



Original Research Article

Aluminium in infant foods: Total content, effect of *in vitro* digestion on bioaccessible fraction and preliminary exposure assessment

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ABSTRACT

The objective of this study was to determine the total concentration and bioaccessible fraction of aluminium (Al) in 95 different baby food samples and estimate the exposure assessment. Total Al content was determined following oxidative microwave digestion by inductively coupled plasma optical emission spectrometry. An *in vitro* digestion method was optimized to evaluate the bioaccessible fraction. Total concentration and bioaccessibility varied according to the sample composition (saltypurees, fruit purees, infant drinks and petitsuisse). Petit suisse, soy-based drink and salty puree samples presented the highest total Al concentrations of 4170 $\mu\text{g kg}^{-1}$, 2860 $\mu\text{g kg}^{-1}$ and 2760 $\mu\text{g kg}^{-1}$, respectively. Bioaccessible fraction varied from 0.5%–48% according to their composition. Exposure to Al was estimated and compared with the tolerable weekly intakes currently established. The results showed that the consumption of 3 portions/day of soy-based drink along the week could represent a concern.

1. Introduction

The introduction of complementary food to infants is a turning point in the development of their eating behavior. The World Health Organization (World Health Organization (WHO), 2009) recommends that infants should be exclusively breastfed for the first 6 months and then start receiving complementary foods to provide critical nutrients (iron and zinc) for their development. Without a balanced diet, an infant might develop a nutritional deficiency (Koo et al., 2018). Moreover, baby foods have special functions in infant diets because they are major source of nutrients and a unique source of food during the first months of life (Ikem et al., 2002). Dietary variety and exposure to fruits and vegetables in infancy have been associated with nutritional benefits (Vasco and Alvito, 2011).

However, it is well-known that foods used to prepare baby foods may contain potentially toxic elements, such as aluminium (Al), which is naturally present in soils, minerals and even in water (Yokel, 2016; Ahmed et al., 2016; Al-Kindy et al., 2007). In addition, its compounds maybe present in the food industry as food additives, in baking powder, processed cheese, meat products, and in cooking and storage utensils (Aydin and Soylyak, 2007; Saracoglu et al., 2007). Al is the most

abundant metallic element and constitutes about 8.13 % of the earth's crust. Elemental Al does not occur in its pure state; it is always combined with other elements (hydroxide, silicate, sulfate, phosphate) (Saracoglu et al., 2007; Aydin and Soylyak, 2007).

Regarding Al children exposure, increased susceptibility is observed in the early stages of life, fetal and early postnatal periods (Makri et al., 2004; Sly and Flack, 2008). Several pieces of evidence suggest that fetuses and infants may be more sensitive to pollutants, including Al (Landrigan et al., 2003). Firstly, children's metabolic pathways are immature, especially during the fetal period and the first months after birth, suggesting that metabolism and detoxification are not as efficient in infants as they are in adults. Development processes during these periods are also more easily disturbed. Lastly, infants are exposed to higher concentrations of chemicals through their diet because the ratio of the quantity consumed to body weight (bw) is higher in infants than in adults (Hulin et al., 2014). Furthermore, human exposure to Al is identified as a possible contributor to neurodegenerative / neurodevelopmental diseases, such as multiple sclerosis (MS) and Alzheimer's disease (Paiva et al., 2019b; Ahmed et al., 2016; Mold et al., 2018).

Therefore, due to the relevant concern about Al toxicity, and considering the particular features of infants to Al exposure, better food

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Table 1
Description of infant foods according to the classification and composition. Infant salty purees samples (SP), fruit purees (FP), infant drinks (ID) and petit suisse (PS).

Sample composition	Brand	n	Classification	Ingredients
Fruits mix	A	3	FP	Water, apple, apple juice, papaya, starch, orange juice, rice flour, vitamin C and acidulant citric acid
Plum	A	3	FP	Water, plum, apple, apple juice, starch, rice flour and acidulant citric acid
Noodles with Chicken breast and vegetables	A	3	FP	Potato, water, chicken breast, carrot, broccoli, onion, squash pulp, starch, canola oil, noodles and corn oil
Meat, vegetables and cassava	A	3	SP	Potato, carrot, water, meat, cassava, onion, canola oil, corn oil, starch, rice flour and tomato pulp
Noodles, meat and vegetables	A	3	SP	Potato, water, carrot, meat, onion, squash pulp, noodles, chayote, starch, canola oil, corn oil, salt and rice flour
Egg yolk, meat and vegetables	A	3	SP	Potato, water, carrot, beef, squash pulp, onion, canola oil, rice flour, starch, egg yolk, corn oil and tomato pulp
Chicken breast with vegetables	A	3	SP	Potato, chicken breast, carrot, water, chicken, squash pulp, onion, chayote, starch, canola oil, corn oil, rice flour, salt and antioxidant ascorbic acid
Vegetables and meat	A	3	SP	Potato, carrot, meat, onion, squash pulp, canola oil, chayote, corn oil, rice flour, starch and salt
Noodles, meat and vegetables	A	3	SP	Potato, water, carrot, meat, chayote, squash pulp, onion, canola oil, rice flour, corn oil, starch and salt
Chopped meat	A	2	SP	Water, potato, carrot, meat, onion, chayote, rice flour, starch, tomato pulp, canola oil, corn oil, low sodium salt, kale, cabbage, garlic and ferrous lactate
Apple and plum	B	3	FP	Apple and plum
Chicken Breast, tomato, potato and broccoli	B	3	SP	Water, tomato, potato, chicken breast, broccoli, onion, extra virgin olive oil, and garlic (organic ingredients)
Zucchini, sweet potato, lettuce, beans and egg yolk	B	3	SP	Water, zucchini, sweet potato, lettuce, beans, egg yolk, onion, extra virgin olive oil, and garlic (organic ingredients)
Meat, carrot, lettuce, black beans and rice	B	3	SP	Water, carrot, meat, lettuce, black beans, rice, onion, garlic and extra virgin olive oil (organic ingredients)
Zucchini, lettuce, lentil, sweet potato and egg	B	3	SP	Water, zucchini, lettuce, lentil, sweet potato, egg, onion, garlic, and extra virgin olive oil (organic ingredients)
Strawberry, raspberry and apple	C	3	FP	Apple puree, raspberry puree, strawberry puree and a drop of lemon
Pear, grape and apple	C	3	FP	Apple puree, pear puree, grape puree and a drop of lemon
Yellow fruits and chia	C	3	FP	Peach puree, banana puree, mango puree, chia seeds, passion fruit juice and a drop of lemon
Banana and apple	C	3	FP	Apple puree, banana puree and a drop of lemon
Dark chocolate milk drink	D	3	ID	Semi-skimmed milk, reconstituted whey powder, cocoa syrup, sugar, quinoa flour, flaxseed meal and chia powder
Apple and papaya milk drink	D	3	ID	Semi-skimmed milk, reconstituted whey powder, reconstituted papaya pulp, reconstituted apple pulp, sugar, quinoa flour, flaxseed flour and chia powder
Banana milk drink	D	3	ID	Semi-skimmed milk, reconstituted whey powder, reconstituted banana pulp, sugar, quinoa flour, flaxseed meal and chia powder
Chocolate milk drink	E	3	ID	Reconstituted milk, whey, water, sugar, cocoa powder, minerals (calcium, magnesium and iron), vitamins (C, B1, B2, niacin, B6, B12, pantothenic acid, biotin)
Strawberry milk drink	E	3	ID	Reconstituted milk, whey, strawberry prepared (water and strawberry pulp), sugar, maltodextrin, minerals (tricalcium phosphate, ferric pyrophosphate and zinc sulfate), vitamins (sodium L-ascorbate, thiamine hydrochloride and cholecalciferol)
Cereal milk drink	E	3	ID	Reconstituted whole milk, reconstituted whey sugar, vitamins and minerals (ascorbic acid, iron, zinc, vitamin B1, vitamin B6 and vitamin B5). May contain soybeans, rye, barley and oats
Chocolate milk drink	F	3	ID	Reconstituted whey or powdered whey or reconstituted concentrated whey, sugar, whole pasteurized milk and / or reconstituted milk powder, milk cream, cocoa powder, vitamins A and D
Soy-based drink	F	3	ID	Water, sugar, soy protein isolate, sunflower oil, maltodextrin, salt, vitamins and minerals (E, Zinc, B2, B6, A, Folic Acid, D and B12)
Oat milk drink	F	3	ID	Whole pasteurized milk, reconstituted concentrated whey, sugar and oatmeal flour
Soy-based drink	G	3	ID	Soybean extract, water, sugar, vitamins (C, E, B2, B6, A, folic acid, D and B12) and minerals (calcium and zinc)
Chocolate soy-based drink	G	3	ID	Water, soybeans, invert sugar, cocoa, calcium and zinc minerals, carob flour, sugar, salt, vitamins C, E, B2, B6, A and folic acid
Petit suisse	H	3	PS	Skimmed milk, sugar, strawberry prepared (water, fructose, strawberry pulp, tricalcium phosphate, calcium citrate, modified starch, zinc, vitamin E, iron and vitamin D)
Petit suisse	I	3	PS	Reconstituted standardized milk, strawberry preparation (sugar syrup, water, sugar, tricalcium phosphate, fructose, modified starch, strawberry pulp, vitamins and mineral (A, D and zinc))
TOTAL		95		

control is needed, especially those destined for children. Additionally, the information about inorganic constituents usually indicates the total concentrations to be ingested and not the amount that will be effectively absorbed by the body. This type of information could be provided by studies related to bioaccessibility (the amount of the element that is released into the gastrointestinal tract from its matrix) and bioavailability (the fraction of the element which is absorbed from the gastrointestinal tract by the human organism, reaches the bloodstream and is then used in biological functions) (Minekus et al., 2014; Machado et al., 2017; Peixoto et al., 2016).

The interest in bioaccessibility of chemical elements has been growing in recent years, and many of them have been assessed in different food matrices, such as baby food (Do Nascimento da Silva et al., 2013; Do Nascimento da Silva et al., 2018), cereal products (Versantvoort et al., 2005; Fu and Cui, 2013; Khouzam et al., 2011; Laparra et al., 2007; do Nascimento da Silva et al., 2013; Peixoto et al., 2016; Bhatia et al., 2013; Domínguez-González et al., 2010; Vitali et al., 2008; Yang et al., 2014), biscuit (Vitali et al., 2008), lettuce (Do Nascimento da Silva et al., 2015), beef (Menezes et al., 2018), coffee (Alongi et al., 2019), berries (Pereira et al., 2018) and cheese (Ayala-Bribiesca et al., 2017; Khouzam et al., 2011), using different experimental models of *in vitro* digestion. Some important issues have been studied such as the influence of the food structures and the synergism or antagonism between food components, as well as the standardization of methods to evaluate the soluble fractions of a food during the gastrointestinal digestion. However, bioaccessibility studies of Al are still limited with regard to food for infants and children. Only a few studies (Souza et al., 2019; Pereira et al., 2018; Melø et al., 2008; Khalifa and Ahmad, 2010) have been found in the literature. Therefore, the aim of the present work was to evaluate the total concentration and the bioaccessible fraction of Al using an optimized *in vitro* digestion method by optical emission spectrometry with an inductively coupled plasma source (ICP OES), and estimate the weekly intake through the consumption of infant foods commercialized in Brazil.

2. Material and methods

2.1. Material, reagents and equipment

All of the reagents used in the research were of or above analytical grade. Water (18.2 MΩ cm) was purified with reverse osmosis (Gehaka, São Paulo, Brazil) while nitric acid purification (Distillacid, Berghof, Eningen, Germany) was achieved with a sub-boiling distiller. To determine Al, the sample was first submitted to acid digestion using distilled HNO₃ and 30 % H₂O₂ (m/v) (Merck, Darmstadt, Germany). The analytical curves were prepared from a certified 100 mg L⁻¹ standard solution (Specsol, Quimlab, Jacareí, Brazil) in a 0.5 % HCl solution (v/v) (Merck, Darmstadt, Germany). Accuracy of the method was evaluated using the following certified reference materials: egg powder (EGGS-1, National Research Council, Canada) and a diet (Typical Diet, NIST SRM 1548a).

The samples were digested in a closed microwave-assisted digestion system (Start E, Milestone, Sorisole, Italy) equipped with 24 teflon flasks with internal volumes of 75 mL. The total Al content was determined using an ICP OES (model 5100 VDV, Agilent Technology, Tokyo, Japan) equipped with a double-step nebulization camera, a sea-spray nebulizer and 99.996 % pure liquid argon (Air Liquide, São Paulo, Brazil). The optimized operational conditions of the equipment were: radio frequency generator power (1400 W); nebulizer argon flow rate (0.5 L min⁻¹); principal argon flow rate (12 L min⁻¹ Ar); auxiliary argon flow rate (1 L min⁻¹ Ar); sample flow rate (0.5 L min⁻¹); axial vision mode; number of replicates (n = 3) and wavelength for Al (396, 152 nm).

The following reagents were used for the bioaccessibility assay: NaHCO₃ (> 99.7 %), alpha-Amylase from *Aspergillus oryzae* (30 U mg⁻¹), lactase (85,300 USP), pepsin porcine gastric mucosa (> 250 U

mg⁻¹), bile from bovine and ovine (bile acid mixture) and pancreatin from porcine pancreas (8 x USP) (Sigma-Aldrich, Saint Louis USA). A dubnoff shaking water bath (NT 230, Nova Técnica, Piracicaba, Brazil) and a pHmeter (Starter 3100, Ohaus, Parsippany, EUA) were also used in the experiments.

2.2. Samples

A total of 95 samples were acquired in the city of Campinas, SP, Brazil from distinct batches and different brands (designated as A to I). The samples were stored in a dry place at room temperature (25 °C) and the determinations of Al carried out in triplicate. The main ingredients contained in the products are shown in Table 1.

The digestion *in vitro* assay was performed in 26 samples, which were selected among the products considered in the present study to represent specific matrix composition. All measurements of the bioaccessible fraction of Al were performed in triplicates and the results were expressed as percentages.

2.3. Determination of total Al

A sample of 1 g of infant purée (solid sample) or 3 g of infant drink (liquid sample) was weighed into a digestion flask, and 8 mL of purified HNO₃ plus 2 mL H₂O₂ were added and maintained in contact overnight. The flasks were then sealed, transferred to the microwave digester and digested using 4 heating ramps applying 1000 W of power: a) room temperature to 70 °C in 5 min; (b) from 70 °C to 120 °C in 5 min; (c) from 120 °C to 170 °C in 5 min; and (d) maintained at 170 °C for 25 min. After cooling, the flasks were opened and the resulting solution transferred to graduated tubes using purified water to 25 mL. The external calibration method was used to determine the Al content by ICP OES through a standard curve in the range from 2 to 200 µg L⁻¹.

2.4. Dialysis method

The *in vitro* digestion model was performed according to Perales et al. (2006), considering the gastrointestinal system of the infant with some adaptations. The quantities and concentrations of the enzymes solution were optimized taking into account the gastrointestinal capacity of the infant. Approximately 5 g of infant purees (solid sample) or 10 mL of infant drink (liquid sample) were transferred to clean erlenmeyer flasks and mixed with water to a final volume of 20 mL. The pH of samples was adjusted to 7.0 and 4 mL of saliva were added for salivary digestion. The mixture was incubated at 37 °C in a shaking waterbath for 5 min. Thereafter, the samples were acidified to pH 2.0 with 6 mol L⁻¹ hydrochloric acid (HCl) with addition of 0.3 mL of porcine pepsin preparation (1.6 g of porcine pepsin in 10 mL 0.1 mol L⁻¹ HCl). The mixture was incubated at 37 °C in a shaking water bath for 2 h. To stop the gastric digestion phase, the samples were maintained for 10 min in an ice bath. The gastric digest solution added by pancreatin-bile was titrated with 0.5 mol L⁻¹ NaHCO₃ solution to determine the volume of base needed to increase pH digests to about 7.5. Dialysis bags (Sigma-Aldrich, Saint Louis, EUA with cut-off from 12.000–16.000 Da and porosity of 25 Å) containing 20 mL of freshly prepared 0.5 mol L⁻¹ NaHCO₃ solution and water, were immersed in pepsin digests and incubated in a shaking water bath at 37 °C. After 30 min, 0.75 mL of pancreatin-bile salt mixture (0.4 g porcine pancreatin and 2.52 mg bile bovine per 100 mL of 0.1 mol L⁻¹ NaHCO₃) were added and incubated in a shaking water bath at 37 °C for more 2 h. The samples were maintained in the ice bath for 10 min to stop intestinal digestion. Dialysates were transferred for weight to the Falcon tube and used to determine Al.

2.4.1. Determination of the optimum concentration of NaHCO₃

The optimum dialysis concentration of NaHCO₃ was calculated using titratable acidity (Shiowatana et al., 2006). Titratable acidity was

defined as the number of equivalents of NaOH required to titrate the amount of digest to a pH of 7.5. It was determined using standard 1 mol L⁻¹ NaHCO₃ as titrant. This concentration of NaHCO₃ changed the pH of the dialysate to 5.0–6.0 after 30 min of dialysis and gradually increased the pH to 7.0–7.5 on the addition of pancreatin bile extract.

2.5. Preliminary exposure assessment and risk characterization

Exposure to Al was calculated as follows:

Estimated aluminium exposure = [Al] x (portion / body weight); where [Al] is the mean Al concentration (mg kg⁻¹) determined in the samples of the present work and the portion is the amount of consumed food (kg). In this study, the portion was calculated from the recommended amount informed on the label of each product, considering 3 servings per day, during seven days, in order to estimate the weekly Al intake. The calculations were done considering the average weight of a child (up to 2 years old) as 11.85 kg, according to the child growth standards of the World Health Organization (World Health Organization (WHO), 2018).

Two parameters were considered to evaluate the potential risk of exposure to Al: the Provisional Tolerable Weekly Intake (PTWI) and the Tolerable Weekly Intake (TWI). The Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a PTWI for Al of 2 mg kg⁻¹ bw applied to all the Al compounds in foods, including food additives (Food and Agriculture Organization/World Health Organization (FAO/WHO), 2011; Codex Alimentarius Commission (CAC), 2018), whilst the European Food Safety Authority (EFSA) established a TWI of 1 mg kg⁻¹ bw for Al in all food sources (European Food Safety Authority (EFSA), 2008). The estimated Al exposure was expressed in terms of % PTWI or TWI according to the equation:

$$\% \text{ PTWI or TWI} = (100 \times \text{Estimated aluminium exposure}) / (\text{PTWI or TWI})$$

2.6. Method validation for Al quantification in infant foods

Accuracy, repeatability, linearity, detection limit (LOD) and quantification limit (LOQ) were evaluated according to the National Institute of Metrology, Standardization and Industrial Quality (INMETRO, 2017). For the concentration range from 2 to 200 µg L⁻¹ of Al, a satisfactory correlation coefficient value was found ($r^2 > 0.999$), showing the linearity of the analytical curve.

The LOD and LOQ were determined using 10 replicates of an analytical blank, multiplied by the sample dilution factor depending on the matrix: (25 x for purees) and (8.3 x for drinks). The values found for purees were: LOD (3s) = 49 µg kg⁻¹ and LOQ (5s) = 92 µg kg⁻¹, whilst for drinks were: LOD (3s) = 16 µg kg⁻¹ and LOQ (5s) = 30 µg kg⁻¹ with "s" being the standard deviation value of the 10 blank replicates.

In order to determine the repeatability of the method, the intra-day coefficients of variation (CV) were evaluated for two different samples (one puree and one drink) (n = 8). The mean values found were 10 % and 8% for infant purees and drinks, respectively, satisfying the conditions recommended by the Official Methods of Analysis (AOAC, 2013) with a maximum CV of 25 % for the range concentration. The accuracy of the method was evaluated in two certified reference materials (CRM): Typical Diet (SRM 1548a - Typical Diet) and Egg Powder (NRC EGGS - Egg Power), containing a mean concentration of 500 and 55 mg kg⁻¹ of Al, respectively. The obtained results varied from 87 % to 97 %, in accordance with the AOAC (2013) guidelines, which establish a range from 75 to 120 %, for the studied concentrations (Table 2).

Table 2

Results obtained in the evaluation of the accuracy of the analytical method for the determination of Al using certified reference materials and recovery tests (n = 3).

Certified Reference Materials	Aluminum		
	Certified Values	Values Obtained	Recovery (%)
Typical diet (µg kg ⁻¹)	72 ± 2	55 ± 2	87 ± 1
Egg power (µg kg ⁻¹)	540 ± 86	500 ± 2	97 ± 1

2.7. Statistical analysis

The means obtained for each batch of the same brand, as well as between different brands, were compared by variance analysis (ANOVA) and Tukey's test (p < 0.05). All statistical analyses were made using the Statistica 7.0 software package (Statsoft, USA).

3. Results and discussion

3.1. Total aluminium content in infant food

The results obtained for total Al concentration in infant food samples from different commercial brands are presented in Table 2. The brand B corresponds to a handmade gourmet formulation of salty and fruit purees commercialized in Brazil, whilst all other brands belong to industrial infant foods.

It was observed that the highest levels of Al are present in: brand A) chopped meat (1170 µg kg⁻¹), followed by fruit mix (925 µg kg⁻¹) and egg yolk, meat and vegetables (880 µg kg⁻¹); brand B) zucchini, lettuce, lentil, sweet potato and egg (2760 µg kg⁻¹), apple and plum (2500 µg kg⁻¹) and zucchini, sweet potato, lettuce, beans and egg yolk (2310 µg kg⁻¹); brand C) pear, grape and apple (1970 µg kg⁻¹) and strawberry, raspberry and apple (1900 µg kg⁻¹); brand D) dark chocolate milk drink (2175 µg kg⁻¹); brand E) chocolate milk drink (1780 µg kg⁻¹); brand F) soy-based drink (2860 µg kg⁻¹); brand G) chocolate soy-based drink (2280 µg kg⁻¹); and brand H) petit suisse (4170 µg kg⁻¹).

It is worth to observe that, in general, the most significant total Al content is present in infant drink, petitsuisse and some salty puree samples. Brand B (handmade claim) notably presents higher levels of Al in comparison to brand A, considering similar compositions of salty and fruit purees. This indicated that handmade production may not have acute control regarding procedures, utensils, storage and raw materials supply. On the other hand, the industrial scale of production of salty and fruit puree samples, represented by brands A and C, showed lower Al levels once that all the steps of manufacturing may be rigorously monitored. Moreover, exposure of the product to a larger amount of Al during manufacture may occur due to the contact with utensils, machines and powder particles, which are more critical in handmade procedures (Yaman et al., 2003).

In the work performed by Khalifa and Ahmad (2010), commercial baby foods based on fruits and vegetable purees available in Saudi Arabia were analyzed. They found mean Al levels of 6450 µg kg⁻¹, whilst in the present work the highest Al concentration in vegetable and fruit preparations was 3450 µg kg⁻¹ (SP).

Melø et al. (2008) developed a study regarding minerals and trace elements in commercial infant food from Norway, analyzing samples of porridge and fruit purees (composition not declared). The mean Al levels among porridge samples was 3450 µg kg⁻¹ which is in agreement with the values found for salty purees in the current work, such as zucchini, sweet potato, lettuce, beans and egg yolk (3320 µg kg⁻¹) and zucchini, lettuce, lentil, sweet potato and egg (3450 µg kg⁻¹) composition. For fruit purees, the mean Al concentration was 559 µg kg⁻¹, whilst a range of 620 to 2080 µg kg⁻¹ was observed in this study.

Regarding infant foods ingredients, Zaida et al. (2007) have

Table 3

Mean values, standard deviation (SD) and concentration range for Al in infant salty purees samples (SP), fruit purees (FP), infant drinks (ID) and petit suisse (PS).

Brand	Composition	Classification	Aluminum ($\mu\text{g kg}^{-1}$)	
			Mean \pm SD	Range
A	Fruits mix	FP	925 \pm 110	845 – 1005
	Plum	FP	750 \pm 160	620 – 930
	Noodles with Chicken breast and vegetables	SP	383 \pm 88	280 – 440
	Meat, vegetables and cassava	SP	530 \pm 150	400 – 700
	Noodles, meat and vegetables	SP	390 \pm 63	325 – 460
	Egg yolk, meat and vegetables	SP	880 \pm 200	690 – 1090
	Chicken breast with vegetables	SP	210 \pm 120	125 – 290
	Vegetables and meat	SP	260 \pm 90	160 – 330
	Chopped meat	SP	1170 \pm 260	960 – 1460
	Apple and plum	FP	2500 \pm 500	2195 – 3070
B	Chicken Breast, tomato, potato and broccoli	SP	1340 \pm 280	1030 – 1550
	Zucchini, sweet potato, lettuce, beans and egg yolk	SP	2310 \pm 1165	1835 – 3320
	Meat, carrot, lettuce, black beans and rice	SP	1404 \pm 542	1070 – 2030
	Zucchini, lettuce, lentil, sweet potato and egg	SP	2760 \pm 980	2070– 3450
C	Strawberry, raspberry and apple	FP	1900 \pm 175	1700 – 2040
	Pear, grape and apple	FP	1970 \pm 105	1850 – 2030
	Yellow fruits and chia	FP	940 \pm 20	950 – 925
D	Banana and apple	FP	1500 \pm 500	1180 – 2080
	Dark chocolate milk drink	ID	2175 \pm 315	2170 - 2720
	Apple and papaya milk drink	ID	890 \pm 130	675 – 900
	Banana milk drink	ID	540 \pm 220	465 - 880
E	Chocolate milk drink	ID	1780 \pm 190	1670 - 2040
	Strawberry milk drink	ID	1050 \pm 65	940 - 1055
F	Cereal milk drink	ID	890 \pm 65	860 - 960
	Chocolate milk drink	ID	1770 \pm 170	1580 - 1920
	Soy-based drink	ID	2860 \pm 152	2760 - 3060
G	Oat milk drink	ID	260 \pm 25	250 - 295
	Soy-based drink	ID	1770 \pm 225	1400 - 1805
	Chocolate soy-based drink	ID	2280 \pm 103	2220 - 2420
H	Petit suisse	PS	4170 \pm 55	4135 - 4210
I	Petit suisse	PS	830 \pm 35	830 - 890

analyzed some matrix such as meat, fruits and vegetables. The study showed high Al levels in beef (12,500 $\mu\text{g kg}^{-1}$), chicken (51000 $\mu\text{g kg}^{-1}$), carrots (41,300 $\mu\text{g kg}^{-1}$), potatoes (73,600 $\mu\text{g kg}^{-1}$), banana (9400 $\mu\text{g kg}^{-1}$) and apple (38,400 $\mu\text{g kg}^{-1}$). In another recent study conducted by Pereira et al. (2018), Al content was quantified in fresh berries, such as blackberries, raspberries, blueberries and strawberries samples. Mean Al concentrations of 8300 $\mu\text{g kg}^{-1}$ and 3100 $\mu\text{g kg}^{-1}$ were found for raspberry and strawberry, respectively. In the current work, maximum Al values for fruit purees was 3070 $\mu\text{g kg}^{-1}$.

With respect to infant drinks, it is possible to note that those with chocolate and soya composition presents the highest Al levels. The presence of Al in cocoa products has already been reported by Bertoldi et al. (2016), who analyzed 61 samples of cocoa beans and chocolates produced in 23 countries in East and West Africa, Asia, and South and Central America. The concentrations found for cocoa beans varied from 41 mg kg^{-1} (Central America) to 275 mg kg^{-1} (East Africa) whereas levels between 10.6 mg kg^{-1} (Central America) and 21.5 mg kg^{-1} (South America) were observed for dark chocolate tablets. These values show that cocoa products, depending on the region cultivated, the proportion used in the formulation and the form of processing, could be an additional source of Al contamination in foods (Bertoldi et al., 2016). Thus, the addition of cocoa powder in infant product should be avoided, in terms of Al contamination.

Similarly, the high Al levels found in soy-based products are in accordance to other studies available in the literature. Kazi et al. (2009) reported mean Al contents in milk-based formulas from 640 $\mu\text{g kg}^{-1}$ to 1520 $\mu\text{g kg}^{-1}$, whereas those of soy-based products varied from 1740 $\mu\text{g kg}^{-1}$ to 2720 $\mu\text{g kg}^{-1}$ for Pakistan infant formulas. Paiva et al. (2019a) found equivalent Al concentrations in soy-based infant formulas commercialized in Brazil with values ranging from 2020 $\mu\text{g kg}^{-1}$ to 4490 $\mu\text{g kg}^{-1}$. In this study soy-based drinks varied from 1400 $\mu\text{g kg}^{-1}$ to 3070 $\mu\text{g kg}^{-1}$ among brands F and G; whilst a sample from brand G - soy-based added by cocoa powder – also presented high

Al concentration (2420 $\mu\text{g kg}^{-1}$) evidencing the potential Al contamination from both sources.

3.2. Bioaccessibility of Al by *in vitro* gastrointestinal digestion

Table 3 shows the bioaccessible fractions of Al present in baby food samples. The bioaccessibility experiments were performed using 5 g of purees and petit suisse and 10 mL of infant drinks.

The results from Table 4 show that the bioaccessibility percentage varied widely from 0.5%–48% which may be related to the composition and characteristics of each sample. Salty purees samples from brand A presented bioaccessibility ranging from 2.1%–18.2%. For fruit and salty purees from brand B, the highest value was 8.4 %, whilst for brand C a maximum value of 8.8 % was found. Among infant drinks from brands D to G, the lower bioaccessibility was observed for soy-based drink (1.4 %) and the highest for cereal milk drink (48 %). Regarding petit suisse samples, relatively high bioaccessibility was found for brand I (37 %), while brand H showed lower percentage (9.2 %). Concerning their composition it is observed that that brand H presents skimmed milk (main ingredient), while brand I is composed by whole standard milk. Therefore, brand I presents higher fat levels than brand H. According to Thakur et al. (2020) studies regarding the digestibility and absorption of phytonutrients such as vitamins, carotenoids, polyphenols, curcuminoids, polyunsaturated fatty acids, proteins, peptides, dietary fibers, oligosaccharides, and minerals, demonstrated that the bioaccessibility of phytonutrients enhanced further by addition of oil, fat and certain enzymes. Moreover, the group states that the bioaccessibility depends on food matrix (dietary fat and fiber), food processing, co-ingested food and nutrients status. Therefore, the distinct composition of fat may explain the difference in Al bioaccessibility observed between the two dairy matrices.

Do Nascimento da Silva et al. (2015) conducted theoretical calculations to determine the hydration energies of the polyphenols and the

Table 4

Al mean values ($n = 3$) for bioaccessible fraction and percentage of bioaccessibility (%) in infant salty purees samples (SP), fruit purees (FP), infant drinks (ID) and petit suisse (PS).

Brand	Composition	Classification	Bioaccessibility	
			Bioaccessible fraction ($\mu\text{g kg}^{-1}$)	%
A	Meat, vegetables and cassava	SP	18.2 \pm 1.5	18.2
	Noodles, meat and vegetables	SP	12.3 \pm 1.4	14.1
	Chicken breast with vegetables	SP	17.2 \pm 1.2	15.6
	Vegetables and meat	SP	3.0 \pm 0.7	7.4
	Chopped meat	SP	2.3 \pm 0.3	2.1
B	Apple and plum	FP	3.0 \pm 0.1	0.5
	Chicken Breast, tomato, potato and broccoli	SP	216 \pm 20	8.4
	Zucchini, sweet potato, lettuce, carioca beans and gem	SP	22 \pm 4	2.6
	Meat, carrot, lettuce, black beans and rice	SP	3.7 \pm 0.3	1.3
	Strawberry, raspberry and apple	FP	8.0 \pm 0.3	1.6
C	Pear, grape and apple	FP	25.3 \pm 2.2	5
	Yellow fruits and chia	FP	4.4 \pm 1.8	1.9
	Banana and apple	FP	27.4 \pm 6.5	8.8
	Dark chocolate milk drink	ID	155 \pm 10	14.2
D	Apple and papaya milk drink	ID	53 \pm 2	24
	Banana milk drink	ID	26 \pm 1	19.3
	Chocolate milk drink	ID	39.0 \pm 3.8	4.4
E	Strawberry milk drink	ID	95.0 \pm 1.8	18.2
	Cereal milk drink	ID	258 \pm 21	48.0
F	Chocolate milk drink	ID	18.0 \pm 2.5	4.0
	Soy milk drink	ID	9.8 \pm 1.4	1.4
	Oat and soy milk drink	ID	24.0 \pm 0.8	37.8
G	Soy milk drink	ID	45.0 \pm 6.2	10.2
	Chocolate soy milk drink	ID	24.0 \pm 1.4	4.2
H	Petit-suisse	PS	97.0 \pm 12.0	9.2
I	Petit-suisse	PS	77.0 \pm 3.8	37.0

binding energies of the cellulose monomer and polyphenols to the metals in order to better understand the effects of certain matrix components on the bioaccessibility of chemical elements. The obtained results from this work regarding the binding energies are also consistent with the concept of hard and soft acids and bases (HSAB), which indicates that Al^{3+} and Fe^{3+} are hard acids. Therefore, Al^{3+} and Fe^{3+} are less polarizable compared to other metals, and polyphenols and cellulose contain OH^- and RCOO^- groups, which are hard bases. Thus, the interactions of fiber sources (cellulose) and polyphenols with Fe^{3+} and Al^{3+} tend to be more stable than the interactions of these compounds with Zn^{2+} and Cd^{2+} , for example (Shriver et al., 1999).

Al exhibits the strongest interaction with cellulose, which explains the fact that in spite of high total Al content, the samples studied in this work presented low bioaccessibility, in general. Especially those purees containing vegetables - zucchini, sweet potato, lettuce, beans and egg yolk (2.6 %), fruits-apple and plum (0.5 %), as well as infant cereal drinks-soy-derived drink (1.4 %) and chocolate milk drink (4.0 %).

In addition to their binding energies, Do Nascimento da Silva et al. (2013) also calculated the hydration energies of the polyphenols to evaluate the solubility of these compounds. Thus, the correlation between the amount of polyphenols and the Al bioaccessibility from different baby foods can be determined. It was observed that lower polyphenol content means a greater interaction between the elements and less soluble structures, such as proteins, fibres and phytates, which leads to smaller bioaccessible fractions (Do Nascimento da Silva et al., 2013). The low bioaccessible fraction observed in Table 4 for meat purees - chopped meat (2.1 %), - chocolate milk drink (4.4 %) and petit

suisse (9.2 %) may be strongly related to the large amount of proteins. In this regard, it is interesting to note that probably brand H (petit suisse sample) presents lower bioaccessible fraction compared to brand I due to the larger amount of proteins, once that the label declares the presence of dairy proteins, besides containing whole milk in its composition. Although the theoretical results show that the interaction between Al and polyphenols is the strongest, which would lead to greater bioaccessibility for this element, its interaction with cellulose is also the strongest. The interaction with cellulose may be the most important factor governing the bioaccessibility of elements in foods that contain larger amounts of cellulose than polyphenols. In addition, the cellulose structures are less soluble, which will more strongly affect the bioaccessibility of Al.

Further, some studies have associated low mineral absorption with the presence of antinutritional factors, such as phytates, which are present in plant seeds, grains and, at high amounts, in wheat bran, acting as the main stored form of phosphate in plant seeds (Schons et al., 2011; Paiva et al., 2019a; Do Nascimento et al., 2015). Regarding total Al content and its bioaccessible fraction, some parameters must be taken into account, such as the element forms (different chemical compounds), the behavior of organometallic species and complexes in the gastrointestinal tract, and the interactions with the food matrix (Khouzam et al., 2011).

3.3. Evaluation of the potential risk related to the intake of Al from infant foods

Table 5 presents the results of Al exposure with regard to the contribution of the PTWI of $2 \text{ mg kg}^{-1} \text{ bw}$ (JECFA, 2011) and the TWI of $1 \text{ mg kg}^{-1} \text{ bw}$ (European Food Safety Authority (EFSA), 2008) considering total Al concentration.

It can be observed that the consumption of three daily portions of soy-based drink along the week exceeded 100 % of the TWI (101 %). Other baby food samples such as chocolate soy-based drink, dark chocolate milk drink, salty puree (zucchini, lettuce, lentil, sweet potato and egg) and fruit puree (apple and plum) contributes up to 80.8 %, 77.1 %, 58.7 % and 53.2 % of TWI, respectively.

This scenario illustrates that the regular consumption of highly contaminated products, such as soy-based drinks, chocolate-based composition, and some salty purees may suggest a potential concern regarding Al exposure. In addition, it should be noted that the results reported in the present study may be under estimated once other foods (e.g. cereals, vegetables, fruits and juices) are incorporated into the diet of children over 6 months, which may increase the total Al intake (Chekri et al., 2019).

When compared to the PTWI established value, the results were reduced by half and a contribution varying from 2.94 % up to 50.7 % among the brands, depending on the composition, was observed.

Paiva et al. (2019a) recent developed a study in infant formulas commercialized in Brazil finding maximum estimated exposure in soy and chocolate based formulas of 36.4 % (6–12 months) and 29.4 % (12–24 months) when compared to TWI. These results are in agreement with the current study which points out that the composition plays an important role in infant food contamination and exposure to Al.

4. Conclusions

In the present study, it was possible to determine the total Al content in baby food samples as well as its bioaccessible fraction through an adapted *in vitro* approach considering the gastrointestinal system of children. The employed analytical method showed suitable values for repeatability and accuracy for baby food samples. The highest total Al content was observed in handmade purees samples containing zucchini, lettuce, lentil, sweet potato and egg, fruit puree containing apple and plum, as well as in soy-based drink, chocolate soy milk drink, and petit suisse. Bioaccessibility, however, presented lower values, varying from

Table 5

Al exposure expressed as % of the Provisional Tolerable Weekly Intake (PTWI) and the Tolerable Weekly Intake (TWI) considering total Al concentration.

Brands	Composition	Al exposure (µg/Kg bw) per day	Al exposure (µg/Kg bw) per week	% PTWI	% TWI
A	Fruits mix	26.9	189	9.43	18.8
	Plum	21.8	152	7.61	15.2
	Noodles with chicken breast and vegetables	11.2	78	3.90	7.81
	Meat vegetables and cassava	15.3	107	5.37	10.7
	Noodles, meat and vegetables	11.3	80	3.97	7.95
	Egg yolk, meat and vegetables	25.5	179	8.93	17.8
	Chicken breast with vegetables	6.06	42.4	2.12	4.24
	Vegetables and meat	7.63	53.4	2.67	5.34
	Chopped meat	33.9	238	11.9	23.8
B	Apple and plum	75.9	531	26.6	53.2
	Chicken Breast, tomato, potato and broccoli	40.8	285	14.3	28.5
	Zucchini, sweet potato, lettuce, carioca beans and egg yolk	70.2	491	24.6	49.1
	Meat, carrot, lettuce, black beans and rice	42.7	299	14.9	29.8
	Zucchini, lettuce, lentil, sweet potato and egg	83.9	587	29.3	58.7
C	Strawberry, raspberry and apple	48.0	336	16.8	33.6
	Pear, grape and apple	49.8	349	17.4	34.9
	Yellow fruits and chia	23.7	166	8.29	16.6
	Banana and apple	38.0	266	13.3	26.6
D	Dark chocolate milk drink	110	771	38.5	77.1
	Apple and papaya milk drink	45.1	316	15.8	31.6
	Banana milk drink	27.2	191	9.53	19.1
E	Chocolate milk drink	89.9	629	31.4	62.9
	Strawberry milk drink	53.2	373	18.6	37.2
	Cereal milk drink	45.1	316	15.8	31.6
F	Chocolate milk drink	89.6	627	31.4	62.8
	Soy milk drink	145	101	50.7	101
	Oat and soy milk drink	13.3	92.9	4.64	9.29
G	Soy milk drink	89.5	626	31.3	62.7
	Chocolate soy milk drink	115	808	40.4	80.8
H	Petit-suisse	42.3	296	14.8	29.6
I	Petit-suisse	8.41	58.9	2.94	5.88

0.5%–48% depends on the sample composition.

It was demonstrated that the consumption of three daily portions of soy-based drinks per week, for example, exceeded the TWI of 1 mgkg^{-1} bw, whereas for chocolate soy-based drink and for a salty puree composition, values of 80.8 % and 58.7 % can be reached, respectively.

The results highlight the importance of evaluating the exposure to Al considering that the incorporation of other foods into the diet may increase its total intake, representing a health concern, especially for frequent consumers. The data obtained in this study will contribute to a global evaluation of possible health risks with respect to Al exposure, indicating the needs to establish ways of controlling the quality of the ingredients used in infant food production and to develop studies to clarify the mechanism of toxicity related to Al.

Declaration of conflicting interest

The authors declare no conflict of interest.

CRedit authorship contribution statement

Esther Lima de Paiva: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing. **Camila Medeiros:** Methodology, Validation, Formal analysis. **Maria Isabel Andrekowisk Fioravanti:** Formal analysis. **Raquel Fernanda Milani:** Software, Data curation. **Marcelo Antônio Morgano:** Conceptualization, Investigation, Resources, Writing - review & editing, Supervision, Project administration, Funding acquisition. **Juliana Azevedo Lima Pallone:** Methodology. **Adriana Pavesi Ariseto-Bragotto:** Conceptualization, Investigation, Writing - review & editing, Supervision, Project administration.

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