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Determination of Metals in Children's Plastic Toys using Inductively Coupled Plasma Optical Emission Spectrometry

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Children's health and safety may be compromised if exposed to metals through contaminated toys. Hence, evaluation of the quality of the toys available in markets is essential to protect children. This study undertaken to identify the presence of metals contents in plastic toys and compares the results with the national and international limits. For this purpose, 56 types of popular children's toys marked were purchased from the UAE markets and the metal contents were estimated in 156 toy samples by inductively coupled plasma-optical emission spectrometry (ICP-OES) technique. The results reveal that Ti, Fe, Zn and Mg were present in all types of toys. At the same time, other metals such as K and Na in 54 types of toys, Cr in 51 types of toys, Al in 50 types of toys, B, Ba and Ca in types of 49 toys, Mn in 41 types of toys, Cd in 40 types of toys, Ag in 39 types of toys, Ni in 25 types of toys, Pd in 23 types of toys, Cu in 18 types of toys, Bi in 4 types of toys and Co in 3 types of toys were also detected. Furthermore, Pd and B concentrations in 3 types of toys, while Zn and Ba concentrations in two types of toys exceeded the permissible limits. Toxic stabilizers and colours added to PVC during manufacturing are responsible for the existence of such elements in plastic toys. However, additional research is required to establish the role of metals in toys, identify potential risks to children's health and propose regulations.

Keywords: Children, Metals, Plastic toys, ICP-OES.

INTRODUCTION

Toxic chemicals in consumer products can have a significant impact on the health of consumers, especially young children who are more vulnerable to exposure due to their physiological and developmental characteristics [1-4]. Children who are exposed to metals run a considerable danger of health problems as different metals have different negative consequences. For example, lead exposure, even at low concentrations, can harm the developing neurological system and lead to behavioural problems [5]. Mercury exposure can also affect cognitive function [6], whereas arsenic exposure may raise the risk of cancer and have an adverse effect on the immune system [7]. Similarly, other toxic elements such as cadmium, chromium, nickel, excessive copper and manganese, *etc*. are associated with various disorders and can also result in excessive damage due to oxidative stress induced by free radical formation [8,9]. These detrimental impacts underscore how vital it is to keep children safe from hazardous metal exposure by enforcing safety regulations, keeping an eye on things and taking steps to reduce the dangers [3,4].

Children are exposed to metals in several manners, including through the environments in which they consume food, air, soil and water. However, hazardous toys should be considered as a way for kids to get exposed to dangerous metals. Children begin to play with a collection of little toys from early on in life and exposure to such toys, children's health could be compromised [10]. It has been reported that certain metals are added during the manufacturing process to provide or improve the softness, brightness, stability and flexibility of the final product to make it more appealing to kids of all ages. These toxic chemicals are already present in contaminated recycled material or plastics materials used to manufacture mostly low-priced consumer goods and products [11]. The most common causes of hazardous metal contamination in children's toys are the use of metals as plastic stabilizers during manufacture,

the use of paint with metal pigment in toys and the use of contaminated recycled plastic, electronic trash or metals in toy production [12,13].

Several studies [14-17] have already documented that toys are contaminated with many toxic metals such as lead (Pd), chromium (Cr), arsenic (As), copper (Cu), nickel (Ni), barium (Ba), cadmium (Cd), zinc (Zn), manganese (Mg), mercury (Hg) and selenium (Se) in various concentrations. As a result, millions of toys have been withdrawn from markets. In June 2010, 12 million drinking cups sold at McDonald's were recollected because the colour coatings contained high levels of cadmium [18]. The U.S. CPSC data indicates that between 2007 and 2018, over 18 million toys were removed from stores due to high palladium contents [13,18,19]. Regarding toy regulations, the U.S. and other countries have issued guidelines and enacted policies to prohibit the addition of toxic elements in children's toys. However, due to the continuation of the non-compliance with such regulations, more toys are being removed from the stores [3,20-22]. Approximately 87% of the toys available in the UAE market are sourced from China. Therefore, this study aimed to assess qualitatively and quantitatively the presence of metals in plastic toys using inductively coupled plasma and optical emission spectrometry (ICP-OES) and to compare the results with the limits recommended by the European Commission [23] and the American Society for Testing and Materials (ASTM) [24] limits approved by the Gulf Standardization Organization [25].

EXPERIMENTAL

Toy samples collection: Different colours of heterogeneous plastic toys (n = 56) were purchased from the UAE local markets, out of which 54 were marked as "Made in China". The toys for ages group 0-10 were randomly bought from the low-cost shops and vendors. The toy prices ranged from less than 2 USD (from low-cost shops) to more than 5 USD (from high-cost shops). The toys were divided into five groups based on their use: teething-related group; figures-related group; cars and trains/sound-generating related group; and games & puzzles related group [26]. To prevent the contamination of trace elements, any materials used in the laboratory methods were washed with dilute HNO_3 (10%). Duplicate analyses in digestion and blanks were used as quality assurance/quality control procedures as reported earlier [10].

Sample preparation: A stratified sampling approach was adopted to ensure accuracy and reliability because samples were mostly heterogeneous in nature (Fig. 1). Initially, each toy was visually evaluated to identify the probable places with a higher metal concentration, including painted or metallic surfaces or soldered parts. Due to the higher probability of hazardous metals being present, these specific places were selected for the purpose of collecting samples. Multiple samples were collected from different areas of each toy to account for potential variations. After that clean blades were used to scrape off the toy's surface paint and labelled. Each toy sample mass were ranged from 5 to 25 mg. The samples of toys were weighed in a TFM® container for a microwave oven and 10 mL of nitric acid was added. The 156 children's toys samples were digested using a closed microwave oven (Multiwave GO, Anton Paar GmbH, Austria) in order to determine the elemental concentrations. After digestion, the microwave vessels were cooled, the digested was transferred to 100 mL volumetric flasks, diluted to 100 mL with Milli-Q® water and then filtered.

Analysis: The digests of 156 samples of children's toys were also analyzed through inductively coupled plasma-optical emission spectrometry (ICP-OES, ICAP 7400 Duo, Thermo, Scientific, USA). The wavelengths used for the detection of the following metals are given in the parenthesis; silver (Ag; 328.068 nm), aluminum (Al; 167.079 nm), boron (B; 249.773 nm), barium (Ba; 455.403 nm), bismuth (Bi; 223.061 nm), calcium (Ca; 393.366 nm), cobalt (Co; 228.616 nm), cadmium (Cd; 226.502 nm), chromium (Cr; 283.563 nm), copper (Cu; 324.754 nm), iron (Fe; 259.940 nm), potassium (K; 769.896 nm), magnesium (Mg; 279.553 nm), manganese (Mn; 257.610 nm), sodium (Na; 818.326 nm), nickel (Ni; 221.647 nm), zinc (Zn; 213.856 nm), lead (Pb; 220.353 nm) and titanium (Ti; 334.941 nm).

Statistical analysis: SPSS version 22 and MS Office were utilized for data representation and statistical analysis. One-way analysis of variance was also used.

Fig. 1. Stratified sampling from the single toy samples

RESULTS AND DISCUSSION

This study emphasizes the possible dangers that children's plastic toys may bring due to the presence of metals, which has significant consequences for children's health. Because of their growing physiology, frequent hand-to-mouth contact and heightened vulnerability to toxins, children–especially those who are young are more susceptible to the negative consequences of heavy metal exposure. This makes their exposure alarming. Children frequently lick, suck or bite their toys. When kids chew or lick painted toys, metals loosely bound to the surface can quickly leach into their bodies. Also, inhalation can pose a risk of toxic metal uptake. Since the body cannot metabolize many toxic metals in the toys, they tend to accumulate and produce metal body burdens in children. Thus, contaminated toys can pose long-term chronic effects on children and thus can hinder children's mental and physical growth [27].

The European Commission [23] has set the migration limit values for 19 elements in three different groups: Group I: Dry, brittle, powder or pliable toy material. Group II: Liquid or sticky toy material. Group III: scraped-off material. In United States, the American Society for Testing and Materials [24] has established limits for 8 soluble migrating elements of toy material. The Gulf Standardization Organization [25] has established general requirements for children's toys and has adopted the limits recommended by the European Commission and the ASTM. The maximum concentration of 19 detected metals and the limits set by the European Commission and ASTM are shown in Table-1.

The percentage of toys containing the detected substance is shown in Fig. 2. Minimum and maximum concentrations (µg/mg) for Ag, Al, B, Ba, Bi, Ca, Co, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Zn, Pd and Ti in different types of children's plastic toys identified by ICP-OES.

TABLE-1
MAXIMUM CONCENTRATION OF NINETEEN DETECTED METALS, AS WELL AS THEIR LIMITS SET BY THE EUROPEAN COMMISSION, ASTM AND THE TOTAL SAMPLES EXCEEDING LIMIT

Element	Maximum concentration (ppm)	EN 71-3 Limits (ppm)-group 3 (scraped-off material)		ASTM Limits (ppm) – General	
		Limit	No and% of samples exceeded limit	Limit	No and% of toys in which the maximum concentrations exceeded limit
Ag	5181.0	No limit	–	No limit	–
Al	4737.8	70000	0	No limit	–
Bo	80028.7	15000	3 (5.4%)	No limit	–
Ba	2498.9	18750	0	1000	2 (3.56%)
Bi	17.9	No limit	–	No limit	–
Ca	27262.5	No limit	–	No limit	–
Co	6.0	130	0	No limit	–
Cd	11.0	17	0	75	0
Cr	59.9	No limit	–	60	0
Cu	668.4	7700	0	No limit	–
Fe	2741.9	No limit	–	No limit	–
K	3603.3	No limit	–	No limit	–
Mg	5565.2	No limit	–	No limit	–
Mn	34.2	15000	0	No limit	–
Na	22461.5	No limit	–	No limit	–
Ni	53.9	930	0	No limit	–
Zn	58675	46000	2 (3.6%)	No limit	–
Pd	1018.2	23	3 (5.4%)	90	1 (1.8%)
Ti	167567.9	No limit	–	No limit	–

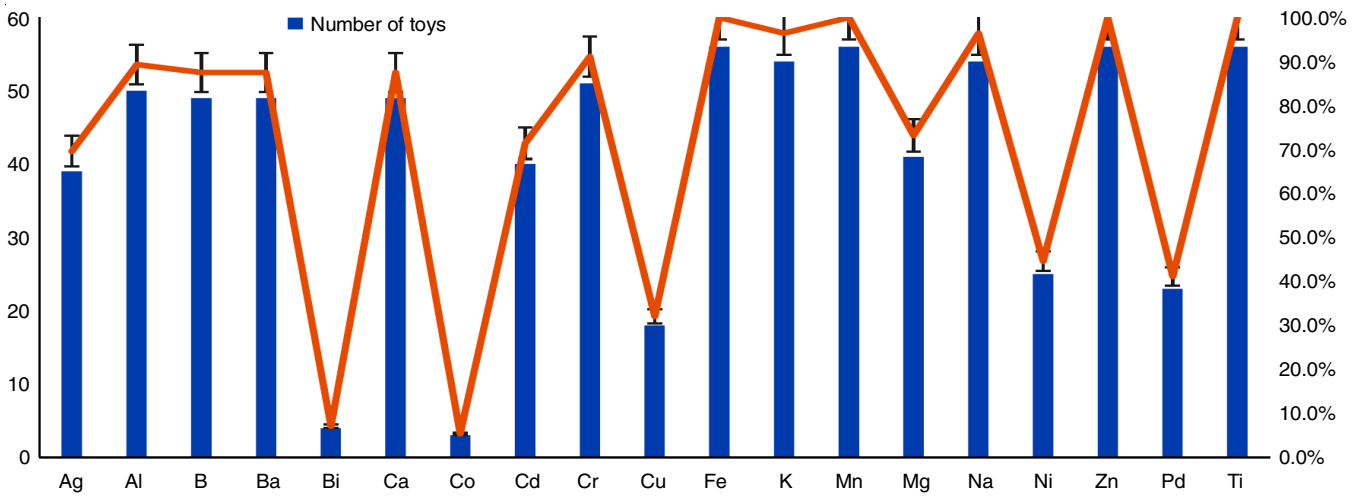

Fig. 2. Number of toys (out of 56) containing the elements of interest

It was found that Ti, Fe, Zn and Mg were present in all toys and their maximum concentrations were 167.5679, 2.7419, 58.6750 and 5.5652 µg/mg, respectively. Potassium and sodium were detected in 54 toys with maximum concentrations of 3.6033 and 22.4615 µg/mg, respectively, while Cr was detected in 51 toys and its maximum concentration was 0.0599 µg/mg. B, Ba and Ca were found in 49 toys at maximum concentrations of 40.3297, 2.4989 and 27.2625 µg/mg, respectively. Also, Al was detected in 50 toys with a maximum concentration of 4.7378 µg/mg. Magnesium, Cd and Ag were found in 41, 40 and 39 toys at maximum concentrations of 0.0342, 0.0110 and 5.1810 µg/mg, respectively. Nickel, Pd and Cu were detected in 25, 23 and 18 toys with maximum concentrations of 0.0539, 1.0182 and 0.6684 µg/mg, respectively, whereas Bi and Co were found in 4 and 3 toys at maximum concentrations of 0.0179 and 0.0060 µg/mg, respectively.

A total of 10 toys were selected in the activity-related group in which Ti, Zn, Mg, Fe and Ba were found in all toys with maximum concentrations of 167.5679, 4.6238, 4.4160, 0.4046 and 0.1692 µg/mg respectively. Al, Ca, Cr, K and Na were present in 9 toys with maximum concentrations of 3.7207, 27.2625, 0.0266, 1.4097 and 10.5299 µg/mg, respectively. Ag,

TABLE-2
METALS DETECTED IN CHILDREN'S PLASTIC TOYS (ACTIVITY-RELATED GROUP)

Toys	# of the samples	Concentration (µg/mg) (min-max)				
		Ag	Al	B	Ba	Bi
Magnetic upper-case letters	(n = 2)	ND-0.0074	0.0052– 0.8017	ND	ND-0.0008	ND
Building blocks of fire truck	(n = 3)	0.0011-0.0023	0.0980-0.4582	0.0115-0.0393	0.0005-0.0020	ND-0.0003
Letter	(n = 2)	ND-0.0042	0.0068-0.7802	0.0016-0.0602	0.0014-0.0186	ND
Military blocks	(n = 2)	0.0102-0.3117	1.4344-3.7207	ND	ND-0.0019	ND
Numbers and capital letters	(n = 2)	ND-0.0024	0.0054-1.5071	0.0002-0.0144	ND-0.0001	ND
Blocks play n learn	(n = 2)	ND-0.1357	ND-1.1032	0.0082-0.1674	0.0064-0.1692	ND
Ball, 12 link loops	(n = 11)	0.0007-0.0017	0.0052-2.8038	0.0109-0.0976	0.00004-0.1413	ND
Hp toys blocks	(n = 3)	0.0042-0.0064	0.2971-1.2452	0.0300-0.4752	ND-0.0014	ND
Yoyo	(n = 1)	ND	ND	0.2844	2.1202	ND
Cubes series	(n = 4)	ND	0.0033-0.0151	0.0227-0.1673	0.0038-0.0102	ND
		Ca	Co	Cd	Cr	Cu
Magnetic upper-case letters	(n = 2)	0.4591-1.2502	ND	ND-0.0007	ND-0.0015	ND
Building blocks of fire truck	(n = 3)	0.4808-0.8441	ND	0.0003-0.0005	0.0008-0.0011	ND
Letter	(n = 2)	0.5331-1.8202	ND	ND-0.0004	ND-0.0028	ND-0.0097
Military blocks	(n = 2)	1.1622-2.2546	ND	0.0014-0.0015	0.0003-0.0029	ND
Numbers and capital letters	(n = 2)	0.8809-1.0134	ND	ND-0.0004	0.0002-0.0015	ND
Blocks play n learn	(n = 2)	2.0860-3.6632	ND	ND-0.0010	0.0025-0.0041	ND-0.0256
Ball, 12 link loops	(n = 11)	0.1604-0.3400	ND	0.00008-0.0003	0.0003-0.0037	0.0087-0.0668
Hp toys blocks	(n = 3)	0.3932-27.2625	ND	0.0005-0.0007	0.0002-0.0266	ND
Yoyo	(n = 1)	ND	ND	ND	ND	ND
Cubes series	(n = 4)	1.0081-1.4303	ND	ND	0.0004-0.0013	ND
		Fe	K	Mg	Mn	Na
Magnetic upper-case letters	(n = 2)	ND-0.0196	ND-0.0543	0.0235-0.0909	ND-0.0008	0.0198-0.1801
Building blocks of fire truck	(n = 3)	0.0044-0.0561	ND-0.1224	0.0256-0.0300	ND-0.000287	0.0084-0.3358
Letter	(n = 2)	0.0215-0.0675	ND-0.1771	0.1872-0.1895	ND-0.0013	ND-0.8495
Military blocks	(n = 2)	0.0025-0.1528	ND	0.104941-0.163109	0.0014-0.0033	ND
Numbers and capital letters	(n = 2)	ND-0.0115	ND-0.0928	0.1262-0.1460	ND-0.0011	ND-0.3772
Blocks play n learn	(n = 2)	0.0131-0.0603	ND-0.4632	0.1785-1.6733	ND-0.0023	0.0914-10.5299
Ball, 12 link loops	(n = 11)	0.0045-0.0353	0.0091-0.0664	0.0135-1.0102	0.0001-0.0013	0.0032-1.2250
Hp toys blocks	(n = 3)	ND-0.3267	ND-0.9937	0.0568-0.5761	ND-0.0016	0.0027-2.2221
Yoyo	(n = 1)	ND-0.4046	ND-1.4097	ND-4.4160	ND	ND-5.8378
Cubes series	(n = 4)	0.0038-0.0276	0.0485-0.1638	0.0319-0.6375	ND	0.247496-3.675012
		Ni	Zn	Pb	Ti	
Magnetic upper-case letters	(n = 2)	ND-0.0048	0.1026-0.2470	ND-0.0040	9.0101-71.7575	
Building blocks of fire truck	(n = 3)	ND	0.0411-0.3857	ND	0.3262-5.7675	
Letter	(n = 2)	ND	0.0126-0.4300	ND	57.6109-70.8768	
Military blocks	(n = 2)	0.0010-.0013	0.3736-0.5792	ND-0.0235	68.4035-159.4178	
Numbers and capital letters	(n = 2)	ND	0.1686-0.1990	ND	8.5667-36.5855	
Blocks play n learn	(n = 2)	ND-0.0143	0.2828-0.8534	ND-0.0090	20.6174-47.9850	
Ball, 12 link loops	(n = 11)	0.0002-0.0006	0.0270-0.3694	0.0005-0.0028	2.1371-78.7925	
Hp toys blocks	(n = 3)	ND	0.1442-1.3841	ND-0.0011	11.9661-167.5679	
Yoyo	(n = 1)	ND	ND-4.6238	ND	ND-24.0221	
Cubes series	(n = 4)	ND	0.185323-0.257974	ND	5.925655-28.15412	

*1 µg/mg = 1000 ppm; ND = Not detected.

B, Cd and Mn were detected in 8 toys with maximum concentrations of 0.3117, 0.4752, 0.0015 and 0.0033 μg/mg, respectively. Pb, Ni and Cu were present in 5, 4 and 3 toys with maximum concentrations of 0.0090, 0.0143 and 0.0668 μg/mg, respectively. Bismuth was present in only 1 toy with a concentration of 0.0003 μg/mg, while Co was not detected in any of the toys in this group (Table-2).

In the teething toys (5 toys), Al, Ca, Cr, Fe, K, Mg, Zn and Ti were identified in all toys and their maximum concentrations were 0.5575, 6.7520, 0.0109, 0.1969, 0.7916, 2.5637, 48.1212 and 60.2351 μg/mg respectively. B, Ba and Na were present in 4 toys with maximum concentrations of 1.5214, 80.0287 and 17.4236 μg/mg, respectively. Ag, Cd and Mn were found in 3 toys, while Cu was detected in 2 toys and the corresponding maximum concentrations were 0.1321, 0.0005, 0.0003 and 0.0062 μg/mg, respectively. Pb was found in only one toy at a concentration of 0.0022 μg/mg, while Bi, Co and Ni were not detected in any of the toys in this group (Table-3).

In the figures group (10 toys), Ti, Zn, Mg, Na, K and Fe were present in all toys and their maximum concentrations were 86.7199, 32.19.57, 1.7067, 22.4615, 0.8921 and 2.7419 μg/mg, respectively. Al and Cr were identified in 9 toys, but Ag, B, Ba and Ca were found in 8 toys at a maximum concentration of 1.9573, 0.0360, 5.1810, 10.3809, 0.1585 and 10.8973 μg/mg, respectively. Cd and Mn were found in 7 toys with a maximum concentration of 0.0034 and 0.0267 μg/mg, respectively. Ni was present in 6 toys, Cu in 4 toys and Pb in 2 toys; the corresponding maximum concentrations were 0.0257, 0.4779 and 0.0081 μg/mg, respectively. Co and Bi were detected in only 1 toy and their maximum concentrations were 0.0060 and 0.0070 0081 μg/mg, respectively (Table-4).

Regarding the six toys in the cars-trains/sound-generating group, Zn, Mg, Fe, Ti, B, Ca, Na and K were detected in all toys, Cr, Ni, Al, Ag and Ba were identified in 5 toys, Cd, Cu and Mn in 4 toys, Pb in 2 toys, Co in 1 toy with a maximum concentration of 58.6750, 3.6896, 0.5618, 76.0292, 0.1982, 1.4236, 1.6661, 0.4564, 0.0599, 0.0261, 4.7378, 0.1970, 0.0963, 0.0006, 0.6684, 0.0342, 0.0124, 0.0009 μg/mg, respectively and Bi was not detected in any of the toys in this group (Table-5).

Table-6 shows the maximum concentrations in the games and puzzles toys group (25 toys); Ti, Zn, Mg, Fe and Na were found in all toys, K in 24 toys and Cr in 23 toys and their maximum concentrations were 131.6089, 23.3857, 5.5652, 1.1149, 11.3734, 3.6033 and 0.015 μg/mg, respectively. Mn was identified in 19 toys, Al and B in 17 toys, Ca and Ba in 16 toys and Ag in 15 toys at maximum concentrations of 0.0560, 4.8639, 80.0287, 13.5191, 2.4989, 1.1672 μg/mg, respectively. Cd was found in 14 toys, Pb in 13 toys, Ni in 9 toys and Cu in 6 toys with maximum concentrations of 0.0110, 1.0182, 0.0539 and 0.2158 μg/mg, respectively. Bi was detected in 2 toys at a maximum concentration of 0.0179 μg/mg, while Co was present in only 1 toy at 0.0004 μg/mg.

Consistent with previous studies, the toy sample analyzed in this research also contained Ag, Al, B, Ba, Bi, Ca, Co, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Zn, Pb and Ti; but in all toys samples Ti, Fe, Zn and Mg were detected. Though, Pb was found in 23 toys, of which three had a concentration exceeding the EN 71-3 limit of 23 ppm (Category 3; scraped-off material) and only one had Pd concentration that exceeded the ASTM limits of 90 ppm and the 3 toys that exceeded the limits belonged to the activity related group (military blocks) and the games and puzzles group (party band, animal world).

TABLE-3
METALS DETECTED IN CHILDREN'S PLASTIC TOYS (TEETHING-RELATED GROUP)

Toys	# of the samples	Concentration (μg/mg) (min-max)				
		Ag	Al	B	Ba	Bi
Deluxe gum soother	(n = 2)	0.0017-0.0026	0.0640-0.0760	0.0049-0.0323	0.00017-0.00023	ND
Water filled teether	(n = 3)	ND	0.00003-0.0133	0.0761-0.1714	0.0159-0.1157	ND
Bruin (musical teething keys)	(n = 2)	ND-0.0471	ND-0.5575	ND	ND	ND
O ball clickity twist	(n = 3)	0.0304-0.1321	0.01822-0.0545	0.0026-0.0173	0.0002-0.3720	ND
Teething fun	(n = 5)	ND	0.0168-0.2154	0.0068-1.5214	ND-0.4608	ND
		Ca	Co	Cd	Cr	Cu
Deluxe gum soother	(n = 2)	0.1723-0.4574	ND	0.0001-0.0002	0.0003-0.0004	ND-0.0029
Water filled teether	(n = 3)	0.6688-2.8665	ND	ND	0.0032-0.0038	ND
Bruin (musical teething keys)	(n = 2)	0.2588-0.7976	ND	ND-0.0005	0.0002-0.0004	ND
O ball clickity twist	(n = 3)	0.9465-0.9728	ND	0.0001-0.0002	0.0006-0.0007	ND-0.0062
Teething fun	(n = 5)	0.3976-6.7520	ND	ND	0.0001-0.0109	ND
		Fe	K	Mg	Mn	Na
Deluxe gum soother	(n = 2)	0.0090-0.0147	ND-0.0701	0.0184-0.0305	0.0002-0.0003	ND
Water filled teether	(n = 3)	0.0167-0.0679	0.0961-0.1975	0.0452-0.1771	ND	0.3039-8.0517
Bruin (musical teething keys)	(n = 2)	0.0013-0.0058	0.0604-0.1471	0.3064-0.3188	ND-0.0002	ND-0.0974
O ball clickity twist	(n = 3)	0.0016-0.0036	ND-0.0124	0.0096-0.0124	ND-0.0001	ND-0.0749
Teething fun	(n = 5)	0.0051-0.1969	0.0233-0.7916	0.0451-2.5637	ND	0.1191-17.4236
		Ni	Zn	Pb	Ti	
Deluxe gum soother	(n = 2)	ND	0.1221-0.2099	ND-0.0022	6.7997-24.1357	
Water filled teether	(n = 3)	ND	0.2159-0.5433	ND	1.1966-2.7152	
Bruin (musical teething keys)	(n = 2)	ND	0.0497-0.1616	ND	1.8364-60.2351	
O ball clickity twist	(n = 3)	ND	0.7248-0.8136	ND	1.8684-16.5251	
Teething fun	(n = 5)	ND	0.1036-48.1212	ND	4.3692-9.8313	

*1 μg/mg = 1000 ppm; ND = Not detected.

TABLE-4
METALS DETECTED IN CHILDREN'S PLASTIC TOYS (FIGURE GROUP)

Toys	# of the samples	Concentration (µg/mg) (min-max)				
		Ag	Al	B	Ba	Bi
Hamtaroo	(n = 2)	ND-0.0801	ND-0.6580	0.0034-1.1246	ND-0.0212	ND
Barbie doll	(n = 2)	ND-0.5737	ND-1.9573	ND-0.0634	ND-0.0080	ND
Alpha company soldiers	(n = 2)	ND-0.0106	0.0279-0.0877	0.0440-0.1110	0.0020-0.0274	ND
Fireman Sam	(n = 2)	ND-0.0007	0.0016-0.5091	0.0030-0.0104	0.00009-0.0576	ND
Power Rangers	(n = 1)	ND	0.0357	ND	ND	ND
Princess Fitment play set	(n = 2)	ND-0.0022	0.1615-0.6267	0.0009-0.0826	0.0307-0.0597	ND
Playgroup, Noah's Ark	(n = 2)	ND-5.1810	0.0124-0.0220	0.0723-10.3809	0.0070-0.1585	ND-0.0070
White happy cute rabbit	(n = 2)	ND-0.1248	ND-0.6777	0.0613-0.0774	ND-0.0011	ND
Debbie beautiful girl	(n = 3)	0.0033-0.1347	0.0013-0.8433	0.0212-0.0734	0.0106-0.0425	ND
Beige and brown lady bird	(n = 1)	ND	ND	ND	ND	ND
		Ca	Co	Cd	Cr	Cu
Hamtaroo	(n = 2)	0.2772-1.0318	ND	ND-0.0002	0.0006-0.0030	ND-0.0101
Barbie doll	(n = 2)	0.7866-3.1006	ND	ND-0.0025	0.0008-0.0030	ND
Alpha company soldiers	(n = 2)	0.6960-2.7508	ND	ND	ND-0.0031	ND
Fireman Sam	(n = 2)	ND-0.1552	ND	ND-0.00004	0.0006-0.0066	ND-0.0105
Power Rangers	(n = 1)	ND	ND	ND	0.0005	ND
Princess Fitment play set	(n = 2)	ND-3.0469	ND	ND-0.0005	0.0009-0.0187	ND-0.4779
Playgroup, Noah's Ark	(n = 2)	3.3738-10.8973	ND-0.0060	ND-0.0034	0.0040-0.0360	ND
White happy cute rabbit	(n = 2)	0.8704-1.1794	ND	ND-0.0008	0.0012-0.0045	ND-0.0020
Debbie beautiful girl	(n = 3)	ND	ND	0.0006-0.0008	0.0011-0.0014	ND
Beige and brown lady bird	(n = 1)	0.4522	ND	ND	ND	ND
		Fe	K	Mg	Mn	Na
Hamtaroo	(n = 2)	0.0137-0.2363	ND-0.3107	0.0534-0.0778	ND-0.0003	ND-0.8385
Barbie doll	(n = 2)	0.1465-0.2707	ND-0.1150	0.0422-0.3938	ND-0.0267	ND-0.3035
Alpha company soldiers	(n = 2)	1.2836-2.7419	0.1504-0.2139	0.0858-0.1436	ND	0.3934-5.2477
Fireman Sam	(n = 2)	0.0167-0.0335	0.0254-0.3025	0.2750-1.2728	ND-0.0018	0.5881-1.0314
Power Rangers	(n = 1)	ND-0.0038	ND-0.0276	ND-0.1270	ND	ND-0.3718
Princess Fitment play set	(n = 2)	0.0186-0.3879	ND-0.8921	0.2165-1.7067	ND-0.0010	0.0403-5.8686
Playgroup, Noah's Ark	(n = 2)	0.0471-0.0859	0.6899-0.7373	0.3988-1.6759	ND-0.0082	6.1368-22.4615
White happy cute rabbit	(n = 2)	0.0203-0.0290	ND-0.1334	0.1775-0.2026	ND-0.0008	0.0328-1.1488
Debbie beautiful girl	(n = 3)	0.0259-0.0821	ND-0.1335	0.0064-0.9109	ND-0.0261	0.7068
Beige and brown lady bird	(n = 1)	0.0041	0.1104	0.2367	ND	0.081
		Ni	Zn	Pb	Ti	
Hamtaroo	(n = 2)	ND-0.0003	0.1233-0.3232	ND	10.6090-14.0578	
Barbie doll	(n = 2)	0.0019-0.0257	0.2050-1.2491	ND	9.3438-53.4776	
Alpha company soldiers	(n = 2)	ND	0.1610-0.2957	ND	ND-4.0961	
Fireman Sam	(n = 2)	ND-0.0011	0.1801-32.1957	ND-0.0081	1.9993-4.6334	
Power Rangers	(n = 1)	ND	ND-0.3376	ND	3.9183-6.2459	
Princess Fitment play set	(n = 2)	ND	0.2284-0.4484	ND	11.4010-86.7199	
Playgroup, Noah's Ark	(n = 2)	ND-0.0097	0.4796-1.8430	ND	2.9236-8.3397	
White happy cute rabbit	(n = 2)	ND-0.0002	0.2928-0.4307	ND	5.8150-8.5337	
Debbie beautiful girl	(n = 3)	ND-0.0015	0.0271-0.2728	ND	9.2496-23.8119	
Beige and brown lady bird	(n = 1)	ND	0.0507	0.0019	2.6614	

*1 µg/mg = 1000 ppm; ND = Not detected.

Additionally, in the selected sample Zn was present in all toy samples, two of which had Zn concentrations above the EN 71-3 limit of 46,000 ppm and the two toys that exceeded the limits were in the teething-related group (teething fun) and cars-trains/sound-generating group (cars). Interestingly, we know Zn can be used as a substitute for the Pd function in PVC (prohibited by specific rules); this explains the presence of Zn in plastic toys in different parts of the world [28,29].

Two of the toys analyzed contained a concentration of Ba that exceeded the ASTM limit of 1,000 ppm in the activity-related group (yoyo) and the games and puzzles group (basketball). The high Ba concentration could be due to the addition of barium sulfate as a plastic filler to increase the density of the polymer [30]. However, in three of the analyzed toys whose B concentration exceeded the EN 71-3 limit of 15,000 ppm were in the games and puzzles group (Lovely little sheep, Fashion doll toys and Animals' World). The use of boron compounds in raw materials (for flexible and stretchy texture) to produce slime and putty toys is the reason B is present in toys [31]. Meanwhile, the maximum concentration of Cr in one toy was 59.9 ppm and was in the cars-trains/sound-generating group (car), almost equal to the ASTM limit of 60 ppm. The findings are consistent with studies on plastic toys [32-34] and fragrances in toy makeup [35]. Furthermore, other metals having a maximum detected concentration in the study were Al (4737.8 ppm; below the EN 71-3 limits of 70,000 ppm),

TABLE-5
METALS DETECTED IN CHILDREN'S PLASTIC TOYS (CARS-TRAINS/SOUND-GENERATING GROUP)

Toys	# of the samples	Concentration (µg/mg) (min-max)				
		Ag	Al	B	Ba	Bi
Paw patrol, air rescue chase	(n = 3)	0.0100-0.0800	2.2987-4.7378	ND-0.0841	0.0003-0.0005	ND
Nert air launcher	(n = 2)	ND-0.0079	0.0675-0.4390	ND-0.0024	ND-0.0003	ND
Cars	(n = 3)	0.00005-0.00012	0.0015-1.5072	0.0004-0.0203	ND-0.0001	ND
Blue car	(n = 1)	ND	ND	0.0689	ND	ND
Aviation international, Airline world	(n = 2)	ND-0.1970	ND-1.4447	ND-0.0356	ND-0.0046	ND
Baby rattles	(n = 7)	0.0002-0.0539	0.0080-2.1127	0.0020-0.1982	0.0001-0.0963	ND
		Ca	Co	Cd	Cr	Cu
Paw patrol, air rescue chase	(n = 3)	0.9092-1.4236	ND	0.0003-0.0004	0.0009-0.0014	ND-0.0210
Nert air launcher	(n = 2)	ND-0.3567	ND	ND-0.0006	ND	ND
Cars	(n = 3)	0.0069-0.1379	ND	ND	0.00006-0.05990	ND-0.0005
Blue car	(n = 1)	0.5046	ND	ND	0.0028	ND-0.0195
Aviation international, Airline world	(n = 2)	0.4114-0.4269	ND	ND-0.0004	ND-0.0006	ND
Baby rattles	(n = 7)	0.1810-0.5216	0.0007-0.0009	0.0002-0.0003	0.0002-0.0108	0.0004-0.6684
		Fe	K	Mg	Mn	Na
Paw patrol, air rescue chase	(n = 3)	0.0102-0.0448	0.0978-0.1477	0.0273-0.3377	0.0003-0.0011	0.0298-0.5975
Nert air launcher	(n = 2)	0.0158-0.0447	ND-0.2539	0.0451-1.0224	ND	ND-1.6661
Cars	(n = 3)	0.0008-0.2665	0.0012-0.0715	0.0042-0.3972	0.0006-0.0029	0.0290-1.4929
Blue car	(n = 1)	ND – 0.0554	ND-0.4564	ND-0.2084	ND	ND-0.2465
Aviation international, Airline world	(n = 2)	ND-0.0767	ND-0.2649	0.0906-0.1777	ND-0.0007	0.0561-0.7262
Baby rattles	(n = 7)	0.0052-0.5618	ND-0.2945	0.0369-3.6896	0.0002-0.0342	0.0219-0.3944
		Ni	Zn	Pb	Ti	
Paw patrol, air rescue chase	(n = 3)	ND-0.0005	0.1102-0.3189	ND	2.7286-64.1807	
Nert air launcher	(n = 2)	ND	0.1782-0.4356	ND	12.1541-61.0925	
Cars	(n = 3)	0.0002-0.0261	0.0257-58.6750	0.0011-0.0124	0.0346-3.9183	
Blue car	(n = 1)	ND-0.0007	ND-0.1451	ND	ND-17.8577	
Aviation international, Airline world	(n = 2)	ND-0.0001	0.0250-0.1106	ND	1.1010-14.9667	
Baby rattles	(n = 7)	ND-0.0022	0.0336-0.2251	0.0007-0.0115	0.6286-76.0292	

*1 µg/mg = 1000 ppm; ND = Not detected.

TABLE-6
METALS DETECTED IN CHILDREN'S PLASTIC TOYS (CARS-TRAINS/SOUND-GENERATING GROUP)

Toys	# of the samples	Concentration (µg/mg) (min-max)				
		Ag	Al	B	Ba	Bi
Basketball (extreme fun)	(n = 2)	ND	0.1016-3.5720	ND-0.0794	0.0046-2.4989	ND
Happy places' Dreamy bear'	(n = 3)	ND-0.0070	ND-0.0171	0.0230-0.7153	0.0006-0.0160	ND
Sylvanian Families	(n = 3)	0.0020-0.0317	1.3241-1.5733	0.0068-0.3327	0.0005-0.1230	ND
Nintendo	(n = 4)	0.0064-0.0247	0.0746-1.9522	0.00003-0.02997	0.0238-0.1858	ND
red wheel and yellow boarder)	(n = 1)	ND	ND	0.0321	ND	ND
Mimi House beauty	(n = 2)	ND	ND-0.0048	0.0058-0.2582	ND	ND
Shopping Cart	(n = 1)	ND	ND	ND	ND	ND
Music	(n = 3)	0.0046-0.0066	0.6374-4.8639	0.0091-0.1298	0.0003-0.0030	ND
Party band	(n = 2)	ND-0.0066	0.008813-0.016366	ND-0.0304	ND-0.0023	ND
Doctor tools doll	(n = 4)	0.2064-1.1672	0.0877-2.5494	0.0039-0.4687	0.0013-0.0593	0.0012-0.0179
Archer	(n = 2)	ND-0.0103	0.0010-0.7312	ND-0.0456	ND-0.1477	ND
Fashion Doll TOYS	(n = 2)	ND	ND-0.0217	0.1945-40.3297	ND-0.4365	ND
Animals World	(n = 4)	ND-0.0011	0.0148-0.0998	0.0043-30.0864	ND-0.4662	ND
Lamaze	(n = 3)	ND-0.1020	0.0317-1.1309	0.0226-0.0365	ND-0.0443	ND
Fishing	(n = 7)	0.0006-0.0064	0.0024-0.1053	0.0156-0.1540	0.00004-0.00565	ND
Happy Valley electronic game	(n = 1)	ND	0.1652	ND	0.0009	ND
Kitchen play set	(n = 11)	0.0013-0.0286	0.0193-1.3886	0.0086-0.1913	0.0014-0.0098	ND-0.0005
Bright starts	(n = 2)	0.0032-0.0088	0.1494-1.2856	ND-0.0382	0.0013-0.0102	ND
Lovely collection	(n = 2)	ND-0.3000	0.0057-0.3017	0.0014-0.0629	0.0003-0.0021	ND
Pink warm	(n = 1)	ND	ND	ND	ND	ND
Toys baby	(n = 3)	0.0013-0.1776	0.0096-0.1442	0.1194-0.1234	0.0039-0.1094	ND
Explore-a-ball playgroup	(n = 3)	0.0015-0.0021	0.0013-0.2712	ND-0.0067	0.0030-0.3494	ND
Bright stars (carry and teeth purse)	(n = 2)	ND	ND-0.0042	ND-0.0037	ND-0.0006	ND
Lovely little sheep	(n = 2)	ND	0.0078-0.0172	0.0016-80.0287	ND-0.0080	ND
King power, BASH wrestle	(n = 2)	ND	0.1109-0.1255	0.0655-0.0926	0.0481-0.1335	ND

		Ca	Co	Cd	Cr	Cu
Basketball (extreme fun)	(n = 2)	ND-0.3648	ND	ND	ND-0.0023	ND
Happy places' Dreamy bear'	(n = 3)	ND	ND	ND	0.0016-0.0058	ND
Sylvanian Families	(n = 3)	0.3091-2.2326	ND	0.0003-0.0004	0.0002-0.0047	ND
Nintendo	(n = 4)	ND	ND	0.0004-0.0010	0.0011-0.0019	0.0131-0.0847
red wheel and yellow boarder)	(n = 1)	0.6297	ND	ND	0.0014	ND
Mimi House beauty	(n = 2)	0.3633-4.0995	ND	ND	0.0006-0.0021	ND
Shopping Cart	(n = 1)	1.4467	ND	ND	ND	ND
Music	(n = 3)	ND-0.6212	ND	0.0003-0.0004	0.0006-0.0063	ND
Party band	(n = 2)	ND-0.7679	ND	ND	ND-0.0015	ND
Doctor tools doll	(n = 4)	0.4494-13.5191	ND	0.0014-0.0110	0.0015-0.0027	ND
Archer	(n = 2)	1.0731-2.4920	ND	ND-0.0015	0.0062-0.0068	ND
Fashion Doll TOYS	(n = 2)	6.9269-7.5024	ND	ND	ND-0.0027	ND
Animals World	(n = 4)	ND	ND	ND-0.0106	0.0015-0.0060	ND-0.0069
Lamaze	(n = 3)	0.9338-3.9673	ND	ND-0.0016	ND-0.0128	ND
Fishing	(n = 7)	ND-0.8882	ND	0.0002-0.0006	0.0020-0.0062	0.0002-0.2158
Happy Valley electronic game	(n = 1)	ND	ND	ND	ND	ND
Kitchen play set	(n = 11)	0.2488-11.5243	ND	0.0007-0.0057	0.0007-0.0150	ND
Bright starts	(n = 2)	0.3822-1.2933	ND	0.0003-0.0007	ND-0.0043	ND-0.0061
Lovely collection	(n = 2)	1.3842-1.7175	ND	ND-0.0005	0.0011-0.0021	ND
Pink warm	(n = 1)	1.1955	ND	ND	0.0009	ND
Toys baby	(n = 3)	ND-0.2740	ND-0.0004	0.0002-0.0003	0.0005-0.0008	ND-0.0154
Explore-a-ball playgroup	(n = 3)	0.1221-0.2621	ND	0.0001-0.0002	0.0002-0.0008	ND-0.0170
Bright stars (carry and teeth purse)	(n = 2)	0.0408-0.5608	ND	ND-0.0002	0.0001-0.0007	ND
Lovely little sheep	(n = 2)	ND-1.0555	ND	ND	0.0014-0.0041	ND
King power, BASH wrestle	(n = 2)	ND-0.6065	ND	ND	0.0035-0.0039	ND

		Fe	K	Mg	Mn	Na
Basketball (extreme fun)	(n = 2)	0.1888-0.2062	0.4836-2.2027	0.2690-5.5652	0.0019-0.0280	0.8854-5.7925
Happy places' Dreamy bear'	(n = 3)	0.0093-0.1776	0.1070-0.3352	0.0927-0.2828	ND-0.0005	0.4837-3.3141
Sylvanian Families	(n = 3)	0.0051-0.0319	0.0210-0.6242	0.0135-0.3594	ND-0.0011	0.1101-2.2765
Nintendo	(n = 4)	0.0078-0.0285	ND-0.0836	0.4037-0.6264	0.0002-0.0035	0.1133-2.1731
red wheel and yellow boarder)	(n = 1)	ND-0.0072	ND-0.1881	ND-0.1401	ND	ND-0.2034
Mimi House beauty	(n = 2)	0.0227-0.0326	0.0510-0.5201	0.1504-1.7612	ND	0.2081-11.3734
Shopping Cart	(n = 1)	ND-0.0007	ND-0.0555	ND-0.1776	ND	ND-0.5667
Music	(n = 3)	0.0256-0.6833	0.1032-0.1331	0.1615-0.4915	0.0026-0.0194	0.0241-0.9595
Party band	(n = 2)	0.0620-0.5269	0.0957-0.2515	0.0671-0.4598	0.0001-0.0105	0.7355-2.1361
Doctor tools doll	(n = 4)	0.0117-0.1857	0.0676-0.1001	0.0428-1.1114	ND-0.0100	ND-2.9801
Archer	(n = 2)	0.0306-0.1087	ND-0.210253	0.059857-0.190058	ND-0.001139	ND-0.679948
Fashion Doll TOYS	(n = 2)	0.5562-1.1149	0.26928-3.60331	0.200523-1.480843	ND-0.012719	10.03382-10.72361
Animals World	(n = 4)	0.0204-0.0882	0.066423-0.106923	0.278931-1.08787	ND-0.001037	0.22925-1.225033
Lamaze	(n = 3)	0.0056-0.1077	ND-0.573221	0.109742-0.313734	ND-0.002414	0.004058-1.509118
Fishing	(n = 7)	0.0231-0.2230	0.026169-0.108102	0.109274-0.226616	0.000255-0.004043	0.017706-0.5971
Happy Valley electronic game	(n = 1)	0.049	0.20881	0.368275	0.007289	0.266611
Kitchen play set	(n = 11)	0.0096-0.0760	ND-0.562644	0.012651-0.924358	0.00023-0.001894	0.076691-1.419719
Bright starts	(n = 2)	0.0180-0.0576	ND	0.043002-0.540627	0.000696-0.002137	0.011806-0.011926
Lovely collection	(n = 2)	0.0125-0.0818	ND-0.150886	0.182217-0.23569	ND-0.000631	0.124537-4.383796
Pink warm	(n = 1)	ND-0.0034	ND-0.115302	ND-0.204846	ND	ND-0.317409
Toys baby	(n = 3)	ND-0.0501	ND-0.049232	ND-0.984125	ND-0.002186	ND-0.271782
Explore-a-ball playgroup	(n = 3)	0.0160-0.0169	ND-0.011166	0.017706-0.14867	0.000235-0.000308	0.010868-0.026584
Bright stars (carry and teeth purse)	(n = 2)	0.0006-0.0136	0.007233-0.020103	0.017787-0.024574	ND	0.087535-0.176589
Lovely little sheep	(n = 2)	0.0214-0.0258	0.162383-0.28716	0.450552-0.666053	ND	0.249173-0.426949
King power, BASH wrestle	(n = 2)	0.2688-0.5246	0.074143-0.313214	ND-0.956109	ND-0.056016	0.380602-4.901812

		Ni	Zn	Pb	Ti
Basketball (extreme fun)	(n = 2)	ND	6.8874-7.5887	ND-0.0077	24.6585-33.7682
Happy places' Dreamy bear'	(n = 3)	ND	0.4357-0.7654	0.0020-0.0028	4.6254-39.9790
Sylvanian Families	(n = 3)	ND-0.0004	0.0481-0.4774	0.0021-0.0076	23.2190-45.0522
Nintendo	(n = 4)	ND	0.3597-0.6412	ND-0.0035	4.2168-85.6840
red wheel and yellow boarder)	(n = 1)	ND	ND-0.0823	ND	ND-5.9861
Mimi House beauty	(n = 2)	ND	0.1186-0.3930	ND	18.1061-18.6470
Shopping Cart	(n = 1)	ND	ND-0.3059	ND	ND-6.0203
Music	(n = 3)	ND-0.0002	0.1251-0.3537	ND-0.0013	3.7269-58.3975
Party band	(n = 2)	ND-0.0539	0.1963-23.3857	0.0003-1.0182	2.8623-8.1879
Doctor tools doll	(n = 4)	ND-0.0048	0.0420-1.2146	ND-0.0178	2.1766-102.9581
Archer	(n = 2)	ND-0.000282	0.616336-1.043709	ND-0.007079	6.625586-36.78309
Fashion Doll TOYS	(n = 2)	ND	0.499923-0.523172	ND	2.473333-3.953465
Animals World	(n = 4)	ND	0.369386-0.603049	0.000639-0.026756	4.035623-20.45472
Lamaze	(n = 3)	ND-0.000616	0.229508-1.759552	ND	4.433496-8.64562
Fishing	(n = 7)	ND	0.098558-0.219775	0.000202-0.012212	2.240309-24.45467
Happy Valley electronic game	(n = 1)	ND	0.121495	ND	7.819665
Kitchen play set	(n = 11)	ND-0.001849	0.062912-1.854388	ND-0.003388	0.062807-131.6089
Bright starts	(n = 2)	ND	0.069316-0.581562	ND-0.000436	4.347347-50.88437
Lovely collection	(n = 2)	ND	0.466268-0.52289	ND	2.99965-3.801068
Pink warm	(n = 1)	ND	ND-0.224263	ND	ND-5.679786
Toys baby	(n = 3)	ND-0.002497	ND-1.151024	ND	ND-22.17498
Explore-a-ball playgroup	(n = 3)	0.000149-0.006297	0.059315-0.10928	ND	10.55748-38.3506
Bright stars (carry and teeth purse)	(n = 2)	ND	0.010245-0.139137	ND	0.013326-0.339905
Lovely little sheep	(n = 2)	ND	0.416029-0.481037	ND-0.004541	0.609528-38.85793
King power, BASH wrestle	(n = 2)	ND	0.222708-2.256408	ND	7.395668-12.59151

*1 µg/mg = 1000 ppm; ND = Not detected.

Co (6 ppm; below the EN 71-3 limits of 130 ppm), Cd (11 ppm, below the EN 71-3 limits of 17 ppm and ASTM limit of 75 ppm), Cu (668.4 ppm; below the EN 71-3 limit of 7,700 ppm) and Ni (53.9 ppm; below the EN 71-3 limit of 930 ppm) and Mn (34.2 ppm; below the EN 71-3 limit of 15,000 ppm). Unfortunately, there were no recommended limits available for plastic toys in any guidelines for metals such as Ag, Bi, Ca, Fe, K, Mg, Na and Ti.

Conclusion

Determination of metals concentration in the popular 56 types of children's toys were successfully analyzed using ICP-OES) technique. The procured children's toys were grouped into five groups *viz*. activity toys, teething, figures, cars-trains/ sound-generating and games & puzzles. The results were compared with European Commission limits (EN 71-3) and ASTM. The results showed that the analyzed toys contain toxic metals in varying concentrations. Majorly, Ti, Fe, Zn and Mg were detected in all the toys. K and Na were detected in 54 toys, while Cr was detected in 51 toys. B, Ba and Ca were found in 49 and Al was detected in 50 toys. Manganese, Cd and Ag were found in 41, 40 and 39 toys, respectively. Nickel, Pb and Cu were detected in 25, 23 and 18 toys, respectively. Bismith and Co were found in 4 and 3 toys, respectively. Moreover, the concentrations of Pd and B in three toys exceeded the limits, while the concentrations of Zn and Ba exceeded the limit in two toys. The reason for the presence of these elements in plastic toys was the stabilizers and dyes that were added to the PVC during processing, which ultimately exposed children to these toxic substances. Therefore, the best approach to prevent many hazardous compounds is to move from plastic toys to those made of safer materials. This study also identified the presence of Ag, Bi, and Ti in the studied children's toys, however, there are no established limits available for comparison. Therefore, further investigations are required to confirm their function in toys, pinpoint related health hazards and suggest limits.

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CONFLICT OF INTEREST

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

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