

Research Paper

Risk Assessment of Dietary Elemental Intakes Contributed by Commercial Baby Foods from Indian Market

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Abstract: Dietary elemental intake has received great attention as it relates to potential health effects on infants. In the present study, the analysis of branded and non-branded packaged baby foods using Inductively Coupled Plasma -Atomic Emission Spectrophotometer (ICP-AES) revealed presence of essential (Ca, Co, Cu, Cr, Fe, K, Mg, Mn, Na, Zn) and non-essential (Al, Ba, B, Li, Ni, P, S, Pb, Sc, Si, Sn, Sr, Ti) elements. Inter-elemental correlation and comparison between levels of elemental contents in milk and cereal based powders (PIF), fruit based infant formulae (FBBF) and milk based foods (MBF) were investigated. The presence of high concentrations of Ba, Ca, Mg, Mn and S in ragi-based infant formulae may be attributed to their highly significant ($p \le 0.01$ *) inter-elemental correlation. The higher average daily intakes of Cu, Mg and Mn were found to be 0.606 mg/day, 129.483 mg/day and 8.815 mg/day, respectively, however, that of Ba and P were found to be 0.461 mg/day and 322.668 mg/day, respectively, in powdered infant formulae of various brands. The higher average daily intakes of Cr (7 µg/day to 69 µg/day) and Pb (39 µg/day to 85 µg/day) were observed in baby foods which may impose risk to health of babies* when consumed frequently. Therefore, this study warrants the monitoring of higher levels of elemental contents, with respect to their recommended limits, along with fortification of essential elements in processed baby foods, in *order to provide balanced and safe nutritive diet.*

Keywords: baby foods, ICP-AES, food contaminants, inter-elemental correlation, ED-XRF, intakes. *© 2018 IJRCE. All rights reserved*

Introduction

Babies go through development stages in every aspect, including the foods they can eat. Though human milk is fundamental food for infants for the first six months of life¹, baby foods are in demand due to several reasons like lack of breast milk, demands of work, demands of urban life circumstances, social problems with breastfeeding in public and need for healthy growth of baby². For elite urban women, the readymade milk and baby food preparations from the market have become handy for infant feeding. Baby foods are available in several forms such as ready to eat, liquids, reconstituted powders and easy to cook form. Apart from breast milk, infant formulae have an important role in diets of babies as they are the major source of nutrients for infants³.

Trace elements in human body are mostly bound to proteins, forming metalloproteins which are part of

enzymatic systems, have structural and storage functions, or use the protein to be transported to their target site in the organism⁴. Trace amounts of some metals, for example, Mn, Cu, Zn, etc. are essential micro nutrients and have a variety of biochemical functions in all living organisms; however, they can be toxic when taken in excess. In addition, some metals like lead (Pb), do not occur naturally in the body, and their presence, usually as a result of occupational or pollution-related exposure, is detrimental to health⁵. As per the reports, children are more susceptible because of their greater intestinal absorption rate than adults, and a lower threshold for adverse effects⁶. Although, baby foods are major source of essential elements for infants and toddlers, the excess intake of trace elements and contaminants may pose health risk to babies. These pollutants may arise from the raw materials used in production, poor quality production processes, adulteration of infant foods and bad practices by

mothers as regards to infant formulation preparation and handling².

Multi-element surveys of baby foods have been published because of growing interest in trace element concentrations in infant foods. The concentrations of Al, Br, Ca, Cl, K, Mg and Na were determined using Instrumental Neutron Activation Analysis (INAA) in baby cereals from Ghanaian market, suggesting constant monitoring and reduction in levels of nonessential elements⁷. In one of the studies, Ni, Pb and Cd contents were determined in chocolates and candies⁸. The concentrations of all analysed elements were highest in cocoa-based chocolates followed by milk based and sugar and fruit flavour-based chocolates which was attributed to their higher contents in the raw material such as cocoa beans, cocoa solids and cocoa butter**.** In India, inadequate low daily intakes of Cu (0.183mg) and Zn (1.537mg) were reported from infant formula consumption⁶ (recommended DRI levels in a range of 0.5-1.0 mg and 3-5 mg for Cu and Zn, respectively⁹). Though the investigations have been conducted worldwide $2,3,5,7,10$ to reveal the levels of elemental contents of infant formulae and the possible health risk, there is dearth of information about inter-elemental correlations present in foods; hence we believe that present study may pave way for newer studies to understand influence of dietary elements on presence of each other. Moreover, there is scarcity of data of elemental concentrations in baby foods marketed in India, which raises concern for Indian babies. Thus, periodical evaluation of baby foods is necessary to comprehend the current scenario of the dietary intakes of elements.

In the present study, the concentrations of elements and their inter-elemental correlations were studied to understand their presence and proportionality in baby foods. The knowledge on dietary elemental intakes with respect to differences in feeding patterns, however, is still limited. Thus, the analysed significant differences in distribution of various elements, depending on their sources, will contribute to assessment of diet plans and serving size as per the requirements. The study represents current market scenario, and will be useful to further reconsider and modify levels of dietary elements. This will help to assure the reliability and safety consumption of branded and non-branded baby foods.

Material and Methods Sampling

All the 17 branded and non-branded baby foods products (four items per product type) were randomly purchased from major supermarket and local market of Mumbai, India. They were segregated into powdered into formulae (PIF – milk powder, milk and cereal based powders), fruit based baby foods (FBBF – fruit

purees and juices) and milk based foods i.e. dairy products (MBF – probiotic milk, yogurt and cheese). The baby foods under study were manufactured in various parts of India viz. Haryana, Maharashtra and Karnataka, except all the fruit based baby foods and cheese products which were imported from Australia, Bahrain, France, UK and USA and available in Indian market.

Elemental analysis of baby foods using Inductively Coupled Plasma-Atomic Emission Spectrophotometer *(ICP-AES)* **Sample preparation**

Each sample $(5 g)$ was digested (in duplicates) with 10 ml of concentrated $HNO₃$ (69%) (Suprapure, Merck) and 2 ml of H_2O_2 (30%) (Suprapure, Merck), in microwave digestion system and diluted up to 25 ml with purified distilled water. Suitable dilutions were made using distilled water for the determination of various elements. The standard solutions of elements used for calibration were produced by diluting a stock solution of 1.023g/ml (20°C) of the given element supplied by Merck (ICP Multi Element Standard Solution IV CertiPUR[®]). These standards and samples were further analysed for elemental composition using ICP-AES.

The validation of digestion method included analysis of Certified Reference Material, NCSZC73009 (GSB-2) wheat (0.5 g). Simultaneously, blank digests were prepared in triplicates, in a similar manner for analytical quality control. The operating conditions for microwave system were: 10 min for 1000W at 220°C, 20 min for 1000W at 220°C and 15 min for 0W at 35°C (Microwave Digestion System, Milestone START D).

Elemental Analysis using ICP-AES

The characteristic wavelengths with high intensity and minimum interference were selected for the elemental determination using an ICP-AES (SPECTRO Analytical Instruments GmbH, ARCOS)

Plasma parameters

Plasma Power: 1400 W; Normal speed of pump: 30 rpm*;* Nebulisation flow-rate: 0.8 l/min; Nebulisation pressure: 1.0 bar

Characteristic wavelength per element in nm

Al(176.641nm), B (249.773nm), Ba (455.404nm), Ca (422.673nm), Co (228.616nm), Cr (267.716nm), Cu (324.754nm), Fe (259.941nm), K (766.491nm), Li (670.780nm), Mg (279.079nm), Mn (257.611nm), Na (589.592nm), Ni (231.604nm), P (213.618nm), Pb (220.353nm), S (182.034nm), Si (251.612nm), Sr (460.733nm), Sn (189.991nm), Sc (361.384nm), Ti (334.941nm) and Zn (213.856nm).

Statistical Evaluation

Statistical analysis of the data was performed using both Microsoft Office Excel and IBM SPSS Statistics 21 Premium x 86 (x 32 bit) statistical software. The distributions of continuous variables in the concentrations of element in baby foods were expressed as mean $+$ standard deviation (SD). The correlation analysis was performed in between the levels of elements occurred in baby foods and their packages using non-parametric Spearman's Rank Correlation Coefficient test. The inter-elemental correlations between elements under the study were also assessed with Spearman's Rank Correlation Coefficient test using SPSS software. A probability level of <0.05 and <0.01 was regarded as 'significant' and 'highly significant', respectively Significant differences in distribution of elemental concentrations between groups viz. powdered (PIF, n=48), fruit based baby foods (FBBF, n=48) and milk products (MBF, n=32) were tested using the non-parametric Kruskal-Wallis 1-way ANOVA test¹¹. The significance of differences in daily elemental intakes was compared to their recommended limits, using non-parametric One-Sample Wilcoxon Signed Rank Test.

Results and Discussion

All the selected packaged baby foods were analyzed for their qualitative and quantitative estimation of elemental concentration. The sample codes were used to represent the data for the respective samples (Table 1).

Elemental analysis of certified reference material

The method was validated using Certified Reference Material, [NCSZC73009 (GSB-2) wheat]. The elemental analysis of reference material was satisfactorily in agreement (recovery 96-99%, as shown in Table 2) with the recommended reference values.

Dietary Elemental Contents from Types and Brands of Baby Foods

The ICP-AES analysis revealed presence of 23 elements in milk powders, milk and cereal based powdered formulae (PIF), fruit based baby foods (FBBF) and milk based food (MBF) from Indian market. The list of elements includes essential (Ca, Co, Cu, Cr, Fe, K, Mg, Mn, Na, Zn) and non-essential (Al, Ba, B, Li, Ni, P, S, Pb, Sc, Si, Sn, Sr, Ti) elements with different concentrations. The sample-wise elemental

contents for each type of packaged baby food are given in Table 3.

Levels of Elements in brands and types of Baby Foods

The non-parametric Kruskal-Wallis 1-way ANOVA statistical analysis of levels of elements showed significant difference ($p < 0.05$) in levels of Al, Ti, Ca, Mn, Ba, Mg, Cr, Ni, P and Si with their higher concentrations in non-branded baby foods (PIF 4-6, MBF 1, 4 and IF-1) compared to the branded (PIF 1-3, FBBF 1-6, MBF 2 and 3) baby foods. Non-branded milk and cereal based powdered formulae (PIF 4-6) held higher concentrations of Mn, Ba, Cr, Al, Ti and Ni than branded powdered infant formulae (PIF 1-3). Non-branded ragi-based baby foods (PIF 4-6) in particular held elevated concentrations of Mn, Ba and Mg ($p < 0.05$). Milk based powdered infant formula (PIF 1) contributed to higher content of Cu, whereas, Ca content of milk and cereal based powdered infant formulae (PIF 2-6) were significantly higher ($p < 0.05$) than plain milk powdered formula (PIF 1). The results showed that milk and cereal based baby foods are rich sources of majority of dietary elemental contents compare to fruit based and milk based baby foods which may be attributed to their uptake from soil to millets 12 . This suggests that various types of baby foods cause potential difference in concentration of elements possibly due to differences in their food sources.

The Ca contents of banana based infant formulae showed no significant difference ($p > 0.05$) in their levels to that of cereal and milk based powdered infant formulae as well as milk based dairy products. Banana based infant formulae held significantly higher ($p <$ 0.05) concentrations of Ca, Fe and S than other fruit based infant formulae. In this study, all other fruit based baby foods except banana based products were found to be poor sources of dietary elements containing significantly lower levels ($p < 0.05$) of P, Mn, S, Zn, Ba, Cu, Sr and Ca than the powdered (PIF 1-6) infant formulae.

In processed cheese products, the high contents of Al and Na were found to be sourced from approved food additives like basic sodium aluminium phosphate and sodium chloride, as reported in previous studies¹³⁻¹⁴. In the present study, milk based dairy products (MBF 1- 4) held higher concentrations ($p < 0.05$) of Al, Na and Pb compared to powdered infant formulae and fruit based baby foods (FBBF 1-6) which may be attributed to difference in processing practices.

Overall, there was significant difference ($p > 0.05$) in distribution of Mn, Ba, Sr, P, S, Zn, Na, Fe and K in powdered infant formulae (PIF 1-6), fruit based infant formulae (FBBF 1-6) and milk based foods (MBF 1-4), with higher concentrations of Mn, Ba, Sr, S, Zn, Fe and K in powdered infant formulae and higher contents of Al, Na and P in milk based foods which contributed to daily dietary elemental consumption in Indian babies. In addition to this, the levels of exposure from dietary intakes also depend upon the serving size and feeding patterns. Hence, the daily dietary elemental intakes were calculated for evaluation of the health risk due to frequent consumption of marketed baby foods.

Element	Certified value (mg/kg)	Observed value (mg/kg)	Recovery $(\%$
Al	$0.0104 + 0.001$	$0.010 + 0.057$	96.8
B	(0.55)	$0.535 + 0.01$	97.2
Ba	$2.4 + 0.3$	$2.378 + 0.25$	99.1
Ca	$0.034 + 0.002$	$0.034 + 0.01$	99.5
$\overline{\text{Cr}}$	$0.096 + 0.014$	$0.094 + 0.09$	98.3
Co	(0.008)	$0.008 + 0.001$	96.9
Cu	$2.7 + 0.2$	$2.668 + 0.021$	98.8
Fe	$18.5 + 3.1$	$18.315 + 0.96$	99
\overline{K}	$0.140 + 0.006$	$0.137 + 0.03$	97.8
Li	$0.024 + 0.005$	$0.023 + 0.005$	96.3
Mg	$0.045 + 0.007$	$0.044 + 0.02$	97.9
Mn	$5.4 + 0.3$	$5.314 + 0.98$	98.4
Na	$\frac{17+5}{2}$	$16.643 + 2.01$	97.9
Ni	$0.06 + 0.02$	$0.058 + 0.006$	96.2
\overline{P}	$0.154 + 0.007$	$0.150 + 0.05$	97.4
Pb	$0.065 + 0.024$	$0.063 + 0.001$	97
S	$0.178 + 0.017$	$0.176 + 0.09$	98.9
\overline{Si}	$0.025 + 0.003$	$0.025 + 0.013$	99.1
Sr	$0.30 + 0.05$	$0.295 + 0.09$	98.4
Sc	(2.5)	$2.443 + 0.8$	97.7
Ti	(2)	$1.938 + 0.20$	96.9
Zn	$11.6 + 0.7$	$11.530+0.89$	99.4

Table 2: Elemental analysis of Certified Reference Material

Concentration in mean value \pm SD, Data in () is for reference only

Sample Code	Al	B	Ba	Ca	$\mathbf{C}\mathbf{r}$	Co	Cu	Fe
PIF-1	\overline{ND}	ND	\overline{ND}	$31.021 + 2.38$	ND	$\overline{0.057} + 0.08$	$10.938 + 0.32$	ND
$PIF-2$	$3.128 + 0.25$	$\rm ND$	$0.284 + 0.4$	$3090.328 + 12.20$	$0.034 + 0.05$	ND	$4.191 + 0.25$	$33.902 + 2.95$
$\overline{PIF-3}$	$\frac{1.054 \pm 0.64}{1.054 \pm 0.64}$	$9.758 + 0.95$	$\overline{0.593} + 0.2$	$6383.04 + 20.77$	$0.042 + 0.05$	ND	$1.823 + 0.45$	$62.631 + 6.14$
PIF-4	$\overline{1.5}03 + 0.67$	$1.507 + 0.12$	$\sqrt{6.719} \pm 0.4$	$7069.732 + 19.22$	$0.107 + 0.09$	\overline{ND}	$2.677 + 0.79$	$46.329 + 4.47$
PIF-5	$8.933 + 1.09$	$3.251 + 0.31$	$8.493 + 0.73$	$2402.365 + 8.21$	$0.1 + 0.02$	$\frac{0.077 + 0.02}{h}$	$2.471 + 0.55$	$31.041 + 2.96$
$\overline{PIF-6}$	$4.756 + 0.33$	$1.588 + 0.67$	$\overline{0.718} \pm 0.09$	$\frac{805.498 + 5.39}{h}$	$0.296 + 0.22$	$0.085 + 0.12$	ND	$10.373 + 0.77$
FBBF-1	ND	ND	ND	$\frac{1}{5.709} + 0.13$	ND	ND	ND	ND
FBBF-2	N _D	$2.972 + 0.12$	ND	$42.004 + 2.58$	\overline{ND}	\overline{ND}	ND	\overline{ND}
FBBF-3	ND	$5.256 + 1.04$	$\rm ND$	$109.51 + 5.61$	$0.381 + 0.12$	$0.141 + 0.03$	ND	$4.516 + 0.29$
FBBF-4	$0.192 + 0.27$	$\rm ND$	$\rm ND$	$\frac{95.659 \pm 4.61}{2}$	$0.025 + 0.04$	$0.027 + 0.04$	$\frac{2.493 + 0.53}{2}$	1.015 ± 0.44
FBBF-5	$\sqrt{2.469} \pm 0.08$	ND	1.396 ± 2.73	$2444.869 + 10.76$	$1.57 + 2.12$	ND	$\overline{0.608} \pm 0.86$	51.005 ± 3.26
FBBF-6	ND	ND	ND	$99.254 + 3.83$	$0.033 + 0.07$	ND	$\overline{1.0}43 + 0.48$	$\overline{1.25} + 0.49$
$MBF-1$	$0.959 + 0.05$	$20.412 + 1.98$	N _D	593.426 \pm 9.98	$\overline{\text{ND}_{\pm}}0.08$	\overline{ND}	$2.791 + 0.95$	$0.025 + 0.57$
$MBF-2$	\overline{ND}	\overline{ND}	N _D	$945.247 + 8.91$	$ND + 0.18$	N _D	$4.408 + 0.23$	ND
MBF-3	$24.036 + 1.53$	$4.655 + 0.77$	ND	$8339.878 + 25.86$	$1.757 + 0.13$	N _D	\overline{ND}	$2.653 + 0.37$
MBF-4	\overline{ND}	ND	\overline{ND}	4489.47 ± 11.05	$0.105 + 0.15$	\overline{ND}	$\overline{6.752} + 0.55$	$0.52 + 0.74$
$IF-1$	\overline{ND}	$0.102 + 0.03$	\overline{ND}	16.194 ± 1.48	$0.012 + 0.01$	\overline{ND}	ND	ND
Sample Code	$\mathbf K$	Li	Mg	Mn	Na	Ni	${\bf P}$	Pb
PIF-1	$\frac{1104.99 + 16.79}{ }$	$\frac{3.139 + 0.39}{2}$	\overline{ND}	\overline{ND}	$544.863 + 10.9$	\overline{ND}	$\frac{211.467 + 17.0}{ }$	ND
$PIF-2$	$2484.99 + 29.54$	$1.908 + 0.70$	$465.677 + 5.85$	1.264 ± 0.78	704.313 ± 12.9	$0.05 + 0.07$	2171.32 ± 28.1	
PIF-3	5413.66 ± 32.94	$\rm ND$	$857.395 + 12.8$	2.215 ± 0.02	$\frac{2098.77}{2098.77}$ + 45.3	0.09 ± 0.02	$3292.02 + 32.0$	$0.034 + 0.004$ $0.758 + 0.049$
PIF-4	7078.36 ± 33.40	ND	1509.54 ± 30.51	$151.784 + 23.74$	2388.06 ± 30.6	0.214 ± 0.06	3731.77 ± 38.3	$\rm ND$
PIF-5	$3464.82 + 29.71$	ND	$1617.04 + 28.82$	$126.968 + 22.58$	ND	0.54 ± 0.13	$2027.44 + 24.0$	
PIF-6	$872.268 + 6.41$	\overline{ND}	$276.001 + 5.71$	$39.472 + 4.47$	\overline{ND}	$0.085 + 0.01$	$\frac{342.184 + 13.9}{ }$	$0.066 + 0.001$
FBBF-1	$123.828 + 3.88$	$0.864 + 0.13$	ND	$\rm ND$	83.416 ± 2.31	\overline{ND}	6.431 ± 0.18	$\rm ND$
FBBF-2	$417.45 + 4.42$	$\rm ND$	\overline{ND}	\overline{ND}	$\sqrt{126.451}$ + 9.68	$\overline{0.054 + 0.01}$	$\frac{37.913 + 0.65}{2}$	ND
FBBF-3	$1148.75 + 21.54$	\overline{ND}	$\frac{387.838 + 11.35}{ }$	$1.44 + 0.59$	ND	$0.386 + 0.06$	$351.251 + 9.0$	$0.519 + 0.02$ $2.238 + 0.02$
FBBF-4	$1494.04 + 26.12$	$0.529 + 0.75$	$140.858 + 19.2$	ND	\overline{ND}	$\overline{0.07} \pm 0.1$	$216.482 + 12.7$	$0.094 + 0.003$
FBBF-5	3460.45 ± 35.37	ND	625.746 ± 23.43	${\rm ND}$	$\overline{956.541}$ + 11.7	0.32 ± 0.31	1132.25 ± 35.5	$\rm ND$
FBBF-6	1219.84 ± 18.59	ND	$110.659 + 11.76$	ND	$12.072 + 0.93$	$0.035 + 0.04$	144.212 ± 5.7	0.033 ± 0.005
$MBF-1$	$540.772 + 19.90$	\overline{ND}	$49.558 + 10.09$	ND	$279.461 + 4.4$	0.036 ± 0.04	$328.087 + 6.3$	0.819 ± 0.097
$MBF-2$	$1047.18 + 18.0$	$\overline{0.878 + 1.24}$	$98.977 + 13.97$	ND	$\frac{315.152 + 5.3}{ }$	$\rm ND$	$610.97 + 13.9$	0.534 ± 0.006
MBF-3	$2469.67 + 23.50$	ND	344.517 ± 21.18	ND	$7149.59 + 23.0$	$\overline{0.092 \pm 0.01}$	6904.75 ± 12.1	$0.267 + 0.038$
MBF-4 $IF-1$	$504.759 + 15.49$ 6.753 ± 0.13	$\frac{3.078 + 5.34}{ }$ $\rm ND$	148.236 ± 10.96 ${\rm ND}$	${\rm ND}$ ND	$\sqrt{6343.23} \pm 21.66$ $\overline{2024.35}$ + 6.5	$0.032 + 0.04$ $\rm ND$	$3533.35 + 32.2$ $0.367 + 0.01$	1.796 ± 0.091 $\rm ND$

Table 3: Elemental concentrations (mg kg-1) in baby foods acquired in Indian market and analyzed herein using ICP-AES analysis

Standard Deviation + , ND- Not Detectable Daily intakes of elements from baby foods

There are no specific guidelines available to determine the food consumption pattern in infants as there is significant variation in dietary intake of babies during the growth period at age up to 3 years. Hence, it is difficult to estimate reliably the levels of dietary elemental intakes with the right approach which considers any contribution to exposure from breast milk or from other foods that may be given from weaning onwards. In earlier studies, it was reported that a 6-12 months old baby would require approximately 5 kg of powdered infant formula every month and in a year around 60 kg^6 . Thus, on the basis of the above study, the daily consumable weight of infant powdered formula was calculated to be 164.4g of infant formula powders. This approach was considered to evaluate the daily elemental exposure for dry powdered infant formulae in the present study. The studies reported that consumption of fruit juices in European infants and

toddlers was found to be on an average $148 \text{ ml/day}^{15-17}$. As per the available guidelines, fruit juice intake should be limited to 4 to 6 oz $(113.39 \text{ g to } 170.09 \text{ g})$ a day until 12 months of age¹⁸. Thus, a daily elemental intake from 170.09 g of fruit-based baby foods was compared to assess the health risk. Also, the approximate daily elemental exposure was assumed on the basis of the one time serving of 100 ml of probiotic milk, 80 g of yogurt and 15 g of cheese, for consumption of milk-based foods, considering the dairy food portion size to fulfill the requirement of Ca in babies as per the experts opinion, The Infant and Toddler Forum, UK, in absence of relevant Indian guidelines on serving size¹⁹. The average daily intakes of essential and non-essential elements from three types of baby foods (PIF, FBBF and MBF) were compared with their recommended safety guidelines (Table 4).

		This Study				
Elements	Powdered Infant Formulae (PIF) in mg/day	Fruit based Baby Foods (FBBF) in mg/day	Milk based foods (MBF) in mg/day	Calculated safety limits in mg/day for 6 months baby (wt 6kg)	Safety guidelines	References
\mathbf{A}	0.488	0.109	0.118	6	UL: 6 mg/day	$\overline{21}$
\mathbf{B}	0.662	0.407	0.531	3	3 mg/day (for age 1-3 yrs)	$\boldsymbol{9}$
Ba	0.461	0.057	ND	$\overline{0.3}$	TDI: 0.05 mg/kg bw/day	$\overline{33}$
Ca	541.601	95.103	81.851	$1,000-1,500$	UL: 1,000mg/day (0-6 months, 1,500 mg/day (7-12 months, 2500 mg/day (1-3 yrs)	$\overline{31}$
Cr	0.016	0.069	0.007	$0.002 - 0.006$	AI*: 0.2 μ g/day (0 to 6 months), 5.5 μ g/day (6 to 12 months), 11 µg/day (1-3 yrs); TDI: ND	$\boldsymbol{9}$
Co	0.006	0.006	ND	ND^*	ND^*	
Cu	0.606	0.141	0.183	$0.2 - 0.22$	AI*: 0.2 mg/day (0 to 6 months), 0.22 mg/day (6 to 12 months), RDA: 0.34 mg/day (1-3 yrs), UL: 1 mg/day (1-3 yrs)	$\boldsymbol{9}$
Fe	5.049	1.966	0.017	(a) $0.27 - 11$; (b) $5.4 - 7.8$	(a) AI^* : 0.27 mg/day (0 to 6 months), RDA: 11 mg/day (6 to 12 months); 7 mg/day (1-3 yrs); UL: 40 mg/day (b) $0.9-1.3$ mg/day $(6-12$ months)	(a) 9; (b) 27
$\mathbf K$	559.483	249.127	45.617	400-700	AI*: 0.4 g/day (0 to 6 months), 0.7 g/day (6 to 12 months), 3.0 g/day $(1-3$ yrs)	39
Li	0.138	0.033	0.032	0.086	Provisional RDI of 1.0 mg/day for a 70kg adult i.e. 14.3 µg/kg bw/day	$\overline{22}$
Mg	129.483	43.038	5.066	$30-75$	AI*: 30 mg/day (0 to 6 months), 75 mg/day (6 to 12 months); RDA: 80 mg/day (1-3 yrs); UL: 65 mg/day (1-3 yrs)	$\overline{31}$
Mn	8.815	0.049	ND	$0.003 - 0.6$	AI*:0.003 mg/day (0 to 6 months), 0.6 mg/day (6 to 12 months), 1.2 mg/day (1-3 yrs); UL: 2 mg/day (1-3 yrs)	9
Na	162.124	37.292	63.888	120-370	AI*: 0.12 g/day (0 to 6 months), 0.37 g/day (6 to 12 months), 1.0 g/day (1-3 yrs); UL: 1.5 g/day (1-3 yrs)	39
Ni	0.027	0.029	0.001	0.2	UL: 0.2 mg/day (for age1-3 yrs)	9
$\overline{\mathbf{P}}$	322.668	63.493	59.564	100-275	AI*: 100 mg/day (0 to 6 months), 275 mg/day (6 to 12 months); RDA: 460 mg/day (1-3 yrs); UL: 3 g/day (1-3 yrs)	39

Table 4: Average daily dietary elemental intake from various groups of baby foods and their safety guidelines

ND – Not Detectable; ND-Not Determinable due to lack of scientific data; RDA- Recommended Dietary Allowance; AI*- Adequate Intake; TDI- Tolerable Daily Intake; UL- Tolerable Upper Intake Level*

Aluminum (Al), Lithium (Li) and Lead (Pb)

There is a concern for the possibility of increased amounts of Al in body resulting high risk of brain dysfunction and bone disorders^{2, 20}. According to several authors, the Al dietary intake must not exceed 6mg/day to avoid potentially toxic levels²¹. Herein, the higher concentration of Al i.e. 24.036 mg/kg (Table 3) was detected in processed cheese (MBF-3), which can be due to leaching of Al into foods during processing of milk. However, the daily intake of Al from cheese was less, due to its smaller serving size i.e. 15 $g/day¹⁹$. The daily intake of Al from various groups of baby foods was found to be 12-50 folds lower than the tolerance limits (Table 4). Lithium is a non-essential element. Upon oral intake, metallic Li may cause speech impairment and damage on central nervous system. Chemically, Li resembles Na but is more toxic with recommended dietary intake as low as 1.0 mg/day for a 70kg adult i.e. 14.3 μ g/kg bw/day²². Accordingly, the recommended dietary intake of Li was derived as 0.086 mg/day for a baby with 6 kg of body weight. In present study of food products intended for infants, the concentration of Li was found to be in the range of $0.529 - 3.139$ mg/kg. It was detected in low concentrations in fruit based (FBBF 1-6, average daily intake 0.033 mg/day) and milk based products (MBF 1-4, average daily intake 0.032 mg/day). The higher concentrations of Li was found to be present in milk and cereal based products (PIF 2-6, average daily intake 0.138 mg/day) of various brands which was comparable to the derived recommended dietary intake of Li in babies.

In another study**,** high negative association was demonstrated between Pb exposure and children's intelligence quotient²³. The present assessment of Pb raises a potential concern for effects on neurodevelopment in infants with an exposure level of 0.033 mg/kg to 2.238mg/kg. The highest concentration of Pb

was detected in branded organic fruit puree FBBF-3 (2.238 mg/kg). Moreover, Pb was found to be present in 65% of overall baby foods. The average daily intakes of Pb for all types of baby foods (PIF 1 to 6, 0.048 mg/day; FBBF 1 to 6, 0.085 mg/day and MBF 1 to 4, 0.039 mg/day) were present 2-4 folds higher than recommended tolerable levels (3.57µg/kg of bw/day i.e. 0.021 mg/day for a baby of average 6 kg of body weight 24) which may impose a potential health risk.

Copper (Cu), Zinc (Zn) and Iron (Fe)

Copper (Cu), zinc (Zn) and iron (Fe) are essential nutrients required for rapid growth and brain development of infants, but chronic metabolic disturbances may result from excess or deficiency of these metals. An excessive intake of Cu has an irritating effect on the gastrointestinal tract and has previously been used to induce vomiting in the event of poisoning in children²⁵. The daily adequate intake of Cu was reported to be 0.2-0.22 mg/day in 0-12 months old babies, whereas the recommended dietary allowance of Cu was noted as 0.34 mg/day with upper limit of 1 mg/day for 1-3 yrs age group of babies⁹. In the present research work, Cu was detected in most of the samples except in fruit juices and gripe water, ranging 0.608 – 10.938mg/kg. The presence of Cu in fruit based and milk based baby foods (0.141 mg/day and 0.183 mg/day, respectively) were found to be equivalent to their daily adequate intakes (i.e. 0.2-0.22 mg/day). Cu was found to be present with highest concentration i.e. 10.938mg/kg in branded milk powdered infant formula and may contribute to higher daily intake of Cu (0.606mg/day) which is close (but lower) to its tolerable upper intake (i.e. 1mg/day). The risk of high Cu exposures via branded milk powdered formula in infancy raises health concern. There was a high variability in the Cu content of different baby foods which may be attributed to the types of food and the places of cultivation, season and the processing techniques.

Deficiency of Zn is associated with dysfunction of the immune system, growth retardation, diarrhea and acute respiratory infections²⁶. The concentrations of Zn were detected in the range of 0.33 – 49.49mg/kg. The tolerable upper levels of Zn range from $4\text{-}7\text{mg/day}^9$. The daily intake of Zn was adequate in powdered formulae (3.407mg/day), whereas, it was found to be considerably low in fruit based baby foods and milk based foods (0.743mg/day and 0.140mg/day respectively).

Iron is essential in oxygen supply as a component of hemoglobin and for oxygen storage as a component of myoglobin. Full term, normal birth weight infants below 6 months do not generally need any Fe in addition to the amounts provided by human milk. The newborn infant has a high blood concentration of hemoglobin, which declines during the first few weeks of life 27 . At around 6 months, additional intake beyond what is available in breast milk becomes necessary²⁸. The daily intakes of Fe $(0.017 - 5.049 \text{mg/day})$ in baby foods were lower than the recommended dietary allowances⁹ (7-11 mg/day) in all the brands. The values for estimated average daily intakes of Fe (5.049mg/day) in powdered infant formulae (PIF 1-6) were found to be lower, however in good agreement with the recommendations (0.9-1.3 mg/kg bw/day for 6-12 months i.e. 5.4-7.8 mg/day for a baby of average 6 kg body weight) suggested by previous study²⁹. Thus, the infants consuming powdered infant formulae as the sole source of nutrition may reach an intake of 5 mg/day Fe from formulas, complying with Fe content regulations, whereas, milk based (1.966 mg/day) and fruit based baby food (0.017 mg/day) products found to be poor source of Fe content.

Manganese (Mn) and Magnesium (Mg)

According to a recent literature search, the risk of development of neurotoxicity due to excess of Mn is considered to be higher in infants than in adults²⁸. In this assessment of food for infants, Mn was found in a variety of brands with concentrations varying considerably, from $1.26 - 151.78$ mg/kg. The highest contribution in the total daily supply came from ragibased non-branded organic fruit mixed powdered formulae (PIF 4, 5 and 6; 39.47 mg/kg to 151.78 mg/kg). The estimated daily intakes of Mn in reconstitutable powdered infant formulae were 8.815mg/day (Table 4), which were around 15 folds higher than the recommended tolerable limits (i.e. 0.6mg/day) raising health concern if consumed frequently.

Magnesium plays a role in regulating potassium fluxes and in the metabolism of Ca. It has not been shown to produce toxic effects when ingested as naturally occurring Mg in food³⁰. Diarrhoea was selected as the critical endpoint as it is the first sign of excess intake.

In this study, Mg was detected in high concentrations (5.70- 8339.88mg/kg), in most of the branded and nonbranded baby foods, except in fruit juices, gripe water and powdered milk formula. The recommended adequate intake of Mg for 0 -12 months age group babies is 30-75 mg/day, whereas, the upper limit is 65 mg/kg for babies of $1-3$ years³¹. The estimated average daily intakes of Mg in powdered infant formulae studied here was 129.483mg/day which is beyond the suggested upper limits.

Ragi (also known as Finger Millet/ *Eleusinecoracana*/ Nachani Satva) is popular and major crop in India and used for infants and health foods both in native and malted conditions 32 . Manganese is an essential nutrient, necessary as a cofactor for several enzymes and important for the normal development of infants 28 . The present study revealed major contents of Mn and Mg in ragi-based formulae which may be attributed to the uptake of these elements from soil to ragi crop. Though Mn and Mg are essential nutrients; it is, however, questionable whether a 2-4 fold higher daily intake of Mg and 10-15 fold higher dietary intake of Mn from baby foods are justifiable.

Barium (Ba) and Strontium (Sr)

Ba and Sr are non-essential elements for human body. Sr content was found in very low concentrations in dry powdered (average daily intake 0.603 mg/day) and fruit based baby foods (average daily intake 0.059 mg/day), whereas it was not detectable in milk based foods. There is lack of data for tolerable recommended limit of dietary intake of Sr.

Ba was detected mainly in cereal and milk based baby foods and banana based fruit puree with concentration ranging from $0.29 - 8.5$ mg/kg. The daily intake of Ba was detected in higher concentrations in powdered infant formulae (0.461mg/day) which was mainly contributed from non-branded ragi-based powdered formulae and found to be beyond its tolerable dietary intake (i.e. 0.05 mg/kgbw/day)³³. The bio-accumulation of Ba in plants has been reported earlier³⁴. Therefore, the higher concentrations of Ba in cereal based and banana based baby foods (Table 3), may be attributed to their uptake from soil to plants. Our study suggests that efforts by baby food manufacturers are required to reduce the dietary intake of Ba which is non-essential for babies health.

Tin (Sn) and Nickel (Ni)

The toxicity of these heavy metals can lower energy levels and damage the functioning of the brain, lungs, kidney, liver, blood composition and other important organs 35 . In the present study, Sn and Ni were found to be present at low concentrations $(0.007 - 0.743 \text{mg/kg})$ and 0.03- 0.54mg/kg respectively) in various baby foods. Branded powdered milk formula (PIF-1) was

detected with highest concentration of Sn (0.743 mg/kg). The dietary allowance of Sn is reported as 50 mg/kg^{36} . No reports are available for Dietary Reference Intakes (DRI) of Sn in dried and powdered infant formulae.

Nickel-sensitive individuals and children are prone to dermatitis due to high intake of $Ni³⁷$. The average daily intakes of Ni in present study $(0.001 - 0.029 \text{ mg/day})$ were found significantly below the recommended limit $(0.2 \text{ mg/day})^9$.

Cobalt (Co) and Chromium (Cr)

Cobalt and Chromium were found in very low concentrations i.e. $0.03 - 0.14$ mg/kg and $0.01 - 1.76$ mg/kg respectively. The adequate intake of Cr ranges from 0.2 μ g/day to 11 μ g/day⁹. The present study showed that the exposure to Cr from processed cheese (1.757mg/kg) was fairly high, whereas the calculated average daily intake of Cr from powdered infant formulae and fruit based baby foods raises concern as it was found to be 2-12 folds higher than the recommended dietary intakes (Table 4).

Phosphorus (P), Sulphur (S) and Silicon (Si)

The average concentrations of non-essential elements P, S and Si were found to be in the range of 0.37 – 6904.75 mg/kg, 11.44 – 594.35 mg/kg and 6.36 – 92.43 mg/kg respectively. The presence of highest concentrations of Si and P were observed in probiotic milk MBF-1 (92.43mg/kg) and processed cheese MBF-3 (6904.75mg/kg), respectively. Phosphorus is a vital ingredient, combined in mineral fertilizers with sulphuric acid, nitrogen and potassium³⁸. Hence, higher daily intakes of P and S in cereal and milk based powdered formulae (322.67 mg/day and 52.91 mg/day, Table 4) may be sourced from soil and use of fertilizers during crop growth. The regular consumption of powdered infant formulae may cause increased burden of P beyond its recommended intakes (100-275 mg/day for 0-12 months of age group) in babies³⁹. The other non-essential elements Scandium (Sc) and Titanium (Ti) were detected in low concentrations ranging from $0.03 - 4.85$ mg/kg and $0.02 - 0.39$ mg/kg, respectively in all types of baby foods.

Sodium (Na), Calcium (Ca) and Potassium (K)

Sodium is an essential nutrient at all stages of life. The level of Na contained in breast milk is adequate to maintain health for infants. In present work, the concentrations of Na in baby foods were found within the recommended limits (Table 4), however, in nonbranded cheese (MBF-3) sample it was detected up to 7,149.59mg/kg which is generally fed to babies after 6 months of age. According to the present intake assessment, the daily intake of Na from branded powdered formulae (183.467mg/day) was measured higher in concentrations ($p < 0.05$) than in non-branded infant powdered formulae (130.866mg/day). On average, the daily intake of Na was found to be adequate in powdered based and milk based (162.124 mg/day and 63.888 mg/day, respectively, Table 4) baby foods.

Calcium is important for structure of bones and teeth. However, higher levels of Ca lead to hypercalcaemia and the formation of kidney stones 40 . Potassium plays an important role in regulating blood flow and blood pressure. However, excessive intake of K may lead to hyperkalemia in low birth weight infants^{$41-42$}. In this study, Ca and K were detected in high concentrations i.e. 49.56 – 1617.04mg/kg and 6.75 – 7078.36mg/kg respectively, in almost all the branded and non-branded baby foods. Calcium was excessively present in milk based branded and non-branded reconstitutable baby foods, banana pulp and cheese samples. However, as shown in Table 4, the average daily intakes of Ca in fruit based baby foods and milk based products (95.103 mg/day and 81.851mg/day, respectively) were found to be significantly less ($p < 0.05$) than the dietary reference intakes (1000 mg/day), however milk and cereal based powdered infant formulae showed adequate daily intake of Ca (average 541.60 mg/day, p > 0.05) compared to remaining product types.

K to Na and Ca to P ratios in baby foods

Sodium (Na) and Potassium (K) are essential nutrients. Excess consumption of Na and inadequate K is one of the major factors contributing to hypertension and related diseases. K:Na ratio is an important factor in cardiovascular disease and mortality⁴³. Though the optimal Na to K ratio has not been derived by WHO, an increased potassium intake and decreased sodium intake is strongly recommended⁴⁴. Figure 1 shows the comparative levels of daily intakes of Na and K of the investigated samples. The plot indicates favourable higher daily intakes K than that of Na in milk and cereal based powdered baby foods (PIF 1-6) and fruit based baby foods (FBBF 1-6) except for processed cheese (MBF-3 and MBF-4). These findings are in agreement with the recent study⁴³. Overall, K:Na ratio was found to be favourable in 82% of the analysed baby foods like powdered infant formulae and fruit based products. Whereas, processed cheese products and gripe water showed excessive Na content.

PIF: Powdered Infant Formulae, FBBF: Fruit Based Infant Formulae, MBF: Milk Based Foods, IF: Infant Formula; Daily Recommended Intake of K for infants (400-700 mg/day); Daily Recommended Intake of Na for infants (120-370mg/day)

PIF: Powdered Infant Formulae, FBBF: Fruit Based Infant Formulae, MBF: Milk Based Foods, IF: Infant Formula; Daily Recommended Intake of Ca for infants (1,000 - 1,500 mg/day); Daily Recommended Intake of P for infants (275mg/day)

Figure 2: Calcium (Ca) and Phosphorus (P) daily intakes in mg/day (mean + SD) in baby foods

A high phosphorous intake had been shown to cause secondary hyperparathyroidism and bone loss in several animal models⁴⁵⁻⁴⁶. The low ratio of Ca to P in diet causes reduction in absorption of Ca and the desired ratio of Ca to P in infant milk powder should be in the range $1.2 - 2.0^{47}$. Herein, we found Ca to P ratios in the range of 0.07 - 2.35, which may be a cause of health concern in babies. A low Ca:P ratio (0.07 – 0.89) was detected in plain milk based powdered formulae (PIF-1) and fruit based baby foods (FBBF-1, 3, 4 and 6) except in banana based baby foods (FBBF-

5). The Ca:P ratio was found to be in the desired range in 53% of the analysed baby foods.

The study of inter-elemental correlation in baby foods

The correlation test was performed to investigate the inter-elemental correlation in examined baby foods. The data obtained from elemental analysis was subjected to statistical analysis and correlation matrices were produced to examine the inter-relationship between the investigated elemental concentrations. The values of correlation coefficients between elemental concentrations are depicted in Figure 3.

Figure 3: Inter-elemental correlation in baby foods acquired in Indian market and analyzed herein

The correlations between the major elements Ca-P, Ca-K, Ca-Mg, Ca-S, K-Mg, K-P, K-S, Mg-Mn and Mg-S were highly significant at the 0.05 and 0.01 levels (r values > 0.721), whereas correlations Ca-Na and K-Mn were significant at middle level (r values > 0.542). A highly significant correlation was observed between occurrences of Si and B (r value $= 0.717$). However, Si and B were significantly but moderately correlated (r values < 0.417) with other elements (Si-Co, Si-Fe, Si-K, B-Ca, B-Fe, B-Ni and B-Sr) in various types of baby foods. Lithium was detected in 35% of analyzed baby foods. A highly significant correlation was observed between Li and Cu at $p < 0.01$ level (r value $=0.643$). Highly significant negative correlations of Li with Al, B, Ca, Cr, Fe, Mg, Ni and Ti, at $p < 0.01$ (r value > 0.507) were observed in various types of baby foods.

There is paucity of earlier literature particularly on inter-elemental correlations in infant formulae to compare the present findings. However, researchers investigated the elemental correlation in maternal blood, cord blood and breast milk⁴⁸⁻⁴⁹. Though Al was detected in trace amounts, it showed highly significant correlations between concentrations of Al and, Ca, Mg and Fe in baby foods (r value > 0.681 , at $p < 0.01$). These pairing of elements may promote contamination of Al from environmental food contact sources.

The presence of high concentrations of Ba, Ca, Mg, Mn and S were observed mainly in ragi-based and other cereal based powdered infant formulae which showed highly significant correlations ($p > 0.01$) between Ba and Ca ($r = 0.505$), Ba and Mg ($r = 0.734$), Ba and Mn ($r = 0.848$) and, Ba and S ($r = 0.618$), as depicted in Fig. 3. The rate of transportation of Ba is soil is dependent on soil properties including high cation exchange capacity, and the presence of sulfate, carbonate, and metal oxides (e.g. oxides of Al, Mn, Si and Ti ³⁴. Thus, higher concentrations of Ba, Ca, Mg, Mn and S in ragi-based and banana-based baby foods (Table 3), may be attributed to availability of these elements in soil and their uptake from soil to plants which may be influenced by their inter-elemental correlations. The inhibitory effect of Ni upon intestinal absorption of Fe, is chemically closely related to Co^{50} . In this study, the significant correlation was observed between Ni and Co (r value $= 0.408$) in baby foods, which may attribute to simultaneous presence of these elements in babies' diet, resulting in further reduction in absorption of Fe in babies.

In one of the studies, significant correlations were reported between Cu and Ca, Cu and Mg, Zn and Mg, Ca and Cd in human breast milk samples⁴⁸; whereas, present research work depicted highly significant correlation only between Zn and Mg (r value $= 0.622$, $p < 0.01$) and weak negative correlations between Cu

and Ca (r value $= -0.246$), Cu and Mg (r value $= -1$ 0.165) from analysis of baby foods. Although phosphorus influences Ca uptake and thus Ca:P ratio is important in bone mineralization and Ca excretion in kidney, no such correlation was reported in analysis of breast milk⁴⁸. On the contrary, the present study demonstrated the highly significant correlation (r value $= 0.858$) between Ca and P in baby foods which may be attributed to varied concentration of these elements from diverse sources.

The increased potassium intake and decreased sodium intake is strongly recommended, as K:Na ratio is an important factor in cardiovascular disease and mortality⁴³⁻⁴⁴. The present study indicated moderate significant correlation (r value = 0.359 , p < 0.05) between K and Na which may help in maintaining their ratio in baby foods for health benefits in babies. The level of significance in inter-elemental correlations may be useful to justify differences in proportionality of dietary elements and to monitor concentrations of these elements by recognizing their potential sources. However, further studies are required to evaluate reliability of these inter-elemental interactions and their application in balancing nutritive value of baby foods.

Conclusion

The analysis of baby foods from Indian market shows that the samples are adequate in some essential elements (Cu, Fe, K, Mg and Na) as well as some of the non-essential trace elements (Al, B, Li, P, S, Si and Sn). The concentrations of the essential trace elements were within the limit specified in international guidelines. Non-branded baby foods were detected with higher concentration of Al, Ba, Ca, Cr, Mg, Mn, Ni, P, Si and Ti. The study indicated the possible health risk as the assessment of Ba, Cr, Cu, Li, Mg, Mn and Pb concentrations were found to be close to or higher than their recommended regulatory thresholds. The dietary intakes of these elements were observed to be higher than the specified upper limits; however, the results must be interpreted with caution due to sample size and to variability in baby food consumptions. The remarkably higher concentrations of Ba, Ca, Mg, Mn and S in ragi-based infant formulae may be attributed to their highly significant inter elemental correlations $(p<0.01)$ and their probable uptake from soil to millets. The contents of K and Na showed significant correlation ($p<0.05$), and highly significant correlation was observed between Ca and P ($p<0.01$). Dietary ratio Ca-P and K-Na ratio were found to be favourable in 53% and 82% of the analysed baby foods, respectively.

Thus, the present study contributes a current scenario of elemental exposure via baby foods with representative wide range of foods for infants from Indian market. Milk and cereals based powdered baby foods were found to be rich source of dietary elements compare to fruit based baby foods and milk products. Further studies are necessary in order to provide balanced and nutritive diets for babies based on their requirements. The awareness regarding dietary intakes of elements should be disseminated among parents and society which will help in healthy growth of babies.

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