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Aluminium intake through the consumption of selected baby foods and risk characterization in a population of Brazilian infants aged 0 to 36 months

Esther Lima de Paiva [a,b,*], Sandy Galvani Lima [c], Nadia Waegeneers [d], Mirjana Andjelkovic [d], Renata Elisa Faustino de Almeida Marques [c], Marcelo Antônio Morgano [b], Adriana Pavesi Arisseto-Bragotto [a]

[a] Faculty of Food Engineering, University of Campinas (UNICAMP), Rua Monteiro Lobato 80, 13083-862 Campinas, SP, Brazil
[b] Institute of Food Technology (ITAL), Avenida Brasil 2880, C. P. 139, 13070-178 Campinas, SP, Brazil
[c] Universidade Paulista (UNIP), Av. Comendador Enzo Ferrari 280, 13045-770 Campinas, SP, Brazil
[d] Sciensano, Rue Juliette Wytsmanstraat 14, 1050 Brussels, Belgium

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ABSTRACT

Aluminium (Al) dietary intake from selected baby foods was estimated for Brazilian infants from the São Paulo State by means of a deterministic and a probabilistic estimation. The deterministic exposure assessment was carried out by combining mean levels of Al in 171 samples of baby foods (infant formula, meat/vegetable purees, fruit purees, petit-suisse and cereals), analytically determined by inductively coupled plasma optical emission spectrometry method, with individual food consumption data. Data on food consumption were generated using a duplicated 24 h recall applied to the parents of 158 infants aged from 0–36 months. The mean Al intakes for the total population, calculated using mean Al concentrations, were estimated to be 0.184 mg/kg body weight (bw) per week, whilst at 95th percentile, the obtained value was 0.474 mg/kg bw per week. In addition, distributions were fitted on the concentration and consumption data for further probabilistic intake estimations. The results corroborated well with the deterministic approach. The highest frequency and daily food consumption (in g/day) were observed for infant formulas and meat/vegetable purees, respectively. Boys presented higher frequency consumption of infant formulas, while meat/vegetable purees intake by girls was higher compared to boys. The baby food that most contributed to Al exposure in the total population, considering mean Al concentration values, was meat/vegetable purees, followed by infant formulas. This study suggests potential concern regarding consumers of highly Al contaminated products and may be used as a basis for the establishment of risk management actions.

1. Introduction

Adequate nutrition in infancy and early childhood is critical for children's full development, growth, and health. A diversified diet can meet physiological and nutritional needs by delivering key vitamins and micronutrients, most of which humans cannot synthesize and must obtain from foods (Milisav et al., 2018).

It is globally recommended that children initiate breastfeeding within the first hour of birth and be exclusively breastfed for the first 6 months of life, with continuous breastfeeding up to 2 years old or beyond. Short-term and long-term benefits on both child and mother can be achieved with breastfeeding, including helping to protect children against various acute and chronic diseases. Breastmilk provides all the nutritional needs for the first months of life, and it continues to provide energy and important nutrients after this period (WHO, 2009). When breastfeeding is not possible, insufficient, or not prioritized for several reasons including the mother's lack of breastfeeding experience, active work life and desire to feed their babies with new tastes, infant formula is usually chosen to supply the nutritional demands of suckling infants (Kazi et al., 2009; Başaran, 2020).

From the age of 6 months, nutritionally adequate and safe complementary foods must be introduced. According to the World Health Organization (WHO, 2009), the introduction of complementary foods marks an important point in their feeding pattern since it coincides with the time when they begin absorbing vital nutrients, such as iron and zinc, being susceptible to nutritional deficiencies in the case of an

* Corresponding author at: Faculty of Food Engineering, University of Campinas (UNICAMP), Rua Monteiro Lobato 80, 13083-862 Campinas, SP, Brazil.
E-mail address: paivel19@gmail.com (E.L. de Paiva).

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unbalanced diet. Infant cereals, purees made from fruit, vegetable and meat, and infant biscuits are only a few examples of complementary foods (Koo et al., 2018).

Despite the diet being an essential source of nutrients, it is widely recognized that raw materials used to prepare foods for infants and young children, such as milk, vegetables, fruits, and cereals, may contain a number of hazardous chemical components known as inorganic contaminants (El-Kady and Abdel-Wahhab, 2018; bPaiva et al., 2019). These include elements such as aluminium (Al), which is the third most abundant element in the earth's crust (8 %) and it is usually released into the environment as a result of the natural weathering of rocks. Food is one of the main sources of Al, which can be found in fresh vegetables and fruits (available in soil), in drinking water (as flocculant) as well as in industrially produced foods (as additive and packaging material). Other sources of Al in the diet include kitchen utensils and tea consumption (Ibrahim et al., 2022).

The primary adverse effects of Al may be the accumulation in lungs, liver, kidney, thyroid and brain (Rebellato et al., 2021). Al has been associated with anemia, impaired bone formation, disorders in the digestive, respiratory, and immune systems, as well as neurotoxic effects (Levin-Schwartz et al., 2019; FAO/WHO, 2018). Human exposure to Al is identified as a possible contributor to neuro-degenerative/neurodevelopmental diseases, such as Alzheimer's disease and multiple sclerosis. Individuals with relapsing remitting multiple sclerosis and secondary progressive multiple sclerosis were shown to excrete large amounts of Al in the urine (Rebellato et al., 2021; Jones et al., 2017).

Increased susceptibility to the toxic effects of Al is observed in the early stages of life, fetal and early postnatal periods (Makri et al. 2004; Sly and Flack, 2008). Children's metabolic pathways are immature, particularly during the fetal period and the first months after birth, implying that metabolism and detoxification are less efficient in infants than in adults. Furthermore, a high intestinal absorption capacity and a low effective excretion imply a higher exposure (Koo et al., 2018; El-Kady and Abdel-Wahhab, 2018). In addition, the low body weight (bw) of children results in a higher amount of ingested Al per unit bw in comparison to adults (Hulin et al., 2014). Recently, Gomes et al. (2020) demonstrated that neonatal Al exposure causes morphological changes in the ventral male prostate and in the female prostate of PN15-gerbils, concluding that the contact of neonates with Al should be strictly avoided.

The Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a Provisional Tolerable Weekly Intake (PTWI) for Al of 2 mg/kg bw applied to all the Al compounds in foods, including food additives, considering renal damage (hydronephrosis, urethral dilatation, obstruction and/or presence of calculi) and reduced grip strength (FAO/WHO, 2011; CAC, 2018). On the other hand, the European Food Safety Authority (EFSA) set a Tolerable Weekly Intake (TWI) of 1 mg/kg bw for Al in all food sources given the persistence of this element in the body and considering combined evidence from a number of animal studies showing adverse effects on testes, embryos and the developing and mature nervous system following dietary administration of Al compounds (EFSA, 2008).

JECFA observed that the PTWI is likely to be exceeded in some population groups, particularly children. The Committee also inferred that exposure to this metal by infants fed some types of infant formulae is high and recommended further studies with these products (FAO/WHO, 2011). EFSA noted that the TWI of 1 mg/kg bw is likely to be exceeded in a significant part of the European population (EFSA, 2008).

These observations have stimulated some exposure studies worldwide. In Spain, Cabrera-Vique and Mesías (2013) estimated Al intake along the day (breakfast, lunch, afternoon meal, and dinner), using analytical data provided by Stahl et al. (2011). An exposure health risk assessment to Al in commonly consumed infant formulas in Nigeria was performed by Igweze et al. (2020). In Turkey, Başaran (2022) estimated Al intake though the consumption of infant formulas and baby biscuits by 348 babies. Al intake was also assessed in children from Lebanon considering the consumption of infant formula and baby food (Ibrahim et al., 2022). Further, Sirot et al. (2018) also conducted a total diet study (TDS) between 2010 and 2016 to assess the risk associated with chemicals in food, including Al, of non-breast-fed children in France.

In Brazil, the population of children aged 0–4 years in 2021 was 14,657 thousand individuals (PNAD, 2021) and, regarding the baby food market, baby milks remain the largest category in value terms with nearly 60 % share of the overall baby food sector, followed by baby cereals and dry meals (GlobalData, 2022). No exposure assessment of children to Al through the consumption of baby food has been reported until recently. In 2019, aPaiva et al. (2019) performed a preliminary exposure assessment through the consumption of infant formulas. In 2020, an intake evaluation through non-cereal baby foods was carried out (aPaiva et al., 2020). Regarding cereal-based baby foods, bPaiva et al. (2020) performed a deterministic calculation using measured Al levels. According to these preliminary studies conducted in Brazil, using theoretical food consumption data based on serving sizes, the intake of Al through the diet suggested a potential concern for infants (aPaiva et al., 2019; aPaiva et al. 2020; bPaiva et al., 2020). This indicated the need to refine these preliminary assessments by using real food consumption data in order to provide a more realistic scenario of Al intake by infants in Brazil. Therefore, the aim of this work was to collect data on food consumption of infants aged 0–36 months in order to perform a refined exposure assessment and risk characterization to Al, considering its occurrence in commercial infant formulas and baby foods sold in Brazil.

2. Material and methods

2.1. Food consumption data

Individual food consumption data were originated from a survey carried out with the parents or guardians of 158 infants and toddlers (81 boys and 77 girls), aged from 0 m to 36 months, using a duplicated 24 h diet recall in which food and beverages consumed in the last two days prior to the interview (48 h) were recorded (McNaughton et al., 2005). The survey was approved by the Ethical Committee of the State University of Campinas and was applied in a private pediatric clinic located in the city of Campinas, São Paulo State, before or after consultation with the pediatrician, between January and May of 2019. A standard explanation of the survey was given to each participant before the beginning of the interview and signing of the informed consent form. The questionnaires were answered in detail by the children's parents or guardians regarding the form of preparation and quantity of consumed foods and beverages, as well as leftovers, time and place of each meal. Measurements of weight and height performed by the pediatrician in the child's last visit were considered in the present assessment, not exceeding a maximum period of 30 days. To convert the data collected in this study, the Avanutri® software for nutritional assessment and prescription was used.

2.2. Aluminium analytical data

Al concentrations in commercial infant formulas and baby foods sold in Brazil were reported in our previous studies (aPaiva et al., 2019; aPaiva et al. 2020; bPaiva et al., 2020). A total of 171 samples, from different brands and batches of the selected products, were purchased in 2018 and 2019 at supermarkets, and pharmacies in the region of Campinas, SP. After homogenization, the samples were submitted to acid digestion using distilled nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) in a closed microwave-assisted digestion system (Start E, Milestone, Sorisole, Italy) equipped with 24 teflon flasks with internal volumes of 50 mL. The total Al content was determined using optical emission spectrometry with an inductively coupled plasma source (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 limits of detection (LOD) and limits of quantitation (LOQ) were, respectively: 49 ng/g and 92 ng/g for purees and petit-suisse; 73 ng/g and 122 ng/g for infant formulas; and 53 ng/g and 89 ng/g for cereals.

2.3. Exposure assessment and risk characterization

The exposure assessment was performed considering the consumption of infant formula, cereal baby foods and non-cereal baby foods (including purees and petit-suisse), for which data on Al concentration were available. Children consuming none of these food items (n = 29) and participants who have not reported body weight (n = 3) were excluded from the data set ("consumers only" calculations). Mean individual food consumption obtained from the duplicated 24 h diet recall was used.

A deterministic approach was applied to estimate Al intake individually, considering the total population as well as subgroups according to the age (0–12 and 13–36 months) and gender (boys and girls). For that, Al concentrations were multiplied by the individual food consumption data for each selected baby food. The intakes from all sources of exposure were then summed and divided by children's weight (Assunção et al., 2015), according to the equation

$$EDI = \frac{1}{bodyweight} \bullet \sum_i \frac{Concentration_i}{1000} \bullet Consumption_i$$

in which EDI is the estimated dietary intake (in µg/kg bw/day), the index i refers to the different types of baby food, $Concentration_i$ is the Al concentration in baby food i (in ng/g) and $Consumption_i$ is the individual daily consumption of baby food i (in g/day). Two scenarios of exposure were assumed: 1) using mean Al concentrations and 2) using maximum Al concentrations (worst case). The estimated daily dietary Al intake data were multiplied by 7 and divided by 1000 to derive the estimated weekly dietary intakes in mg/kg bw. Dietary intakes were reported as mean, maximum, and at 50th, 95th and 97.5th percentiles.

The probabilistic exposure estimation was applied to estimate Al intake by infants (0–12 months) and toddlers (13–36 months). Hereto, different distributions were fitted to the Al concentrations measured in infant formulae, non-cereal based baby food and cereals, and to the consumption data on infant formulae, non-cereal based baby food and cereals by either infants or toddlers, expressed in g/kg bw/day. The distributions were fitted using the Fitdistrplus package in the Open Source R Studio software (version 1.4.1717), available at the CRAN (Comprehensive R Archive Network) repository. The Aikaike Information Criterion (AIC) was used to determine the best fitting distribution. Table 1 lists the best fitted distributions of the consumptions and Al concentrations, while Figs. 1 and 2 show how the distributions fitted over the available data.

Based on these distributions, a Monte-Carlo simulation was performed using R Studio. The daily dietary Al intake was calculated according to the equation

$$EDI = \sum_i \frac{Concentration_i}{1000} \bullet Consumption_{bw,i}$$

in which EDI is the estimated dietary intake (in µg/kg bw/day), the index i refers to the different types of baby food, $Concentration_i$ is a randomly selected Al concentration in baby food i (in ng/g) and $Consumption_{bw,i}$ is a random individual daily consumption of baby food i (in g/kg bw/day). Six exposure scenarios were assumed: 1) consumption of infant formulae only, 2) consumption of infant formulae and non-cereal based baby food, 3) consumption of infant formulae, non-cereal based baby food and cereals, and for each of these three scenarios a scenario assuming a negligeable Al concentration in water (0 ng/mL) needed to prepare the infant formulae out of the powdered product, and a scenario

Table 1
Fitted distributions of Al concentrations in different types of food, and fitted distributions of the consumptions of these foods by infants and toddlers for the probabilistic assessment of aluminium intake.

	Distribution	AIC	Applied min	Applied max
Al concentration in:				
Infant and follow-on formulae	Weibull (1.622669; 1854.817)	1258	126 ng/g	6534 ng/g
Non-cereal based babyfood	Log-normal (6.894036; 0.833037)	931	113 ng/g	4631 ng/g
Cereals	Weibull (2.043079; 4272.736)	629	828 ng/g	9702 ng/g
Consumption of:				
Infant/follow-on formulae by infants	Weibull (1.822337; 10.79622)	253	0.96 g/kgbw/day	28.2 g/kgbw/day
Infant/follow-on formulae by toddlers	Weibull (2.183559; 8.371312)	339	1.10 g/kgbw/day	21.1 g/kgbw/day
Infant/follow-on formulae by infants	Normal(7.011073; 6.336774)	370	0.96 g/kgbw/day	28.2 g/kgbw/day
Non-cereal based babyfood by infants	Exponential (0.05077)	448	0 g/kgbw/day	85.4 g/kgbw/day
Infant/follow-on formulae by toddlers	Normal(5.366879; 4.504189)	513	1.10 g/kgbw/day	21.1 g/kgbw/day
Non-cereal based babyfood by toddlers	Exponential (0.094715)	586	0 g/kgbw/day	76.7 g/kgbw/day
Infant/follow-on formulae by infants	Normal(7.011073; 6.336774)	370	0.96 g/kgbw/day	28.2 g/kgbw/day
Non-cereal based babyfood by infants	Exponential (0.05077)	448	0 g/kgbw/day	85.3 g/kgbw/day
Cereals by infants	Exponential (11.33326)	-158	0 g/kgbw/day	2.75 g/kgbw/day
Infant/follow-on formulae by toddlers	Normal(5.187983; 4.532061)	531	1.10 g/kgbw/day	21.1 g/kgbw/day
Non-cereal based babyfood by toddlers	Exponential (0.097981)	600	0 g/kgbw/day	76.7 g/kgbw/day
Cereals by toddlers	Exponential (2.261779)	35	0 g/kgbw/day	10.8 g/kgbw/day

assuming an Al concentration in water equal to 0.2 mg/L (200 ng/g), which is the maximum permitted level in drinking water (ANVISA, 2011). In both scenarios, a water consumption of 5 mL per gram powdered infant formula was assumed.

The Monte Carlo simulation was performed 250 times with 2000 iterations. The estimated daily dietary Al intake data were multiplied by 7 and divided by 1000 to derive the estimated weekly dietary intakes in mg/kg bw.

Two health-based guidance values were considered to evaluate the potential risk of exposure to Al: the PTWI defined by the JECFA as 2 mg/kg bw (FAO/WHO, 2011; CAC, 2018) and the TWI established by the EFSA at 1 mg/kg bw (EFSA, 2008). The estimated Al exposure was expressed in terms of % PTWI or TWI according to the equation: % PTWI or TWI = (100 x Estimated Al exposure) / (PTWI or TWI). A risk may exist if Al exposure is above 100 % of the PTWI or TWI. Uncertainties on the exposure estimation were evaluated qualitatively according to the EFSA guidance document (2018).

2.4. Statistical analysis

The means obtained for each baby food type, regarding the frequency and consumption (g/day) between different age group and gender, were compared by variance analysis (ANOVA) and Tukey's test ($p < 0.05$). All statistical analyses were made using the Statistica 7.0

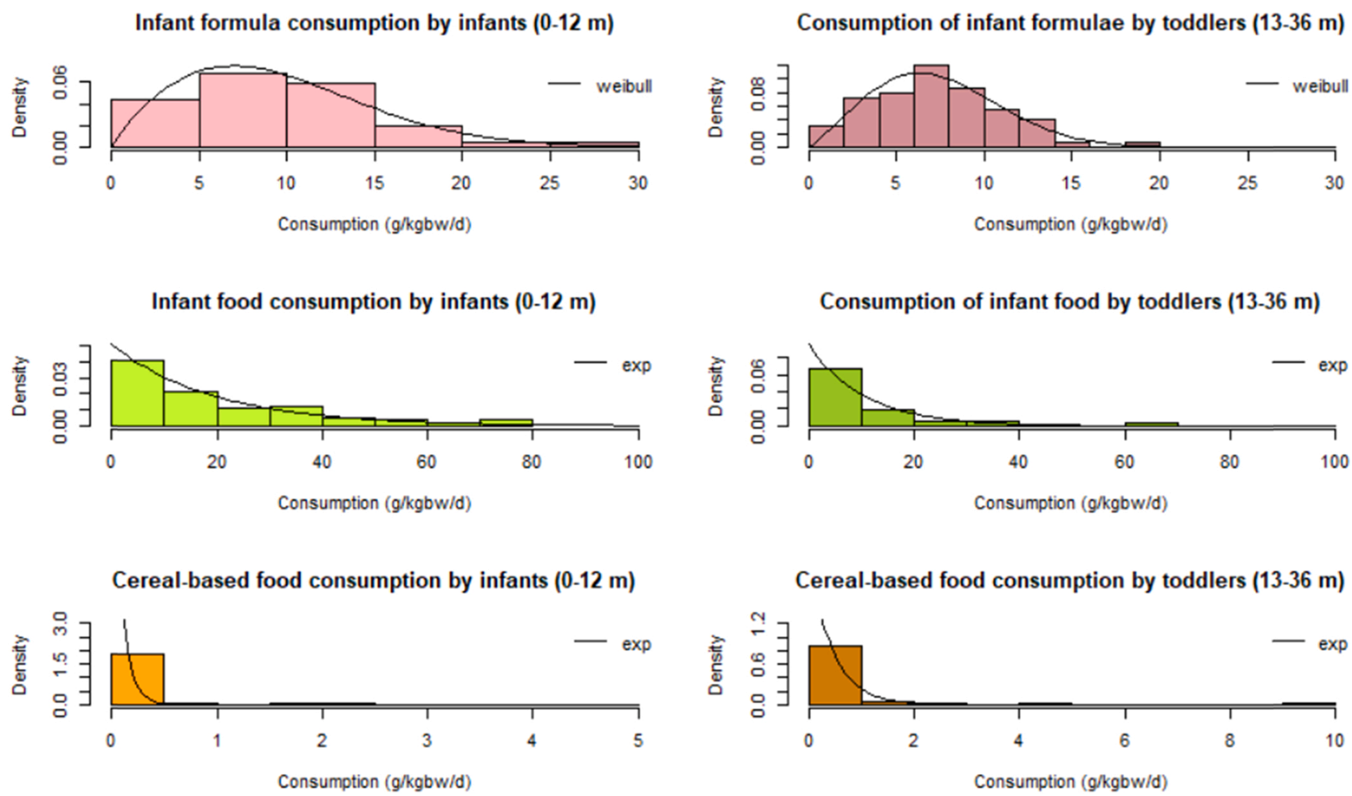

Fig. 1. Distributions fitted over the consumption data for infant and follow-on formulae, non-cereal based infant food and cereals by children aged 0–12 months and 13–36 months.

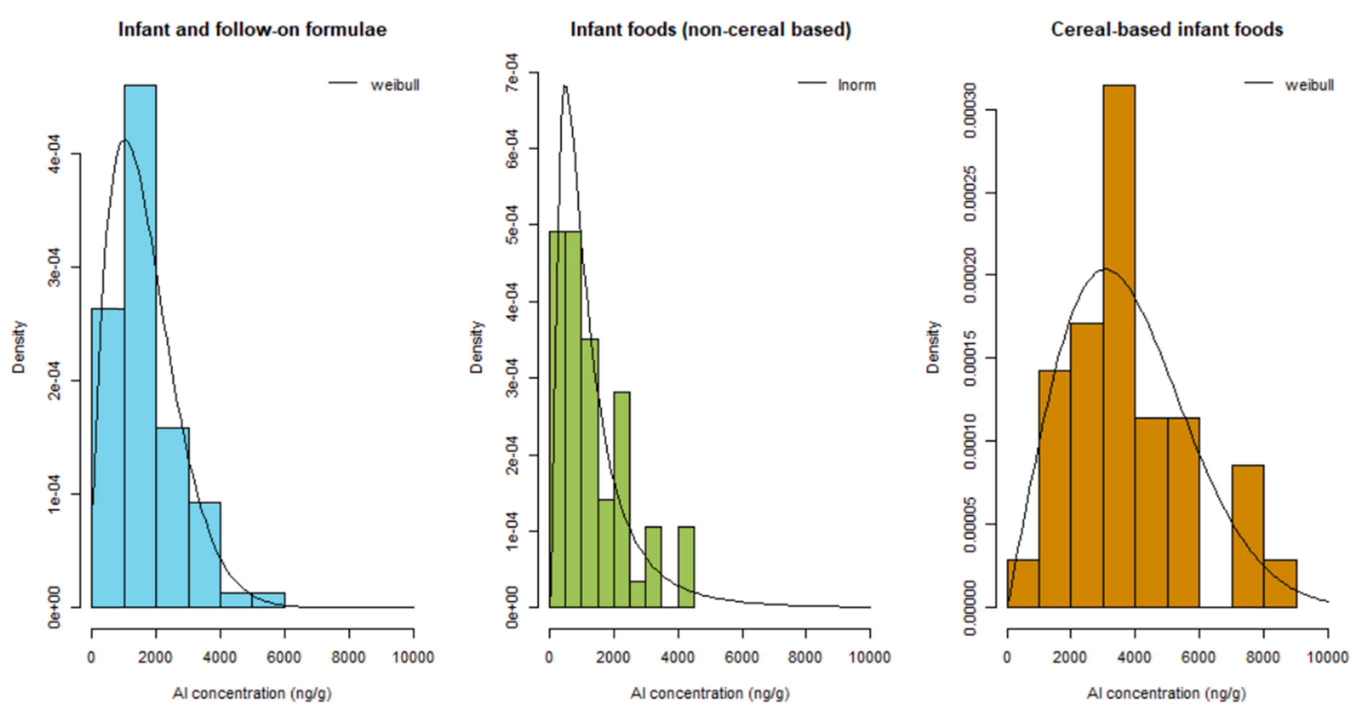

Fig. 2. Distributions fitted over the concentration data for infant and follow-on formulae, non-cereal based infant food and cereals.

software package (Statsoft, USA).

3. Results and discussion

3.1. Food consumption

Table 2 presents the frequency of consumption of the selected baby

Table 2
Consumption frequency of selected baby foods among the studied children.

Baby foods	Frequency (%)				
	All children (n = 126)	Infants 0–12 m (n = 57)	Toddlers 13–36 m (n = 69)	Girls (n = 61)	Boys (n = 65)
Infant formula[a]	58.7	68.4[c]	50.7[c]	54.1[e]	63.1[e]
Non-cereal					
Meat/ vegetable puree[a,b]	55.6	80.7[c,d]	34.8[c,d]	54.1[e]	56.9[e]
Fruit puree[b]	5.6	8.8[d]	2.9[d]	8.2[f]	3.1[f]
Petit-suisse[a,b]	21.4	8.8[c,d]	31.9[c,d]	23.0[f]	20.0[f]
Cereal[a,b]	22.2	14.0[c,d]	29.0[c,d]	24.6[e,f]	20.0[e,f]

Values followed by different letters on the same column differ significantly by the Tukey test ($p \leq 0.05$),. Distinctions related to baby foods category (a,b), age group (c,d), and gender (e,f).

Table 3
Mean consumption of selected baby foods by the studied children.

Baby foods	Consumption (g/day)				
	All children (n = 126)	Infants 0–12 m (n = 57)	Toddlers 13–36 m (n = 69)	Girls (n = 61)	Boys (n = 65)
Infant formula (powder)[a,b]	48.5	56.6[c.d]	41.8[c,d]	41.4[e,f]	55.1[e,f]
Non-cereal					
Meat/ vegetable puree[a]	111	143[c]	85.3[c]	117[e]	106[e]
Fruit puree[b]	5.0	7.5[d]	2.9[d]	7.6[f]	2.6[f]
Petit-suisse[b]	9.8	2.9[d]	15.5[d]	8.5[f]	11.0[f]
Cereal[b]	4.8	2.0[d]	7.2[d]	7.2[f]	2.6[f]

Values followed by different letters on the same column differ significantly by the Tukey test ($p \leq 0.05$),. Distinctions related to baby foods category (a,b), age group (c,d), and gender (e,f).

foods in the studied population. Considering the total population, infant formulas presented the highest value of frequency (58.7 %), followed by meat/vegetable puree (55.6 %), which were significantly higher than the frequency consumption of fruit puree, petit- suisse and cereal. Among the subgroups, the highest consumption frequencies of infant formula (68.4 %), meat/vegetable puree (80.7 %) and fruit puree (8.8 %) were found for 0–12 months group, while the highest frequency values for petit-suisse (31.9 %) and cereals (29.0 %) were observed in the 13–36 months subgroup. Regarding the gender subgroups, it is possible to observe that girls presented higher consumption frequency of cereal (24.6 %) and petit-suisse (23.0 %) followed by fruit puree (8.2 %), while boys showed significantly higher frequency values for infant formula (63.1 %) and meat/vegetable purees (56.9 %) compared to cereals (20.0 %), petit-suisse (20.0 % and fruit puree (3.1 %).

A Brazilian study performed by Karnopp et al. (2017) considering children younger than 6 years showed that, among infants below 12 months, the frequency mean value for the consumption of purees, cereal and juice was 12.9 % followed by biscuits (2.6 %), breads (1.9 %) and candies (1.8 %). For infants above 12 months, the obtained mean value was 14.9 % with a higher frequency for biscuits (6.2 %), breads (5.6 %) and candies (5.4 %).

Hurley and Black (2010) reported an increase of 70 % in the vegetable consumption by American infants between 3 and 5 months and 6–8 months, with frequency values of 33.3 % and 55.4 %, respectively. However, after 12 months old the consumption frequency decreased to 32.8 %, which is similar to the present study value of 34.8 % for infants aged 13–36 months.

Frizzo et al. (2021) conducted a study regarding the adequate introduction age of complementary food for Brazilian infants from 4 to 18 months. The findings showed that after 12 months old, the consumption frequency of meat purees was 12 %, which is lower in comparison to the current study (Table 2).

A review concerning infant and preschool children feeding in Brazil reported a consumption frequency of 85.9 % for milk and dairy products for infants aged 6–12 months. However, only 6.7 % of infants older than 6 months consumed infant formula as a breast milk substitute (Mello et al., 2016). Meat puree consumption assessed by 24 h-recall showed frequency values of 35 % and 47.3 % for 6–12 months and 12–24 months, respectively. No frequency data was reported for infants above 24 months old.

Regarding daily food consumption (Table 3) of the total population, meat/vegetable purees presented the highest value (111 g/day), which was significantly different from the other baby food types. Considering the subgroups, the highest mean consumption of infant formula (56.6 g/day) and meat/vegetable purees (85.3 g/day) was observed at 0–12 months, while at 13–36 months the highest mean consumption was observed for petit-suisse (15.5 g/day). Girls presented higher consumption of meat/vegetable puree (117 g/day), fruit puree (7.6 g/day) and cereal (7.2 g/day) while for boys the higher obtained consumptions were infant formula (55.1 g/day) and petit-suisse (11.0 g/day).

It is possible to observe that the frequency and daily consumption of purees (meat/vegetable and fruit) and infant formulas declines from 0 to 12 months to 13–36 months, whilst petit-suisse and cereal consumption increases. These findings suggested that infants from 13 to 36 months are in the period of transition to the family diet, with the inclusion of solid foods. For petit-suisse, the frequency values increased from 8.8 % to 31.9 % and the daily consumption from 2.9 g to 15.5 g. Cereal baby food presented more than double frequency value, rising from 14 % to 29 %, whilst daily consumption increased from 2.0 g to 7.2 g.

Concerning to the gender subgroups, it is possible to note that boys are reported as more frequent consumers of infant formula (63.1 %) and meat/vegetables (56.9 %) purees, whilst girls presented higher values for fruit puree (8.2 %), petit-suisse (23.0 %) and cereal (24.6 %). However, a different scenario is obtained if the gender daily consumption is considered. Boys presented higher consumption of infant formula (55.1 g/day) and petit-suisse (11 g/day) in comparison to the girls, whilst girls showed high daily consumption of meat/vegetables and fruits purees and cereals, with values of 1167 g/day, 7.6 g/day and 7.2 g/day, respectively.

Lee et al. (2019) performed questionnaires collected repeatedly from 64 Korean families. Means of the total amounts of solid baby food consumed were: 221 g/day for infants aged 9 months, 286 g/day for 12 months, and 348 g/day for 15 months, considering cooked rice, egg/-dairy products and others, which also supports higher consumption of solid food along infants' age group.

Hulin et al. (2014) worked on French total diet studies (TDS) project focusing on exposure to food chemicals in the population aged up to 3 years. The reported daily consumption values were: 212 g of infant formula, 37.8 g of meat puree, 37.8 g of fruit puree and 7.4 g for cereal-based food. In terms of comparison to the present findings, these values are higher for infant formulas and fruit purees; however, similar consumption is observed for the girl's intake of cereal-based foods (7.2 g/day). According to Mello et al. (2016), cereal daily consumption of 28 g and 82 g were obtained for Brazilian infants between 6 and 12 months and above 12 months, respectively, which is in accordance with the trend of increased consumption of solid foods presented in the current study.

A Japanese study developed by Sato et al. (2014) regarding Al exposure and food analysis from 189 commercialized samples in Tokyo reported mean daily consumption of 0.56 mg/kg bw for cereal-based food, 0.08 mg/kg bw of legumes and nuts, 0.09 mg/kg bw of milk and milk products, and 0.01 mg/kg bw of fruits and vegetables, considering infants from 1 to 6 years old.

3.2. Aluminium concentration in selected baby foods

Table 4 presents the concentrations of Al in the selected baby foods considered in the calculations, which were previously reported by the group (aPaiva et al., 2019, aPaiva et al., 2020, bPaiva et al., 2020). The highest mean levels of Al are present in cereal (3783 ng/g) and petit-suisse (2500 ng/g), followed by infant formula (1599 ng/g), fruit puree (1497 ng/g) and meat/vegetables puree (1057 ng/g).

The study developed by bPaiva et al. (2020) regarding Al content in cereal-based baby foods allowed the observation of high total Al concentrations in the samples (max 8820 ng/g), which may be related to the brand, type of cereal used in the formulation (oat, corn, rice, wheat, barley, among others) and flour composition. Other authors also reported that the presence of Al may be related to the manufacturing processes of these foods or to the packaging and emphasized concern about Al levels found in the samples (Ayivor et al., 2011).

Considering infant formula samples, Paiva et al. (2019) found Al levels varying from 330 to 1460 ng/g for starters and concentrations between 170 and 5940 ng/g for follow-up ones (non-reconstituted product). In Pakistan, Kazi et al. (2009) reported mean Al contents in milk-based formulas from 640 ng/g to 1520 ng/g, similar to the mean Al levels presented in the current study (1599 ng/g). Other authors (Woollard et al.,1990) carried out a study in New Zealand about the presence of Al in infant formulae from different countries and found concentrations varying from 250 ng/g to 5000 ng/g in milk-based formula.

Regarding baby purees, Paiva et al. [a](2020) reported that brands with a handmade claim notably presented higher levels of Al in comparison to industrial ones considering similar compositions of salty and fruit purees. This indicated that handmade production might not have accurate control regarding procedures, utensils, storage and raw materials supply.

El Daouk et al. (2020) performed Al analysis in Lebanese foods and high mean concentrations were observed for vegetables (9370 ng/g), followed by candies (7970 ng/g), ready meals including purees (7580 ng/g), and potatoes (7280 ng/g). In Japan, values of 6750 ng/g for cereal, 670 ng/g for infant beverages, 1130 ng/g for potatoes and legumes, 460 ng/g for meats, 1210 ng/g for milk and dairy products and 1560 ng/g for fruits and vegetables were reported (Sato et al., 2014).

The major factors of food contamination by Al are not specified, but soil and irrigation water appear to be the primary cause. According to El Daouk et al. (2020), the content of Al in food showed large dissimilarities between countries and this was due to the variation in the study design, selected food, food processing (diverse quantities used of Al containing food additives), cooking, storage (contact with aluminum utensils, containers, Al foil and others). 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).

Table 4
Aluminium concentrations in selected baby foods.

Sample	N	Aluminum (mg/kg)			References
		Mean	Median	Min - Max	
Infant formula (powder)	76	1599	1710	140 – 5940	Paiva et al.(2019)
Non-cereal					
Meat/vegetable puree	33	1057	880	125 – 3450	aPaiva et al.(2020)
Fruit puree	21	1497	1500	620 – 3070	aPaiva et al.(2020)
Petit-suisse	6	2500	2500	830 – 4210	aPaiva et al.(2020)
Cereal	35	3783	3460	920 – 8820	bPaiva et al.(2020)

3.3. Estimated intake of Al from selected baby foods and risk characterization

3.3.1. Deterministic approach

The estimated Al intake for scenario 1 (mean Al concentrations) and scenario 2 (maximum Al concentrations) are presented in Table 5.

In scenario 1, a mean intake of 0.18 mg/kg bw/week was estimated for the total population of infants considered in this study. At 50th percentile, the dietary intake was estimated to be 0.14 mg/kg bw/week, which means that 50 % of the total population had an intake of Al at or below 0.14 mg/kg bw/week. At high percentiles, the Al intake was approximately 2.5- (95th percentile) and 2.8-fold (97.5th percentile) higher than the mean, and the maximum estimated intake was 0.85 mg/kg bw/week.

Mean values of 0.22 mg/kg bw/week and 0.15 mg/kg bw/week were observed for consumers from 0 to 12 months and 13–36 months, respectively. Concerning the gender, girls presented higher mean intake in comparison to boys, with values of 0.19 mg/kg bw/week and 0.18 mg/kg bw/week, respectively. At 95th and 97.5th percentiles, girls Al intake were also higher while at 50th percentile boys presented 0.15 mg/kg bw/week and girls 0.13 mg/kg bw/week.

If a critical scenario is proposed considering maximum Al concentrations (scenario 2), illustrating consumers of highly contaminated products, a mean intake of 0.58 mg/kg bw/week was observed for the total population, which is approximately 3-fold higher than scenario 1. At 95th percentile, the estimate Al intake was 1.50 mg/kg bw/week. The maximum value reported was 2.79 mg/kg bw/week.

Mean values of 0.73 mg/kg bw/week and 0.46 mg/kg bw/week were obtained for infant (0–12 months) and toddler (13–36 months) subgroups, in this order, whilst at 95th percentile the Al intake were 1.63 mg/kg bw/week and 1.32 mg/kg bw/week, respectively. For the girls group, a mean value of 0.60 mg/kg bw/week was observed and 0.57 mg/kg bw/week was the mean intake estimated for boys. At 95th percentile, girls also presented a higher Al intake than boys.

3.3.2. Probabilistic approach

The estimated Al intake for infants and toddlers according to different consumption scenarios are presented in Tables 6 and 7. Diversification in food consumption results in an increasing intake of Al. For infants, the introduction of non-cereal based baby food and cereals besides the consumption of infant formulae, increases the mean and 95th percentile Al intake by 60–80 % depending on the assumed Al concentration in water to prepare the infant formulae. For toddlers, the consumption of non-cereal based baby food and cereals increases the mean Al intake by 60–100 %, and the 95th percentile Al intake by 60–90 % compared to the Al intake through the consumption of infant/follow-on formulae only. The mean Al intake by toddlers consuming infant/follow on formulae and baby food is, however, 21–31 % lower than the mean Al intake by infants consuming these foods, which can be explained by the decreasing consumption of food per unit bodyweight. At the 95th percentile, the intake decreases by 12–36 %. The concentration of Al in water is a source of uncertainty in the dietary intake estimation of Al. At the two extreme concentrations (0 mg/L and the maximum allowed concentration) the mean Al intake varies by 29 %–45 %, depending on the diversification in food consumption, and by 37–57 % at the 95th percentile Al intake.

3.3.3. Comparison of deterministic and probabilistic approaches

The mean deterministic intake calculations for infants and toddlers according to scenario 1 (based on mean Al concentrations), 0.22 and 0.15 mg/kg bw/week respectively, correspond well to the mean probabilistic intake calculations (0.23 and 0.18 mg/kg bw/week). Likewise, the mean deterministic intake calculations for infants and toddlers according to scenario 2 (based on maximum Al concentrations), 0.73 and 0.46 mg/kg bw/week respectively, correspond well to the 97.5th percentile probabilistic intake calculations (0.61 and 0.55 mg/kg bw/

Table 5
Estimated aluminium intake (mg/kg body weight per week) according to the deterministic approach.

Intake	Scenario 1 (using mean Al concentrations)					Scenario 2 (using maximum Al concentrations)				
	All children (n = 126)	Infants 0–12 m (n = 57)	Toddlers 13–36 m (n = 69)	Girls (n = 61)	Boys (n = 65)	All children (n = 126)	Infants 0–12 m (n = 57)	Toddlers 13–36 m (n = 69)	Girls (n = 61)	Boys (n = 65)
Mean	0.18	0.22	0.15	0.19	0.17	0.58	0.73	0.46	0.60	0.56
P50	0.13	0.19	0.11	0.13	0.14	0.45	0.69	0.32	0.38	0.47
P95	0.48	0.49	0.43	0.49	0.45	1.49	1.63	1.32	1.62	1.43
P97.5	0.53	0.56	0.50	0.56	0.49	1.67	1.82	1.47	1.86	1.53
Maximum	0.85	0.85	0.55	0.85	0.53	2.79	2.79	1.82	2.79	1.67

Table 6
Probabilistic intake estimates of aluminium (mg/kg body weight per week) by Brazilian infants.

Scenario	Mean	P50	P95	P97.5
Al concentration in water = 0 mg/L				
Consumption of infant formulae only	0.14	0.12	0.31	0.36
Consumption of infant formulae & non-cereal based babyfood	0.22	0.18	0.49	0.61
Consumption of infant formulae, non-cereal based babyfood & cereals	0.23	0.19	0.49	0.61
Al concentration in water = 0.2 mg/L				
Consumption of infant formulae only	0.18	0.15	0.42	0.51
Consumption of infant formulae & non-cereal based babyfood	0.32	0.26	0.77	0.96
Consumption of infant formulae, non-cereal based babyfood & cereals	0.33	0.27	0.77	0.96

Table 7
Probabilistic intake estimates of aluminium (mg/kg body weight per week) by Brazilian toddlers.

Scenario	Mean	P50	P95	P97.5
Al concentration in water = 0 mg/L				
Consumption of infant formulae only	0.09	0.07	0.23	0.28
Consumption of infant formulae & non-cereal based babyfood	0.17	0.13	0.43	0.55
Consumption of infant formulae, non-cereal based babyfood & cereals	0.18	0.14	0.44	0.55
Al concentration in water = 0.2 mg/L				
Consumption of infant formulae only	0.14	0.12	0.31	0.37
Consumption of infant formulae & non-cereal based babyfood	0.22	0.18	0.49	0.61
Consumption of infant formulae, non-cereal based babyfood & cereals	0.22	0.19	0.49	0.61

week). The high percentiles (95th and 97.5th) of the deterministic calculations in scenario 2 tend to overestimate the dietary Al intake by children. These results justify the use of the deterministic approach, except for the high percentiles of scenario 2.

3.4. Risk characterization

Fig. 3 presents the results of the risk characterization of Al exposure as percentage of the PTWI and TWI, for both evaluated scenarios in the deterministic approach. In scenario 1 (mean Al concentrations), maximum contributions in relation to the PTWI were: 43 %, 27 %, 43 % and 26 % for infant, toddler, girls' and boys' subgroups, respectively. Regarding the TWI, maximum values were observed for the girls and infant subgroup (85 %) whilst 55 % was obtained for toddlers and 53 % for boys. Considering scenario 2 (maximum Al concentrations) at 95th percentile, values of 81 %, 66 %, 81 % and 72 % were obtained for PTWI for the respective subgroups of infants, toddlers, girls and boys; whilst 163 %, 132 %, 162 % and 143 % were observed for TWI, in these subgroups. This assumption may suggest a potential concern regarding Al exposure in particular cases.

Fig. 4 presents the risk characterization of Al exposure for infants and toddlers according to the probabilistic approach. At negligeable Al concentrations in water, the dietary exposure to Al reaches up to 30 % of the PTWI or 60 % of the TWI for infants (0–12 months) and up to 28 % of the PTWI or 55 % of the TWI for toddlers (13–36 months). If water at the maximum allowed Al concentration would be used to prepare the infant and follow-up formulae, these percentages increase to almost 100 % of the TWI for infants.

It should be noted that the results reported in the present study may be underestimated due to some reasons. For children over 6 months, other foods (e.g. fruit juices, rice, beans, pasta, cassava and biscuits) are incorporated into the diet and may increase the intake of Al (Chekri et al., 2019).

Sato et al. (2014) reported that the average weekly dietary Al exposure from processed foods is estimated to be 0.86 mg/kg bw/week, considering children from 1 to 6 years old. The 95th percentile was 2.0 mg/kg bw/week, which amounted to 101 % of the PTWI. The authors supported that in order to limit Al intake in young children, especially in the population with the highest Al exposure, the Japanese government should regulate the use of food additives, such as leavening agents (including baking powder) in the manufacture of confectionaries.

In a duplicate diet sampling study considering the main meals along the day (breakfast, lunch, afternoon meal, and dinner), a total dietary intake of Al was estimated in two population groups from southern Spain (family and university students) (Cabrera-Vique and Mesías, 2013). Mean values of dietary exposures to Al were 0.34 and 0.12 mg/kg bw/week, which amounted to 17 % and 6 % of the PTWI. The results obtained in this study (scenario 1) were similar to those reported by Navarro-Blasco and Alvarez-Galindo (2003), who found mean values of 8–10 % of the PTWI for the age range of 0–6 months. In a recent study reported by Sirot et al. (2018) regarding French infant total diet, the lowest TWI of 1 mg/kg bw/week was exceeded only in high consumers of spinach (excluding jars of baby food) which displayed higher concentration than the average of vegetables. Başaran (2022) estimated mean ingestion values of 3.30 and 2.73 μg/kg bw/week for Al through the consumption of infant formulas and infant biscuits, respectively, corresponding to < 3 % and < 6 % of PTWI and TWI contribution, in the same order.

Other sources of uncertainty in the exposure estimation may result in an under- or overestimation of the risk. These uncertainties include uncertainties related to the consumption data (limited number of children; two 24 h-recalls; consumers only estimation), the food sampling (limited number of samples with regard to the high diversification of available food; absence of home-prepared meals), the analytical measurements (measurement uncertainty), the multiplication by a factor 7 to estimate the Al intake on a weekly basis, and uncertainties on the distributions applied in the probabilistic approach (models based on a limited number of data, remaining discrepancy between variation of real data and fitted models).

3.5. Contribution of selected baby foods to Al intake

Fig. 5 illustrates the average contribution of the selected baby foods

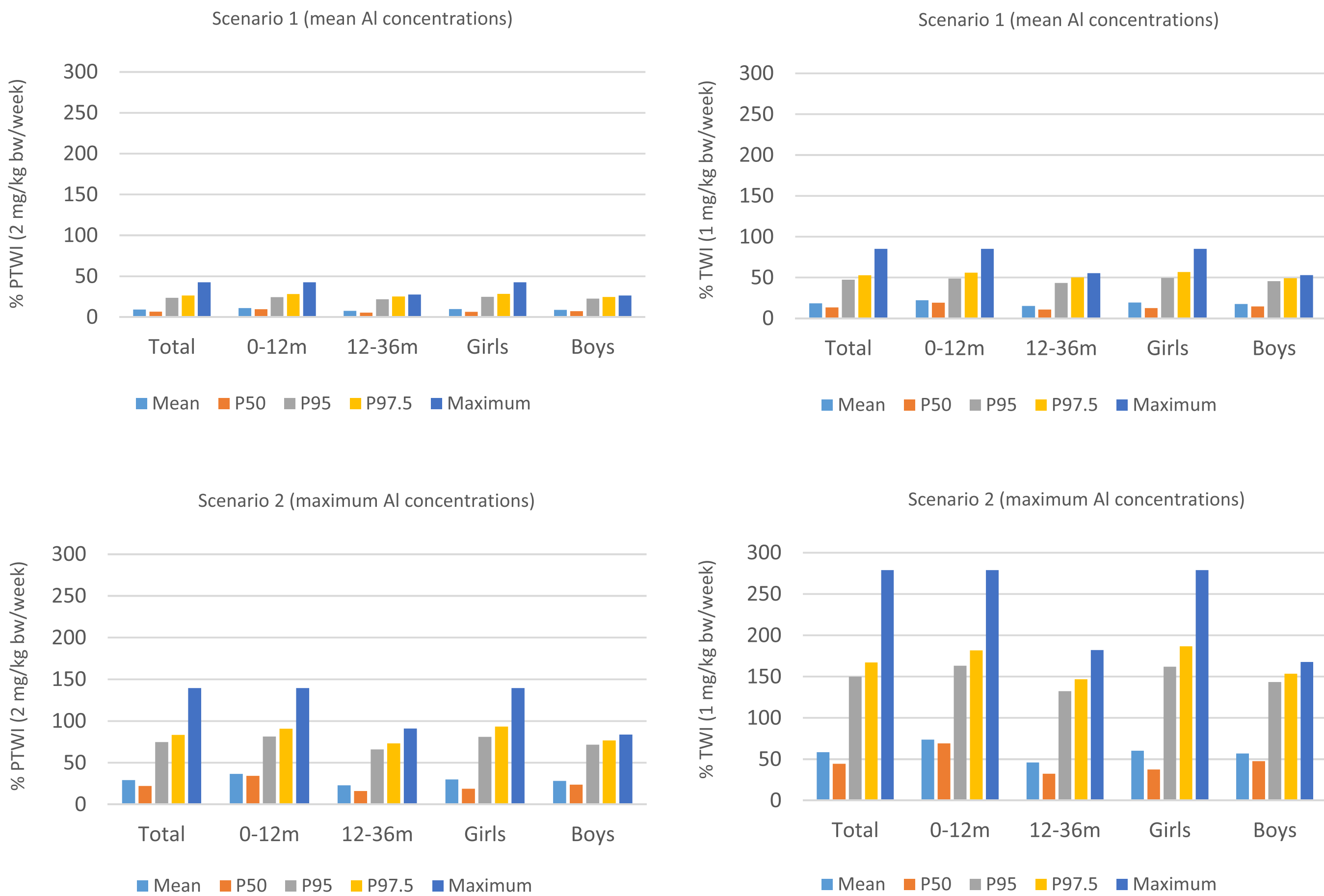

Fig. 3. Aluminium exposure (deterministic approach) in terms of % of the Provisional Tolerable Weekly Intake (PTWI) and Tolerable Weekly intake (TWI).

in relation to the total exposure to Al according to the deterministic approach.

Considering scenario 1 (mean Al concentrations), meat/vegetable purees and infant formulas were the most important source of Al in the diet, contributing up to 38 % and 36 % of the total average intake, respectively, followed by petit-suisse (15 %) and cereal (9 %). In relation to age groups, the highest contribution for infants was observed for meat/vegetables purees (55 %) and infant formulas (36 %), whilst low contributions were obtained for petit-suisse (2 %), fruit puree (3 %) and cereal (4 %). Concerning toddlers, a different profile can be seen in comparison to infants. Significant reduction of meat/vegetable (24 %) contribution occurs, whilst an increase of petit-suisse (25 %) and cereal (12 %) is observed, which is in agreement with solid food introduction to the diet. For the girl's subgroup, the highest Al contribution to the diet came from meat/vegetable purees (40 %) followed by infant formulas (32 %), whilst for boys the main contribution was observed for infant formulas (40 %). Girls' subgroup also presented more significative contribution of cereals (10 %) and fruit purees (4 %) in comparison to the boy's subgroup, with respective values of 7 % and 1 %.

The second scenario (maximum Al concentrations) presented equal contribution values for infant formulas and meat/vegetable purees (38 %), followed by petit-suisse (14 %), cereal (8 %) and fruit puree (2 %) in the total population. Main differences were observed for infant formulas increased contributions, with higher values obtained for the 13–36 months age group (39 %), which might be related to the high maximum concentration found for this food category (5940 ng/g) combined to consumption frequency and daily intake presented in the current study.

Filippini et al. (2019) estimated the dietary intake of Al in a Northern-Italy adult population, based on data from a validated food-frequency questionnaire. Median total Al intake was 4.1 mg/day and the food categories which mainly contributed to total intake were vegetables (25 %), mainly leafy vegetables (15 %) and tomatoes (4 %), followed by beverages (28.6 %), and finally cereal products (17 %).

The results of dietary Al intake estimates obtained in this study were similar to recent results from other national investigations, such as in Greece and China, although the survey measures of each investigation are unique and different. In Greece, researchers reported that the major Al contributors for adult population (no age group category) were cereal products and vegetables, corresponding to 47 % and 26 % of total Al intake, respectively (Bratakos et al., 2012). A similar study in South China showed that steamed flour products typically contain significant quantities of Al and are a major contributor to Al intake by urban residents (no age group was mentioned) with values of 53 % for wheat-made products, followed by vegetables (12 %) (Jiang et al., 2013). The Chinese study suggests that steamed flour products in South China contain food additives, including Al as leavening agents.

In general, the literature reports that cereals and cereal products, vegetables and beverages appeared to be among the highest Al contributors (Cabrera et al., 2003; Fung et al., 2009). In contrast, Biego et al. (1998) reported as main Al sources milk and dairy products followed by fish and crustaceans. Other reports denote that the major dietary Al sources include products containing Al additives (grain products, processed cheese and salt), as well as several products that are naturally high in Al (tea, herbs and spices) (Lopez et al., 2000; Saiyed and Yokel, 2005).

Pereira et al. (2020) reinforce metals, such as Al, as naturally mobile in the environment being accumulated by plants. Therefore, relevant contributors to human metals exposure through the diet might be grain and grain-based products, such as cereals, nuts and oilseeds, and vegetables and vegetable products (including fungi) (EFSA, 2015).

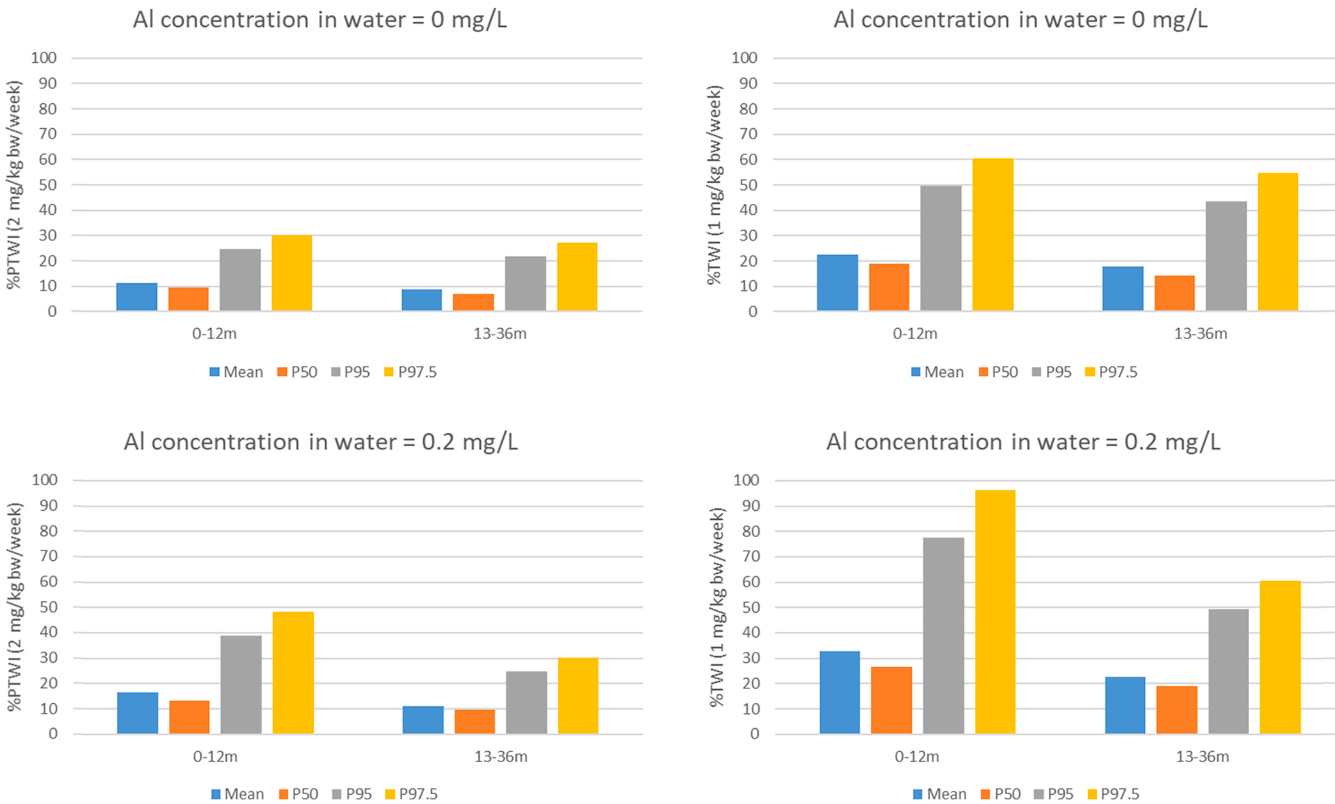

Fig. 4. Aluminium exposure (probabilistic approach) in terms of % of the Provisional Tolerable Weekly Intake (PTWI) and Tolerable Weekly intake (TWI).

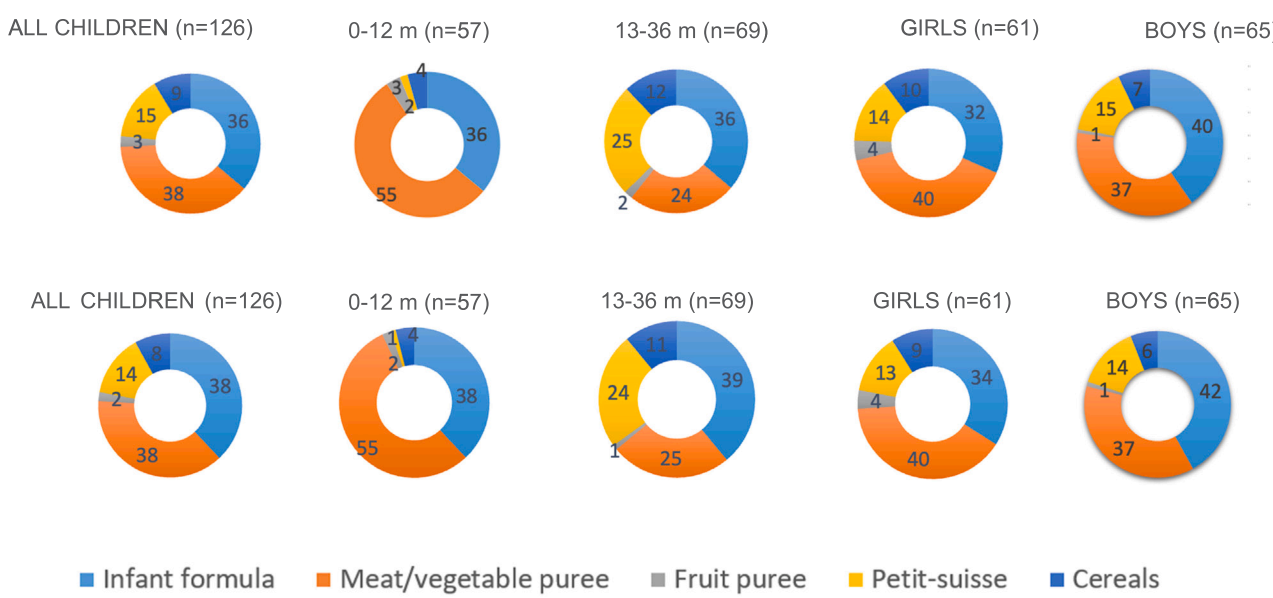

Fig. 5. Average contribution (%) of the selected baby foods in relation to the total exposure to aluminium for different scenarios. (1) mean Al concentration and (2) maximum Al concentration.

4. Conclusion

This work reports the results of young children's exposure assessment from Brazil to Al via selected baby foods. In general, the comparison values between the deterministic and probabilistic approaches corresponded well with regard to the mean intake calculation, except at high percentiles where the deterministic calculations tended to overestimate the concentrations. The risk characterization of Al exposure presented as percentage of the PTWI and TWI, also reported similar mean values for both deterministic and probabilistic methods, depending on the considered scenario. At the 97.5th percentile, the Al intake by infants aged 0–12 months is almost equal to the tolerably weekly intake derived by EFSA, and there may be a health concern for some of these children. Diversification in food consumption resulted in an increasing intake of Al. The foods that most contributed to Al exposure by infants considering mean Al concentrations were meat/vegetable purees and infant formulas. Further research is proposed in order to both compare and reinforce relevant data and methodology used in the present study.

CRediT authorship contribution statement

Esther Paiva: Writing – Original draft preparation**,** Writing – review & editing, Investigation. **Nadia Waegeneers**: Data curation. **Adriana Pavesi Arisseto-Bragotto:** Supervision, Investigation, Conceptualization. **Marcelo Antônio Morgano:** Validation, Co-Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jfca.2022.105013.

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