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# **Trace element contents in foods from the first French Total Diet Study on infants and toddlers**

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## ABSTRACT

Occurrence data for aluminium, antimony, arsenic, barium, cadmium, chrome, cobalt, gallium, germanium, nickel, strontium, silver, tellurium, tin and vanadium were compiled during the first French Total Diet Study on infants and toddlers. For infant foods, meat-/fish-based and vegetable-based ready-to-eat meals were among the most contaminated food categories for most trace elements, except for gallium, antimony and vanadium, for which the concentrations were relatively similar in all food categories. Soups/purees and cereal-based foods had the highest levels of aluminium (653 and 630  $\mu\text{g kg}^{-1}$ , respectively), whereas fruit purees had the highest level of tin (424  $\mu\text{g kg}^{-1}$ ). Infant and follow-on formulae and growing-up milks had relatively low mean contents of trace elements compared with the other infant food categories: e.g. aluminium (220  $\mu\text{g kg}^{-1}$ ), arsenic (1.80  $\mu\text{g kg}^{-1}$ ), cadmium (0.51  $\mu\text{g kg}^{-1}$ ). Chocolate-based foods contributed substantially to the higher levels of aluminium, cadmium, cobalt, chromium and nickel in sweet and savoury biscuits and bars, dairy-based desserts and croissant-like pastries. Only the contribution of chromium and barium levels were statistically different between infant and common foods, with median concentrations being slightly higher in infant foods. The results were largely comparable to those from other surveys on baby food.

**Keywords:** Trace elements; ICP-MS; occurrence data; total diet study; infants and toddlers; food analysis; food composition

## 1. Introduction

Adequate feeding during early childhood is essential for ensuring healthy growth and development. A varied diet can satisfy physiological needs and nutritional requirements, but can also potentially pose health risks due to the higher vulnerability of infants and toddlers. During the first months of life, breast milk is recommended as the most suitable food. However, according to the World Health Organization (WHO), it is estimated that, worldwide, only 34.8% of infants are exclusively breastfed for the first 6 months of their life, the majority consuming infant formula in their early months (WHO, 2009). In France, 23% of infants are still breastfed at 6 months old and less than 2% are exclusively or predominantly breastfed (INVS, 2016) at the same age. Moreover, various foods are gradually introduced at a few months of age. The composition of foods, their raw ingredients, the manufacturing process, the cooking methods, etc. are all sources that can lead to the accumulation of certain chemicals, including trace metals, in baby foods, thus altering their quality. Some trace elements play significant roles in numerous biological functions. Cobalt is one of the components of the B12 vitamin, related to brain activity and the nervous system (Pedron et al., 2016). Vanadium is associated with glucose metabolism and with improving insulin receptivity (Lopez-Garcia et al., 2009). On the other hand, some trace elements (e.g., aluminium, arsenic, etc.) are food contaminants with cumulative properties, and are considered 'potentially' toxic. Moreover, renal immaturity and greater intestinal absorption in children under 12 months of age make infants and toddlers more vulnerable to trace elements (Ikem et al., 2002; Kazi et al., 2010). Widely used in many countries, the Total Diet Study (TDS) is one of the methods recommended by the WHO (WHO, 2006a) for risk assessment. It provides realistic exposure data, because foods are analysed 'as consumed by the consumer', thus facilitating international comparisons of consumer exposure owing to a standardised methodology. However, TDSs are usually carried out on the general population including adults and children, and much less frequently on infants and toddlers. Consequently, there are few data on this specific population. The UK conducted a TDS involving

young children (1.5 - 4.5 years) and adults (Rose et al., 2010), but the occurrence data were not specifically attributed to infant or adult foods.

Moreover, the range of foods is constantly growing and changing for the general populations and for infants, and infant diets are particularly composed of a more restricted range of foods, which can lead to specific dietary exposure. Therefore, collecting contamination data and periodical food monitoring for nutritional and toxicological purposes is important for this young population. Thus, in 2011, the French Agency for Food, Environmental and Occupational Health & Safety (ANSES) undertook the first TDS on infants (from 0 to 12 months old) and toddlers (from 12 months up to 3 years old) to estimate their dietary exposure to food chemicals, including essential and non-essential elements (Hulin et al., 2014).

The aim of this study was to determine the contamination levels of several trace elements (aluminium, antimony, arsenic, barium, cadmium, chromium, cobalt, gallium, germanium, nickel, strontium, silver, tellurium, tin and vanadium) in 291 infant foods and common foods representative of the diet of non-breastfed children during their first three years. The inorganic elements were monitored using a fully validated in-house ISO 17025-accredited method based on microwave digestion and inductively coupled plasma-mass spectrometry (ICP-MS) determination. The analytical methods used have already been described in detail (Chevallier et al., 2015; Guérin et al., 2017, 2018). The separate contribution of common and infant foods to the total average elemental concentrations is given. We also compared our occurrence data with those from other international studies.

## **2. Materials and methods**

### *2.1 Sample collection and preparation*

The establishment of the sampling plan and the preparation of food samples have been described elsewhere (Hulin et al., 2014). Briefly, the food list was set according to the survey on food consumption in children under three years of age conducted by the *Syndicat Français des Aliments*

*de l'Enfance et de la Nutrition Clinique*, © Etude SOFRES 2005/Université de Bourgogne – Pr M. Fantino pour le *Syndicat Français des Aliments de l'Enfance* and described in (Fantino and Gourmet, 2008). The list was based on two main criteria: the most consumed food in terms of quantity and/or percentage of consumers and the foods that are known or supposed to be the main contributors to the exposure to one or more substances of interest. For trace elements, 291 food samples were defined, including 219 infant foods and 71 common foods or bottled water and one sample of reference water. The whole list, covering 89.5 to 99.5% of the diet depending on the age class, is available in (Hulin et al., 2014). Foods were sampled in the region of Clermont-Ferrand (central France) and prepared as consumed by the same operator (i.e. peeled, cooked using tap water, etc. to be 'ready to consume') based on the result of a national study on parents' food preparation/cooking practices carried out by ANSES (Hulin et al., 2014). Therefore, the foods 'as sold' that were not ready to consume were cooked (i.e. pasta, fish) and all the sampled foods were analysed 'as consumed'. To take into account any potential seasonal variability, 12 subsamples of equal weight (80 to 400 g) of the same food were bought every month for one year, frozen and then pooled together by cryogrinding at the end of the sampling period, and put in containers for analysis. The sample of 'reference water' (a specific brand of mineral water, the most used by parents, conditioned in a glass bottle) was used in food preparation for foods that must be diluted (mainly infant and follow-on formulae). Products were diluted according to manufacturers' indications. Therefore, results are expressed for ready-to-eat products and include the trace element contents in the powdered formula and the reference water used for the reconstitution of the product as consumed. To evaluate trace element contents in the reference water, a specific sample composed of every batch of the bottle used was analysed. These individual composite samples (n=291), representative of French infant food habits, were categorised into a total of 36 food categories (11 food categories containing only infant foods and 25 food categories of common foods) for analysis and listed in supplementary material Table **SM-1**.

## *2.2 Reagents and gases*

All solutions were prepared with analytical reagent-grade chemicals and ultra-pure water (18 M $\Omega$ -cm) generated by purifying distilled water with a Milli-QTM PLUS system associated with an Elix 5 water purification system (Millipore S.A., St Quentin en Yvelines, France).

For nitric acid, Suprapur HNO<sub>3</sub> (67% v/v) was purchased from VWR (Fontenay sous Bois, France). The standard solutions of analytes for the calibration procedure were prepared by diluting standard stock solutions of 1000 mg L<sup>-1</sup> of each element purchased from Analytika (Prague, Czech Republic). Working standards were prepared daily in 6% (v/v) HNO<sub>3</sub> and were used without further purification. For internal standard solutions, 1000 mg L<sup>-1</sup> standard stock solutions of scandium, yttrium and rhenium were purchased from Analytika. Regarding certified reference materials (CRM), TORT-2 (Lobster hepatopancreas) and DOLT-4 (Dogfish liver) from the National Research Council Canada and SRM 1548a (Typical diet) from the National Institute of Standards and Technology were purchased from Courtaige Analyses Services (Mont Saint Aignan, France) and from LGC Standards (Molsheim, France). Ultrapure grade carrier (99.9995% pure argon and helium) was supplied by Linde (Montereau, France).

## *2.3 Sample digestion and ICP-MS analysis of samples*

The analyses were carried out using an ICP-MS with a 7700x (Agilent Technologies, Courtaboeuf, France), equipped with a third-generation octopole reaction system (ORS3) using helium as the collision gas. Further details of the instrument settings are given in Table 1. This method has been described elsewhere (Chevallier et al., 2015). Briefly, 0.2 to 2 g of sample was weighed precisely in a quartz digestion vessel and then wet-oxidised with a mixture of 3 mL of ultra-pure water and 3 mL of ultra-pure HNO<sub>3</sub> (67% v/v) in a closed microwave digestion system Multiwave 3000 (Anton-Paar, Courtaboeuf, France). After cooling to room temperature, the digested samples were transferred into 50 mL polyethylene tubes to which a solution of a mixture of internal standards (yttrium, scandium and rhenium) at 2  $\mu$ g L<sup>-1</sup> and ultrapure water were added to the final volume

before analysis using ICP-MS. One randomly selected vessel was filled with reagents only and taken through the entire procedure as a blank.

#### 2.4 Quality control

Several internal quality controls (IQC) and associated criteria were used to ensure the reliability of the results. Each run included a standard calibration, three blanks, three CRMs, two different spiked sample solutions, 33 food samples including 2 samples analysed in duplicate and a mid-range standard analysed for every eight samples and at the end of the sequence. The set criteria were as follows: calibration ( $r^2 > 0.995$ ), blanks (values  $<$  limit of quantification (LOQ)), internal standards (values within 70 and 130% of the target value), mid-range standards (values within 80 and 120% of the target value), spiked standard solutions (spike recovery within 70 and 130% of the theoretical spiked standard value), CRMs (Z-score  $< \pm 2$ ) and duplicates (acceptable if the relative standard deviation (RSD)  $\leq 20\%$  when mean value  $\geq 5 \times$  LOQ or RSD  $\leq 40\%$  when mean value  $\geq$  LOQ and  $< 5 \times$  LOQ). When acceptance criteria were not met, the results were discarded and the samples were re-analysed (Millour et al, 2010).

To check trueness, three CRMs (TORT-2, DOLT-4 and SRM 1548a) were chosen to cover the maximum of elements of interest (12 out of 15) and were analysed every run throughout the study (10 runs in total), except for gallium, germanium and tellurium (no CRMs were available, but trueness of these elements was monitored using spiked test samples). The results of CRMs were corrected for moisture content. Each individual result, as well as the measured means, was compared to the confidence interval (CI) around the reference or indicative value calculated as:

$$CI = M \pm \left[ k * \frac{CV_R * M}{100 * \sqrt{n}} \right]$$

with  $k=3$ ,  $n=1$  sample,  $M$ , the reference or indicative value of the CRM and  $CV_R$ , the coefficient of variation of reproducibility estimated previously (Chevallier et al., 2015). All the results were within the CI (**Table 2**), except one value for silver in DOLT-4, the results related to this run were

discarded and the samples were re-analysed. For gallium, germanium and tellurium, three different spiked standard solutions, covering the concentration range of the method (1 - 5 - 10  $\mu\text{g L}^{-1}$ ) were included in random samples in the runs. The samples were spiked before digestion and underwent the complete analytical procedure. The analysis was carried out on the samples before and after spiking. All the results fell in the range of 70-130% of the target value (**Table 3**).

### *2.5 Statistical analysis and data management*

The descriptive statistics including the arithmetical means, standard deviation (SD), minimum (min), maximum (max) of samples of food categories were calculated using Microsoft Excel 2010 software. The management of left-censored (LC) data (results reported below the limit of detection (LOD) and/or LOQ) were managed by adapting the WHO recommendations (WHO, 2013), i.e. by using a lower-bound hypothesis (LB) and an upper-bound hypothesis (UB). Values lower than LOD were replaced by 0 under LB or by the LOD under UB, and values lower than the LOQ, but higher than the LOD were replaced by the LOD under LB or the LOQ under UB. Because mean LB and UB concentrations were generally quite similar or equal due to the low LOD/LOQ values in this study, we present and discuss the results according to the UB approach. The LB results are available in the supplementary material (Table **SM-2**). For each element, a mean value was calculated for infant foods and for common foods. Their sum represents 100% of the total element content in infant feeding. Then the percentage contribution (%) of each food type (i.e. infant foods and common foods) to the total element content was calculated. Moreover, we carried out statistical comparisons of the median element contamination values in infant and common foods and some of food categories. Given that the infant and common foods were composed of distinct food categories, that the sample populations were of different sizes and the non-normal distribution of the data, data analysis was performed using a non-parametric test on the median contamination values. Mann-Whitney and Kruskal-Wallis tests were applied for comparison of two or more

groups, using StatGraphics Software Centurion XVII.I. purchased from Francestat (Neuilly-sur-Seine, France).

All analytical results are reported in micrograms per kilogram on a fresh weight basis (f.w.) (foods as consumed).

### 3. Results and Discussion

**Table 4** shows the limits of quantification and detection ( $LOQ = 2 \times LOD$ ), based on 21 reagent blanks analysed in accordance with the NF EN 13804 standard (AFNOR, 2013)), as well as the percentage of detection and quantification of the investigated elements in the analysed samples. The mean concentrations and SD of the trace elements in the 36 food categories are given in **Table 5**. Depending on the element and the food category, SD can vary considerably, due to the composition of different food samples belonging to the same category. The results are discussed for each element separately and compared with those from other countries, when available. Considering that only two samples were quantified for tellurium (rice ( $2 \mu\text{g kg}^{-1}$ ) and baby cereal with vegetables ( $0.14 \mu\text{g kg}^{-1}$ )), four samples for germanium (instant hot chocolate drink, baby cereal with biscuit and cocoa, baby cereal with vegetables and rice ( $0.041 - 0.072 - 0.072 - 1.00 \mu\text{g kg}^{-1}$ , respectively)) and no samples for silver on the 291 analysed food samples, these elements are not discussed below. No results for these elements have been reported in previous studies, except for silver which was not detected in a USA survey (Ikem et al., 2002) and similar low germanium levels ( $0.4$  to  $2 \mu\text{g kg}^{-1}$ ) were reported in a UK TDS (Rose et al., 2010).

#### 3.1 Aluminium

Of the 291 food samples analysed, 85.9% had quantifiable aluminium levels and the mean levels ranged from 42 to  $10,000 \mu\text{g kg}^{-1}$  for common foods and from 189 to  $653 \mu\text{g kg}^{-1}$  for infant foods. The highest mean levels in common foods were observed in decreasing order in sweet and savoury

biscuits and bars ( $10,000 \mu\text{g kg}^{-1}$ ;  $n=1$  in dry chocolate biscuits), dairy-based desserts ( $5950 \mu\text{g kg}^{-1}$ ;  $n=2$ , with the highest level of  $9350 \mu\text{g kg}^{-1}$  found in refrigerated chocolate mousse), croissant-like pastries ( $4300 \mu\text{g kg}^{-1}$ ;  $n=2$ , with the highest level of  $6710 \mu\text{g kg}^{-1}$  found in chocolate croissants), and cheese ( $4220 \mu\text{g kg}^{-1}$ ;  $n=1$ ). Among infant foods, the highest aluminium contents were found in soups/purees ( $653 \mu\text{g kg}^{-1}$ ;  $n=11$ ) and cereal-based foods ( $630 \mu\text{g kg}^{-1}$ ;  $n=17$ ). This latter category showed the highest aluminium levels due to two samples (of different brands) of biscuits for infants ( $3800$  and  $2580 \mu\text{g kg}^{-1}$ ) and two samples of baby cereal with biscuit and cocoa ( $1090$  and  $695 \mu\text{g kg}^{-1}$ ). Three food categories – fruit purees, meat-/fish-based ready-to-eat meals, and vegetable-based ready-to-eat meals – had comparable aluminium contents ( $556$ ,  $597$  and  $575 \mu\text{g kg}^{-1}$ , respectively) ( $p>0.05$ ). Particularly high contents were found in some green vegetable-based foods in the two ready-to-eat meal categories (e.g.  $2590 \mu\text{g kg}^{-1}$  in a spinach and salmon meal and  $2480 \mu\text{g kg}^{-1}$  in a spinach and pasta dinner). The other remaining food categories had aluminium levels ranging from  $189 \mu\text{g kg}^{-1}$  (growing-up milks) to  $328 \mu\text{g kg}^{-1}$  (milk-based beverages). Mean aluminium levels in infant and follow-on formulae in this study ( $196$  and  $276 \mu\text{g kg}^{-1}$ ) were respectively similar and higher compared with those from a USA study in the same food categories ( $168$  and  $35 \mu\text{g kg}^{-1}$ ) (Ikem et al., 2002) which reported higher levels in soy-based powder formulae ( $460 \mu\text{g kg}^{-1}$ ) as in our study in a sample of a soy-based follow-on powder formula ( $713 \mu\text{g kg}^{-1}$ ). A Norwegian study (Melo et al., 2008) reported similar aluminium contents in fruit purees category ( $559 \mu\text{g kg}^{-1}$ ) and mean aluminium contents ranged from  $350$  to  $1690 \mu\text{g kg}^{-1}$  in samples of ready-to-eat dinners (containing vegetables, meats, etc.). The results are comparable to or lower than those reported in the 2008 European Food Safety Authority scientific (EFSA) opinion (EFSA, 2008), which indicated that high aluminium mean levels ( $5000$  to  $10,000 \mu\text{g kg}^{-1}$ ) were often found in breads, cakes and pastries (with biscuits having the highest levels), some vegetables such as spinach, and manufactured foods, especially for those intended for infants. EFSA (2008) also reported that, in some brands of infant formula (both milk-based and soy-based), the aluminium content was around four times higher than the mean concentrations estimated above, which may lead to higher

aluminium exposure in infants. Aluminium contamination can be attributed to the use of additives (EFSA, 2008) such as baking powder, firming agents and anti-caking agents in many food products (Yeh et al., 2016) and calcium salts and soy protein in some fortified infant formulae (Ikem et al., 2002). Moreover, the contamination through aluminium utensils, containers and manufacturing processes cannot be excluded (EFSA, 2008). In our study, cocoa-/chocolate-based foods contained high aluminium levels compared with other common and infant foods, which is probably related to the aluminium content in chocolate, as also reported previously (Stahl et al., 2011; Yeh et al., 2016).

### *3.2 Antimony*

Of the 291 food samples analysed, antimony values were quantified in only 11.3% of them, (6.4% for infant foods and 26.3% for common foods). Antimony levels were quantified in individual samples at relatively low concentrations, from LOQ ( $1 \mu\text{g kg}^{-1}$ ) to 3 x LOQ ( $3 \mu\text{g kg}^{-1}$ ) for the infant foods, and ranged on average from 0.50 to  $0.75 \mu\text{g kg}^{-1}$  in these foods. The highest mean levels were found in fruit juices, cereal-based foods and fruit purees ( $0.75$ ,  $0.72$  and  $0.65 \mu\text{g kg}^{-1}$ , respectively), the other remaining food categories had mean contents ranging from 0.50 to  $0.58 \mu\text{g kg}^{-1}$ . For the common foods, mean antimony content was measured from  $0.37 \mu\text{g kg}^{-1}$  to 6 x LOQ, with the highest mean levels found in sugar and sugar derivatives and cheese ( $6.00$  and  $5.00 \mu\text{g kg}^{-1}$ , respectively). Only a few studies have reported data on antimony. Our results in infant/follow-on formulae and cereal-based foods categories, were respectively higher than and lower than values from Sweden (Ljung et al., 2011) ( $0.03$  to  $2.8 \mu\text{g kg}^{-1}$  in infant formulae and infant cereal-based foods samples) and slightly lower than those from the USA (Ikem et al., 2002), where antimony levels ranged from ND to  $14 \mu\text{g kg}^{-1}$  in milk-based first liquid formulae and soy-based powder formulae categories. Our results corroborate the report that antimony is present in foods, including vegetables, grown on antimony-contaminated soils, mostly in the low  $\mu\text{g kg}^{-1}$  wet weight range or less (WHO, 2003). This contamination can lead to a low dietary exposure. However, considering

that antimony is a toxic element and in light of the lack of data, particularly on infant foods, it is important to collect occurrence data to obtain an associated risk assessment.

### *3.3 Arsenic*

Arsenic was quantified in 44.3% of the 291 food samples analysed. Apart from a relatively high arsenic content measured in fish ( $2750 \mu\text{g kg}^{-1}$ ), the mean highest concentrations were found, as expected, in rice-/cereal-based and fish-based foods. The highest arsenic levels were found, in decreasing order, in rice and wheat products (common food) ( $30.0 \mu\text{g kg}^{-1}$ ), followed by meat-/fish-based ready-to-eat meals (infant food) ( $27.5 \mu\text{g kg}^{-1}$ ) and in the following common foods: bread and dried bread products ( $12.5 \mu\text{g kg}^{-1}$ ), mixed dishes ( $11.0 \mu\text{g kg}^{-1}$ ) and eggs and egg products ( $11.0 \mu\text{g kg}^{-1}$ ). In the meat-/fish-based ready-to-eat meal food category, the highest mean levels were found in salmon with sorrel and rice, hake and rice, vegetable and hake vegetable and tropical sole and carrots and cod ( $78, 95, 123, 216$  and  $411 \mu\text{g kg}^{-1}$ , respectively). For the remaining food categories, mean concentrations were quite low and ranged from  $0.49 \mu\text{g kg}^{-1}$  (hot beverages) to  $8.50 \mu\text{g kg}^{-1}$  (poultry and game) for the common foods and from  $1.17 \mu\text{g kg}^{-1}$  (milk-based desserts) to  $4.82 \mu\text{g kg}^{-1}$  (soups/purees) for the infant foods. The arsenic concentrations in the infant foods in this study (range  $1.17$  to  $27.5 \mu\text{g kg}^{-1}$ ) were slightly higher (around 2-fold) than data from Brazil (Pedron et al., 2016) (ranging from  $0.1$  to  $11.6 \mu\text{g kg}^{-1}$  in rice-based and non-rice-based baby food categories) and from the USA (Jackson et al., 2012) (ranging from  $0.3$  to  $12.6 \mu\text{g kg}^{-1}$  in infant formulae and fruit/vegetable purees categories), but much lower than those reported from Spain (ranging from  $49$  to  $479 \mu\text{g kg}^{-1}$  in infant rice and pureed infant food samples and from  $40$  to  $2610 \mu\text{g kg}^{-1}$  in fish-based baby food samples) (Burlo et al., 2012; Vinas et al., 1999). The highest contributors to arsenic contamination were rice-based and fish-based ready-to-eat-meal foods. However, risk assessment generally involves inorganic arsenic, which is the most toxic arsenic species and is classified as carcinogenic for humans (IARC, 2012). Consequently, infant foods showing the highest inorganic arsenic content should be taken into consideration for risk assessments.

Rice/cereal-based products are known to contribute to inorganic arsenic content more than fish in which a non-toxic species – arsenobetaine – is dominant (Sirot et al., 2009). In fact, several studies have reported that non-rice-based infant foods contain less inorganic arsenic compared with rice-based foods (Carbonell-Barrachina et al., 2012; Meharg et al. 2008). An EFSA opinion also reported that rice-containing foods, cereal-based food for infants and young children (with rice) and ready-to-eat meals for children and cereal-based meals (with rice) have the highest mean inorganic arsenic contents (middle bound: MB=133.1 and 107.5  $\mu\text{g kg}^{-1}$ , respectively) and that the consumption of three portions (90 g day<sup>-1</sup>) of rice-based infant food represents a substantial source of inorganic arsenic. Thus, the elevated levels of total arsenic/inorganic arsenic in rice-based infant foods require special attention because, although they are not the most consumed foods, they can lead to high inorganic arsenic exposure (EFSA, 2014a). Another EFSA study (EFSA, 2009a) reported mean LB/UB arsenic levels of 149.6/157.5  $\mu\text{g kg}^{-1}$  in rice-based infant foods versus 2.3/2.9  $\mu\text{g kg}^{-1}$  in infant and follow-on formulae without cereal products, and 33.2/50.7  $\mu\text{g kg}^{-1}$  in cereal-based foods without rice. Finally, in 2015, the European Commission (Council of the European Union, 2015) established maximum levels (ML) of inorganic arsenic in rice and rice-based products (rice waffles, rice cakes, parboiled and husked rice, etc.) from 0.10 to 0.30 mg kg<sup>-1</sup>, depending on the product. A specific ML of 0.10 mg kg<sup>-1</sup> was set for rice intended for the production of foods intended for infants and young children, but we could not compare our results with this value because MLs apply only to raw commodities.

### *3.4 Barium*

Of the 291 food samples analysed, 87.6% had quantifiable barium levels. The mean concentration of barium found in this study ranged from 25.0  $\mu\text{g kg}^{-1}$  in butter, sugars and sugar derivatives, meat and poultry and game to 1080  $\mu\text{g kg}^{-1}$  in the sweet and savoury biscuits and bars. Mean barium levels were similar ( $p>0.05$ ) and ranged from 417 to 615  $\mu\text{g kg}^{-1}$  in the following food categories: bread and dried bread products, croissant-like pastries, dairy-based desserts, pasta, fruits and,

finally, vegetables (excluding potatoes). In the latter two food categories, the highest levels were found in kiwi fruit ( $1096 \mu\text{g kg}^{-1}$ ) and in carrots ( $1116 \mu\text{g kg}^{-1}$ ), respectively. These high concentrations in kiwi and carrot are in accordance with previous studies (Hadayat et al., 2018; Millour et al., 2012). Among the infant foods, barium content ranged from  $58.9 \mu\text{g kg}^{-1}$  in growing-up milk, to  $337 \mu\text{g kg}^{-1}$  in vegetable-based ready-to-eat meals, with the highest mean level found in carrots:  $1450 \mu\text{g kg}^{-1}$ . The barium levels in fruit purees, soups/purees, meat-/fish-based ready-to-eat meals and fruit juices ranged from 184 to  $317 \mu\text{g kg}^{-1}$ , and in follow-on and infant formulae, milk-based desserts and cereal-based foods, they were similar ( $p > 0.05$ ) and ranged from 111 to  $118 \mu\text{g kg}^{-1}$ . Contamination of vegetables may occur via root uptake, which is influenced by environmental pollution and industrial and human activities. This occurrence in soil can lead to higher contamination of the food categories containing fruits and vegetables. In surveys conducted in the UK (Zand et al., 2012) and the USA (Ikem et al., 2002), barium concentrations range from  $< 53$  (LOQ) to  $290 \mu\text{g kg}^{-1}$  (chicken- and fish-based infant food samples) and from 30 to  $68 \mu\text{g kg}^{-1}$  (infant formulae category), respectively, which is in agreement with the present study (meat-fish based ready-to-eat meals and infant/follow-on formulae food categories). Most of foods contain less than  $2000 \mu\text{g kg}^{-1}$  but, due to the accumulation of barium in plants, some plants such as legumes, cereals and nuts may contain high barium levels (e.g. pecans,  $6700 \mu\text{g kg}^{-1}$ ) (ATSDR, 2007; WHO, 2004), which is consistent with our findings. Furthermore, the barium levels in foods and drinking waters are typically too low to be of concern; consequently, the dietary barium exposure of the general population is typically low. However, the lack of data regarding infant exposure does not allow a reliable barium exposure assessment for this population (ATSDR, 2007).

### *3.5 Cadmium*

Of the 291 food samples analysed, 57% had quantifiable cadmium levels. The mean concentration of cadmium found in this survey ranged from  $0.30 \mu\text{g kg}^{-1}$  in fruit juices, butter, bottled water and milk to  $25.0 \mu\text{g kg}^{-1}$  in potatoes and potato products. In the common foods, the highest mean levels

were found, in decreasing order, in potatoes and potato products, sweet and savoury biscuits and bars, pasta, bread and dried bread products and, finally, vegetables (excluding potatoes) (25.0, 22.0, 16.0, 15.5 and 15.5  $\mu\text{g kg}^{-1}$ , respectively). In the infant foods, the most contaminated categories were meat-/fish-based ready-to-eat meals and vegetable-based ready-to-eat meals (9.31 and 9.26  $\mu\text{g kg}^{-1}$ , respectively), followed by soups/purees (7.36  $\mu\text{g kg}^{-1}$ ) and cereal-based foods (2.79  $\mu\text{g kg}^{-1}$ ). The other remaining food categories (infant formulae, fruit juices, fruit purees and milk-based products) had low contents ranging from 0.30 to 1.88  $\mu\text{g kg}^{-1}$ . Vegetables and wheat/rice products, identified to be among the highest contributors to cadmium intake by the Joint FAO/WHO Expert Committee on Food Additives and Contaminants (WHO, 2001), were the most contaminated categories in the common and infant foods. Sweet and savoury biscuits represented by dry chocolate biscuits were also high contributors to cadmium content, probably due to their chocolate content. Cadmium in infant and follow-on formulae and growing-up milks presented low levels (0.39, 0.43 and 0.71  $\mu\text{g kg}^{-1}$ , respectively), as well as milk-based desserts and milk-based beverages (0.65 and 1.88  $\mu\text{g kg}^{-1}$ , respectively). Although the cadmium content was low in these food categories, they may contribute to dietary cadmium intake. Indeed, (EFSA, 2009b) reported that the high consumption of milk and dairy products contributes to high cadmium intake among toddlers. However, cereals and cereal-based products, vegetables, nuts and pulses still dominate as cadmium sources, even for this age group. In addition, infant formulae and cereal foods contribute significantly to exposure in infants and young children (EFSA, 2012). Infant formulae have been reported to contain cadmium at concentrations lower than 1.2  $\mu\text{g kg}^{-1}$  (Ikem et al., 2002; Ljung et al., 2011; Pedron et al., 2016) in various studies from different countries which is in agreement with the present study. Higher mean levels have been measured in infant formulae (10.75  $\mu\text{g kg}^{-1}$ ) in an Italian survey (Bargellini et al., 2018) and in chicken-based infant food samples (22  $\mu\text{g kg}^{-1}$ ) in a UK survey (Zand et al., 2012). Finally, the EFSA opinion (EFSA, 2012) estimated average LB/UB cadmium in food for infants and small children at 14.2/14.8  $\mu\text{g kg}^{-1}$ , with the highest level found in cereal-based foods for children (11.9/12.6  $\mu\text{g kg}^{-1}$ ) and a lower level in liquid follow-on formulae

(1.0/1.4  $\mu\text{g kg}^{-1}$ ), which is consistent with our results. All cadmium concentrations observed in individual samples in the follow-on formula, growing-up milk and infant formula categories were far below the ML of 0.005  $\text{mg kg}^{-1}$  set by Commission Regulation (EC) No 488/2014 (Council of the European Union, 2014), in liquid formulae manufactured from cows' milk proteins or protein hydrolysates. This ML (the lowest among infant formulae and follow on-formulae MLs) is the most suitable regarding the samples of the present study. Similarly, no results exceeded the ML of 0.040  $\text{mg kg}^{-1}$  set for processed cereal-based foods and baby foods for infants and young children.

### *3.6 Chromium*

Chromium was present in almost all samples with 93.2% of quantification (69.4% in the common foods and 95.8% in the infant foods), and ranged from 6.25 to 232  $\mu\text{g kg}^{-1}$  in the common foods and from 20.8 to 68.9  $\mu\text{g kg}^{-1}$  in the infant foods. In the common foods, the highest mean chromium levels were quantified, in decreasing order, in sweet and savoury biscuits and bars, dairy-based desserts, cheese and croissant-like pastries at 232, 178, 111 and 95  $\mu\text{g kg}^{-1}$ , respectively. These high levels can be attributed to the samples containing chocolate in these food categories (dry chocolate biscuit: 178  $\mu\text{g kg}^{-1}$ , refrigerated chocolate mousse: 280  $\mu\text{g kg}^{-1}$  and chocolate croissant: 162  $\mu\text{g kg}^{-1}$ ), except for the cheese category. This is consistent with the high chromium concentrations observed in chocolate products which can increase the mean occurrence values in certain food categories (EFSA, 2014b). In the infant foods, the two most contaminated food categories were the meat-/fish-based ready-to-eat meals (68.9  $\mu\text{g kg}^{-1}$ ; n= 45) and the vegetable-based ready-to-eat meals (50.4  $\mu\text{g kg}^{-1}$ ; n=27), for which all the samples showed contents ranging from 20 to 155  $\mu\text{g kg}^{-1}$  and from 16 to 92  $\mu\text{g kg}^{-1}$ , respectively. For the remaining food categories, concentrations ranged from 20.8  $\mu\text{g kg}^{-1}$  (infant formulae) to 42.7  $\mu\text{g kg}^{-1}$  (fruit purees). Previous studies from Norway (Melo et al., 2008), USA (Ikem et al., 2002), Spain (Sola-Larranaga and Navarro-Blasco, 2006) and Italy (Bargellini et al., 2018), reported maximum chromium levels in infant formulae of 0.2, 15, 15.02 and 37.5  $\mu\text{g kg}^{-1}$ , respectively, which is in agreement with our findings (milk-based

products and infant/follow-on formulae food categories), except for the low value of  $0.2 \mu\text{g kg}^{-1}$ . Chromium levels reported in infant ready-to-eat dinners category (ranging from  $32.3$  to  $101 \mu\text{g kg}^{-1}$ ) (Melo et al., 2008) and rice-based and non-rice-based babyfood categories in Brazil ( $3.2$  -  $35.2 \mu\text{g kg}^{-1}$ ) (Pedron et al., 2016) were also comparable to those in the present study (meat-/fish- and vegetable-based ready-to-eat meals), whereas those reported in fish-based and chicken-based ready-to-eat infant food samples in the UK (range  $50$  to  $310$  and  $50$  to  $1900 \mu\text{g kg}^{-1}$ , respectively) (Zand et al., 2012), were higher. The 2014 EFSA opinion (EFSA, 2014b) indicated that the main sources of chromium in foods were products for special nutritional use, herbs, spices and condiments, sugar and confectionery – due to the presence of chocolate/cocoa products –, and vegetables and vegetable products. It also stated that the contamination during food processing (contact with stainless steel) may contribute to chromium levels. The chromium LB/UB level of  $52/64 \mu\text{g kg}^{-1}$  and  $0.9/3.4 \mu\text{g L}^{-1}$  determined in foods for infants and small children and bottled water respectively, were consistent with the values in the present study. The dietary exposure to chromium has been assessed for chromium (III) and chromium (VI) content and the CONTAM panel (EFSA, 2014b) considered the analytical results in food as chromium (III) (because food is by and large a reducing medium) and chromium present in drinking water as chromium (VI) (due to oxidative water treatments to make water potable). The IARC (2012) classified chromium (VI) as carcinogenic in 2012. Moreover, EFSA, 2014b stated that although the chromium (III) had been recognised as a beneficial nutrient for a long time, it is no longer considered as an essential element.

### *3.7 Cobalt*

Of the 291 food samples analysed, 62.2% had quantifiable levels of cobalt, with the highest mean levels found, in decreasing order, in the common food categories of sweet and savoury biscuits and bars ( $58.0 \mu\text{g kg}^{-1}$ ;  $n=1$ ), dairy-based desserts ( $40.0 \mu\text{g kg}^{-1}$ ;  $n=2$ ) and croissant-like pastries ( $19.5 \mu\text{g kg}^{-1}$ ;  $n=2$ ). These high levels were likely due to the presence of chocolate in some samples:  $58 \mu\text{g kg}^{-1}$  in dry chocolate biscuits,  $62 \mu\text{g kg}^{-1}$  in refrigerated chocolate mousse and  $35 \mu\text{g kg}^{-1}$  in

chocolate croissants from the three food categories cited above, respectively. The other foods categories had contents varying from 0.70 to 8.69  $\mu\text{g kg}^{-1}$ . In the infant foods, the highest mean levels were found in milk-based desserts (5.62  $\mu\text{g kg}^{-1}$ ; n=6), with the highest contents found in two brands of dairy-based desserts (10 and 20  $\mu\text{g kg}^{-1}$ ), followed by meat-/fish-based ready-to-eat meals (3.82  $\mu\text{g kg}^{-1}$ ; n=45) and vegetable-based ready-to-eat meals (3.69  $\mu\text{g kg}^{-1}$ ; n=27), for which all the samples were quantified between 1.00 and 7.00  $\mu\text{g kg}^{-1}$ . The other foods categories had contents lower than 3.05  $\mu\text{g kg}^{-1}$  with the lowest mean cobalt content found in a growing-up milk category (0.90  $\mu\text{g kg}^{-1}$ ). The values of cobalt are similar to those previously reported in Norway (in infant formulae, ready-to-eat dinners and fruit purees categories) and Brazil for baby food puree category (Melo et al., 2008; Pedron et al., 2016) with a maximum level of 6.3  $\mu\text{g kg}^{-1}$ . Cobalt is an essential micronutrient that must be consumed in adequate amounts to maintain physiological functions, but may be toxic if taken up in excessive amounts. In this study, among the infant foods, growing-up milks and infant formulae which are mostly consumed during early childhood were the foods that contributed the least to this intake. However, chocolate-based products (common or infant foods) seem to be among the highest source of cobalt content.

### 3.8 Gallium

Only 6.9% of the samples had quantifiable gallium levels in this study. The highest mean levels in the common foods were found, in decreasing order, in sweet and savoury biscuits and bars, dairy-based desserts, croissant-like pastries and vegetables (excluding potatoes) (3.00, 2.00, 1.25 and 1.19  $\mu\text{g kg}^{-1}$ , respectively), and, for the infant foods, in cereal-based foods, vegetable-based ready-to-eat meals and growing-up milks (0.62, 0.57 and 0.56  $\mu\text{g kg}^{-1}$ , respectively). The remaining food categories had mean gallium contents not exceeding 0.53  $\mu\text{g kg}^{-1}$  in the infant foods and 1.00  $\mu\text{g kg}^{-1}$  in the common foods. The lowest levels were 0.45  $\mu\text{g kg}^{-1}$  (hot beverages) and 0.50  $\mu\text{g kg}^{-1}$  (milk-based beverages, milk-based desserts, fruit juices, soups, purees and infant formulae), in the common foods and infant foods, respectively. No data on gallium have been reported in surveys

from other countries. Gallium has been quantified at low levels in only a few food categories in infant and common foods, but because it is considered carcinogenic (IARC 2006), it is important to collect contamination data for dietary risk assessment.

### 3.9 Nickel

Of the 291 food samples analysed, 37.1% of nickel levels were quantifiable, providing a high percentage of censored data. The highest mean levels were found in the common food categories, mainly due to the contribution of samples containing chocolate: sweet and savoury biscuits and bars ( $527 \mu\text{g kg}^{-1}$  in dry chocolate biscuit), dairy-based desserts ( $388 \mu\text{g kg}^{-1}$ ), with the highest level found in a sample of refrigerated chocolate mousse ( $605 \mu\text{g kg}^{-1}$ ), croissant-like pastries ( $173 \mu\text{g kg}^{-1}$ ), with the maximum found in a chocolate croissant ( $295 \mu\text{g kg}^{-1}$ ) and in hot beverages ( $96.2 \mu\text{g kg}^{-1}$  in an instant hot chocolate drink). In the infant foods, the highest nickel levels were found, in decreasing order, in meat-/fish-based ready-to-eat meals ( $75.7 \mu\text{g kg}^{-1}$ ), vegetable-based ready-to-eat meals ( $71.5 \mu\text{g kg}^{-1}$ ), soups/purees ( $57.7 \mu\text{g kg}^{-1}$ ) and fruit purees ( $54.7 \mu\text{g kg}^{-1}$ ). In these food categories, nickel levels in individual samples were quite low and ranged from 25.0 to  $143 \mu\text{g kg}^{-1}$ , as in the remaining food categories, where values were lower than  $51.0$  and  $69.5 \mu\text{g kg}^{-1}$  in infant foods and common foods, respectively. Several studies have shown similarly low levels: not detected (ND) to  $25.5 \mu\text{g kg}^{-1}$  in infant formulae (Bargellini et al., 2018), from ND to  $125 \mu\text{g kg}^{-1}$  in infant foods (infant formulae, ready-to-eat dinners and fruit purees categories) (Melo et al., 2008) and from  $100$  to  $200 \mu\text{g kg}^{-1}$  in solid food and beverages categories (Pandelova et al., 2012). Slightly higher levels were found in ready to-eat infant food samples (from  $< 80$  to  $410 \mu\text{g kg}^{-1}$ , Zand et al., 2012) and much lower levels in infant formulae category (from ND to  $2 \mu\text{g kg}^{-1}$ , Ikem et al., 2002). Our results were also in agreement with an EFSA opinion (EFSA, 2015) that reported nickel concentrations in common foods less than  $500 \mu\text{g kg}^{-1}$ , nickel levels in baby foods (reported in previous European studies) ranging from  $100$  to  $1300 \mu\text{g kg}^{-1}$  and in breast milk and water concentrations generally below  $10 \mu\text{g L}^{-1}$ . Current data from the EFSA opinion indicate that nickel

mean LB/UB are 126/152  $\mu\text{g kg}^{-1}$  in food for infants and small children ( $n = 309$ ) and 1/2  $\mu\text{g kg}^{-1}$  in drinking water ( $n = 25,700$ ). These values are two to four times higher than those of the present work for the infant foods included in the study (range: 25 - 75.7  $\mu\text{g kg}^{-1}$ ). Among the common foods, the main contributors to nickel content were sugar and confectionary (related to chocolate-based products) and legumes, nuts and oilseeds (mean LB/UB: 1504/1586 and 1862/1880  $\mu\text{g kg}^{-1}$ , respectively). Nickel contamination in certain foods and more generally in processed foods can be attributed to food preparation techniques (cooking utensils in stainless steel) (IARC, 2012).

### *3.10 Strontium*

The only unquantified sample in the present work was sugar (the 'sugar and sugar derivatives' food category). In the other food categories, strontium mean levels ranged from 50  $\mu\text{g kg}^{-1}$  (poultry and game) to 2180  $\mu\text{g kg}^{-1}$  (bottled water) in the common foods and from 221  $\mu\text{g kg}^{-1}$  (cereal-based foods) to 695  $\mu\text{g kg}^{-1}$  (milk-based beverage) in the infant foods. Strontium contents were widely found in the 12 samples of bottled water and ranged from 18  $\mu\text{g kg}^{-1}$  to 9270  $\mu\text{g kg}^{-1}$ , due to its natural occurrence in surface water (geologically variable). In the other common food categories, strontium levels were quantified with the highest mean levels found, in decreasing order, in cheese (1980  $\mu\text{g kg}^{-1}$ ;  $n=1$ ), vegetables (excluding potatoes) (1590  $\mu\text{g kg}^{-1}$ ;  $n=8$ ), with the highest levels found in spinach and broccoli samples (3470 and 2120  $\mu\text{g kg}^{-1}$ , respectively), fish (1410  $\mu\text{g kg}^{-1}$ ;  $n=3$ ), sweet and savoury biscuits and bars (1300  $\mu\text{g kg}^{-1}$ ;  $n=1$ ) and fruit (1160  $\mu\text{g kg}^{-1}$ ;  $n=6$ ), with the highest levels found in fresh orange and clementine or mandarin samples (2860 and 2250  $\mu\text{g kg}^{-1}$ , respectively). These results corroborate the fact that the primary strontium exposure sources are drinking water, vegetables, grains and dairy products, and the strontium concentrations in these ingredients can vary widely (WHO, 2010). For the infant foods, categories similar to those cited above had the highest strontium levels. Mean strontium contents were quantified in milk-based beverages, soups/purees, meat-/fish-based ready-to-eat meals, vegetable-based foods and vegetable-based ready-to-eat meals at 695, 581, 580 and 568  $\mu\text{g kg}^{-1}$ , respectively. The other food categories

had strontium contents ranging from 221 to 512  $\mu\text{g kg}^{-1}$ . In some food categories, the strontium results varied up to six times between the lowest and the highest value, likely reflecting, the variability of strontium content in the raw ingredients (fruits, vegetable, cereals, water, etc.) used in the composition of the final product. A previous study reported concentrations ranging from 160 to 220  $\mu\text{g kg}^{-1}$  in milk-based and soy-based infant formulae categories (Ikem et al., 2002), which are lower than our findings in the same category.

### *3.11 Tin*

Tin concentrations were by and large quite low: tin was quantified only in 4.5% of the samples in this study. The most contaminated food category was an infant food: fruit purees, with a mean content of 424  $\mu\text{g kg}^{-1}$ . In this food category (n=30), the quantified results were quite similar and varied from 98 to 244  $\mu\text{g kg}^{-1}$ , except for four samples with relatively high contents of 1970, 2280, 3320 and 3330  $\mu\text{g kg}^{-1}$  in mixed (exotic) fruit purees (2 different brands) and apple-kiwi purees, respectively. These high values could not be attributed to possible contamination from packaging, because these four samples were packaged in plastic, glass or a mixture of both; only metal cans often contain tin. Similarly, contamination cannot be traced to a specific brand, because the samples were of different brands. The other food categories with quantifiable tin contents showed means varying from 42.0 to 62.5  $\mu\text{g kg}^{-1}$ . In the common foods, apart from two food categories – vegetables (excluding potatoes) and potatoes and potato products with mean values of 63.9 and 63.3  $\mu\text{g kg}^{-1}$  respectively – tin was not detected in the other food categories. A previous study reported low tin concentrations ranging from 29.0 to 85.0  $\mu\text{g kg}^{-1}$ , in milk-based and soy-based infant formulae categories (Ikem et al., 2002), which is consistent with the present work, if we exclude the extreme high values measured in fruit purees. Previous European studies have reported tin levels ranging from ND to 300  $\text{mg kg}^{-1}$  and generally less than 1  $\text{mg kg}^{-1}$  in unprocessed foods. Tin intake from the diet depends greatly on the type and amount of canned food consumed (WHO, 2005; 2006b). However, in this study, few samples were packaged in metal cans (growing-up milks, infant

formulae categories); likewise, their tin concentrations were far below the ML of  $50 \text{ mg kg}^{-1}$ , set by Commission Regulation (EC) No 1881/2006 (Council of the European Union, 2006) in canned infant formulae and follow-on formulae (including infant milk and follow-on milk), excluding dried and powdered products.

### *3.12 Vanadium*

Vanadium was present in 75.3% of the samples, albeit at low levels. The highest vanadium contents were found in the common food categories, in decreasing order, of sweet and savoury biscuits and bars, cheese, dairy-based desserts, and croissant-like pastries ( $37.0$ ,  $36.0$ ,  $23.5$  and  $12.0 \text{ } \mu\text{g kg}^{-1}$ , respectively). Other levels in the common foods varied from  $0.50 \text{ } \mu\text{g kg}^{-1}$  (butter, sugars and sugar derivatives) to  $7.00 \text{ } \mu\text{g kg}^{-1}$  (bread and dried bread products;  $n=2$ ). For the infant foods, the mean contents did not exceed 3 LOQ ( $\text{LOQ} = 1 \text{ } \mu\text{g kg}^{-1}$ ), except for cereal-based foods for which the mean contents were slightly higher ( $3.18 \text{ } \mu\text{g kg}^{-1}$ ;  $n=17$ ). The highest levels were found in samples of different brands of biscuits for infants ( $14.0$  and  $20.0 \text{ } \mu\text{g kg}^{-1}$ ) and cereals with cocoa ( $2.88$ ,  $3.17$  and  $5.26 \text{ } \mu\text{g kg}^{-1}$ ). The other infant food categories had low levels with mean contents ranging from  $0.88$  (fruit juices) to  $2.63 \text{ } \mu\text{g kg}^{-1}$  (infant formulae). Only some data have been reported, also at relatively low concentrations, in milk-based and soy-based infant formulae categories ( $0.0822$  to  $3 \text{ } \mu\text{g kg}^{-1}$ ) in a USA survey (Ikem et al., 2002).

### *3.13 Mean contribution (%) of infant foods and common foods to the total element contents*

Infants' and toddlers' diets vary during their first three years and are defined by crucial phases. After the first six months, during which non-breastfed children usually consume infant formula, follow-on formula and various foods (such as infant foods with specific regulations) are progressively introduced, as well as common foods (vegetables, fruit, etc.). Because common foods are an important part of infants' and toddlers' diets (Hulin et al., 2014), it is important to evaluate their contribution – in addition to that of infant foods – to trace element contents. The contribution

of common and infant foods to the total element concentration (**Fig. 1**) shows that common foods presented a higher contribution (94%) on average, to arsenic content than infant foods, due to the high arsenic levels found in the fish food category, whereas infant foods contributed to 69% of tin content due to the fruit puree food category. Regarding the other trace elements, infant foods contributed, on average, to 49%, 45%, 43% and 42% to chromium, barium, nickel, gallium and cadmium contents, respectively, and less than 36% for the other elements, with common foods contributing the rest. However, regarding median concentrations, infant foods presented significantly higher contamination levels ( $p < 0.05$ ) than common foods only for chromium and barium and no statistical differences were found for the other elements (**Fig. 2**); no statistical comparisons were carried out for antimony, gallium, silver, tellurium, tin or germanium, due to the high percentage of left-censored data. Therefore, although there are no statistical differences for most of elements in common and infant foods, some specific food categories may have high contamination values and may also potentially lead to higher levels of exposure to trace elements depending on daily food consumption (common foods or infant foods). The dietary exposure assessment of infant and toddlers to these trace elements and their associated risks have been described in detail elsewhere (Sirot et al., 2018).

#### **4. Conclusions**

This TDS presented occurrence data on food mainly consumed by French infants and toddlers, data that are rarely available in other TDSs.

The contamination data for infant foods indicated higher levels of trace elements in processed foods and relatively low contents in infant formulae, which are widely consumed by infants. The contribution of trace element levels was not statistically different between infant and common foods, except for chromium and barium with median concentrations slightly higher in infant foods.

Despite the differences observed for some elements and food categories, the results of the first French TDS on infants and toddlers were generally comparable to those from other surveys.

Caution must be taken when comparing with other studies, because food composition may differ in many instances.

These results, combined with individual food consumption data, were used to evaluate the dietary exposure to trace elements of the French population of infants and toddlers (Sirot et al., 2018). Moreover, the output of this first French TDS on infants and toddlers will be useful for refining future health risk assessments.

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### **Declaration of interest**

Declarations of interest: none

## **Figure Captions**

**Figure 1:** Mean contribution (%) of infant foods and common foods to the total element contents (calculated as the mean element content of each food type (infant foods or common foods) normalized to the sum of element content of all food types)

**Figure 2:** Distribution of element contamination levels in infant and common foods (median values in  $\mu\text{g kg}^{-1}$ /box plots)

## **Supplementary material**

**Table SM-1:** List of food categories and the associated sample foods

**Table SM-2:** Lower bounds (LB) levels of trace elements in foods ‘as consumed’ by French infants and toddlers (in  $\mu\text{g kg}^{-1}$  fresh weight)

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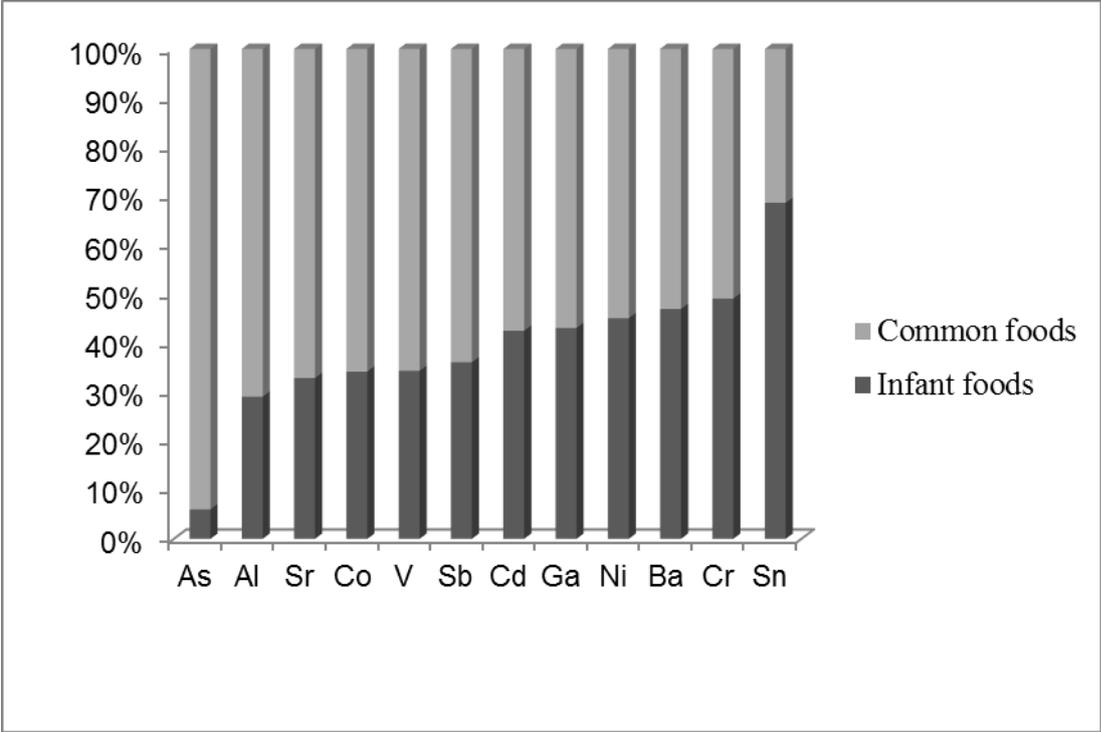
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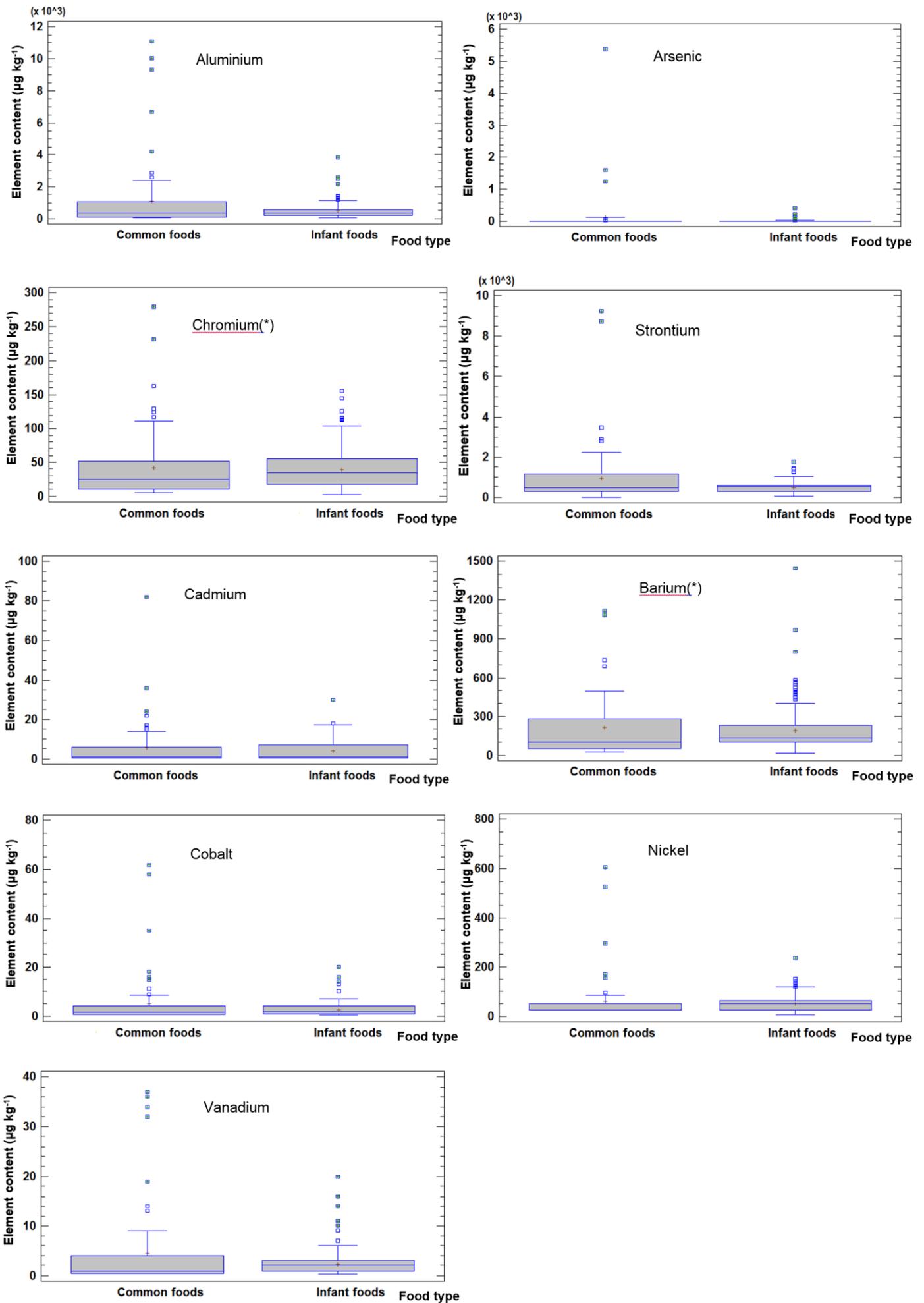
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\*Mann Whitney statistical test significant at  $p < 0.05$

**Table 1:** ICP-MS operating conditions and acquisition parameters

Operating conditions	Agilent 7700 Series ICP-MS	
Nebulizer	Quartz concentric (Micromist) 400 $\mu\text{L min}^{-1}$	
Spray chamber	Scott-type double-pass water cooled	
Cell geometry	Octopole	
Sampling cone	Nickel, 1.0 mm orifice	
Skimmer cone	Nickel, 0.75 mm orifice	
RF power	1400 - 1500 W	
Reflected power	< 10 W	
Standard mode		
Plasma gas flow	15 $\text{L min}^{-1}$	
Nebulizer gas flow	0.95-1.00 $\text{L min}^{-1}$	
Auxiliary gas flow	0.99 $\text{L min}^{-1}$	
Expansion stage	2.0 mbar	
Intermediate stage	$2.0 \times 10^{-4}$ - $3.0 \times 10^{-4}$ mbar	
Analyser stage	$1.0 \times 10^{-6}$ - $2.0 \times 10^{-6}$ mbar	
Octopole bias	-8 V	
Quadrupole bias	-3 V	
He mode (collision cell mode)		
He gas flow	4.3 $\text{mL min}^{-1}$	
Octopole bias	-18 V	
Quadrupole bias	-15 V	
Acquisition parameters		
Mass range	2-260 a.m.u	
Number of channels	500	
Dwell time	100 $\mu\text{s}$	
Number of sweeps	500	
Total acquisition time	219 s	
Isotopes measured	Analysis Mode	Internal standard
$^{27}\text{Al}$ , $^{51}\text{V}$ , $^{52}\text{Cr}$ , $^{59}\text{Co}$ , $^{71}\text{Ga}$ , $^{74}\text{Ge}$ , $^{60}\text{Ni}$ , $^{75}\text{As}$	He	$^{45}\text{Sc}$
$^{88}\text{Sr}$	He	$^{89}\text{Y}$
$^{107}\text{Ag}$ , $^{111-114}\text{Cd}$ , $^{118}\text{Sn}$ , $^{125}\text{Te}$ , $^{135-137}\text{Ba}$	Standard	$^{185}\text{Re}$
$^{121}\text{Sb}$	Standard	$^{185}\text{Re}$

**Table 2:** Evaluation of trueness of the method: multi-elemental analyses of certified materials TORT-2 (Lobster hepatopancreas), DOLT-4 (Dogfish liver) and SRM 1548a (Typical diet)

Element	CV <sub>R</sub> (%)	TORT-2				DOLT-4				SRM-1548			
		Reference value (mg kg <sup>-1</sup> )	Confidence interval (mg kg <sup>-1</sup> )	Mean ± SD measured value (n=10) (mg kg <sup>-1</sup> )	Min-max (mg kg <sup>-1</sup> )	Reference value (mg kg <sup>-1</sup> )	Confidence interval (mg kg <sup>-1</sup> )	Mean ± SD measured value (n=10) (mg kg <sup>-1</sup> )	Min-max individual value (mg kg <sup>-1</sup> )	Reference/indicative value (mg kg <sup>-1</sup> )	Confidence interval (mg kg <sup>-1</sup> )	Mean ± SD measured value (n=10) (mg kg <sup>-1</sup> )	Min-max individual value (mg kg <sup>-1</sup> )
Al	12	-	-	-	-	-	-	-	-	72.4	46.3 - 98.5	65.5 ± 9.6	56.8 - 90.6
Sb	8	-	-	-	-	-	-	-	-	0.009*	0.007 - 0.011	0.008 ± 0.001	0.007 - 0.009
As	12	21.6	13.8 - 29.4	20.1 ± 2.3	17.7 - 23.1	9.66	6.18 - 13.1	9.46 ± 0.90	7.61 - 10.83	0.2	0.1 - 0.3	0.2 ± 0.01	0.2 - 0.2
Ba	20	-	-	-	-	-	-	-	-	1.1*	0.4 - 1.8	1.0 ± 0.1	0.9 - 1.0
Cd	10	26.7	18.7 - 34.7	25.1 ± 2.5	20.3 - 23.1	24.3	17.0 - 31.6	24.7 ± 2.0	21.0 - 28.3	0.035	0.025 - 0.046	0.034 ± 0.002	0.030 - 0.037
Cr	10	0.77	0.54 - 1.00	0.68 ± 0.07	0.54 - 0.75	-	-	-	-	-	-	-	-
Co	10	0.51	0.36 - 0.66	0.45 ± 0.04	0.37 - 0.52	-	-	-	-	-	-	-	-
Ni	15	2.5	1.4 - 3.6	2.1 ± 0.2	1.7 - 2.4	0.97	0.43 - 1.41	0.98 ± 0.20	0.76 - 1.37	-	-	-	-
Ag	12	-	-	-	-	0.93	0.60 - 1.26	0.75 ± 0.13	0.59 - 1.01	-	-	-	-
Sr	15	45.2	24.9 - 65.2	34.9 ± 3.3	29.1 - 39.9	-	-	-	-	-	-	-	-
Sn	10	-	-	-	-	-	-	-	-	17.2	12.0 - 22.4	15.1 ± 0.8	13.4 - 16.0
V	8	1.64	1.25 - 2.03	1.60 ± 0.15	1.31 - 1.77	-	-	-	-	-	-	-	-

\* Not certified values, indicative values regarding the element concentration

**Table 3:** Evaluation of trueness of the method: multi-elemental analyses of spiked samples

	Theoretical value ( $\mu\text{g L}^{-1}$ )	Mean $\pm$ SD measured value (n= 6) ( $\mu\text{g L}^{-1}$ )	Mean recovery (%)	Min-Max measured value ( $\mu\text{g L}^{-1}$ )	Min-Max recovery (%)
Ga	1	$1.0 \pm 0.1$	100	0.9 - 1.0	90 - 100
	5	$4.8 \pm 0.2$	96	4.5 - 5.1	90 - 102
	10	$9.5 \pm 0.2$	95	9.3 - 9.8	93 - 98
Ge	1	$1.0 \pm 0.1$	100	0.9 - 1.0	90 - 100
	5	$5.1 \pm 0.2$	102	4.8 - 5.4	96 - 108
	10	$9.5 \pm 0.3$	95	9.2 - 9.8	92 - 98
Te	1	$1.0 \pm 0.2$	100	0.9 - 1.1	90 - 110
	5	$5.1 \pm 0.2$	102	4.8 - 5.8	96 - 116
	10	$10.0 \pm 0.3$	100	9.8 - 10.5	98 - 105

**Table 4:** Limits of detection and quantification (LOD/LOQ), percentage of detected and quantified results

<b>Element</b>	<b>LOD* µg kg<sup>-1</sup> fw</b>	<b>LOQ* µg kg<sup>-1</sup> fw</b>	<b>% &gt; LOD</b>	<b>% &gt; LOQ</b>
<b>Aluminium</b>	42	83	93.8	85.9
<b>Antimony</b>	0.5	1	22.8	11.3
<b>Arsenic</b>	1	2	70.3	44.3
<b>Barium</b>	25	50	93.8	87.6
<b>Cadmium</b>	0.3	0.5	64.5	57.0
<b>Chromium</b>	5.0	10	98.6	93.2
<b>Cobalt</b>	0.7	1	84.1	62.2
<b>Gallium</b>	0.5	1	14.1	6.9
<b>Germanium</b>	0.5	1	7.2	1.4
<b>Nickel</b>	25	50	54.5	37.1
<b>Strontium</b>	5.0	10	99.7	99.7
<b>Silver</b>	25	50	0	0
<b>Tellurium</b>	1	2	1.4	0.7
<b>Tin</b>	42	83	13.8	4.5
<b>Vanadium</b>	0.5	1	89.7	75.3

\*LOD and LOQ were estimated as three and six standard deviation respectively, of 21 sample blanks.

**Table 5:** Upper bound (UB) levels of trace elements in foods “as consumed” by French infants and toddlers (in  $\mu\text{g kg}^{-1}$  fresh weight)

Food category	Al			Sb		As		Ba		Cd	
	N	Mean $\pm$ SD	Min-Max								
<b>Infant foods</b>											
Cereal-based foods	17	630 $\pm$ 1030	44.0 - 3810	0.72 $\pm$ 0.54	0.04 - 2.00	3.13 $\pm$ 2.85	0.19 - 8.00	118 $\pm$ 213	14.0 - 762	2.79 $\pm$ 4.68	0.14 - 17.0
Follow-on formulae	34	276 $\pm$ 231	83.0 - 1140	0.57 $\pm$ 0.28	0.50 - 2.00	1.68 $\pm$ 0.59	1.00 - 3.00	111 $\pm$ 28.4	50.0 - 156	0.43 $\pm$ 0.33	0.30 - 2.00
Fruit juices	4	191 $\pm$ 112	42.0 - 314	0.75 $\pm$ 0.29	0.50 - 1.00	2.00 $\pm$ 0.00	1.00 - 2.00	317 $\pm$ 164	91.0 - 483	0.30 $\pm$ 0.00	0.30 - 0.30
Fruit purees	30	556 $\pm$ 254	260 - 1420	0.65 $\pm$ 0.48	0.50 - 3.00	2.00 $\pm$ 1.39	1.00 - 8.00	184 $\pm$ 57.0	110 - 303	0.66 $\pm$ 0.49	0.30 - 2.00
Growing-up milks	9	189 $\pm$ 220	42.0 - 724	0.56 $\pm$ 0.17	0.50 - 1.00	2.11 $\pm$ 0.78	1.00 - 4.00	58.9 $\pm$ 31.8	25.0 - 139	0.71 $\pm$ 1.23	0.30 - 4.00
Infant formulae	28	196 $\pm$ 135	42.0 - 585	0.54 $\pm$ 0.13	0.50 - 1.00	1.61 $\pm$ 0.69	1.00 - 4.00	115 $\pm$ 25.9	25.0 - 157	0.39 $\pm$ 0.19	0.30 - 1.00
Meat/fish-based ready-to-eat meals	45	597 $\pm$ 436	116 - 2590	0.56 $\pm$ 0.16	0.50 - 1.00	27.5 $\pm$ 70.4	1.00 - 411	286 $\pm$ 141	85.0 - 584	9.31 $\pm$ 4.33	3.00 - 30.0
Milk-based beverages	8	328 $\pm$ 140	150 - 561	0.50 $\pm$ 0.00	0.50 - 0.50	3.13 $\pm$ 2.85	0.00 - 10.0	65.8 $\pm$ 39.9	50.0 - 164	1.88 $\pm$ 1.13	1.00 - 4.00
Milk-based desserts	6	306 $\pm$ 289	104 - 859	0.58 $\pm$ 0.20	0.50 - 1.00	1.17 $\pm$ 0.41	1.00 - 2.00	117 $\pm$ 14.7	91.0 - 136	0.65 $\pm$ 0.38	0.30 - 1.00
Soups/purees	11	653 $\pm$ 595	207 - 2140	0.55 $\pm$ 0.15	0.50 - 1.00	4.82 $\pm$ 2.27	1.00 - 9.00	259 $\pm$ 100	102 - 483	7.36 $\pm$ 2.69	5.00 - 15.0
Vegetable-based ready-to-eat meals	27	575 $\pm$ 511	83.0 - 2480	0.52 $\pm$ 0.10	0.50 - 1.00	3.33 $\pm$ 3.63	1.00 - 17.0	337 $\pm$ 316	76.0 - 1450	9.26 $\pm$ 4.48	1.00 - 18.0
<b>Common foods</b>											
Bottled waters	12	66.0 $\pm$ 59.5	42.0 - 248	0.58 $\pm$ 0.20	0.50 - 1.00	1.17 $\pm$ 0.58	1.00 - 3.00	37.8 $\pm$ 19.8	25.0 - 74.0	0.30 $\pm$ 0.00	0.30 - 0.30
Bread and dried bread products	2	1800 $\pm$ 499	1450 - 2150	1.00 $\pm$ 0.00	1.00 - 1.00	12.5 $\pm$ 3.54	10.0 - 15.0	615 $\pm$ 170	495 - 735	15.5 $\pm$ 2.12	14.0 - 17.0
Butter	1	42.0	-	0.50	-	1.00	-	25.0	-	0.30	-
Cheese	1	4220	-	5.00	-	5.00	-	178	-	2.00	-
Compotes and cooked fruit	2	618 $\pm$ 110	540 - 696	1.00 $\pm$ 0.00	1.00 - 1.00	1.00 $\pm$ 0.00	1.00 - 1.00	168 $\pm$ 85.6	107 - 228	0.75 $\pm$ 0.35	0.50 - 1.00
Croissant-like pastries	2	4300 $\pm$ 3400	1890 - 6710	1.00 $\pm$ 0.00	1.00 - 1.00	4.00 $\pm$ 0.00	4.00 - 4.00	568 $\pm$ 173	446 - 690	12.5 $\pm$ 0.71	12.0 - 13.0
Dairy-based desserts	2	5950 $\pm$ 4800	2550 - 9350	1.50 $\pm$ 0.71	1.00 - 2.00	3.00 $\pm$ 1.41	2.00 - 4.00	471 $\pm$ 303	257 - 685	7.00 $\pm$ 5.66	3.00 - 11.0
Delicatessen meats	2	712 $\pm$ 889	83.0 - 1340	0.75 $\pm$ 0.35	0.50 - 1.00	4.50 $\pm$ 0.71	4.00 - 5.00	51.0 $\pm$ 36.8	25.0 - 77.0	0.75 $\pm$ 0.35	0.50 - 1.00
Eggs and egg products	1	42.0	-	0.50	-	11.0	-	261	-	0.50	-
Fish	3	1380 $\pm$ 1340	266 - 2860	3.00 $\pm$ 1.00	2.00 - 4.00	2750 $\pm$ 2290	1250 - 5390	136 $\pm$ 84.5	62.0 - 228	4.33 $\pm$ 2.52	2.00 - 7.00
Fruit	6	300 $\pm$ 290	83.0 - 851	0.50 $\pm$ 0.00	0.50 - 0.50	1.17 $\pm$ 0.41	1.00 - 2.00	418 $\pm$ 356	122 - 1100	0.73 $\pm$ 0.68	0.30 - 2.00
Hot beverages	1	1390	-	0.37	-	0.49	-	107	-	1.85	-
Meat	2	326 $\pm$ 242	155 - 497	2.00 $\pm$ 0.00	2.00 - 2.00	5.50 $\pm$ 6.36	1.00 - 10.0	25.0 $\pm$ 0.00	25.0 - 25.0	0.75 $\pm$ 0.35	0.50 - 1.00
Milk	3	69.3 $\pm$ 23.7	42.0 - 83.0	0.50 $\pm$ 0.00	0.50 - 0.50	1.00 $\pm$ 0.00	1.00 - 1.00	77.7 $\pm$ 15.2	64.0 - 94.0	0.30 $\pm$ 0.00	0.30 - 0.30
Mixed dishes	1	1290	-	1.00	-	11.0	-	303	-	13.0	-
Non-alcoholic beverages	6	348 $\pm$ 382	42.0 - 1030	0.58 $\pm$ 0.20	0.50 - 1.00	1.17 $\pm$ 0.41	1.00 - 2.00	117 $\pm$ 60.1	25.0 - 205	0.42 $\pm$ 0.29	0.30 - 1.00
Pasta	1	1040	-	2.00	-	5.00	-	470	-	16.0	-
Potatoes and potato products	3	607 $\pm$ 400	311 - 1060	0.83 $\pm$ 0.29	0.50 - 1.00	2.00 $\pm$ 0.00	2.00 - 2.00	93.3 $\pm$ 24.2	76.0 - 121	25.0 $\pm$ 10.5	15.0 - 36.0
Poultry and game	2	271 $\pm$ 26.9	252 - 290	0.50 $\pm$ 0.00	0.50 - 0.50	8.50 $\pm$ 0.71	8.00 - 9.00	25.0 $\pm$ 0.00	25.0 - 25.0	0.75 $\pm$ 0.35	0.50 - 1.00
Rice and wheat products	2	718 $\pm$ 38.9	690 - 745	2.00 $\pm$ 1.41	1.00 - 3.00	30.0 $\pm$ 33.9	6.00 - 54.0	238 $\pm$ 266	50.0 - 426	10.0 $\pm$ 7.07	5.00 - 15.0
Soups/broths	1	443	-	0.50	-	5.00	-	177	-	8.00	-
Sugar and sugar derivatives	1	83.0	-	6.00	-	1.00	-	25.0	-	1.00	-
Sweet and savoury biscuits & bars	1	10000	-	3.00	-	6.00	-	1080	-	22.0	-
Ultra-fresh dairy products	5	298 $\pm$ 169	83.0 - 470	0.50 $\pm$ 0.00	0.50 - 0.50	1.60 $\pm$ 0.89	1.00 - 3.00	64.4 $\pm$ 12.4	54.0 - 85.0	0.48 $\pm$ 0.30	0.30 - 1.00
Vegetables excluding potatoes	8	2380 $\pm$ 3580	585 - 11000	0.75 $\pm$ 0.54	0.50 - 2.00	3.25 $\pm$ 2.71	1.00 - 9.00	417 $\pm$ 355	62.0 - 1120	15.5 $\pm$ 27.3	1.00 - 82.0

Food category	Cr			Co		Ga		Ni		Sr	
	N	Mean $\pm$ SD	Min-Max								
<b>Infant foods</b>											
Cereal-based foods	17	23.0 $\pm$ 38.0	2.38 - 125	3.05 $\pm$ 4.67	0.14 - 16.0	0.62 $\pm$ 0.63	0.07 - 2.00	43.0 $\pm$ 61.5	4.00 - 234	221 $\pm$ 407	36.9 - 1380
Follow-on formulae	34	22.1 $\pm$ 13.5	10.0 - 78.0	0.98 $\pm$ 0.29	0.70 - 2.00	0.53 $\pm$ 0.12	0.50 - 1.00	26.5 $\pm$ 5.97	25.0 - 50.0	495 $\pm$ 134	199 - 721
Fruit juices	4	21.0 $\pm$ 10.8	5.00 - 29.0	0.93 $\pm$ 0.15	0.70 - 1.00	0.50 $\pm$ 0.00	0.50 - 0.50	25.0 $\pm$ 0.00	25.0 - 25.0	263 $\pm$ 84.7	191 - 367
Fruit purees	30	42.7 $\pm$ 15.8	13.0 - 84.0	2.87 $\pm$ 1.04	1.00 - 5.00	0.52 $\pm$ 0.09	0.50 - 1.00	54.7 $\pm$ 26.7	25.0 - 121	273 $\pm$ 150	94.0 - 756
Growing-up milks	9	27.7 $\pm$ 17.3	10.0 - 61.0	0.90 $\pm$ 0.15	0.70 - 1.00	0.56 $\pm$ 0.17	0.50 - 1.00	25.0 $\pm$ 0.00	25.0 - 25.0	325 $\pm$ 183	165 - 791
Infant formulae	28	20.8 $\pm$ 9.28	5.00 - 38.0	0.91 $\pm$ 0.26	0.70 - 2.00	0.50 $\pm$ 0.00	0.50 - 0.50	25.9 $\pm$ 4.72	25.0 - 50.0	512 $\pm$ 116	113 - 735
Meat/fish-based ready-to-eat meals	45	68.9 $\pm$ 30.6	20.0 - 155	3.82 $\pm$ 1.32	2.00 - 7.00	0.53 $\pm$ 0.13	0.50 - 1.00	75.7 $\pm$ 25.7	25.0 - 143	580 $\pm$ 203	250 - 1280
Milk-based beverages	8	31.8 $\pm$ 12.5	20.0 - 51.0	2.75 $\pm$ 1.58	1.00 - 5.00	0.50 $\pm$ 0.00	0.50 - 0.50	32.4 $\pm$ 13.9	25.0 - 59.0	695 $\pm$ 607	216 - 1750
Milk-based desserts	6	33.0 $\pm$ 24.0	10.0 - 75.0	5.62 $\pm$ 7.93	0.70 - 20.0	0.50 $\pm$ 0.00	0.50 - 0.50	41.2 $\pm$ 29.1	25.0 - 97.0	509 $\pm$ 124	356 - 660
Soups/purees	11	39.0 $\pm$ 12.7	19.0 - 57.0	2.73 $\pm$ 1.10	1.00 - 4.00	0.50 $\pm$ 0.00	0.50 - 0.50	57.7 $\pm$ 20.6	25.0 - 106	581 $\pm$ 92.0	454 - 756
Vegetable-based ready-to-eat meals	27	50.4 $\pm$ 18.1	16.0 - 92.0	3.69 $\pm$ 2.67	0.70 - 14.0	0.57 $\pm$ 0.18	0.50 - 1.00	71.5 $\pm$ 28.0	25.0 - 137	568 $\pm$ 157	226 - 864
<b>Common foods</b>											
Bottled waters	12	6.25 $\pm$ 2.26	5.00 - 10.0	0.70 $\pm$ 0.00	0.70 - 0.70	0.50 $\pm$ 0.00	0.50 - 0.50	27.1 $\pm$ 7.22	25.0 - 50.0	2180 $\pm$ 3290	18.0 - 9270
Bread and dried bread products	2	80.5 $\pm$ 51.6	44.0 - 117	6.00 $\pm$ 0.00	6.00 - 6.00	1.00 $\pm$ 0.00	1.00 - 1.00	69.5 $\pm$ 3.54	67.0 - 72.0	644 $\pm$ 4.24	641 - 647
Butter	1	30.0	-	1.00	-	0.50	-	25.0	-	101	-
Cheese	1	111	-	2.00	-	1.00	-	51.0	-	1980	-
Compotes and cooked fruit	2	41.5 $\pm$ 21.9	26.0 - 57.0	2.50 $\pm$ 0.71	2.00 - 3.00	0.50 $\pm$ 0.00	0.50 - 0.50	40.5 $\pm$ 21.9	25.0 - 56.0	305 $\pm$ 194	168 - 442
Croissant-like pastries	2	95.0 $\pm$ 94.8	28.0 - 162	19.5 $\pm$ 21.9	4.00 - 35.0	1.25 $\pm$ 1.06	0.50 - 2.00	173 $\pm$ 173	50.0 - 295	611 $\pm$ 314	389 - 833
Dairy-based desserts	2	178 $\pm$ 144	76.0 - 280	40.0 $\pm$ 31.1	18.0 - 62.0	2.00 $\pm$ 1.41	1.00 - 3.00	388 $\pm$ 307	171 - 605	861 $\pm$ 431	556 - 1170
Delicatessen meats	2	38.5 $\pm$ 33.2	15.0 - 62.0	3.50 $\pm$ 3.54	1.00 - 6.00	0.50 $\pm$ 0.00	0.50 - 0.50	37.5 $\pm$ 17.7	25.0 - 50.0	170 $\pm$ 156	60.0 - 280
Eggs and egg products	1	21.0	-	0.70	-	0.50	-	25.0	-	352	-
Fish	3	81.7 $\pm$ 17.9	61.0 - 93.0	2.67 $\pm$ 0.58	2.00 - 3.00	0.50 $\pm$ 0.00	0.50 - 0.50	41.7 $\pm$ 14.4	25.0 - 50.0	1410 $\pm$ 530	868 - 1930
Fruit	6	28.0 $\pm$ 13.9	10.0 - 46.0	4.50 $\pm$ 5.75	1.00 - 16.0	0.58 $\pm$ 0.20	0.50 - 1.00	44.0 $\pm$ 15.4	25.0 - 62.0	1160 $\pm$ 1140	113 - 2860
Hot beverages	1	42.4	-	8.69	-	0.45	-	96.2	-	153	-
Meat	2	91.5 $\pm$ 46.0	59.0 - 124	2.00 $\pm$ 0.00	2.00 - 2.00	0.50 $\pm$ 0.00	0.50 - 0.50	41.0 $\pm$ 22.6	25.0 - 57.0	109 $\pm$ 53.7	71.0 - 147
Milk	3	10.7 $\pm$ 1.15	10.0 - 12.0	1.23 $\pm$ 0.68	0.70 - 2.00	0.50 $\pm$ 0.00	0.50 - 0.50	25.0 $\pm$ 0.00	25.0 - 25.0	358 $\pm$ 61.7	315 - 429
Mixed dishes	1	86.0	-	6.00	-	0.50	-	65.0	-	839	-
Non-alcoholic beverages	6	13.2 $\pm$ 5.49	5.00 - 20.0	1.40 $\pm$ 0.92	0.70 - 3.00	0.50 $\pm$ 0.00	0.50 - 0.50	29.2 $\pm$ 10.2	25.0 - 50.0	511 $\pm$ 307	161 - 971
Pasta	1	33.0	-	1.00	-	0.50	-	50.0	-	646	-
Potatoes and potato products	3	35.0 $\pm$ 23.8	17.0 - 62.0	8.33 $\pm$ 3.06	5.00 - 11.0	0.50 $\pm$ 0.00	0.50 - 0.50	60.7 $\pm$ 13.6	50.0 - 76.0	344 $\pm$ 101	238 - 438
Poultry and game	2	41.0 $\pm$ 21.2	26.0 - 56.0	1.00 $\pm$ 0.00	1.00 - 1.00	0.50 $\pm$ 0.00	0.50 - 0.50	25.0 $\pm$ 0.0	25.0 - 25.0	50.0 $\pm$ 1.41	49.0 - 51.0
Rice and wheat products	2	28.0 $\pm$ 17.0	16.0 - 40.0	1.50 $\pm$ 0.71	1.00 - 2.00	0.75 $\pm$ 0.35	0.50 - 1.00	50.0 $\pm$ 0.00	50.0 - 50.0	406 $\pm$ 136	310 - 502
Soups/broths	1	46.0	-	4.00	-	0.50	-	25.0	-	583	-
Sugar and sugar derivatives	1	12.0	-	0.70	-	0.50	-	25.0	-	5.00	-
Sweet and savoury biscuits & bars	1	232	-	58.0	-	3.00	-	527	-	1300	-
Ultra-fresh dairy products	5	17.2 $\pm$ 7.92	10.0 - 27.0	0.88 $\pm$ 0.16	0.70 - 1.00	0.50 $\pm$ 0.00	0.50 - 0.50	25.0	25.0 - 25.0	379 $\pm$ 58.9	328 - 477
Vegetables excluding potatoes	8	45.3 $\pm$ 40.0	5.00 - 129	4.88 $\pm$ 4.70	1.00 - 15.0	1.19 $\pm$ 1.16	0.50 - 4.00	62.4 $\pm$ 44.8	25.0 - 157	1590 $\pm$ 853	792 - 3470

Food category	Sn			V	
	N	Mean $\pm$ SD	Min-Max	Mean $\pm$ SD	Min-Max
<b>Infant foods</b>					
Cereal-based foods	17	49.2 $\pm$ 16.1	42.0 - 83.0	3.18 $\pm$ 5.47	0.29 - 20.0
Follow-on formulae	34	42.0 $\pm$ 0.00	42.0 - 42.0	2.31 $\pm$ 2.74	0.50 - 16.0
Fruit juices	4	62.5 $\pm$ 23.7	42.0 - 83.0	0.88 $\pm$ 0.25	0.00 - 1.00
Fruit purees	30	42.4 $\pm$ 9.46	42.0 - 33.30	1.40 $\pm$ 0.81	1.00 - 4.00
Growing-up milks	9	42.0 $\pm$ 0.00	42.0 - 42.0	2.50 $\pm$ 3.32	0.00 - 11.0
Infant formulae	28	42.0 $\pm$ 0.00	42.0 - 42.0	2.63 $\pm$ 2.83	0.50 - 10.0
Meat/fish-based ready-to-eat meals	45	49.3 $\pm$ 15.9	42.0 - 83.0	2.56 $\pm$ 1.32	1.00 - 7.00
Milk-based beverages	8	42.0 $\pm$ 0.00	42.0 - 42.0	2.13 $\pm$ 1.25	1.00 - 5.00
Milk-based desserts	6	42.0 $\pm$ 0.00	42.0 - 42.0	1.50 $\pm$ 1.34	0.00 - 4.00
Soups/purees	11	42.0 $\pm$ 0.00	42.0 - 42.0	2.18 $\pm$ 0.75	1.00 - 3.00
Vegetable-based ready-to-eat meals	27	59.5 $\pm$ 27.7	42.0 - 143	2.19 $\pm$ 1.27	1.00 - 6.00
<b>Common foods</b>					
Bottled waters	12	42.0 $\pm$ 0.00	42.0 - 42.0	0.88 $\pm$ 1.00	0.50 - 4.00
Bread and dried bread products	2	42.0 $\pm$ 0.00	42.0 - 42.0	7.00 $\pm$ 2.83	5.00 - 9.00
Butter	1	42.0	-	0.50	-
Cheese	1	42.0	-	36.0	-
Compotes and cooked fruit	2	42.0 $\pm$ 0.00	42.0 - 42.0	1.50 $\pm$ 0.71	1.00 - 2.00
Croissant-like pastries	2	42.0 $\pm$ 0.00	42.0 - 42.0	12.0 $\pm$ 9.90	5.00 - 19.0
Dairy-based desserts	2	42.0 $\pm$ 0.00	42.0 - 42.0	23.5 $\pm$ 14.8	13.0 - 34.0
Delicatessen meats	2	42.0 $\pm$ 0.00	42.0 - 42.0	4.00 $\pm$ 0.00	4.00 - 4.00
Eggs and egg products	1	42.0	-	1.00	-
Fish	3	42.0 $\pm$ 0.00	42.0 - 42.0	4.00 $\pm$ 0.00	4.00 - 4.00
Fruit	6	42.0 $\pm$ 0.00	42.0 - 42.0	1.00 $\pm$ 0.55	0.50 - 2.00
Hot beverages	1	42.0	-	4.22	-
Meat	2	42.0 $\pm$ 0.00	42.0 - 42.0	1.00 $\pm$ 0.00	1.00 - 1.00
Milk	3	42.0 $\pm$ 0.00	42.0 - 42.0	0.67 $\pm$ 0.29	0.50 - 1.00
Mixed dishes	1	42.0	-	4.00	-
Non-alcoholic beverages	6	42.0 $\pm$ 0.00	42.0 - 42.0	3.17 $\pm$ 5.34	0.50 - 14.0
Pasta	1	42.0	-	3.00	-
Potatoes and potato products	3	63.3 $\pm$ 37.0	42.0 - 106	4.00 $\pm$ 4.36	1.00 - 9.00
Poultry and game	2	42.0 $\pm$ 0.00	42.0 - 42.0	1.50 $\pm$ 0.71	1.00 - 2.00
Rice and wheat products	2	42.0 $\pm$ 0.00	42.0 - 42.0	3.50 $\pm$ 0.71	3.00 - 4.00
Soups/broths	1	42.0	-	3.00	-
Sugar and sugar derivatives	1	42.0	-	0.50	-
Sweet and savoury biscuits & bars	1	42.0	-	37.0	-
Ultra-fresh dairy products	5	42.0 $\pm$ 0.00	42.0 - 42.0	0.90 $\pm$ 0.65	0.50 - 2.00
Vegetables excluding potatoes	8	63.9 $\pm$ 47.5	42.0 - 176	6.25 $\pm$ 10.5	1.00 - 32.0