed that the highest and lowest concentrations in a food item even out over a longer period of time and thus the most probable concentration during a long time period is close to the average [24].

Empirical data on the concentrations (i.e., the measured heavy metal concentrations in various foods at sample level) were used in the calculations, and a beta-binomial-normal (BBN) model and logarithmic transformation were chosen among the options given by the MCRA program. The BBN model uses betabinomial distribution for frequency of exposure and (transformed) normal distribution for the amounts. It corrects the variation in long-term exposure for the within-person variation in the study days [25]. For consumption frequency and for consumed amount, the gender of the child was taken into account as a covariate. Other covariates were not considered in this study as background data on the individuals only detailed their age, weight, breastfeeding status and gender. Altogether, 100,000 Monte Carlo iterations were calculated for each analysis.

Model based approach was used in the assessment. In uncertainty analysis with bootstrap approach for each run, 10,000 iterations were carried out from resampled data of individual consumption using 100 resampling cycles. The uncertainty analysis shows the uncertainty relative to the MCRA calculations, but it does not cover uncertainty from limited datasets, assumptions and unknown data.

In the concentration data, some of the analysis results were nondetects, i.e., below the limit of quantification (LOQ) of the analysis method. If a food did not have any numerical results (i.e., all results were below the LOQ), it was not taken into account in the calculations. Foods with numerical as well as below LOQ results were calculated with two scenarios: lower bound (LB: <LOQ = 0) and middle bound (MB: <LOQ = 0.5 LOQ).

The fraction of the LOQ used for nondetect values is the same for all foods in the analysis, irrespective of their LOQ values. Therefore, data with high LOQ values in one or few food groups may potentially lead to an overestimation of the relative importance of these food groups to the total exposure in the middle bound scenario. The percentage of nondetects in each food category and for each heavy metal has been published in [26].

A more comprehensive discussion on the mathematical background of the assessment is presented in the reference manual of the MCRA 8 program [23].

Gender differences in exposure were estimated through two-sided t-test (in Microsoft Excel) from individual daily exposure values produced in MCRA analysis.

3. Results

3.1. Main dietary sources of heavy metal exposure for 1-year-olds

The sources of dietary heavy metal exposure for the breastfed and non-breastfed 1-year-old children are shown in Figure 1. Despite their low levels of heavy metals, commercial baby foods, especially ready-to-eat foods and porridges, were the main source of cadmium, lead and inorganic arsenic exposure. This food group is often consumed in relatively large portions by young children, which explains its importance as a source of heavy metals.

Among the porridges for children, those containing either rice with fruit or non-rice cereals with fruit were the most consumed types, and their contributions to the exposure from porridges were therefore the highest for all heavy metals. Porridges containing rice contribute approximately half of the inorganic arsenic exposure from porridges. For cadmium exposure, porridges containing other cereals (e.g., wheat) were the biggest contributor among porridges. Ready-to-eat foods with meat as the protein source were the biggest contributor to cadmium and lead exposure among this food group, and they were also one of the largest sources of the cadmium and lead exposure from all foods. Fish-based or vegetable-based ready-to-eat foods were not consumed as frequently, so their contribution is lower although the concentrations in them were slightly higher. However, 75–100% of the measured ready-to-eat foods had lead levels or total arsenic levels below the limit of detection, except for the fish-based ready-to-eat foods, which had high total arsenic levels (average 511 mg/kg). In fish, however, most of the arsenic is found in relatively nontoxic organic forms, and the inorganic arsenic content is low.

After the commercial baby foods, cereals were the second largest source of exposure to cadmium, lead and inorganic arsenic. The levels of cadmium in cereals in this study were not very high, with average middle bound concentrations varying between 12 µg/kg in rye and 32 µg/kg in wheat. Still, the cadmium content in many other frequently consumed food groups was lower. For lead, the average middle bound concentrations in cereals varied between 20 µg/kg in rice and 32 µg/kg in barley, but 39% to 63% of the cereal data were below the LOQ. The arsenic content of rice was approximately ten times higher than that of the other cereals. Although the children ate other cereals

more, the inorganic arsenic exposure from rice alone was approximately 1/8 of the total exposure in the middle bound scenario.

The contribution of all of the other food groups than cereals and commercial baby foods (i.e., groups B to J in Figure 1) to the total inorganic arsenic exposure was 10% for the non-breastfed children. For cadmium, in comparison, these food groups contributed 39% of the total exposure, with potatoes and milk both at 10% and drinks at 7%. For lead, the food groups B to J in Figure 1 contributed 17% of the total exposure, out of which potatoes contributed 5% of the total exposure and the other food groups less.

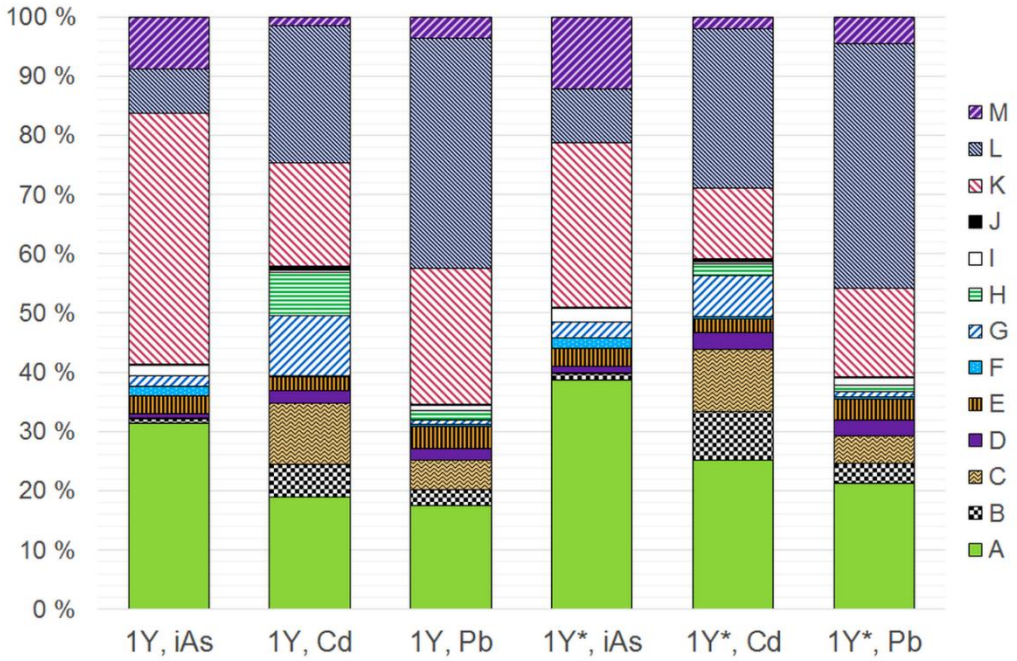

Figure 1. Contribution of different food groups to the total dietary heavy metal exposure of non-breastfed (1Y) and breastfed (1Y*) 1-year-old children at population group mean level. Middle bound scenario was used for exposure assessment. Inorganic arsenic (iAs) was calculated from total arsenic content in foods. The food groups are: A, Cereals and cereal products; B, Vegetables and vegetable products, C, Potatoes and other starchy roots; D, Fruits and berries; E, Meat and meat products; F, Fish and seafood; G, Milk and dairy products; H, Drinks (juices, rice drinks and follow-on formulae); I, Tap water; J, Chocolate and cocoa; K, commercial baby food/porridges; L, Commercial baby food/ready-to-eat meals; M, Commercial baby food/Desserts and fruit or berry purees.

Children with the highest dietary exposure to heavy metals had slightly different sources compared with children at population group mean level. The breastfed children with the highest cadmium exposure got a larger percentage of their total exposure from vegetables and a lower percentage from meat-based baby foods than the average member of the breastfed group average shown in Figure 1. For the children with the highest lead exposure, the relative contribution of potatoes to the total lead exposure was higher than for the average child in the corresponding age

group. For inorganic arsenic, the contribution of rice-based porridges and other rice products to the total exposure was higher for the children with the highest exposure than for the age group average.

3.2. Levels of dietary heavy metal exposure

The dietary exposure of Finnish 1-year-olds to the heavy metals was generally lower for the breastfed than non-breastfed group. At least partially this is explained by the fact that the exposure via breast milk was not included in the assessment since the consumed amount of breast milk was not available. Figure 2 shows the median exposure and the exposure at 95th percentile of the distribution (high users) for breastfed 1-year-olds as well as for non-breastfed 1-year-olds. The exposure level is shown compared with the toxicological reference value (tolerable weekly intake TWI or benchmark dose BMDL) of the heavy metal.

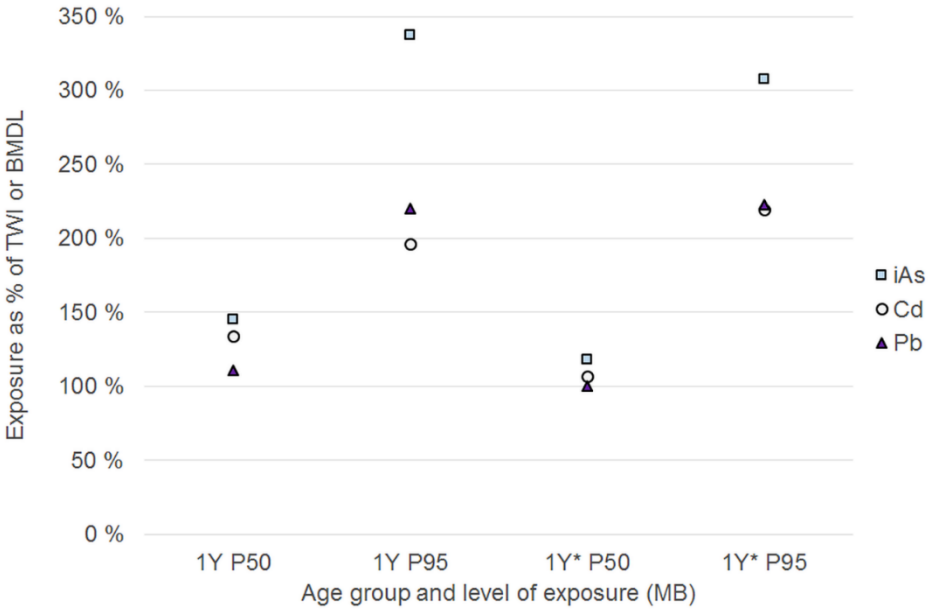

Figure 2. Heavy metal exposure at the age group median (P50) and 95th percentile (P95). The middle bound scenario was used for nondetect values. The exposure is given as % of the tolerable weekly intake (Cd, 2.5 µg/kg bw/week) or the lowest benchmark dose (iAs, $BMDL_{01}$ 0.30 µg/kg bw/day; Pb, $BMDL_{01}$ 0.50 µg/kg bw/day). Above the 100% mark the estimated exposure exceeds these toxicological reference values. 1Y*, breastfed 1-year-olds; 1Y, non-breastfed 1-year-olds.

Table 2 shows the part of the studied population group exceeding the toxicological reference value under lower bound (LB, results below LOQ calculated as zeros) and middle bound (MB, results below LOQ calculated as 50% of LOQ) estimates. Since the inorganic arsenic content of the foods was mainly estimated from the total arsenic by use of fixed fractions, the estimate for this heavy metal contains additional uncertainty not visible in the confidence interval shown in the table. The LOQs were relatively high for many food matrices, and therefore the data for many foodstuffs were left censored, i.e., included values below LOQ. This results in differences between the LB and MB estimates.

Table 2. Lower bound (LB) and middle bound (MB) estimates of the part of the age group exceeding the TWI or the $BMDL_{01}$ values determined by EFSA. The 95% CI of the estimate is given in parentheses. The $BMDL_{05}$ for iAs determined by JECFA [7] is included in the table. 1Y*, breastfed 1-year-olds; 1Y, non-breastfed 1-year-olds.

Compound	Ref. value	1Y, LB	1Y, MB	1Y*, LB	1Y*, MB
iAs	0.30 μg/kg bw/day	29% (20–48%)	77% (69–81%)	21% (14–33%)	61% (53–70%)
iAs, JECFA	3.0 μg/kg bw/day	0% (0–0.01%)	0% (0–0.11%)	0% (0–0.07%)	0% (0–0.07%)
Cd	2.5 μg/kg bw/week	53% (44–64%)	89% (81–94%)	42% (35–50%)	56% (49–65%)
Pb	0.50 μg/kg bw/day	49% (14–81%)	60% (22–79%)	34% (7–65%)	50% (20–74%)

As evident in Table 2, the cadmium exposure exceeded the TWI for most of the children. Exceeding the TWI means that the risk of adverse effects is low, as the TWI includes a safety factor, but above zero, especially for sensitive groups.

Since lead and inorganic arsenic do not have a safe threshold level of exposure, a margin of exposure (MOE) was calculated for these heavy metals by dividing the benchmark dose with the exposure. For lead, the benchmark dose was the one for developmental neurotoxicity, $BMDL_{01}$ 0.50 μg/kg bw/day [3]. For inorganic arsenic, the benchmark dose value determined by JECFA for 0.5% cancer risk increase [7] was used in the MOE calculations as the value contained less uncertainty than the value range determined by EFSA. At population group median exposure, the MOE for lead was 0.96 for the breastfed and 0.86 for the non-breastfed children. The MOE for inorganic arsenic was 8.3 for the breastfed and 6.8 for the non-breastfed children. Thus, the lead exposure as well as the inorganic arsenic exposure are both at a level where the risk of adverse effects is possible.

Figure 3 shows the distribution of exposure among non-breastfed 1-year-olds for cadmium, lead and inorganic arsenic. The reference values also used in Table 2 are marked on the picture to illustrate the part of the studied population with dietary exposure exceeding these values.

3.3. Differences between boys and girls in the levels of heavy metal exposure

Individual daily exposure values produced in MCRA analysis for each of three study days were subjected to two-sided t-test in Microsoft Excel to estimate differences by sex in the heavy metal exposure of the children. The boys in both groups were significantly heavier than the girls.

Among both groups of Finnish 1-year-olds, the girls had higher exposure than the boys in relation to the individual body weight, but the difference was only significant for cadmium, lead and inorganic arsenic exposure of the still breastfed children (Table 3). However, the absolute exposure as μg/d was significantly higher for the non-breastfed boys (Cd and Pb, not shown) than the girls in the same group. This signifies that the boys consumed more foods containing these heavy metals, or their consumption of foods with higher heavy metal levels was more frequent, but because of their higher body weight, the exposure relative to the body weight was not significantly different.

In our previous study [8], Finnish boys aged 3 years had significantly higher cadmium and lead exposure than the girls of the same age, and 6-year-old boys had significantly higher cadmium, lead

and inorganic arsenic exposure than the girls of the same age. As the exposure in the previous study was calculated with identical concentration data and identical methods to what were used in the current study, the change from the girls having the higher heavy metal exposure as toddlers to the boys having the higher exposure among the older children must originate from a change in the food consumption of the children as a group.

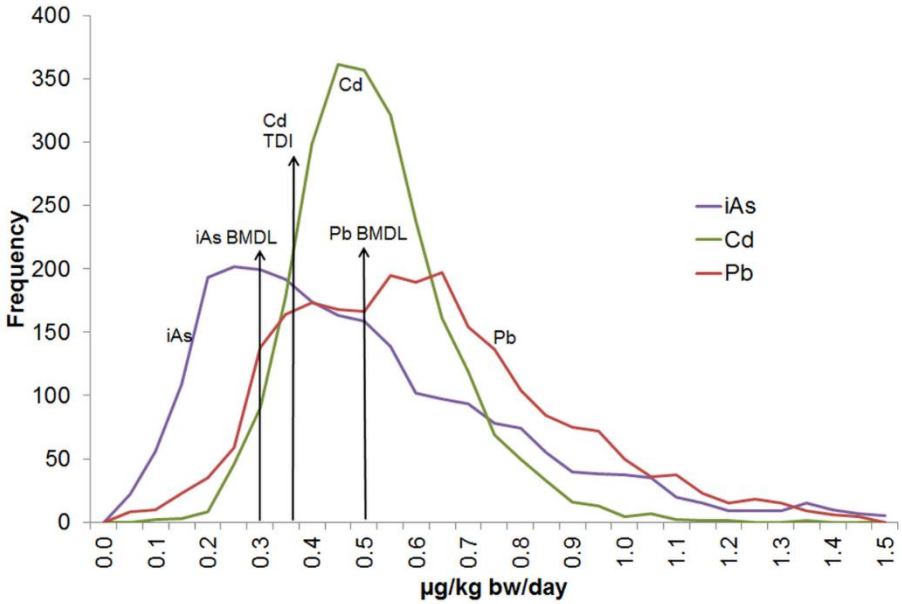

Figure 3. Heavy metal exposure distribution of non-breastfed 1-year-olds under middle bound scenario. $BMDL_{01}$ values of iAs (lower limit of the EFSA BMDL, 0.3 µg/kg bw/day) and Pb (0.5 µg/kg bw/day) as well as TWI/7 value of Cd (0.36 µg/kg bw/day) are marked by arrows.

Table 3. Mean weights and mean dietary heavy metal intakes by sex and breastfeeding. Middle bound scenario was used for nondetects. P values are given for two-sided t-test where variances were assumed to be different. SD = standard deviation.

Age group	Factor	Weight (kg)	iAs exposure (µg/kg bw/d)	Cd exposure (µg/kg bw/d)	Pb exposure (µg/kg bw/d)
1Y	Female mean	9.76	0.49	0.50	0.60
	SD (Female mean)	1.12	0.30	0.14	0.25
	Male mean	10.34	0.48	0.49	0.59
	SD (Male mean)	1.06	0.31	0.15	0.28
	P	7.50 E−38	0.49	0.18	0.27
1Y breastfed	Female mean	9.54	0.43	0.43	0.57
	SD (Female mean)	1.03	0.33	0.18	0.30
	Male mean	10.21	0.38	0.39	0.53
	SD (Male mean)	1.04	0.24	0.15	0.25
	P	8.82 E−16	0.03	0.001	0.05

The dietary cadmium exposure for breastfed and non-breastfed girls and boys is shown as boxplots in Figure 4. As in Table 3, the exposure in the smaller (by number of individuals) breastfed group is slightly lower than that of the non-breastfed group, partly due to the additional, unknown exposure from breastfeeding.

The consumption of chocolate and cocoa-based products was roughly as common among boys and girls. There were some differences in the consumed amounts of legumes, nuts and seeds. For non-breastfed children their consumption was more common for boys, and the consumed amounts were also higher. In the breastfed group the consumption of this food group was equally common between the sexes and the consumed amounts were slightly higher for girls.

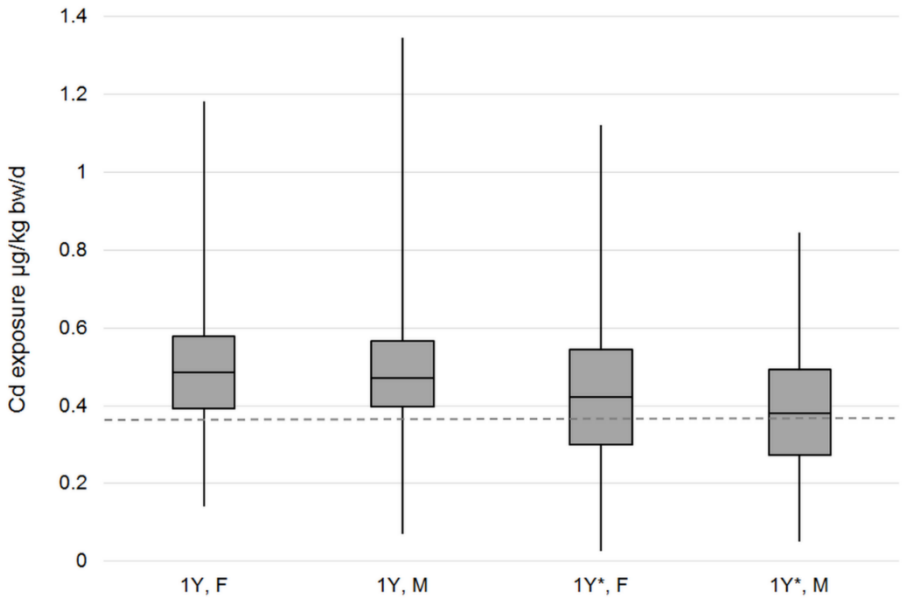

Figure 4. Boxplot of the dietary cadmium exposure (MB) for breastfed (1Y*) and non-breastfed (1Y) girls (F) and boys (M). The dashed line at 0.36 µg/kg bw/d marks the daily exposure corresponding to the tolerable weekly intake of cadmium. Individual daily exposure levels were used, i.e., each of the three study days for each child was included as a separate data point. Lower and upper ends of the box are 25th and 75th percentile of exposure and median is shown as line inside the box.

4. Discussion

The current study shows that the dietary heavy metal intake of Finnish children exceeds the tolerable weekly intake of cadmium and the lowest benchmark doses ($BMDL_{01}$) of lead and inorganic arsenic for a part of the 1-year-old population, even though the national estimates are lower than the estimates published previously by EFSA [14–16] from concentration data collected from all EU Member States.

The dietary exposure to iAs worldwide was estimated by Oberoi et al. [27] from GEMS cluster diets that mainly take into account the adult population. Finland is part of diet cluster F, together with most of the Nordic and Baltic countries. The exposure in this cluster (LB, bioavailability 50%–UB, bioavailability 100%) was estimated to be 1.84–2.19 µg/kg bw/day for

total arsenic and 5.25–57.27 μg/day for inorganic arsenic. The resulting burden of cancers caused by foodborne arsenic was estimated to be between 17 and 270 additional cases per year (LB, UB) for different cancers [27]. Cluster F had total arsenic exposure above the median but iAs exposure below the median of the 13 clusters. The estimates of the exposure of Finnish 1-year-olds to total arsenic and iAs in the current study are lower than the estimates presented in [27], possibly due to use of more specific data on food consumption and concentration levels relevant to Finland. The median exposure to total arsenic at LB level was estimated in the current study to be 0.80 μg/kg bw/day for breastfed and 1.00 μg/kg bw/day for non-breastfed children. The median exposure to inorganic arsenic at MB level was 0.36 μg/kg bw/day and 0.44 μg/kg bw/day for the breastfed and non-breastfed children, respectively, while the respective values at LB level were 0.19 μg/kg bw/day and 0.23 μg/kg bw/day. With these lower exposure levels, the resulting burden of cancers is also lower than estimated in the literature. The dietary arsenic exposure of Finnish adults relative to body weight is lower than that of children.

The levels of heavy metals, especially lead, in Finnish tap water are low in comparison with many other European countries. The change to lead-free gasoline in the early 1990s already has decreased Pb levels in many food ingredients. In addition, steps have been taken to decrease the heavy metal content in foods by improvement of agricultural practices. As a result, the concentrations of heavy metals in many foods with high consumption were found to be on the average lower than the averages reported by EFSA from monitoring data collected predominantly from Central European Member States [14–16]. The lower heavy metal concentrations in the national data are at least part of the explanation of why this study shows lower exposure than previous assessments by EFSA, as seen in Table 4. In addition, the current study did not cover the entire diet. The consumption data used in the current study covered most of the diet and contained all of the main sources of exposure, but the exclusion of low-concentration foods such as eggs, soft drinks and sugar means that the estimated exposure is slightly lower than would have been estimated from the entire diet.

The available data for this study were limited, which may also affect the results. The concentration data were mainly from monitoring samples collected with a targeted plan and so they may be different from the national average levels. Most likely the concentrations in the monitoring samples are above the national average levels if there is a difference. Further, if there were no Finnish heavy metal concentration data available for some foods, EU averages were used, which may not accurately reflect the levels in Finland.

The sensitivity of the analytical measurement techniques also affects the exposure estimates. Particularly for cases where the data are from different laboratories or the LOQ values within a food group are different for other reasons, such as development of new methods, the use of the middle bound estimation may overestimate the relative importance of a food group if it contains quantifiable results as well as some nondetects with high LOQ values. On the other hand, the lower bound estimation is likely to underestimate the exposure to compounds like the heavy metals, which are ubiquitous in nature.

Table 4. Comparison of heavy metal exposure results of this study with exposure reported by EFSA [14–16] for toddlers (1 to <3 years). The exposure is given as lower bound values, and for the current study, the exposure of non-breastfed 1-year-olds is given with the CI of 95% in parentheses. The EFSA calculations for Finnish children are based on the same consumption data as the current study, but the concentration data used by EFSA are predominantly Central European.

Compound, area	Average (μg/kg bw/day)	P95 (μg/kg bw/day)	Reference
Cd, Finland	0.60	1.18	[14]
Cd, median of EU Member State averages	0.56	0.73	[14]
Cd, Finland	0.38	0.64 (CI 0.58–0.73)	This study
Pb, Finland	1.14	2.40	[15]
Pb, median of EU Member State averages	1.08	1.44	[15]
Pb, Finland (*	0.52	0.87 (CI 0.56–1.69)	This study
iAs, Finland	0.32	0.73	[16]
iAs, median of EU Member State averages	0.39	0.86	[16]
iAs, Finland	0.25	0.49 (CI 0.42–0.76)	This study

Note: (*Unlike the middle bound results shown in the Figures, this value was calculated using also milk concentration data from years 2000–2006 in addition to the data from 2006–2012 which had no numerical results. The current lower bound exposure of Finnish toddlers with the same consumption habits would therefore be somewhat lower than this number.

Some foods in this study were represented by only a few samples, which may skew the distribution from the 'real' one. In addition, most of the arsenic data were of total arsenic, and the relative proportion of inorganic arsenic from the total arsenic was estimated based on fixed percentages. While these error sources undoubtedly had an effect, it is impossible to know whether the effect would lead to overestimation or underestimation of the exposure.

The food consumption data also has limitations, as they were collected more than a decade ago and only from one part of the country. All of the children had genetically increased risk of type 1 diabetes, although they were still nondiabetic. The group of still breastfed 1-year-olds was small, which results in a large amount of uncertainty in the probabilistic exposure estimates. The studied children of South-Western Finland were not a representative sample of their age group at national level. However, the used food consumption data were the largest and most recent one collected from Finnish children at the time this assessment was done.

In addition to dietary exposure, heavy metals can enter the body also through pulmonary tract and, especially in young children, as a result of putting non-food things into mouth. A rough estimate of non-dietary exposure was made to put the dietary exposure into context [26]. In a non-smoking environment, the pulmonary exposure to heavy metals is low: less than 1% of TWI or $BMDL_{01}$ for cadmium and lead, and 2.6% of the lower limit of the $BMDL_{01}$ defined by EFSA for inorganic arsenic. Passive smoking will increase the pulmonary exposure, as the smoke of one cigarette, according to Serdar et al. [28], contains 7–350 ng cadmium, 17–980 ng lead and 12–22 ng arsenic.

When playing outside, small children often swallow soil either by accident or deliberately. The exposure from eating Finnish playground soil was estimated in [26] and the arsenic and lead exposure through this habit was found to be comparable to the dietary intake, assuming that the daily

intake of playground soil was 150 mg. It is not likely, however, that the heavy metals in playground soil would be absorbed into the body quite as efficiently as from food.

5. Conclusions

Foods for children, such as ready-to-eat baby foods, porridges for children and follow-on formulae, were an important source of exposure to heavy metals as these foods comprised a large part of the diet of the studied children. However, EU legislation sets lower acceptable limits for heavy metals in these kinds of foods than in other food ingredients, and the baby foods in this study were low in heavy metals. If the children had consumed homemade foods, where the ingredients are subject to the acceptable limits of general food (non-baby food), their heavy metal exposure would likely have been higher because of the higher legislative limits.

Exposure to cadmium from food and tap water exceeded the tolerable weekly intake for a large part of the studied children: according to MB estimates, 56–89% of the studied population were in danger of exceeding the TWI. Exposure to lead in Finland continues to decrease, but still more than half of the studied children (MB estimate) exceeded the benchmark dose for developmental neurotoxicity and the margin of exposure at age group median was below one. Exposure to inorganic arsenic is to a large part due to consumption of rice-based foods like rice, rice porridges and rice drinks. The margin of exposure for inorganic arsenic against the benchmark dose determined by JECFA [7] was at a level corresponding to at least moderate risk.

Breastfed girls had significantly higher exposure to cadmium and inorganic arsenic than the breastfed boys, and the difference in lead exposure also approached significance. The non-breastfed girls also had higher heavy metal exposure than the boys, but the difference was not significant. The boys were significantly heavier than the girls in both studied groups. In comparison, while the 3-year-old and 6-year-old Finnish boys studied in [8] were also significantly heavier than the girls of the same age, the exposure to cadmium and lead among the boys was significantly higher than among the girls.

Decreasing the levels of heavy metals in foods and food ingredients is not easy, as local soil characteristics have a large role in determining the concentrations ending up in primary products. New or tighter permissible levels set in EU legislation for many foods have helped in decreasing the heavy metal concentrations and, as a consequence, also the dietary exposure of Finnish children. In addition, based on the results of this project, a risk management decision was made to communicate to the consumers that a varied (use of different food groups), versatile (use of different foods in a food group) and moderate (serving sizes) diet will both decrease their heavy metal intake and provide adequate nutrition, which will aid in decreasing the absorption of heavy metals.

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Conflict of interest

All authors declare no conflicts of interest in this paper.

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