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Evaluation of Some Toxic and Essential Trace Elements in Children Foods and Infant Formulae by Using ICP-OES

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In Turabah province of Saudi Arabia, infant formula and children′s food products were investigated to determine the concentrations of essential trace elements, namely Mn, Ni, V and Si, and toxic trace elements, namely Al, Ba, Pb, Cd, and As, present in them. Their daily intake amounts were calculated and compared with provisional tolerable daily intake (PTDI), which was calculated based on the provisional tolerable weekly intake (PTWI), which was recommended by joint FAO/WHO. The results obtained were found to be below the PTDI levels. Some samples exhibited a large amount of Al. The PTWI of Si and V is not established; however, their concentrations were compared with those reported in other studies. In some samples, a high concentration of Si was observed. The method employed for evaluation was based on the limit of quantification (LOQ), limit of detection (LOD), and recovery percent of 92.3–101.6%. Accuracy and precision were employed for evaluating the results.

Keywords: Children foods, ICP-OES, Infant formulae, Trace elements.

INTRODUCTION

Commercial food products such as popcorn, potato chips, cocoa, biscuits, cocoa sweets, indomie, cheese, powder milk and cerelac are widely consumed by children and infants. Moreover, mothers use food products that provide nutrients for babies. These food products contain different concentrations of trace elements and some of these trace elements, including potassium, sodium, copper, calcium and zinc, are act as essential nutrients when taken in micro amounts as they have different biochemical functions. However, high concentrations of these elements can be toxic. Elements including nickel, manganese, silicon and vanadium are classified as probably essential nutrients. Small amounts of Mn are required by humans and many enzymes comprise Mn and are activated by it [1]. The oral uptake of manganese and deficiency of some minerals were considered to enhance the incidences of neurological symptoms [2-4].

Silicon is essential because of its function in connective tissues and bones [5-9]. It′s deficiency can reduce bone concentrations of Mg, Ca, K, Cu and Zn [10]. Furthermore, silicon deficiency can influence the absorption and behaviour of minerals such as Mg, Al and Cu [11].

Cadmium, lead, barium, aluminium and arsenic are some of potentially toxic elements. Cadmium can demineralize bones and is toxic to kidneys [12]. The effects of aluminium toxicity are severe in infants because their gastrointestinal tracts are more permeable to aluminium than those of adults [13]. Aluminium exhibits potent neurotoxic properties, and its long-term consumption may impair the mental development and negatively influence bone health in infants [14]. International Agency for Research on Cancer has classified lead(II) as a group 2B carcinogen and Pb toxicity leads to sterility, miscarriage, and infant morbidity and mortality [1]. However, lead deficiency adversely influences the intellect of children [15]. Lead(II) toxicity disrupts certain cellular signal processes and affects the function of various enzymes and proteins [16]. Barium is not an essential element; however, barium chloride and barium carbonate leads to hypopotassaemia, gastroenteritis, cardiac arrhythmias, hypertension and skeletal muscle paralysis [17]. The biological effects of arsenic related its chemical form. Inorganic arsenic is group 1 carcinogen that causes lung, skin, and urinary bladder cancer [18].

Analytical studies have increasingly focused on detecting the trace elements in baby foods and infant formulae for health concerns. Research on this topic is increasing because of increasing awareness regarding trace element concentrations in baby foods [19]. Optical emission spectroscopy is commonly employed to detect trace elements present in infant formulae and baby foods, because it provides large linear dynamic ranges, and accurate and precise results. Before the determination of analytes in dissolved aqueous samples, the samples should be acid preserved and filtered [20]. Inductively coupled plasma optical emission spectroscopy (ICP-OES) functions using the interaction between electromagnetic radiation and molecules. ICP-OES can be used for a simultaneous analysis of approximately 60 elements by using a single source (plasma). Accurate sample preparation is crucial, because it influences the results. Before ICP-mass spectroscopy and ICP-OES analysis microwave digestion must be performed to prepare and clean the samples.

In this study, four probably essential (Ni, Mn, Si, and V) and five toxic elements (Pb, Al, Cd, Ba and As) present in infant formulae and baby foods of 19 different brands were determined and evaluated through ICP-OES after microwave digestion.

EXPERIMENTAL

Samples collection and preparation: A total of 57 infant food samples (3 brands) and baby-food (16 brands) samples were purchased from various pharmacies and local supermarkets in Turabah province of Saudi Arabia. All samples were popular infant formulae and baby foods, such as popcorn, potato chips, cocoa sweets, biscuits, cerelac, cream cheese, baby powder milk, and indomie. The shelf lives, production dates and expiry dates of the samples were recorded.

To prepare the samples, PTFE-TFM digestion vessels with liners were employed. Approximately 0.5 g of each purchased product was weighed and then was transferred into different vessels. A total of 12 and 2 mL of conc. HNO_3 and H_2O_2, respectively, were added in each vessel with the sample. In some vessels, only 12 and 2 mL of conc. HNO_3 and H_2O_2, respectively, were added to obtain reagent blanks. The vessels were sealed and placed in a microwave-digestion rotor. When the samples were completely digested, each sample was transferred into a different volumetric flask of 50 mL and diluted to the mark with deionized-distilled water (DDW).

The samples and standards of trace elements, namely Al, Ba, Pb, As, Ni, Cd, Mn, V and Si, were analyzed using Plasma Quant® PQ 9000 ICP-OES, Dual View PLUS plasma observation of Analytik Jena′s (Germany). The conditions of ICP-OES operation were optimized to achieve maximum sensitivity (Table-1).

TABLE-1
ICP-OES OPERATING PARAMETERS FOR DETERMINATION OF SELECTED TRACE ELEMENTS

Parameters	Setting
Power	1200 W
Plasma gas flow	15 L/min
Auxillary gas flow	0.5 mL/min
Nebulizer gas flow	0.4 L/min
Sample pump rate	0.4 mL/min

Statistical analysis: For result evaluation, Q, F and ANOVA tests ($p = 0.05$) were performed using Microsoft Excel. Moreover, Microsoft Excel was used to determine variable differences in the nutrient samples. The concentrations obtained were reported as the average value ± confidence interval (at 95% confidence).

RESULTS AND DISCUSSION

The results were expressed as x ± s , where x and s denote the mean values and standard deviation, respectively. To evaluate the analysis method used, the LOD, LOQ and confirmation reliability were determined using H_2O_2 and conc. HNO_3 (Table-2). The results of microwave acid digestion indicated that the recovery percentage (%) of certified reference materials was in the range of 92.3-101.6%.

TABLE-2
LOD AND LOQ (ppm) FOR DETERMINATION OF SELECTED TRACE ELEMENTS

Element	Wavelength (nm)	LOD	LOQ
Si	251.611	0.0085	0.021
Al	308.212	0.0055	0.064
Mn	257.610	0.0005	0.006
V	292.401	0.0016	0.017
Ni	231.604	0.0011	0.012
Ba	233.527	0.0052	0.085
Pb	220.353	0.0062	0.086
Cd	226.499	0.0051	0.013
As	188.979	0.0054	0.046

Daily exposures of trace elements were determined based on the dosages recommended by the manufacturers of infant formulae and the food behaviour of the children obtained from studies conducted in the local region. In this study, the age of infants and children was up to 3 years and in the range of 6-12 years (average age = 9 years), respectively. The daily intake of analyzed trace elements from nutrients formulae was calculated as µg/kg bw/day (Tables 3B-7B) and then compared with PTDI, which is calculated using PTWI that recommended by FAO/WHO Expert Committee on Food Additives (GECFA) (Table-8) [12,21,22].

Tables 3A-7A and Figs. 1-5 present the Al concentration (ppm) obtained in various food brands, whereas Tables 3B-7B present the average daily intakes of Al (µg/kg bw/day) for different food brands. The results indicated that brand 6 (biscuit)

TABLE-3A
AVERAGE CONCENTRATIONS OF ELEMENTS OF DIFFERENT BRANDS OF POTATO CHIPS AND POPCORN

Element	Concentration (ppm)			
	Potato chips (different flavor)			Popcorn
	Brand 1	Brand 2	Brand 3	Brand 4
Si	275.4±1.02	374.5±1.59	163.4±0.54	74.76±1.06
Al	27.93±0.73	38.45±0.72	19.65±0.58	24.05±0.81
Mn	9.73±0.46	3.84±0.29	1.24±0.056	3.12±0.18
V	1.48±0.061	1.13±0.058	0.430±0.038	0.459±0.041
Ni	0.649±0.043	3.074±0.31	2.23±0.33	2.87±0.36
Ba	0.999±0.046	0.699±0.049	0.639±0.044	0.569±0.039
Pb	0.010±0.003	0.110±0.004	0.030±0.003	0.050±0.004
Cd	ND	ND	ND	ND
As	ND	ND	ND	ND

Mean value ± standard deviation (n = 3), ND = not detected

TABLE-3B
AVERAGE DAILY INTAKES OF ELEMENTS IN DIFFERENT BRANDS OF POTATO CHIPS AND POPCORN

Element	Daily intakes of elements (μg/kg bw/day)*			
	Potato chips (different flavor)			Popcorn
	Brand 1	Brand 2	Brand 3	Brand 4
Si	177.7	241.6	121.2	120.6
Al	18	24.8	14.6	38.8
Mn	6.3	2.5	0.52	5.03
V	0.95	0.73	0.32	0.74
Ni	0.40	2.0	1.65	4.63
Ba	0.65	0.45	0.47	0.92
Pb	0.006	0.071	ND	0.08
Cd	ND	ND	ND	ND
As	ND	ND	ND	ND

*Calculated according to body weight average = 31 kg

TABLE-4A
AVERAGE CONCENTRATIONS OF ELEMENTS OF DIFFERENT BRANDS OF BISCUITS

Element	Concentration (ppm)		
	Brand 5	Brand 6	Brand 7
Si	79.86±1.06	278.60±1.13	105.50±1.05
Al	20.91±0.74	291.90±1.14	3.57±0.31
Mn	8.30±0.46	9.43±0.45	8.53±0.31
V	0.940±0.05	1.42±0.05	0.720±0.03
Ni	1.03±0.04	3.28±0.29	3.00±0.18
Ba	2.34±0.19	0.440±0.03	0.690±0.04
Pb	0.08000.03	0.2299±0.01	ND
Cd	ND	ND	ND
As	ND	ND	ND

Mean value ± standard deviation (n = 3), ND = not detected

TABLE-4B
AVERAGE DAILY INTAKES OF ELEMENTS IN DIFFERENT BRANDS OF DIFFERENT BISCUITS

Element	Daily intakes of elements (μg/kg bw/day)*		
	Brand 5	Brand 6	Brand 7
Si	128.8	462.0	184.5
Al	33.7	484.0	6.2
Mn	13.40	15.60	14.9
V	1.52	2.35	1.26
Ni	1.70	5.44	5.30
Ba	3.80	0.73	1.20
Pb	0.13	0.38	ND
Cd	ND	ND	ND
As	ND	ND	ND

ND = Not detected; *Calculated according to body weight average = 31 kg

TABLE-5A
AVERAGE CONCENTRATIONS OF ELEMENTS OF DIFFERENT BRANDS OF SWEETS WITH COCOA

Element	Concentration (ppm)		
	Brand 8	Brand 9	Brand 10
Si	ND	44.76±0.42	43.18±0.39
Al	0.30±0.01	14.76±0.49	8.30±0.43
Mn	6.47±0.37	1.72±0.25	ND
V	1.23±0.22	0.359±0.040	0.690±0.042
Ni	1.14±0.224	ND	0.320±0.031
Ba	2.34±0.24	0.440±0.037	0.690±0.051
Pb	ND	ND	ND
Cd	ND	ND	ND
As	ND	ND	ND

Mean value ± standard deviation (n = 3), ND = not detected

TABLE-5B
AVERAGE DAILY INTAKES OF ELEMENTS IN DIFFERENT BRANDS OF SWEETS WITH COCOA

Element	Daily intakes of elements (μg/kg bw/day)*		
	Brand 8	Brand 9	Brand 10
Si	ND	30.84	0.80
Al	0.21	10.20	0.15
Mn	4.43	1.19	ND
V	0.85	0.25	0.01
Ni	0.79	ND	0.01
Ba	1.62	0.30	0.01
Pb	ND	ND	ND
Cd	ND	ND	ND
As	ND	ND	ND

ND = Not detected
*Calculated according to body weight average = 31 kg

TABLE-6A
AVERAGE CONCENTRATIONS OF ELEMENTS OF DIFFERENT BRANDS OF COCOA WITH DIFFERENT ADDITIVES

Element	Concentration (ppm)			
	Brand 11	Brand 12	Brand 13	Brand 14
Si	21.37±0.73	26.62±0.69	137.25±0.58	328.14±1.61
Al	13.86±0.49	5.76±0.37	36.96±0.70	143.49±0.67
Mn	1.82±0.066	6.49±0.37	5.04±0.29	10.25±0.38
V	0.589±0.043	0.888±0.051	1.050±0.06	2.010±0.21
Ni	0.030±0.003	0.559±0.045	0.520±0.050	6.55±0.39
Ba	0.700±0.247	1.698±0.060	1.630±0.05	3.26±0.28
Pb	ND	ND	ND	ND
Cd	ND	ND	ND	ND
As	ND	ND	ND	ND

Mean value ± standard deviation (n = 3), ND = not detected

TABLE-6B
AVERAGE DAILY INTAKES OF ELEMENTS OF DIFFERENT BRANDS OF COCOA WITH DIFFERENT ADDITIVES

Element	Daily intakes of elements (μg/kg bw/day)*			
	Brand 11	Brand 12	Brand 13	Brand 14
Si	39.40	25.20	129.70	158.80
Al	25.0	5.4	34.90	69.44
Mn	3.35	6.14	4.76	4.96
V	1.08	0.84	1.00	0.97
Ni	0.05	0.53	5.00	3.20
Ba	1.30	1.60	1.54	1.58
Pb	ND	ND	ND	ND
Cd	ND	ND	ND	ND
As	ND	ND	ND	ND

ND = Not detected
*Calculated according to body weight average = 31 kg

contained a high daily intake of Al (484 μg/kg bw/day), which was followed by brand 19 (baby powder milk) with an Al daily intake of 391.12 μg/kg bw/day. The daily intake values for other brands were low (up to 69.44 μg/kg bw/day). Brands 6 and 19 exhibited considerably high concentration of Al, exceeding the PTDI-recommended value of 285.7 μg/kg bw/day (Table-8), and the Al concentration in other brands was lower than the PTDI-recommended value. Excluding brands 6 and 19, the daily intakes noted for other brands were less than those reported in a study conducted in Turkey, in which a concentration range of 108.3-191 μg/kg bw/day was obtained for infant

TABLE-7A
AVERAGE CONCENTRATIONS OF ELEMENTS OF DIFFERENT BRANDS INCLUDED CHEESE AND BABY POWDER MILK

Element	Concentration (ppm)				
	Cream cheese	Infant cereal	Instant noodles	Baby powder milk	
				6-12 month	1-3 years
	Brand 15	Brand 16	Brand 17	Brand 18	Brand 19
Si	76.76±1.07	23.35±0.74	170.1±0.56	21.66±0.49	49.44±0.42
Al	8.62±0.47	6.82±0.24	ND	ND	8.69±0.46
Mn	0.40±0.037	6.14±0.23	9.17±0.41	0.459±0.040	1.16±0.05
V	0.279±0.029	0.770±0.046	ND	0.640±0.038	0.809±0.043
Ni	ND	ND	ND	ND	ND
Ba	0.549±0.044	0.889±0.063	1.19±0.07	0.160±0.06	0.630±0.045
Pb	0.070±0.0023	0.060±0.0028	ND	0.057±0.003	0.079±0.0031
Cd	ND	ND	ND	ND	ND
As	ND	ND	ND	ND	ND

Mean value ± standard deviation (n = 3), ND = not detected

TABLE-7B
AVERAGE DAILY INTAKES OF ELEMENTS OF DIFFERENT BRANDS INCLUDED CHEESE AND BABY POWDER MILK

Element	Daily intakes of elements (µg/kg bw/day)				
	Cream cheese	Infant cereal	Instant noodles	Baby powder milk	
				6-12 month	1-3 years
	Brand 15*	Brand 16**	Brand 17*	Brand 18****	Brand 19***
Si	39.6	84.11	192.0	151.21	222.43
Al	4.3	68.20	ND	ND	391.12
Mn	6.4	61.38	10.4	32.03	52.18
V	0.15	7.65	ND	44.78	36.43
Ni	ND	ND	ND	ND	ND
Ba	0.28	8.89	1.3	11.37	28.42
Pb	0.036	0.60	ND	4.13	3.62
Cd	ND	ND	ND	ND	ND
As	ND	ND	ND	ND	ND

ND = Not detected; *Calculated according to body weight average = 31 kg; **Calculated according to body weight average = 15 kg; ***Calculated according to body weight average = 12 kg; ****Calculated according to body weight average = 9 kg

TABLE-8
PROVISIONAL TOLERABLE WEEKLY INTAKES (PTWIs) [Ref. 12,21,22]

Element	PTWIs (µg/kg bw week)	PTDI (µg/kg bw/ day)
Si	Not available	Not available
Al	2000	285.7
Mn	500	71.42
V	Not available	Not available
Ni	273	39
Ba	Not available	Not available
Pb	25	3.6
Cd	7	1
As	15	2.14

formulae [12]. However, the average of Al daily intakes was closer to that reported by Popovic-Dordevic *et al.* [21] (56 µg/kg bw/day). Except brands 6 and 19, the concentration of Al did not exceed the PTDI-recommended value based on JECFA, and all products studied (except brands 16, 18 and19) were not the main food sources for children. Daily intakes obtained from main meals, drinking water, and other food products may cause the concentration to exceed the PTDI-recommended value, which can present the health risks in children.

Tables 3A-7A and 3B-7B also present Pb concentrations (ppm) and Pb daily intakes (µg/kg bw/day), respectively, obtained from different food brands (Figs. 1-5). Lead was not detected in brands 3, 7-14 and 17. The daily intake values of 4.13 and

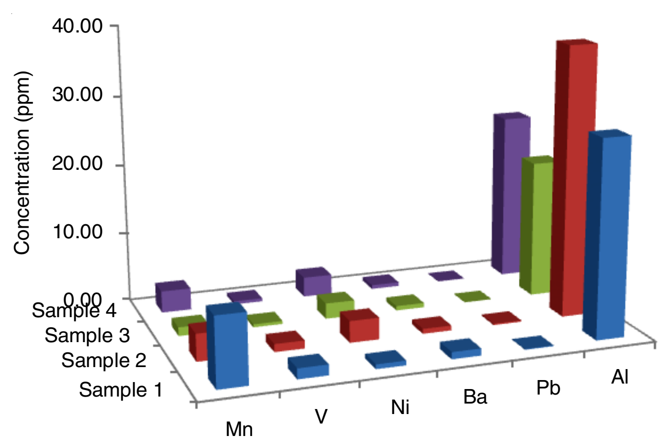

Fig. 1. Concentration of selected trace elements of different brands of potatochips and popcorn

3.62 µg/kg bw/day were obtained for lead in brands 18 and 19 (baby powder milk), respectively, which were higher than the PTDI-recommended value of 3.6 µg/kg bw/day. Lead intake values for brands 6, 16, 18, and 19 were higher than those reported by Sipahi *et al.* [12], which was 0.10-0.17 µg/kg bw/day, obtained for 63 infants formulae in Turkey. However, the highest lead daily intake values obtained in this study (3.62 and 4.13 µg/kg bw/day) were considerably lower than those obtained in Serbian vegetables (37.14 µg/kg bw/day) [21]. In all samples, barium was detected (Tables 3A-7A and Figs. 1-5). Excluding

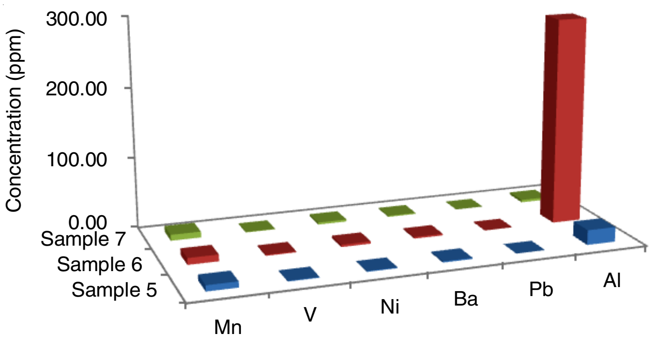

Fig. 2. Concentration of selected trace elements of different brands of biscuits

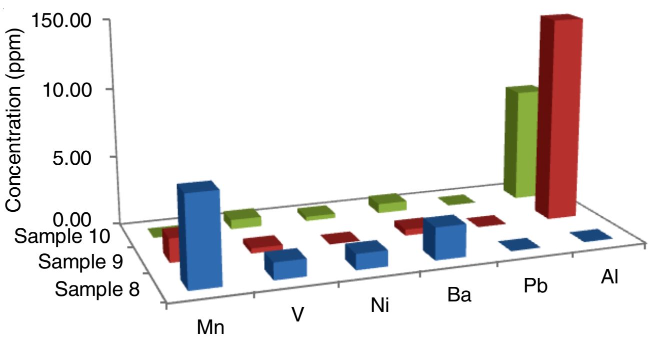

Fig. 3. Concentration of selected trace elements of different brands of sweets with cocoa

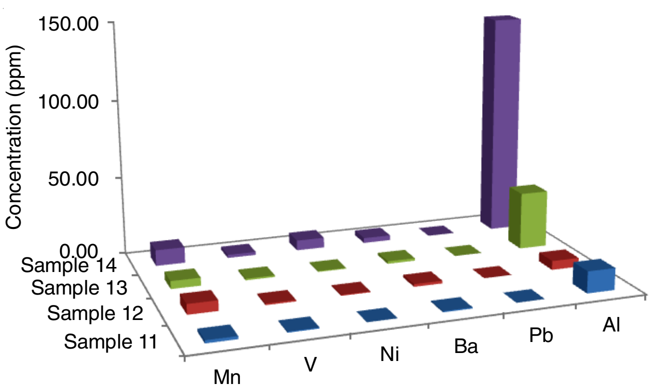

Fig. 4. Concentration of selected trace elements of different brands of cocoa with different additives

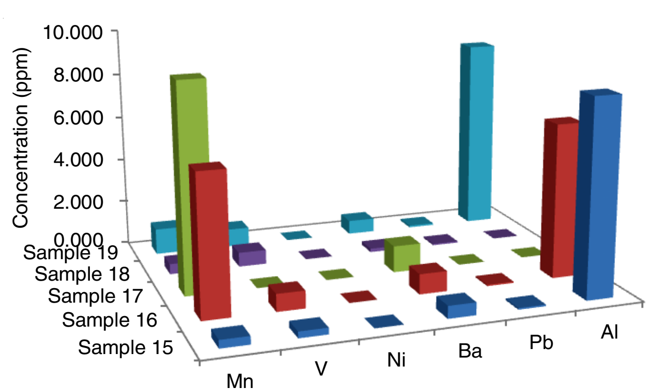

Fig. 5. Concentration of selected trace elements of different brands included cheese and baby powder milk

brands 5, 16, 18, and 19, barium concentration and intake values for other brands were closer to or lower than those obtained in infant formulae from UK, USA and Nigeria by using ICP-OES with microwave digestion (30-68 ppm and 23.8-57.5 µg/day) [23]. Brand 19 exhibited the highest barium daily intake of 341.04 µg/day, which was lower than the mean value of 550 µg/day reported by Orecchio *et al*. [24] for gluten-free food. Arsenic and cadmium were not present in any sample (Tables 3A-7A).

Probably essential elements (Ni, Mn, Si and V) present in the samples were evaluated (Tables 3A-7A and 3B-7B and Figs. 1-5). Brands 16 and 19 exhibited the highest and second highest Mn daily intakes of 920.7 and 626.16 µg/day, respectively. These values were lower than the Mn daily intake range of 1700-6800 µg/day reported by Koubová *et al*. [25]. Brands 16 and 19 exhibited the highest daily intake values of 61.38 52.18 µg/kg bw/day, respectively, which were lower than those reported in a study on essential and toxic metals present in infant formulae (77.23-136.21 µg/kg bw/day) [12] and higher than the range of 3.00-24.71 µg/kg bw/day of daily intake values reported by Popovic-Dordevic *et al*. [21] for vegetables. The Ni daily intake value for our samples was in the range of 0.01-5.44 µg/kg bw/day, which concurred with that of vegetables (0.86-6.00 µg/kg bw/day) obtained from Serbia [21]. However, our value range was lower than that for commercial infant foods from Iran (52.70-75.47 µg/kg bw/day) and the WHO-recommended value (39 µg/kg bw/day) [22]. The vanadium daily intake was determined (µg/day), and it was not detected in any brand [17]. The range obtained for the vanadium intake value was 0.31-403.02 µg/day, but most samples exhibited a range of 4.5-73 µg/day, which was higher than the 0.05-3.5 µg/day range reported by Ikem *et al*. [23] by using different samples from UK, Nigeria, and USA. The range obtained in the present study was lower than that obtained in a study on trace elements in different types of fishes in Ghana [26]. The range of vanadium daily intake value (Tables 3B-7B) was from 0.01 to 44.78 µg/kg bw/day, which was higher than the range reported by Iwegbue *et al*. [27] from 0.1 to 3.0 µg/kg bw/day.

Silicon was present in all samples except in brand 8 (Fig. 6) and obtained in a wide intake value range of 0.8-462 µg/kg bw/day and the daily intake range of the elements was 24.8-4322.0 µg/day. A range of 20000-40000 µg/day was the estimated daily intake of Si [28]. In the United States, 19000 and 40000 µg /day of Si was obtained in the diets for women and men, respectively, which are substantially higher than present results.

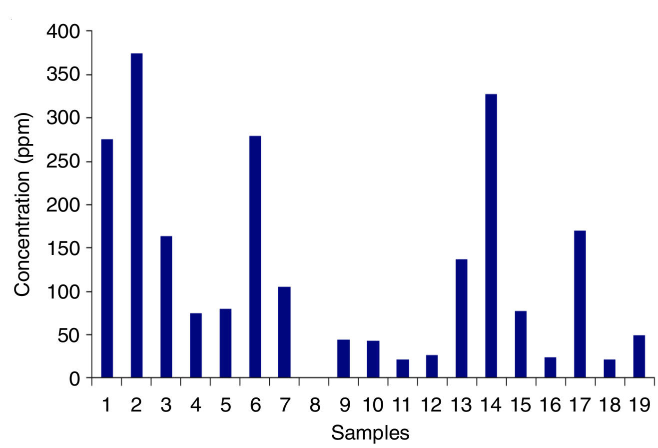

Fig. 6. Concentration of Si in different brands samples

Conclusion

The daily intakes and concentrations of Al were high in some samples, which leads to an increase in health risks caused by Al, such as nephrotoxicity and neurotoxicity. The concentrations and lower daily intakes of other elements studied was less than those recommended by JECFA or WHO. The samples studied (except brands 16, 18, and 19) are not the main food sources for children. However, drinking water and main meals may contain trace elements investigated in this study. Thus, the overall daily intake of these elements, especially of Al, may be higher than that recommended by JECFA or WHO.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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