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Nickel in foods sampled on the Belgian market: identification of potential contamination sources

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ABSTRACT

Nickel can occur in plant-based, animal-based foods and drinks. It can either naturally occur in plants or it could originate from contamination. The natural occurrence of nickel arises from the fact that the element plays an essential role in the functioning of enzymes involved in the nitrogen fixation process. Besides, contamination can occur at any stage of the production, processing or packing of the foods. More specifically, nickel can leach from contact materials to foods or drinks before their consumption by humans. In recent years, the European Food Safety Authority expressed concern regarding the chronic and acute exposure of the European population to nickel. This study aimed to screen foods available on the Belgian market for their nickel content and to identify potential sources of the contamination. In total, 708 samples were collected from three different main categories of foods, including plant-based products, animal-based products and drinks. Elevated nickel concentrations were found in plant-based products such as chocolate, legumes, nuts, figs, peanut butter, chocolate spreads and breakfast cereals. The nickel concentrations in the animal-based products and drinks were significantly lower compared to the plant-based products. In the beer samples, no correlation between the alcohol percentage and nickel concentration was found. Higher nickel concentrations were found in the tea drinks in comparison to other drinks. Furthermore, the effect of packaging, e.g. storage in cans, on the final nickel concentration of the foods was investigated. No effect of the packaging was found, demonstrating that leaching of nickel from packaging materials is not significantly contributing to the nickel content in foods. The results demonstrate high concentrations of nickel in some plant-based food products and further exposure assessment studies are needed to evaluate the risk due to intake of nickel-enriched food products.

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Introduction

The essential role of nickel (Ni) in bacteria, plants and animals has been shown by Welch (1981). He investigated the role of Ni in growth of bacteria, e.g. *Alcaligenes*, and green alga, and showed that Ni is indispensable for the growth of these species. Furthermore, he clarified the important role of Ni in the growth of some trees, e.g. pine tree (Welch 1981). In nitrogen-fixing plants, e.g. legumes such as soy bean and peanut, Ni is involved in nitrogen metabolism as a structural component of essential enzymes, i.e. urease and hydrogenases (Lavres et al. 2016). Thus, Ni content of these types of nitrogen fixating plants and their derived foods can be considered as naturally occurring Ni.

Different types of foods have been studied in different countries to evaluate their trace elements

content, including Ni. The probable factors playing an effective role in increasing the Ni content of these foods have been reported, such as composition of the cultivation soil and contamination at the production and processing sites (Quintaes et al. 2007; Zhu et al. 2011). In aquatic systems, sewage discharge and non-ferrous metal smelters are responsible for Ni pollution, leading to Ni contamination in seafood (Cempel and Nickel 2006). For vegetables and plant-based foods, physical and chemical characteristics of the soil, and nature and absorption capacity of the plant itself can affect their final Ni content (Li et al. 2012). Besides, crops irrigated with contaminated water generally appear to be more contaminated with trace elements (Yang and Li 1998; Li et al. 2012). De Brouwere et al. (2012) estimated the human

exposure to Ni, through oral routes, at regional scale in the EU. They investigated the effect of Ni which is transferred from the soil (originated from aerial Ni deposition) to the plant food growing in districts near Ni emitting industries. They stated that plant-origin foods are considered to be the key source of Ni exposure in humans (De Brouwere et al. 2012). The food sources mainly contributing to the daily intake of Ni by humans have been identified, e.g. chocolate, cereals and nuts (Leblanc et al. 2005; Noël et al. 2012). According to Noël et al. (2012), cereals, dark chocolate and tofu are administering the highest Ni amounts to the human body. In another study conducted by Leblanc et al. (2005), nuts, oilseeds, chocolate and breakfasts cereals were identified to have elevated Ni contents. Also, leaching of Ni from metal-containing food contact materials, e.g. equipment, tanks, and packaging materials, has previously been reported as potential source of Ni in foods (Kamerud et al. 2013; Blondeel et al. 2014).

A too-elevated Ni exposure disturbs the magnesium (Mg) and zinc (Zn) metabolism (Anke et al. 1995). In pigs, Zn deficiency symptoms similar to parakeratosis may occur upon elevated Ni exposure. In humans, gradual filling of the Ni pools in the body occurs through Ni intake and can induce dermatitis (eczema) reactions in Ni-sensitised individuals (Anke et al. 1995). The prevalence of Ni allergy is three times higher in females in comparison to males (Torres et al. 2009).

In experimental animals, Ni is able to pass the placental barriers and directly affect embryo or foetus development. For the female rat's offspring, perinatal mortality can increase when they ingest Ni salts (EFSA 2015). In a risk assessment conducted by the EFSA (2015), reproductive toxicity in experimental animal was identified as a critical effect for chronic oral exposure to Ni in risk characterisation approach. A tolerable daily intake of $2.8 \mu\text{g Ni kg body weight (bw)}^{-1}$ was obtained through performing a benchmarks dose (BMD) modelling. This was performed on a dose range finding of 1-generation and 2-generation studies. The average chronic dietary exposure to Ni was ranged from $2 \mu\text{g Ni kg bw}^{-1}$ (for elderly) to $13 \mu\text{g Ni kg bw}^{-1}$ (for toddlers) in the exposure values reported for different European countries. At the

95th percentile chronic dietary exposure was ranged from $3.6 \mu\text{g Ni kg bw}^{-1}$ (for elderly) to $20 \mu\text{g Ni kg bw}^{-1}$ (for toddlers) which were all exceeded the TDI of $2.8 \mu\text{g Ni kg bw}^{-1}$. EFSA Panel on Contaminants in the Food Chain (CONTAM Panel) indicated concerns for the chronic dietary exposure to Ni for the general population of Europe.

Besides, CONTAM Panel stated that eczematous flare-up skin reactions may develop in Ni-sensitised individuals when they are orally exposed to Ni as well (EFSA 2015). For acute exposure to Ni, they stated that TDI of $2.8 \mu\text{g Ni kg bw}^{-1}$ may not be enough to protect these highly sensitised individuals. Therefore, they selected lowest benchmark dose (BMDL_{10}) of $1.1 \mu\text{g Ni kg bw}^{-1}$ which was obtained from the dose-response analysis. They concluded that Ni-sensitised individuals may develop eczematous flare-up skin reactions at the current level of acute dietary exposure. In Europe, between 2003 and 2012 18885 food and 25700 drinking water samples were analysed for Ni content. These samples were collected from 15 different countries across Europe but 80% of these samples were collected from only one member state. Therefore, more data are needed to be collected on the Ni concentration in different food/drinks which are available in European markets. As previously mentioned, the origin of Ni in food can be diverse, from contamination of the raw materials to processing conditions as well as release from contact materials. Stainless steel in processing facilities and cans containing Ni is widely abundant in the food chain (Mochizuki et al. 1985; Keyvani 2002; Imai 2015). Hence, this research aimed to investigate the extent of Ni occurrence/concentration in food products available on the Belgian market and their potential contamination routes.

Material and methods

Food samples, sampling plan and collection procedure

Prior to starting the sampling and analysis campaign, a risk-based sampling plan was fine-tuned after literature review. Not all food products available on the Belgian market were monitored as the objective was to focus on those food products

which were not yet investigated in other research projects and/or where the presence of Ni could be expected.

In total, 708 samples were collected for further analysis, divided into three main categories, plant-based foods ($N = 406$), animal-based foods ($N = 113$) and drinks ($N = 189$). All food products were purchased in supermarkets or specialised retailer shops in Belgium and were analysed before their expiration date. Within each category, further subcategories are identified; for plant-based products: legumes, soy products, chocolates, coffee, tea, tomato sauces, dried fruits, nuts, chocolate spreads, peanut butter, vegetables, breakfast cereals and canned fruits. Animal-based products were collected and divided into gelatins, emulsified sauces (including mayonnaise), eggs, dairies (including milk, yoghurt) and seafood. Drinks included beer, coffee, tea and ice tea. For each subcategory, circa 20 products were sampled ($N = 20$) in order to be able to have insight into the variability within each subcategory. Characteristics including the brand name, shop, purchase date, expiring date, unit price, and origin and package type (if applicable) were inventoried. Specifically, for beers and chocolate, the alcohol percentage and cacao content were recorded, respectively. The products were photographed in their original package.

Coffee and tea beverages preparation

For the coffee beverages prepared according to the domestic practices, 3.5 g of the coffee powder was weighted on a filter paper of 90-mm diameter. Unground coffee beans were first ground manually with a ceramic mortar prior to weighing into the filter. After adding 30 mL of boiling ultrapure water, the filtrates were collected in falcon tubes and the volume of the solutions was increased until 40 mL. Hence, an amount of 87.5 g solid coffee per litre of beverage was generally used. The coffee ingredients were selected from the commercially available ground coffees and the roasted coffee beans available in the market. For the coffee beverage prepared according to the Golden Cup Standard protocol (SCA 2017), the ground coffee/water ratio was adjusted to $55 \text{ g L}^{-1} \pm 10\%$ and temperature was adjusted to $93 \pm 3^\circ\text{C}$. Coffee-

water contact time was 1 to 4 min and filter paper of 90 mm was used for coffee preparation. These coffee beverages were prepared to compare their Ni content with the Ni content of the coffee beverages prepared through a domestic protocol.

For tea beverages, prepared according to the domestic practices, 2.5 g of the solid tea was weighted on filter paper of 90 mm in diameter. A volume of 20 mL boiling ultrapure water was added to every sample. After filtration in falcon tubes, the total volume of every tea beverage was adjusted to 20 mL. Hence, a mass of 125-g solid tea was used per litre of tea beverage. The ISO 3130 (The Standardized Method for Brewing Tea 1985) protocol was used to prepare a standard tea beverage. The tea/water ratio was $20 \text{ g L}^{-1} \pm 2\%$ and freshly boiled ultrapure water was used for preparation. The contact time was adjusted to 5 min (ISO 1985).

Cooking of lentils

The dry lentils were soaked in ultra-pure water overnight prior to cooking. After throwing away this water, 5 g of the soaked lentils were weighed into a 50 mL polypropylene falcon tube, and 15 mL of ultra-pure water was added. Tubes were closed and lentils were cooked at 100°C in a water bath, i.e. Memmert WNE14, GmbH, for 30 min.

Drying the legumes

Fresh/frozen, canned and bottled legumes were dried to calculate dry weight/wet weight ratio for them. Tray cardboard Nr75 (100x70x35mm) from food packaging at AVA (Belgium) was used to weigh around 20 g of every original legume sample. After overnight drying in the oven (Mettler oven, ULM-700, GmbH, Germany), the dried samples were weighed again.

Centrifugation of the beer samples

A centrifuge (Eppendorf AG 5804 R) Hamburg was used to separate the yeast cells from the selected beer samples. The centrifugation was conducted at g-force of 13440 for 10 min for every sample.

Analysis of Ni

Determination of Ni was performed by a microwave-assisted acid digestion, followed by measurement using Inductively Coupled Plasma–Mass Spectrometry (ICP-MS). ICP-MS device was NexION™ 350D, Pekin Elmer from USA. The legumes were first dried at 60°C for 24 h. Solid food samples were ground or chopped, according to their nature, initially. A ceramic mortar was used to grind and homogenise the dried foods, e.g. breakfast cereals. The fresh foods were chopped using a ceramic knife and polypropylene chopping board into fine particles prior to weighing. The ceramic knife and polypropylene chopping board were used to prevent Ni contamination from the contact materials to the food samples. Ceramic food contact materials may release trace elements into foods but Ni release from these materials to food appears to be insignificant under the conditions of use (Demont et al. 2012). Generally, amounts of 0.2 g (for solid food samples) and 5 g (for liquid samples) were weighted and transferred to pre-rinsed and inert polytetrafluoroethylene digestion tubes. Subsequently, a volume of 10 mL 65% HNO₃ of the highest analytical purity (Chem-Lab, Belgium) was added to every sample. The suspensions were placed in a sonicator (Sonorex RK103H, Germany) for 15 min to guarantee a complete mixing, before being introduced in a microwave digestion system (Mars 6, CEM, US). According to the type of food matrix, some adaptations were made on the initial mass of the food and volume of added nitric acid. For canned fruits, eggs and mayonnaises the initial weight was lowered to 0.5 g in order to ensure complete digestion.

After completion of the microwave digestion, the clear solutions were diluted with ultrapure water to a total volume of 50 mL in the case of solid food samples and 25 mL in the case of liquid food samples. To assess the reproducibility of the method, all samples were digested in duplicate. Subsequently, ICP-MS analysis was performed for duplicate samples.

A Perkin Elmer Nexion 350D (US) quadrupole ICP-MS instrument was used. The sample was introduced by a PrepFAST auto-sampler system (ESI, US) which continuously added 10 µg L⁻¹ rhodium (¹⁰³Rh) solution, serving as an internal standard. A set of platinum cones was installed to further exclude circumstantial Ni

contamination that may be introduced by Ni cones. The Ni was monitored at its most sensitive m/z ratio of 58 after the addition of 0.4 L min⁻¹ He collision gas to eliminate various polyatomic interferences, including ²³Na³⁵Cl⁺ and ⁴⁰Ar¹⁸O⁺. Highly sensitive detection was further assured by the Quadrupole Ion Deflector (QID) to remove neutral species and photons. Quantification of Ni was established using external calibration with Ni standards in the range of 0–200 µg L⁻¹. The RSDs, i.e. relative standard deviations, of the duplicates were checked after measuring Ni to evaluate the variability of the analytical method.

Quality control of Ni analysis

Limit of detection and limit of quantification

The limit of detection (LOD) and limit of quantification (LOQ) were determined according to Equations (1) and (2) in which S_{blank} was determined as the standard deviation of 10 blank measurements and a the sensitivity from calibration.

$$LOD = 3 \times \frac{S_{blank}}{a} \quad (1)$$

$$LOQ = 10 \times \frac{S_{blank}}{a} \quad (2)$$

Initial weight of a sample and the final volume of the digest after microwave digestion were applied to calculate LOD and LOQ values in µg kg⁻¹. Generally, LOD values were 12.04 µg kg⁻¹ and 0.36 µg kg⁻¹ for solid and liquid samples, respectively. The LOQ value was 40.12 µg kg⁻¹ for solid samples and 1.2 µg kg⁻¹ for liquid samples. Similarly, LOD value of 0.04 µg kg⁻¹ and 2 µg kg⁻¹ was obtained for beer, coffees/chocolates, respectively. A LOQ value of chocolate and coffees was 6.7 µg kg⁻¹. For the beers, LOQ value was 0.13 µg kg⁻¹. For canned fruits, eggs and mayonnaises, LOD and LOQ values of 4.8 µg kg⁻¹ and 16.05 µg kg⁻¹ were obtained, respectively (Table 1).

Accuracy of the analysis

For the chocolate and coffee, spikes of known Ni concentrations in the form of dissolved Ni chloride were added prior to the microwave digestion. The concentration range of NiCl₂ spikes varied from 0.8

Table 1. The LOD and LOQ values of Ni calculated for different food samples.

Food sample	LOD $\mu\text{g kg}^{-1}$	LOQ $\mu\text{g kg}^{-1}$
Coffee and chocolate	2.0	6.7
Canned fruits, eggs and mayonnaises	4.8	16.1
Rest of solid foods	12.0	40.1
Beer	0.04	0.13
Rest of liquid foods	0.36	1.2

to $8 \mu\text{g kg}^{-1}$ for coffees and from 5 to $20 \mu\text{g kg}^{-1}$ for chocolates. Results from the elemental analysis were traced back to the original amount of Ni that was added to the food sample (Table 2). The precision of the analyses was verified by analysing duplicates of each product type. Furthermore, spiking after microwave digestion into the food digests was also performed for several food categories (Table 3).

The accuracy of the analysis was also verified by analysing four different certified reference materials (CRMs). The CRM analysis was performed through the same method as described for solid samples and this was done in duplicate for each CRM (Table 4). As it is clear, a large number of the quality control analyses were performed to check the accuracy of the Ni analysis. CRMs were chosen from different types to check the accuracy of the method for different food

Table 2. Average recovery of Ni spiked to the products prior to the microwave digestion.

Product category	Ni concentration of spike ($\mu\text{g kg}^{-1}$)	Recovery (%)	
		N	$\pm\text{SD}$
Chocolate	5	3	98 ± 2
	10	3	95 ± 6
	20	3	95 ± 5
Coffee	0.8	4	83 ± 29
	4	4	93 ± 9
	8	4	102 ± 3

Table 3. Recovery of Ni spiked to different food products after microwave digestion.

Food type	Sample	Ni concentration of spike ($\mu\text{g L}^{-1}$)	Recovery (%) $\pm\text{SD}$
Legumes	Lentil	10, 50	104 ± 1
Soy products	Soy dessert	10, 50, 100, 200	109 ± 1
	Soy drink	10, 50, 100, 200	98 ± 5
Solid tea ingredients	Black tea	10, 50	107 ± 3
	Green tea	10, 50	103 ± 3
Gelatine		5, 10, 50	99 ± 1
Tomato passata		10, 50, 100, 200	106 ± 2
Dried fruits	Banana	1, 5, 10, 50	109 ± 1
	Walnut	10, 50	106 ± 2
	Hazelnut	10, 50	94 ± 10
Choco-hazelnut paste		10, 50	107 ± 1
Vegetables	Carrot	1, 5, 10, 50	107 ± 4
	Potato	1, 5, 10, 50	108 ± 4
Plain yoghurt		1, 5, 10, 50	107 ± 2
Fish		1, 5, 10, 50	105 ± 5
Minced meat		1, 5, 10, 50	97 ± 7
Salami		1, 5, 10, 50	103 ± 6

Table 4. Average recovery of Ni obtained for certified reference materials (CRMs). Analysis of the CRMs conducted in duplicate.

Reference material	CODE	concentration (mg kg^{-1})	Recovery (%)
Lobster	TORT2	2.50 ± 0.19	98
Hepatopancreas			
Rye grass	ERM CD 281	15.2 ± 0.6	104
White cabbage	BCR-679	27.0 ± 0.8	107
Spinach leaves	1570a	2.14 ± 0.06	99

types. The recovery percentage of Ni in CRMs ranged from 98% to 107% which confirms high accuracy of the analysis. Furthermore, different types of food samples were chosen for the spiking experiments after microwave digestion. This was done again to check the method accuracy with respect to the different food types and matrixes. In these spiking experiments, the recovery percentage of Ni was from 94% to 109%. Except for a single spike with low recovery (83%), the remainder of the recoveries ranged from 93% to 102% for the spiking experiment before microwave digestion. Accordingly, all obtained recovery values indicate the accuracy of the Ni analysis in the current study.

Statistical analysis

Statistical analysis of the data was performed in SPSS (IBM) or Sigma Plot 12. Results were plotted using Sigma Plot 12, Microsoft Excel and SPSS. According to the data distribution found (normal or not), parametric (independent sample T-test, paired sample T-test and one-way ANOVA) and non-parametric tests (Kruskal-Wallis test) were performed in SPSS ($\alpha = 0.05$).

Results and discussion

Plant-based products

In total, 406 plant-origin samples were collected from the Belgian market. This major category comprises 13 subcategories, i.e. legumes, soy products, chocolates, chocolate spreads, coffees (beans and ground coffees), tea (loose tea and tea bags), tomato sauces, dried fruits, nuts, vegetables, breakfast cereals and canned fruits. Compared to other major categories (animal-based products and drinks), the plant-based products showed the highest Ni concentrations. Chocolates, legumes, nuts, figs, peanut butters, chocolate spreads and breakfast cereals showed to have elevated Ni concentrations with average Ni contents of 3380, 2089, 1594, 1566, 1348, 1226 and 908 $\mu\text{g kg}^{-1}$, respectively (Table 5). Except for a few vegetables

and canned fruits all other samples were far below the detection limits.

As displayed in Table 5, Ni contents of the legumes were generally high which could be explained by contribution of Ni to the structure of enzymes involved in nitrogen fixation processes in the plant. Cabrera et al. (2003) reported Ni concentrations ranging from 20 $\mu\text{g kg}^{-1}$ to 350 $\mu\text{g kg}^{-1}$ in legumes. Previous studies also reported that legumes are food products with a high Ni content and one of the major sources of the dietary exposure to Ni (Nielsen and Flyvholm 1984; Cabrera et al. 2003). For peanuts, Ni concentration ranged from 594 to 1841 $\mu\text{g kg}^{-1}$. A Ni content of 3600 $\mu\text{g kg}^{-1}$ was reported for peanuts by Duda-Chodak and Blaszczyk (2008). In the current study, a mean concentration of 1348 $\mu\text{g kg}^{-1}$ (N = 10) was obtained for peanut butters.

Table 5. Summary statistics for the Ni content ($\mu\text{g kg}^{-1}$) in different plant-based food products purchased from the Belgian market (N = 406).

Category	Product	N	Ni content ($\mu\text{g kg}^{-1}$) FW				Weight basis ^a	
			Mean	Minimum	P ₅₀	Maximum		
Legumes ^b	Beans ^c	33	2892	867	2170	10050	DW	
	Lentils	14	1883	734	2099	3694		
	Peas	16	1413	552	1135	4162		
	Legumes with pods	8	2169	695	2067	4673		
	Peanut	3	1356	594	1631	1841		
Soy products	Drinks	15	227	110	170	482	FW	
	Desserts & Creams	12	170	94	155	406		
	Tofu	7	425	89	425	942		
Chocolate	Sugar-based	24	4140	2204	3955	8457	FW	
	Polyol-based	20	2620	883	3161	4912		
Coffee (beans and ground coffee)	Coffee beans	20	723	394	548	1507	DW	
	Ground coffee	20	992	312	581	4268		
Tea (loose tea and tea bags)	Black tea	11	6271	3518	5723	9758	DW	
	Green tea	11	6194	3704	6643	8504		
Tomato sauces	Passata	20	124	36	106	281	FW	
Dried fruits	Fig	8	1566	861	1249	3683	DW	
	Raisin	12	125	72	92	253		
	Almond	7	869	577	896	1092	DW	
Nuts	Hazelnut	6	2383	1196	2280	3846		
	Pistachio	4	950	406	827	1740		
	Walnut ^f	6	2411	720	2253	4623		
	Chocolate spreads (hazelnut)	11	1226	661	1280	1502	FW	
Peanut butter	10	1348	227	1401	3106	FW		
Vegetables (fresh and frozen)	Carrot	21	LB ^d	21	0.0	0.0	96	FW
			UB ^e	42	12	40	96	
	Spinach	21	LB	109	0.0	154	267	
			UB	123	12	154	267	
	Tomato	20		241	110	221	438	
	Potato	20	LB	199	0.0	233	370	
			UB	204	12	233	370	
Breakfast cereals (Not containing nuts, raisins)		20	908	166	814	2262	DW	
Canned fruits		6	LB	42	0.0	0.0	163	FW
			UB	48	5.0	16	163	

^aIn this column, DW and FW refer to the dry weight base and fresh weight base respectively.

^bThe Ni content of fresh/frozen, canned and in glass legumes were originally obtained on FW. Recalculation was done to make them available on DW.

^cThese bean samples were collected from different types bean including white bean (N = 11), kidney bean (N = 10), broad bean without pod (N = 3), black bean (N = 3), green bean without pod (N = 2), black eyed bean (N = 1), mung bean (N = 1) and calypso bean (N = 1).

^dLB = Lower bound scenario at which results below LOD/LOQ were substituted with zero.

^eUB = Upper bound scenario at which results below LOD were replaced with reported value as the LOD and those lower than LOQ were substituted with the LOQ.

^fSamples were collected from different trees located in different areas of Flemish region.

In comparison to previous studies, the mean concentrations of Ni in the current study were still high. There is considerable variation of the Ni concentration among legumes. The peas and peanuts showed the lowest variation and overall the lowest Ni content. Especially beans show high variation in this study (Figure 1).

The effect of the package type, i.e. plastic bag, metal can and glass, on the Ni contamination in beans, legumes with pods and peas was assessed. For the same types of the aforesaid legumes, no significant differences were observed in Ni content of dry packed, conserved in glass, canned and fresh-frozen legumes. However, the mean concentration of Ni in dry samples of lentil ($N = 10$) was significantly higher (p -value = 0.021) than that of the canned lentils ($N = 4$) on dry weight basis (DW). Lentils have small seeds which are soaked easily. Thus, the probability of washing Ni off during the hydration stage of the canning process may be quite high. However, the sample size is not large enough ($N = 4$) to draw a conclusion having sufficiently strong statistical power.

To investigate this in more detail, a few dry lentil samples ($N = 5$) were soaked, cooked and analysed. The results show that only between 15% and 17% of the total Ni was retained in the soaked and cooked lentils. This confirms that Ni could be washed out easily during the hydration stage of the canning process.

The Ni content in soy products varied from 89 $\mu\text{g kg}^{-1}$ to 942 $\mu\text{g kg}^{-1}$ (on fresh weight basis, i.e. FW) in the current study. The mean concentration of Ni in the four different types of soy products ($N = 34$) was 274 $\mu\text{g kg}^{-1}$. This mean Ni concentration was

lower than a previously reported average Ni content of 5100 $\mu\text{g kg}^{-1}$ for soy products (EVM 2002). Having the smallest sample size ($N = 7$), tofu products demonstrated the highest variation and highest overall Ni content. The category of desserts and creams ($N = 12$) had the lowest variation, as well as the lowest overall content of Ni. Although the mean concentration of Ni in tofu was slightly higher than in drinks, desserts and creams, no significant differences were observed among these three categories (p -value = 0.197). Soy products originated from the soybean which is a nitrogen-fixing plant. So Ni presents in the structure of the enzymes in these products. According to the nutrition facts of the products, tofu has more protein content, e.g. 8 g per 100 g (FW), than soy drinks, e.g. 3 g per 100 g (FW) and soy creams, e.g. 2 g per 100 g (FW). This might explain the observed differences in the Ni content of these products. A mean Ni concentration of 227 $\mu\text{g kg}^{-1}$ was obtained for soy drinks (imitation milk product). Aforesaid mean Ni content was higher than a mean Ni content of cow's milk, i.e. 1.9 $\mu\text{g kg}^{-1}$ (UB), in current study and a reported Ni content for cow's milk, i.e. 1 $\mu\text{g L}^{-1}$ to 100 $\mu\text{g L}^{-1}$, in US (WHO 2005).

Almond, pistachio and hazelnut were selected initially to investigate the Ni content of nuts. These nuts were chosen due to their high frequency of consumption by the Belgian population (Van de Perre et al. 2015). In addition, a few walnut samples ($N = 6$) from the Flemish region of Belgium were also analysed to investigate their Ni content as well. The Ni concentration ranged from 577 to 1092 $\mu\text{g kg}^{-1}$, from 1196 to 3846 $\mu\text{g kg}^{-1}$, from 406 to 1740 $\mu\text{g kg}^{-1}$ and from 721 to 4624 $\mu\text{g kg}^{-1}$ (DW) for almond, hazelnut, pistachio and walnuts, respectively (Figure 2).

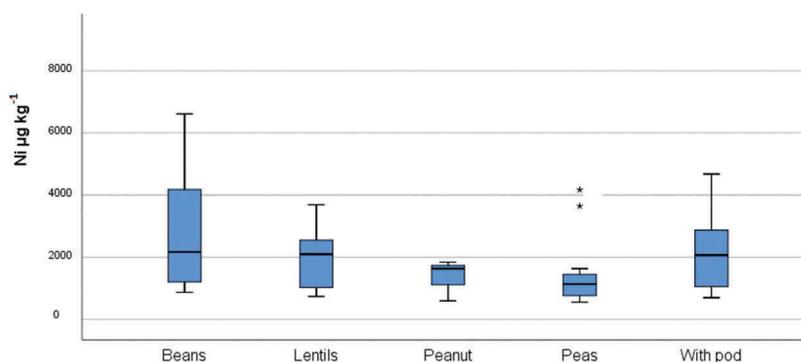


Figure 1. Box plot of the Ni content, $\mu\text{g kg}^{-1}$ (DW), for five different types of legumes including beans ($N = 33$), legumes with pods ($N = 8$), lentils ($N = 14$), peas ($N = 16$) and peanuts ($N = 3$).

An average Ni content of $2200 \mu\text{g kg}^{-1}$ was found for 48 hazelnut samples in the EFSA report (2015). For almonds, an average Ni concentration of $830 \mu\text{g kg}^{-1}$ was reported by Ščančar et al. (2013). This perfectly fitted in the range that was found for almonds in our current study. In another study conducted by Cabrera et al. (2003), a range of Ni content from 100 to $640 \mu\text{g kg}^{-1}$ was reported for 51 samples of different types of nuts. An average Ni concentration of $1800 \mu\text{g kg}^{-1}$ was reported by Ysart et al. (2000) for a group of nuts. In the current study, an overall average concentration of $1653 \mu\text{g kg}^{-1}$ was found for nuts, which is in line with the previously reported values.

The Ni content in chocolate varied from $883 \mu\text{g kg}^{-1}$ to $8457 \mu\text{g kg}^{-1}$ (FW). The wide concentration range covers the value of $1090 \mu\text{g kg}^{-1}$ reported by Kohiyama et al. (1992), $1173 \mu\text{g kg}^{-1}$ reported by Dohnalova et al. (2017), $1600 \mu\text{g kg}^{-1}$ reported by Rehman and Husnain (2012) and $2763 \mu\text{g kg}^{-1}$ reported by Dahiya et al. (2005). The Ni content was checked for correlation with the minimum cacao content that is indicated on the product package (Figure 3). For all chocolate products ($N = 44$), a significant positive correlation found between the percentage of cacao (Pearson $R = 0.784$) and the Ni content. The cacao beans contain carbohydrates in the form of soluble starch and insoluble dietary fibres. Cacao's dietary fibres have a high affinity for trace elements and can be a probable source for these elements (Valiente et al. 1996). In this way, by increasing

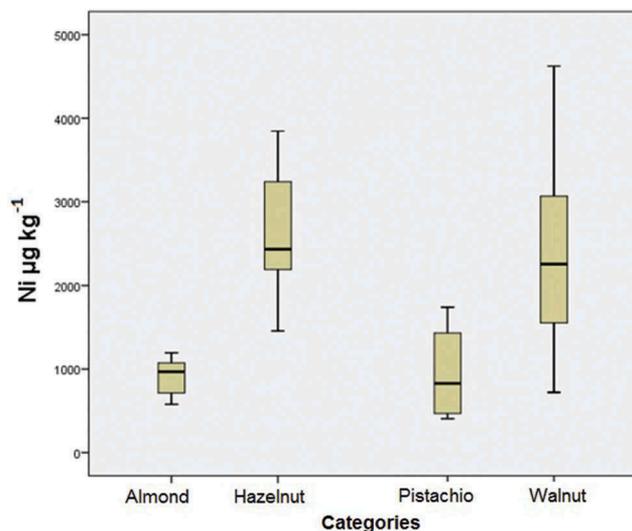


Figure 2. Box plot of the Ni content $\mu\text{g kg}^{-1}$ (DW), in almond ($N = 8$), hazelnut ($N = 6$), pistachio ($N = 4$) and walnuts ($N = 6$).

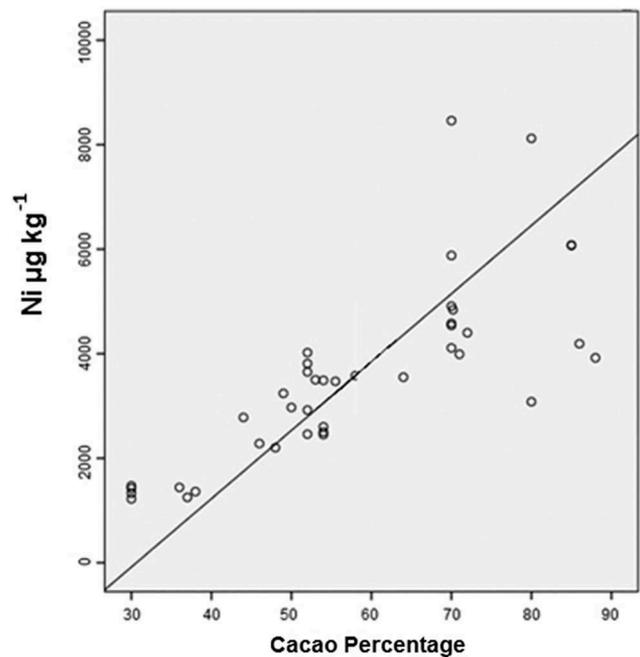


Figure 3. Correlation between the Ni content, $\mu\text{g kg}^{-1}$ (FW), and cacao percentage in chocolate ($N = 44$). The Pearson correlation coefficient R amounted 0.784.

the cacao content of the chocolate, their dietary fibre and trace element content may increase as well.

In addition, two groups of chocolate products were considered in the current study, i.e. sugar-based and polyol-based (= reduced energy) chocolate. According to a sampling plan of this research, chocolate with polyols should be analysed to check their Ni content since there is a possibility of the Ni contamination during production of polyols. However, it was observed that the Ni content in the sugar-based chocolate ($4140 \mu\text{g kg}^{-1}$) was significantly higher (p -value = 0.002) compared to the polyol-based chocolate ($2620 \mu\text{g kg}^{-1}$). This may be attributed to the fact that the cacao content of the sugar-based chocolate is higher (65% on average) than that of the polyol-based chocolate (48% on average).

A mean concentration of $1226 \mu\text{g kg}^{-1}$ (FW) for Ni was found in chocolate spreads. This fully met the expectations for this kind of food product due to the fact that high Ni concentrations were also found in their major ingredients coca and hazelnuts.

The Ni content of the coffee products (dry coffee ingredients for preparing beverages) ranged from $312 \mu\text{g kg}^{-1}$ to $4268 \mu\text{g kg}^{-1}$ (Table 5) with results expressed on DW basis. These results exhibited a broader range compared to the values

between $627 \mu\text{g kg}^{-1}$ and $931 \mu\text{g kg}^{-1}$ reported by Nędzarek et al. (2013). Such high variation in Ni content probably could not be attributed to any difference in the packaging nor grinding of the coffee. Instead, it is believed that the geographical origin of the coffee beans plays an important role. Thus, the trace element profile of coffee is used as the indicator to specify its geographical origin (Anderson and Smith 2002; Kelly et al. 2005; Bertrand et al. 2008; Gonzalez et al. 2009). More specifically, the Ni content in the soil on which the trees were grown, the extent of the metal uptake by the plant and the extent of leached Ni during a washing step of the green coffee beans processing (Vincent 1987) are affecting the Ni content of the coffee beans.

The mean concentration of Ni in the solid tea ingredients ($N = 22$) ranged from 3500 to 9700 $\mu\text{g kg}^{-1}$ (Table 5), expressed on DW basis. An average Ni content of $6200 \mu\text{g kg}^{-1}$ and $6300 \mu\text{g kg}^{-1}$ was observed in the green ($N = 11$) and black tea ($N = 11$), respectively. A study conducted by Szymczycha-Madeja et al. (2013) reported the concentration range of Ni in four different herbal teas as between 3080 and 8840 $\mu\text{g kg}^{-1}$. Another study on six different tea types, including white tea, green tea, oolong tea, and black tea (all *Camellia sinensis*), flowers of herbal chamomile (*Matricaria chamomilla*) and hibiscus (*Hibiscus sabdariffa*), was conducted by Ščančar et al. (2013). They reported a range of Ni concentration from 1210 to 1440 $\mu\text{g kg}^{-1}$ for aforementioned tea samples. In our current study, the overall level of Ni in the black and green tea categories was similar and no significant differences were observed between these two categories.

The effect of the package type on the Ni content in tomato sauces ($N = 12$) was evaluated by collecting from the same brands three different types of packaging including can ($N = 4$), glass ($N = 4$) and laminated carton ($N = 4$). No significant differences were observed (p -value = 0.572) among the Ni content of the samples with different types of packages. The range of the Ni concentration in 20 samples of tomato passata was 36 to 281 $\mu\text{g kg}^{-1}$ (FW). Data in literature regarding the Ni concentrations in tomato passata are currently lacking. Nevertheless, a concentration of $54 \mu\text{g kg}^{-1}$ was reported by Li et al. (2012) in a study conducted on tomatoes in China. When taking the concentration process into

account during the preparation of the sauces (Van de Perre et al. 2014), it is logical that the Ni concentration may reach a maximum of $281 \mu\text{g kg}^{-1}$.

According to Van de Perre et al. (2015), raisins and figs are the most consumed dried fruits by the Belgian population, and for this reason, they were included in the sampling plan. The Ni concentration ranged between 861 and 3683 $\mu\text{g kg}^{-1}$ (DW) for figs ($N = 8$). For the raisins ($N = 12$), this range varied from 72 to 253 $\mu\text{g kg}^{-1}$ (DW). In EFSA (2015), a mean concentration of 1800 $\mu\text{g kg}^{-1}$ was reported for figs ($N = 3$), and an overall mean Ni content of $160 \mu\text{g kg}^{-1}$ was reported for dry fruits ($N = 13$). For now, it is unclear where the Ni in figs originates (from the environment during growth, e.g. the soil, or from other sources). Further investigation is necessary to address this.

Fresh and frozen vegetables, including carrot ($N = 21$), spinach ($N = 21$), tomato ($N = 20$) and potato ($N = 20$), were collected from the Belgian market. The mean concentrations of Ni in carrot, spinach, tomato and potato were 21.3, 109, 241 and 199 $\mu\text{g kg}^{-1}$ (FW), respectively. All mean values are calculated on lower bound (LB) scenario bases. The Ni concentrations in the carrot were very low, with lots of samples having Ni contents below LOD and LOQ. A mean concentration of $160 \mu\text{g kg}^{-1}$ (LB) was mentioned in the EFSA report (2015) for 303 samples of carrots. Generally, the mean Ni concentration of the fresh spinach ($N = 8$), i.e. $193 \mu\text{g kg}^{-1}$, was higher than that of its frozen counterpart i.e. $56.5 \mu\text{g kg}^{-1}$. In the production process of frozen spinach, there is a blanching step that can lead to leaching Ni from the leaves. This can explain the higher Ni content of the fresh spinaches in comparison to the frozen ones. For most frozen spinach products ($N = 13$) available in Belgian supermarkets, the Ni content was lower than LOD/LOQ. An average Ni concentration of $264 \mu\text{g kg}^{-1}$ (LB) was reported (EFSA 2015) for potato samples ($N = 205$) as the main crop representing "Starchy root and tuber vegetables". This average concentration of Ni is compatible with the mean value obtained in the current study, i.e. $199 \mu\text{g kg}^{-1}$ (LB). In this study, the results that obtained for fresh and frozen vegetables are in discrepancy with the reported results by Smart

and Sherlock (1987) for Ni concentration in canned vegetables in UK. They reported the highest concentration of Ni in canned vegetables, sugars, bread and cereals. Smart and Sherlock (1987) stated that the use of Ni in utensils, food-processing equipment and in catalysts can lead to the contamination of food by Ni in the UK diet (Smart and Sherlock 1987). Packaging materials were not found to have an effect on the final Ni concentration in this study.

The focus in the subcategory of breakfast cereals was laid only on pure breakfast cereals and not the products containing also other ingredients, i.e. chocolate, nuts and/or dried fruits. This was done to only evaluate the Ni content of cereals since other ingredients such as chocolate and dried fruits were investigated as separate categories in the current study. In total, 20 samples of the breakfast cereals were analysed, i.e. mixed cereals ($N=1$), oat ($N=13$) and wheat ($N=6$). In general, an average Ni content of $908 \mu\text{g kg}^{-1}$ (DW) was obtained in these samples. In the EFSA (2015) report, an average Ni content of $630 \mu\text{g kg}^{-1}$ (LB) was reported for 313 samples of breakfast cereals. It was not specified in the EFSA report (2015) whether this mean Ni content was obtained for the plain breakfast cereals or mixture of the cereals with other ingredients such as chocolate and nuts. The mean Ni contents of the oat ($1104 \mu\text{g kg}^{-1}$) and wheat samples ($540 \mu\text{g kg}^{-1}$) were marginally different (p -value of 0.058) (Figure 4). Sharma (2013) included wheat and oat in the list of high Ni containing foods when providing a guideline for Ni sensitive patients with specific dietary recommendations.

For the previously analysed samples in this study, no significant effect of canning on the Ni content of the food products was observed. Mainly two types of cans are available on the market, i.e. aluminium and steel cans. Aluminium cans are using for packaging soft drinks, while steel cans are more using for the foods. Two types of coating layers, i.e. a tin coating and a lacquer layer, can typically be used inside the steel cans. From these two coating layers, the tin coating is always present. Therefore, an uncoated can refers to a can without a lacquer layer. In the European market, the number of canned products without a lacquer layer is limited. In this study, in total, 6 samples of foods in

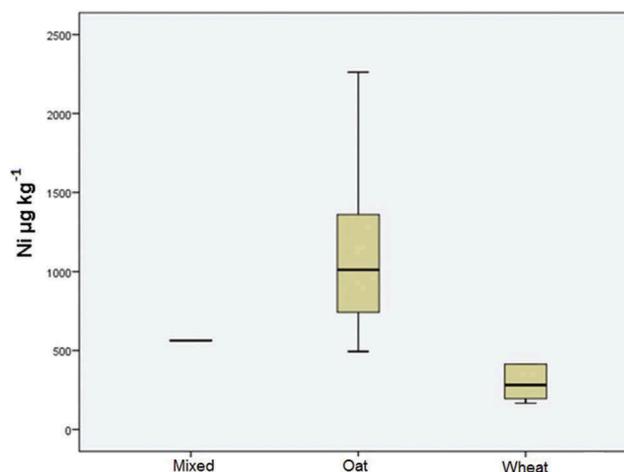


Figure 4. Box plot of the Ni content, $\mu\text{g kg}^{-1}$ (DW), in mixed ($N=1$), oat ($N=13$) and wheat ($N=6$) breakfast cereals.

uncoated cans, i.e. peaches ($N=2$), apricots ($N=2$) and pears ($N=2$), were analysed. No detectable Ni was found in the pears. According to the LB scenario, the mean Ni content of the apricots and peaches was 15 and $134 \mu\text{g kg}^{-1}$ (FW) respectively. Comparing to the other plant-based products such as legumes and nuts, Ni concentration in canned fruit samples was not high.

Animal-based food products

In total, 113 food samples of animal origin were collected and analysed in this study. In general, the Ni content of the animal-based products was found to be much lower than that of the plant-based food products. Among the analysed animal-based products, gelatins, mayonnaises (as a representative of the emulsified sauces), fresh eggs, UHT milk with direct steam injection, fishes and shrimps showed average Ni concentrations lower than LOD/LOQ (Table 6).

For dairy products, UHT milk ($N=13$), yoghurt ($N=9$) and UHT milk with direct steam injection ($N=2$) were sampled. The latter milk samples were included to check the effect of direct steam injection at the time of processing on the final Ni content of the milk. Direct steam injection may provoke a higher corrosion from processing equipment and therefore could be a source of increased Ni in foods due to leaching from food contact materials. No effect of the direct steam injection on the Ni content of the milk products was observed. In the current

Table 6. Summary statistics for the Ni content ($\mu\text{g kg}^{-1}$) in different animal-based food products purchased from the Belgian market (N = 113). Results are based on the fresh weight of the edible portion (FW).

Category	Product	N	Ni content ($\mu\text{g kg}^{-1}$) FW				
			Mean	Minimum	P50	Maximum	
Gelatines	Pure gelatines	6	0.0 ^a	NA ^d	NA	NA	
	Gelatine products ^c	14	0.0 ^a	NA	NA	NA	
Emulsified sauces	Mayonnaise	20	0.0 ^a	NA	NA	NA	
Eggs	Fresh eggs	15	LB	0.0 ^a	0.0	0.0	26
			UB	15.5 ^b	4.8	16.1	26
Dairy	Milk	13	LB	0.9	0.0	0.0	3.4
			UB	1.9	0.36	1.2	3.4
	Yoghurt	11	LB	3.8	0.0	3	10
			UB	3.9	0.3	3	10
Fish and Seafood	Milk – direct steam injection	2	0.0 ^a	NA	NA	NA	
	Fatty fish	5	0.0 ^a	NA	NA	NA	
	Lean fish	6	0.0 ^a	NA	NA	NA	
	Mussels	7	LB	68	0.0	57	227
			UB	71	12	57	227
	Shrimps	8	LB	0.0 ^a	0.0	0.0	61.5
UB			38.9 ^b	12	40	61.5	

^aNi was below the level of reliable detection or it was not detected (ND).

^bNi was present at trace level that was below the limit of reliable quantification (TR).

^cGelatine containing candies (N = 14).

^dAbbreviation of not applicable.

study, the mean Ni content of the milk and yoghurts ranged from 0 to 3.9 (LB-UB) $\mu\text{g kg}^{-1}$. EFSA (2015) reported a Ni content of 93 $\mu\text{g kg}^{-1}$ (LB) for milk and other dairy products (N = 631).

In total, 26 samples of seafood were analysed in the current study. The samples were collected from fatty fish (N = 5), lean fish (N = 6), shrimps (N = 8) and mussels (N = 7). Apart from the mussels, the Ni content in the other types of seafood samples, i.e. fishes and shrimps, was below the LOD/LOQ. The Ni concentration in mussels ranged from 0.0 to 227 $\mu\text{g kg}^{-1}$ (LB). Guérin et al. (2011) reported a mean Ni concentration of 299 $\mu\text{g kg}^{-1}$ for 159 samples of seafood.

In another study conducted by Skibniewska et al. (2009) in Poland, a low mean Ni concentration of 40 $\mu\text{g kg}^{-1}$ was reported for 9 samples of fresh water fishes (roach, bream and carp).

Drinks

In total, 189 drink samples were analysed in the current study. The samples collected were beer, hot beverages of tea, iced tea drinks, commercially available coffee drinks and hot coffee beverages (Table 7).

The Ni concentration in beers ranged from 1.5 $\mu\text{g kg}^{-1}$ to 33.8 $\mu\text{g kg}^{-1}$. Distinction between three different subcategories of beers was made based on

Table 7. Summary statistics for the Ni content ($\mu\text{g kg}^{-1}$) in different drinks purchased/prepared in the current study (N = 189).

Category	Product	N	Ni content ($\mu\text{g kg}^{-1}$)			
			Mean	Minimum	P50	Maximum
Beer	Pilsener	46	4.5	1.5	4.4	8.1
	Top-fermented beer	67	7.7	2.0	6.7	21.4
	Sour beer	35	12.9	2.0	10.5	33.8
Coffee beverages ^a (prepared with ultrapure water through domestic protocol)	Ground	5	16.8	6.0	8.1	36
	Unground	5	7.0	3.0	5.4	13
Coffee beverages ^b (prepared with ultrapure water through Golden coffee protocol)	Ground	5	12	2.0	7.8	26
Commercial coffee drinks ^c		3	17	4.0	9.0	38
Tea beverages ^d (prepared with ultrapure water through domestic protocol)	Black tea	4	85	72	73	121
	Green tea	4	194	112	207	252
	Black tea	2	56	28	56	84
Tea beverages ^e (prepared with ultrapure water through ISO 3130 protocol)	Green tea	2	85	66	85	105
	Without flavour	6	34	13	32	58
Ice tea		5	26	14	28	34
	Lemon					

^aRefers to the coffee beverages made through non-standard (domestic) practices.

^bRefers to the coffee beverages made through standard (std) protocols.

^cEspresso macchiato.

^dRefers to the tea beverages made through non-standard (domestic) practices.

^eRefers to the tea made through standard protocols.

the brewing process and ingredients, namely top-fermented beers of high alcohol percentage, pilsener beers and sour beers (Figure 5). No correlation of the Ni concentration with the alcohol percentage was found in beers, even within the different subcategories.

In order to trace source of the Ni contamination in beers, four top-fermented beer samples were subjected to centrifugation in order to separate the yeast cells. Comparing to the bulk supernatants, it was verified that the separated *Saccharomyces cerevisiae* (yeast) cell fractions contained a higher amount of Ni. Bioaccumulation of Ni in the yeast cells can occur through the uptake in the cell or bio-sorption on the cell's surface. Results indicate that indeed an enrichment of Ni was found in the filtration residue (Figure 6).

Generally, both coffee and tea beverages were prepared by two different protocols (domestic and standard). This was done to see how the preparation protocol can affect the final Ni concentration in beverages. Furthermore, an effect of standardising a preparation protocol on the final Ni concentration in beverages was assessed as well.

For the coffee beverages prepared through domestic practices, the concentration range of Ni

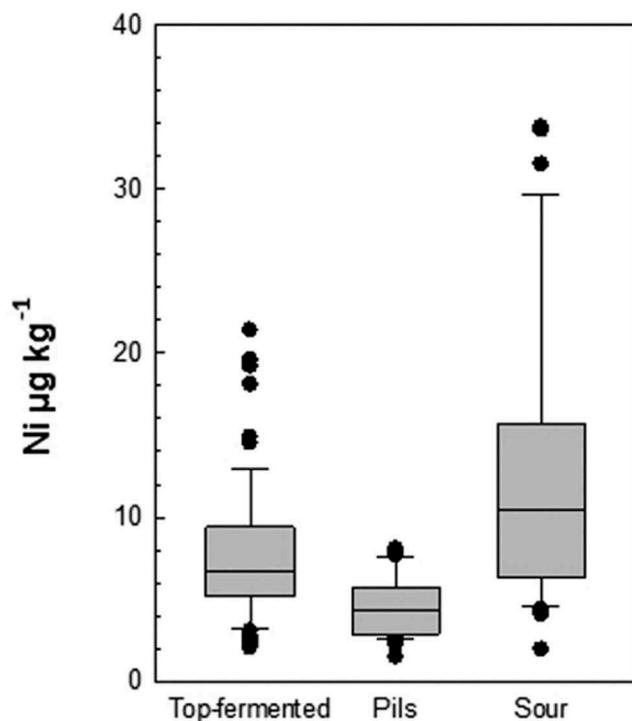


Figure 5. Box-plot of the Ni content in three categories of beers including top-fermented ($N = 67$), pils ($N = 46$) and sour beers ($N = 35$). Analysis conducted in duplicate per sample.

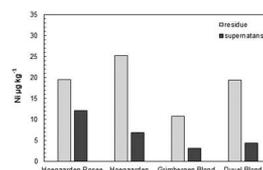


Figure 6. The Ni content ($\mu\text{g kg}^{-1}$) in different fractions of four top-fermented beers. The Ni content in the yeast residue (light grey) is higher (two-tailed p -value = 0.022) than the bulk liquid (dark grey).

was from $3 \mu\text{g kg}^{-1}$ to $36 \mu\text{g kg}^{-1}$. During a brewing process of coffees, Ni extraction rate (from the coffee ingredients, i.e. solid, into the corresponding coffee beverages) was calculated for 10 samples of coffees. The extraction rates were ranged from 7% to 20%. A few samples of coffee beverages ($N = 5$) were prepared through the Golden cup standard protocol (SCA 2017). The Ni concentration for these samples ranged from 2 to $26 \mu\text{g kg}^{-1}$, while for the coffees beverages prepared from the same coffee ingredients using an own (domestic) protocol, this value ranged from 3 to $36 \mu\text{g kg}^{-1}$. According to a paired sample T-test, no significant difference was observed (p -value = 0.182) between the coffee prepared through the domestic and the standard protocol. In the study conducted by Müller et al. (2015), the effect of the machine types on the extent of the Ni release was reported. They reported the highest levels of the Ni leaching for portafilter machines (beyond the release limits proposed by European council). Therefore, a careful rinsing routine, especially after decalcification, was recommended for these machines (Müller et al. 2015).

Furthermore, three coffee drinks (espresso macchiato) that are commercially available on the Belgian super markets were analysed for their Ni content. The Ni concentration ranged from 4 to $38 \mu\text{g kg}^{-1}$ which is still comparable with the concentration range obtained for the other prepared coffee beverages in our current study.

For the tea beverages prepared through domestic practices ($N = 8$), the range of Ni concentration was found to vary from $72 \mu\text{g kg}^{-1}$ to $252 \mu\text{g kg}^{-1}$. Mean concentrations of Ni in the beverage prepared from green tea and black tea were $194 \mu\text{g kg}^{-1}$ and $84.9 \mu\text{g kg}^{-1}$, respectively. The mean concentrations of Ni in green and black tea beverage were not significantly different (p -value = 0.057), although the samples size

is too small to draw strong conclusions. A few tea beverage (N = 4) were prepared using the ISO 3103 standardised protocol for brewing tea (ISO 1985). The concentration of Ni ranged from 28 to 105 $\mu\text{g kg}^{-1}$. No significant differences were observed (p -value = 0.188) between the tea beverage prepared through the standard and the domestic protocols. Besides, the Ni concentration varied from 13 to 58 $\mu\text{g kg}^{-1}$ for the ice tea samples without flavourings (N = 6). The Ni content in the ice tea samples with lemon (N = 5) ranged from 14 to 34 $\mu\text{g kg}^{-1}$.

Conclusions

Samples from three main categories, i.e. plant-based food products, animal-based food products and drinks, were collected according to a risk-based sampling plan. Elevated Ni concentrations were observed in plant-based foods: chocolates, legumes, nuts, figs, peanut butters, chocolate spreads and breakfast cereals. The observed high Ni concentration in the legumes is attributed to the presence of Ni in the structure of the enzymes involved in the nitrogen fixation process in the plants. The Ni content of the soy drinks was found to be higher than cow's milk. A positive correlation was observed between the cacao percentage and the Ni content of chocolate. Lowest Ni contents among the plant-based products were observed for vegetables and the canned fruits.

Across the three main food categories, lowest average Ni concentrations (< LOD/LOQ) were observed for animal-based products including gelatins, mayonnaises, eggs, milk with direct steam injection, fishes and shrimps. Compared to the Ni concentration of the plant-based products, the Ni content of drinks was lower as well. No correlation was found with the indicated alcohol percentage of the beers. Furthermore, no effect of the package type on the Ni contamination of the products was generally found in this study.

Ultimately, it can be concluded that animal-based foods and drinks are not priority groups to be focused on when evaluating Ni in foods. However, plant-based foods are generally of potential concern due to their high Ni contents and their high frequency of consumption by population. An exposure assessment study still needs to be conducted to further investigate the real daily intake of Ni through

consuming different foods and their potential impact on human health.

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