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HEAVY METALS CONTENT IN LOW-PRICED TOYS

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ABSTRACT

This study determines the level of selected heavy metals in 42 toy samples and its health risk to children obtainedfrom various convenience shops in the urban area of central region Peninsular Malaysia. High Definition X-ray Fluorescence (HDXRF®) technique (Model 800701-01) was used to analyse the sample and the health risk was calculated using the USEPA method. Most of the samples in this study were detected with Zn (97%, N = 41), Sr (90%, N = 38) and Cu (79%, N = 33) and half of the samples were detected with Ni (64%, N = 27), Cr (59%, N=25), Ba (57%, N = 24) and Pb (50%, N = 21). The highest element detected in toys was Bain materials of polymer (2255.00 ppm), printed ink (1698.00±758.02 ppm), plastic (1160.82±898.06 ppm) and textile (1284.00 ppm). Plastic and metal toys contained the most heavy metals. Based on the level detected, Co and Ni pose carcinogenic health risk while Hg, Sb and Sn pose non-carcinogenic health risks in this study. Heavy metals in toys are varies by its material and bring a significant health risk to children.

Keywords: heavy metals, toys, materials, health risk, children.

1. INTRODUCTION

Toy is defined as any goods designed or intended for the use in play by children age below 14 years [1].Toys play an important role in children development at their young age as it's develop the physical, intelligence and social abilities of children besides bring fun, joy and entertainment [2].

News on contamination of heavy metals and other dangerous substances in toys continue to be alarming headlines to the public especially the effects to children. Some heavy metals such as lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), selenium (Se), mercury (Hg), barium (Ba) and zinc (Zn) were commonly detected in toys [3-6]. These elements were added to improve the softness, brightness and flexibility of toys and to make it more attractive for the children. Toys also may contain paint which rubs off with time. Omalaoye *et al*. (2010) reported greater levels of Cr in children's toys (ND to 191.67 lg/g). Zn also was detected at the level of 266.67-2,043 lg/g. Cr is mainly concentrated in the toy's paint rather than the plastic itself [7] while Zn and Cd in the children's toys was used as a stabilizer and catalyst in PVC [8-11].Hg was used as a catalyst to enhance the chemical reaction among ingredients to made plastic toys [12]. Metal such as Se is used as coloring agents for several toy materials such as in paints, ceramics, plastics, and glazes [3].

Most toys are licked, sucked or bite by children. Metals in painted toys are loosely bound to the surface and can leach easily when they are chewed or sucked by children, thus posed chronic effects in the long term [3, 6]. Heavy metals also may pose threat to children through inhalation [13]. Cases of heavy metal poisoning were widely reported all around the globe. For instance, a study conducted by Abdullah (2015) found high Pb concentration exceeding the permissible level in cheap yellow colour toys in Malaysia. In North America, childhood Pb poisoning was linked to the ingestion of old paint chips [15]. Exposure to high levels of Pb can damage the brain, kidney and eventually cause death [16]. Cd is released as fine airborne particles which react with oxygen to form respirable cadmium oxide which may cause disruption in the nephritic system [4].

Heavy metals have a density of at least 5 times than water and cannot be metabolized by the body. This can cause metals accumulation and produce metals body burden. Heavy metal poisoning can disrupt the functions of mental, energy, nervous system, kidneys, lungs and other organs [17]. For example, inorganic As is carcinogen and can cause skin, lungs, liver, and bladder cancer. Lower level of exposure can cause nausea and vomit, decreased production of red and white blood cells, abnormal heart rhythm, and damage to blood vessels. Chronic exposure to as can cause skin darkening and the appearance of small warts on the palms, soles, and torso. Ba in paint can cause vomiting, abdominal cramps, and diarrhea. It may also cause difficulties in breathing, numbness around the face, and muscle weakness. Large exposure to Ba will cause high blood pressure, changes in heart rhythm, paralysis, and death. Cd is human carcinogens than can cause severe damage to lungs through inhalation. Accidental ingesting of very high levels of Cd irritates the stomach which leads to vomiting and diarrhea. Chronic exposure to Cd can buildup in the kidneys and possible to cause kidney disease. Cr is carcinogens that can cause irritation to the nose lining through inhalation. In addition, this element is able to cause breathing problems such as asthma, cough, and wheezing. High Pb levels can severely damage the brain, kidneys and cause death [3, 16, 17].

A study shows that age differences affect the susceptibility to Pb poisoning in children. Alimentary absorption is about 50% in children. Together with their greater volume of air inhaled in relation to body size, the total Pb absorption from the environment is around three times higher than adults. Besides, iron deficiency and low dietary calcium promote the chance to increase rate of lead absorption [13]. According to ISO 8124: 2010 Safety of Toys Part 3: Migration of Certain Elements, there are eight metals in concern. The permissible levels of these metals were based on the bioavailability of the elements in toys. They are Sb (0.2 µg), as (0.1 µg), Ba (25.0 µg), Cd (0.6 µg), Cr (0.3 µg), Pb (0.7 µg), Hg (0.5 µg) and Se (5.0 µg). These elements should not exceed the permissible level per day.

In exercise of the powers conferred by Section 19 of the Consumer Protection Act 1999 [Act 599] in Malaysia, the Minister had made the Regulation of the Consumer Protection (Safety Standards for Toys) in 2009. This regulation had come into force on 30 January 2010 and had imposed the safety standards for toys based on the Standards of Malaysia Act 1996 [Act 549] as specified in the First Schedule which is designated as safety standard for toys for the of Section 19 of the Consumer Protection Act 1999. This guideline assist the businesses that supply toys in Malaysia to ensure compliance with the legislation and requirements by the Ministry and helps the industry to understand the main features of the legislations and requirements to be compiled. A few amendments have been made to the regulation thus, the Ministry of Domestic Trade, Co-operatives and Consumerism (MDTCC) had imposed the Guideline on Mandatory Safety Standards for Toys in 2010.

The market value of toys and games industry has been expanding from 2008 to 2012 at an average growth of 9.5% [19, 20]. One possible reason for this growth is due to the successful collaboration of toy manufacturers with the animation industry such as Disney, Marvel and Universal Studio through character tie-ins based on latest movies which have been released making the toys more appealing to the customers. However, there are almost no descriptive data available on the growth of toys industry and the origin of toys available in Malaysian market. The emerging trend of importing toys from China is observed in Malaysia and was sold at low price in many convenience shops. Similar trend was reported in the United States where 87% of the toys are produced abroad which 74% are manufactured in China. Toys from China were reported contained high Pb in its paint. A study by Scott Clark, found that 50% of the paint sold in China, India, and Malaysia had Pb concentrations 30 times higher than the Consumer Product Safety Commission (CPSC) standard [21].

Routine inspection by the CPSC has issued a first recall in 2007 of nearly 130,000 toys made by a Hong Kong company (i.e. Toy Century Industrial) and imported by Toys R Us due to high Pb levels. Given that most of its wares are made in China, the toy industry ramped up its inspections for Pb and found that high levels were a lot more common than they had assumed. By the end of 2007, 42 recalls involving nearly 6 million toys had been issued because of excessive Pblevels [21].

Due to limited research on the safety status of toys in Malaysian market, there is a need to assess the level of heavy metals in these toys. Study determines the compliance of toy manufacturers and importers in the country to the safety standard and level provided by the regulation. It is important to ensure that the concentration of heavy metals present in toys is below the safe limit as what is addressed by the law. The absence of any known study on heavy metals in toys coupled with the fact that these materials dominate the children environment propels the need for this study.Finding of this study provide baseline information of the toys in the local market and also acts as an evidence-based to create preventive action to ensure toy safety in Malaysia.

2. MATERIALS AND METHODS

2.1 Sample collection

Study was performed from December 2015 to March 2016 to measure the level of heavy metals in cheap toys and the potential health risk to children in urban area, central region of Peninsular Malaysia. Low-priced toys imported from China (less than MYR 10) were selected randomly from several convenience shops in urban area in Selangor. These shops are commonly visited by the people and it has wide various types of toys at low price.

To obtain a homogenized sampling, a total of 42 toys were sampled by five categories highlighted in the ISO 8124: 2010 Safety of Toys. There are physical activity toys (N=10), intellectual (N=11), technical (N=5), creative (N=9) and social (N=7). Physical activity toys allow children to exercise, building strong bones and muscles and aiding in physical fitness. The examples of this toy are Frisbees, foot bags and jumping rope. Intellectual development toy allow children to think, processes information and understand them. The example of this toy is Lego, toy blocks, puzzle and robot kits. Technical toys allow the children to improve the skills relating to the practical use of machines and how its work. The example of this toy is metal do-it-yourself (DIY) vehicle and bicycle. Creative toys allow the children to be creative such as cooking tray and brick game while the social toys give children to establish social relationship among peers. These toys were categorized as paint coated (N = 4), printed ink (N = 3), polymer (N = 3), textile (N = 2), paper (N = 2), paperboard (N = 5), metal (N = 3) and plastic (N = 21). Most of the toys in this study are made of plastic.

2.2 Sample analysis

Heavy metals concentration in the toys was analyzed using High Definition X-ray Fluorescene (HDXRF®) HD Rocksand XOS's (Model 800701-01). The XRF technology is a rapid and convenient high technology tool that enables instant detection of heavy metals. XRF determines the total amount of heavy metals in toys.

2.3 Permissible concentration of heavy metals

Heavy metals concentration were compared to the permissible concentration as highlighted in ISO 8124:2010 Part 3 and European Union (EU) New Toy Safety Directive (EN 71-3: 2013). The maximum permissible

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concentration of migrated heavy metals in children toys under category III - the scrapped off toy material type was used for comparison (Table-1).

2.4 Statistical analysis

Data were analyzed using SPSS Version 22. Descriptive analysis was used to determine the mean, standard deviation, and range value. The comparison between mean of heavy metals in types of toys and materials were analyzed using Kruskal Wallis test. Spearman Correlation test is used to determine the association between heavy metal concentration in different types and materials of toys.

Table-1. The maximum permissible concentration of migrated heavy metals.

Elements	EN 71-3: 2013			EN 71-3: 1994/ A1: 2000/ AC: 2000/ AC: 2002	
	Category 1 (mg/kg)	**Category II** (mg/kg)	**Category III** (mg/kg)	**Any toy material except for modeling clay**(mg/kg)	**Modelling clay**(mg/kg)
Aluminium	5 625	1 406	70 000	-	-
Antimony	45	11.3	560	60	60
Arsenic	3.8	0.9	47	25	25
Barium	4 500	1 125	56 000	1.000	250
Boron	1 200	300	15 000	-	-
Cadmium	1.9	0.5	23	75	50
Chromium	-	-	-	60	60
Chromium (III)	37.5	9.4	460	-	-
Chromium (VI)	0.02	0.005	0.2	-	-
Cobalt	10.5	2.6	130	-	-
Copper	622.5	156	7 700	-	-
Lead	13.5	3.4	160	90	90
Manganese	1 200	300	15 000	-	-
Mercury	7.5	1.9	94	60	25
Nickel	75	18.8	930	-	-
Selenium	37.5	9.4	460	500	500
Strontium	4 500	1.125	56 000	-	-
Tin	15 000	3.750	180 000	-	-
Organic Tin	0.9	0.2	12	-	-
Zinc	3 750	938	46 000	-	-

Note: Category I - in dry, brittle, powder like or pliable toy material, Category II – in liquid or sticky toy material, Category III – in scraped off toy material

2.5 Health risk assessment

Health risk assessment for carcinogenic and non-carcinogenic health risk was adapted from USEPA (2009). Elements such as As, Cd, Co, Cr, Ni, and Pb are known to be carcinogens while other elements (i.e. Sb, Ba, Cu, Mn, Hg, Se, Sr, Sn, Zn) are non-carcinogens to human [24].Health risk assessment models were developed by the the United States [25]. These models have been developed in all details and are available through Risk Assessment Information System (RAIS) and are supported by the Toxicological profiles developed and gathered by the USEPA Integrated Risk Information System (IRIS). It is also supported by the US Agency for Toxic Substances and Disease Registry (ATSDR).

2.6 Non carcinogenic health risk

The non-carcinogenic health risk was calculated as average daily dose (ADD) in Eq. (1). The concentrations of elements (Cs) were taken from value detected in this study while other parameters used the default value applied in Grzetic&Ghariani (2008). Table 2 shows the description of the default value for all parameters in health risk calculation. The hazard quotient (HQ) was calculated as the ratio of ADD to the Reference Dose (RfD) (Eq. 2). RfD is an estimate of a daily exposure to the human population which includes sensitive subgroups that is likely to be without an appreciable risk of deleterious effects during a lifetime. The RfD is generally expressed in units of milligrams per kilogram of bodyweight per day (mg/kg/day). The RfD can be used as reference point to identify the potential effects of the chemical at different doses. Usually, doses less than the RfD are not likely to be associated with adverse health. While doses more than the RfD are likely to be associated with adverse health risks which is a concern [28]. The RfD value was quoted from several references as in Table-3. The HQ >1 indicates significant non-carcinogenic risk while HQ<1 indicates no significant risk.

$$\text{ADD (Ingestion)} = \frac{C_S\ x\ IR\ x\ ED\ x\ EF}{BW\ x\ AT_{nc}} \quad (1)$$

$$\text{HQ (Ingestion)} = \frac{ADD}{RfD} \quad (2)$$

Carcinogenic health risk

Carcinogenic health risk was determined as a lifetime cancer risk (LCR) where the lifetime average daily dose (LADD) times by cancer slope factor (CSF). The LADD was calculated as in Eq. (3). CSF is used to estimate the risk of cancer which is associated with the exposure to a carcinogenic or potentially carcinogenic substance. CSF for each element is expressed in units of proportion of a population affected per milligrams per kilogram of bodyweight per day (mg/kg/day) (Table-3). Risk between $1\ x\ 10^{-6}$and $1\ x\ 10^{-4}$is considered as acceptable risk while risk higher than $1\ x\ 10^{-4}$ is considered as significant risk.

$$\text{LADD} = \frac{C_S\ X\ IR\ x\ EDc\ x\ EF}{BW\ x\ CSF} \quad (3)$$

$$\text{LCR} = \text{LDD x CSF} \quad (4)$$

Table-2. The description of the variables in the health risk assessment.

Symbol	Description	Value	Unit	Source
HQ	Hazard quotient	-	-	-
ADD	Average daily dose	-	-	-
Cs	Concentration of elements	-	ppm	From this study
Ing	Ingestion rate (child)	0.0002	kg/day	Grzetic&Ghariani (2008)
EDnc	Expected exposure Duration for non-carcinogenic	6	years	Grzetic&Ghariani (2008)
EDc	Expected exposure Duration for carcinogenic	70	years	USEPA 2009
EF	Exposure frequency	365	Days/year	USEPA 2009
ATnc	Average period of exposure for non-carcinogen element	ED_{nc} x 365 days/year = 2190	days	Grzetic&Ghariani (2008)
ATc	Average period of exposure for carcinogen element	ED_c x 365 days/year = 25550	days	Grzetic&Ghariani (2008)
BW	Body weight	15	Kg	Grzetic&Ghariani (2008)
RfD	Reference dose	For ingestion	mg/kg-day	RAIS (2013)
CSF	Cancer slope factor		(mg/kg-d)	RAIS (2013)

Table-3. Oral reference dose (RfD) for non-carcinogenic risk and cancer slope factor.

Heavy metals	**RfD** (mg/kg-d)	Source	Heavy metals	**CSF** (mg/kg-d)	Source
Sb	$4.0x10^{-4}$	RAIS (2013)	As	1.50	RAIS (2013)
Ba	$6.0x10^{-1}$	RAIS (2013)	Cd	$3.8x10^{-1}$	Walkes&Rehm (1994)
Cu	$4.0x10^{-2}$	RAIS (2013)	Cr	$5.00x10^{-1}$	RAIS (2013)
Mn	$1.4x10^{-1}$	RAIS (2013)	Co	3.00^{a}	RAIS (2013)
Hg	$1.0x10^{-4}$	RAIS (2013)	Ni	$2.6x10^{-1\ a}$	CaIEPA (1997)
Se	$5.0x10^{-3}$	RAIS (2013)	Pb	$8.50x10^{-3}$	RAIS (2013)
Sr	$6.0x10^{-1}$	RAIS (2013)			
Sn	$3.0x10^{-4}$	RAIS (2013)			
Zn	$3.0x10^{-1}$	RAIS (2013)			

Source: US EPA = United State Environmental Protection Agency, 2009 [23], RAIS= The Risk Assessment Information System, 2013, CaIEPA=California Environmental Protection Agency 1997 [45], Walkes&Rehm, 1994 [48], [a] Inhalation Cancer Slope Factor

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3. RESULTS

3.1 Heavy metals concentration by toy categories and materials

Most of the samples in this study were detected with Zn (97%, N = 41), Sr (90%, N = 38) and Cu (79%, N = 33) and half of the samples were detected with Ni (64%, N = 27), Cr (59%, N=25), Ba (57%, N = 24) and Pb (50%, N = 21). One third of the samples were detected with Mn (43%, N=18) and As (24%, N = 10). Other metals such as Cd, Hg, and Sn also detected in the samples. Only Cd, Co and Pb exceed the EN 71-3: 2013 Category III limit (Table-4).

Ba was high in most of the toy material such as polymer (2255.00 ppm), printed ink (1698.00±758.02 ppm), plastic (1160.82±898.06 ppm) and textile (1284.00 ppm) (Table-4). Mn was high in metal (1707.50±761.55 ppm), paint coated (786.43±658.84 ppm) and paperboard material (110.62±69.20 ppm). Zn was detected in metal (465.67±446.58 ppm), plastic (327.13±567.79 ppm) and paperboard (139.80±112.38 ppm).

Co (3690.00 ppm), Cu (449.57± 460.00 ppm), Ni (257.00 ppm) and Pb (171.67± 83.11 ppm) were high in metal toys. Pain coated material was high with Ni (223.60± 154.15 ppm), Pb (169.93±130.29 ppm), Cr (215.48±160.93 ppm) and Hg (66.90 ppm). Printed ink (574.85±773.79 ppm) and paper (148.00 ppm -1 sample) materials were detected with high Cu. Plastic toys contain high Sb (221.75±233.70 ppm) and Pb (109.85±216.81 ppm). Cd also was detected in metal (28.30 ppm) and plastic (26.07 ±24.35 ppm) toys. Se was detected in metal (33.40 ppm) and plastic (11.90±14.22 ppm).

Most of the heavy metals were detected in plastic toys with 14 elements followed by metal (13 elements). The least material detected with metals was paper with only 5 elements (Figure-1).

Table-4. Mean (±SD) heavy metal concentration (ppm) by toy materials (N=42).

Metals	Toy materials								No of samples detected (n) with metals	EN 71-3: 2013 Category III (mg/kg)	No. of Samples exceed EN 71-3: 2013 Category III limit
	Paint coated (n = 4)	Printed ink (n = 2)	Polymer (n = 3)	Textile (n = 2)	Paper (n = 2)	Paperboard (n = 5)	Metal (n = 3)	Plastic (n = 21)			
Sb	ND	ND	ND	74.90	ND	ND	ND	221.75±233.70	3	560.00	-
As	16.25±8.70	ND	ND	ND	ND	1.70±0.56	ND	23.42±26.30	10	47.00	-
Ba	599.50±374.06	1698.00±758.02	2255.00	1284.00	ND	221.75±67.69	190.00	1160.82±898.06	24	56 000	-
Cd	ND	6.60	ND	ND	ND	5.00	28.30	26.07±24.35	6	23	3
Cr	215.48±160.93	30.60	30.70	62.35±70.22	14.60	41.32±20.57	116.85±117.59	94.40±186.29	25	-	-
Co	ND	ND	ND	ND	ND	ND	3690.00	ND	1	130.00	1
Cu [a]	210.70±230.31	574.85±773.79	6.10	23.35±17.04	148.00	61.94±34.88	449.57±460.00	32.85±44.25	33	7 700	-
Pb [b]	169.93±130.29	ND	3.60	1.50	ND	11.44±6.00	171.67±83.11	109.85±216.81	21	160.00	3
Mn	786.43±658.84	ND	ND	8.80	78.85±10.68	110.62±69.20	1707.50±761.55	53.74±30.09	18	15 000	-
Hg	66.90	2.90	ND	ND	ND	2.50	15.60	7.10±3.59	8	94	-
Ni [c]	223.60±154.15	ND	3.80	4.80±0.42	ND	7.40±3.54	257.00	4.99±1.97	27	930	-
Se	ND	ND	ND	ND	ND	ND	33.40	11.90±14.22	4	460	-
Sr	87.45±118.16	34.30±38.89	31.43±10.00	29.70±37.34	52.30±4.81	49.86±16.72	19.80	46.53±48.86	38	56 000	-
Sn	64.10	113.00	ND	ND	ND	ND	58.50	47.67±19.73	6	180 000	-
Zn	157.58±65.21	62.15±64.84	24.85±29.63	38.55±13.22	33.95±21.85	139.80±112.38	465.67±446.58	327.13±567.79	41	46 000	-

Note: ND-Not Detected, [a]Kruskalwallis test significant at X^2=16.076, p = 0.024,[b]Kruskalwallis test significant at X^2=11.166, p = 0.048,
[c]Kruskalwallis test significant at X^2=- 11.896, p = 0.036

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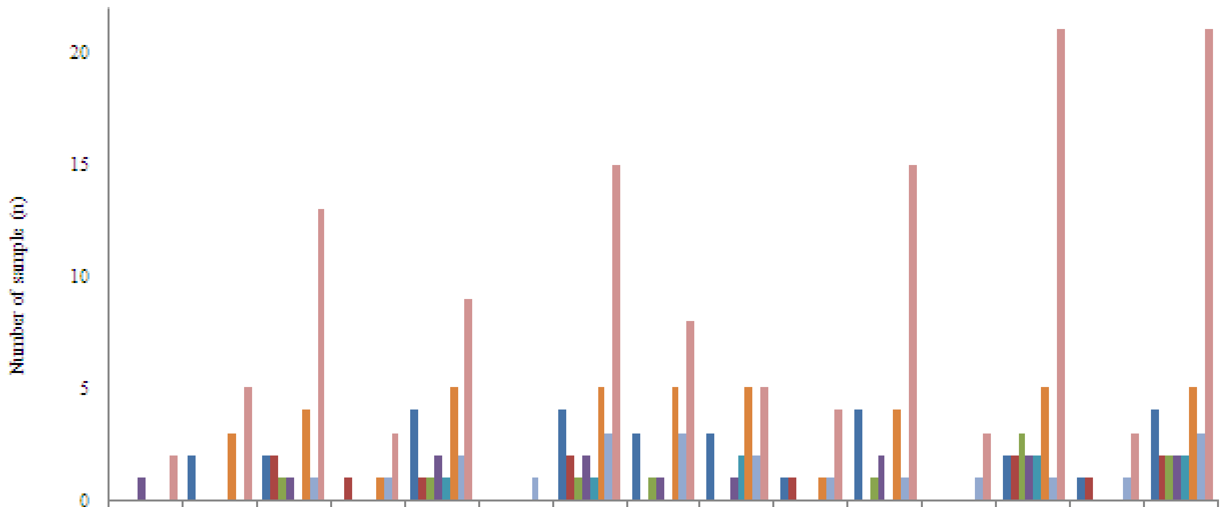

	Sb	As	Ba	Cd	Cr	Co	Cu	Pb	Mn	Hg	Ni	Se	Sr	Sn	Zn
Paint Coatings (n=4)		2	2		4		4	3	3	1	4		2	1	4
Printing Ink (n=3)			2	1	1		2			1			2	1	2
Polymer (n=3)			1		1		1	1			1		3		2
Textile (n=2)	1		1		2		2	1	1		2		2		2
Paper (n=2)					1		1		2				2		2
Paperboard (n=5)		3	4	1	5		5	5	5	1	4		5		5
Metal (n=3)			1	1	2	1	3	3	2	1	1	1	1	1	3
Plastic (n=21)	2	5	13	3	9		15	8	5	4	15	3	21	3	21

Figure-1. Number of samples detected with heavy metals in different toy materials (N = 42).

3.2 Health risk of heavy metals in toy samples

Figure-2 shows the mean HQ and LCR for all metals by toy materials. Only 3 metals were determined with HQ >1 which is Sb in textile (HQ = 2.39) and plastic (HQ = 4.73± 6.68), Hg in paint coated (HQ = 8.55) and metal (HQ = 1.99) and Sn in paint coated (HQ = 2.732), printed ink (HQ = 4.82) and plastic (HQ = 1.52±1.23). This indicates a significant health risk from heavy metals exposure in toys. HQ for other metals was < 1 indicates no significant risk. All metals studied were within a range of generally acceptable risk for cancer except for Co (1.30E-02) and Ni (2.90E-02±4.10E-02) both found in metal.

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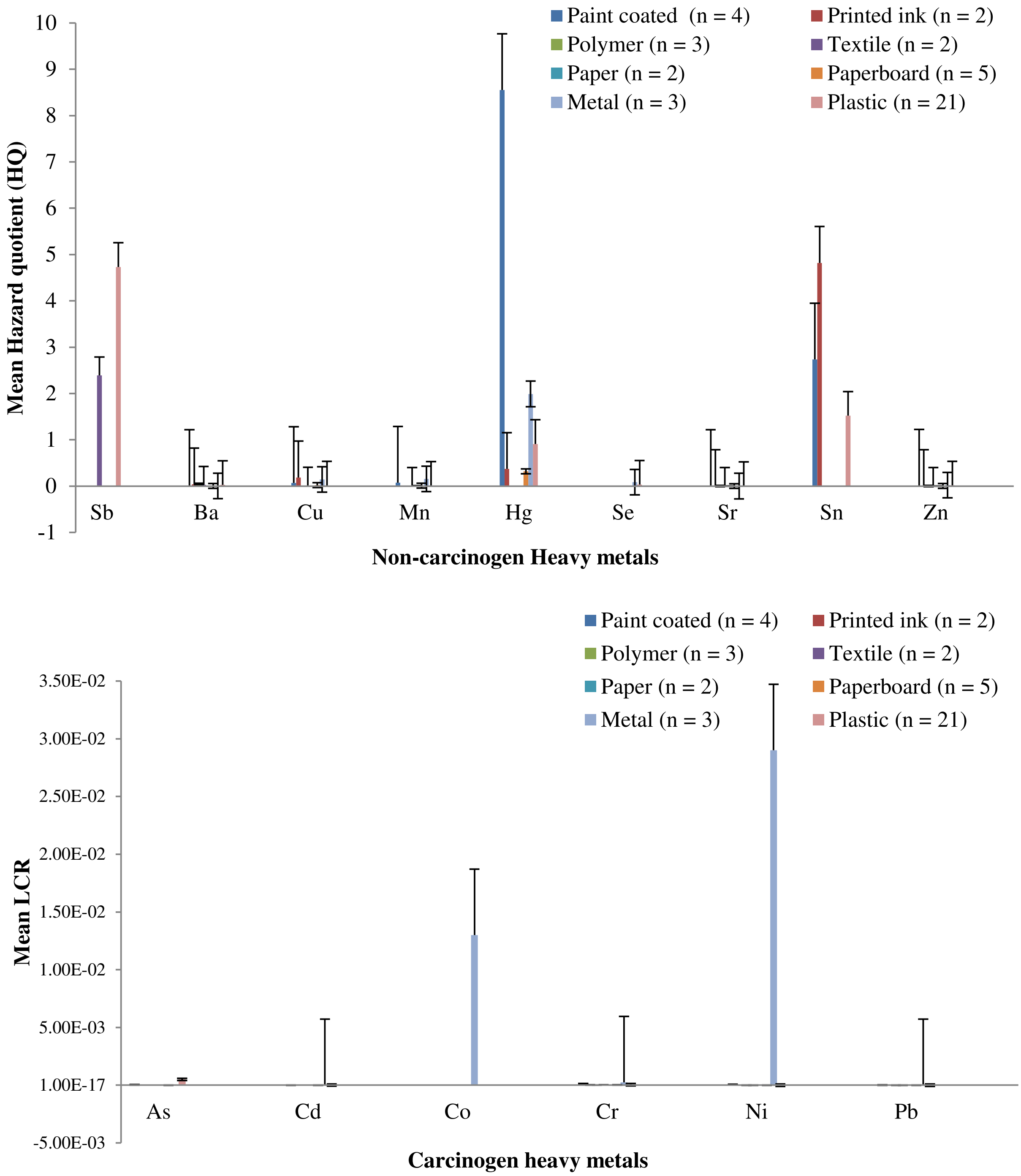

Figure-2. The mean HQ and LCR for heavy metals exposure by toy material.

4. DISCUSSIONS

Ba was commonly detected and among the highest in all toy categories in this study. This element also highly detected in toys made of polymer, printed ink, plastic and textile. Toys may contain barium sulfate ($BaSO_4$) synthetic which is used as a component of white pigment for paints and often mixed with other colors to vary the shade [29]. This possibly explains why Ba was commonly detected in printed ink and textile toys. $BaSO_4$ is also filler for plastics to increase the density of the polymer. For example, 70% of $BaSO_4$ is used as filler in polypropylene and polystyrene plastics [30]. This causes high Ba content in plastic and polymer toys.

Mn and Zn also among the highest metals detected in this study. It was highly detected in technical, physical and intellectual toy categories. These elements were high in metal, paint coated, plastic and paperboard material. Pain coated material also detected with high Ni, Pb and Hg. Previous studies had proven that the concentration of Pb is significant in paint coated toys. Based on Omalaoye *et al.*, (2010), 17% of the toys samples were detected with high level of Pb, Cr, and Cd. According to Van Alphen, (1999) there is a correlation between toy colour and Pb concentration. Most of the yellow samples in his study were determined with the highest concentration of Pb attributed mainly to lead (II) chromate ($PbCrO_4$) or chrome yellow used as a pigment [31].

Toys made of metal material in this study contain high Co, Zn, Cu, Ni and Pb. These toys usually came as silver, grey, and white color that may contain high heavy metals. Cu is significantly high in intellectual and ink printed toys. This can be due to the fact that Cu is one of the elements that is use to make printing inks. Besides, Cu used to dominate the printed conductive electronic market [32].Pb, Mn, Zn, Cr, Cd, and Cu also used as coloring agents, catalysts, or filling material [8, 9].Co and Mn also the main additives used in paints [27, 33]. This explains why heavy metals were obviously high in the toy samples in this study.

Se also was detected in metal and plastic toys in this study. Se is used as a pigment (coloring agents) for paints, plastics, ceramics, and glazes [34]. Depending on the form of selenium used the color ranges from deep red to light orange. It can be used to decolorize glass and to impart a scarlet red color to glass, glazes, and enamels. High Cr was determined in paint coated material toy in this study. According to Adelantado *et al.* (1993), Cr is mainly concentrated in the toy's paint rather than the plastic itself. Ba with Cr and Pb were combined to be used as colorants or pigments [3]. Printed ink and paper material contain high Cu which mainly used as a pigment in paints [8, 9].

Toys are made from many types of plastic and one of the most used plastic materials is polyvinyl chloride (PVC) [11]. Half of the samples in this study were made of plastic (N=21).Toys made of plastic in this study contain high Zn, Sb, Pb and Cd. Zn is mainly used as a stabilizer and catalyst in PVC [8, 9]. PVC is the potential source of heavy metals poisoning as Pb and Cd were added as stabilizers to prevent free chlorine radicals in PVC from reacting with hydrogen radicals to form hydrochloric acid. Hydrochloric acid weakens the structure of PVC [10]. In addition, lead sulfate ($PbS0_4$) and lead oxide (PbO) were mixed with the lead chromate ($PbCr0_4$) a standard colorant for plastic toys [35].Cd is used as a thermal stabilizer for polyvinyl chloride (PVC) plastic during the manufacturing process. It replaces labile chlorine atoms in the polymer and modifies chain reactions. It inhibits the elimination of hydrogen chloride and interrupts the formation of polyene sequences in the polymer which is attributed to its formation of the gradual demise of yellow and its conversion to orange and then red and brown [36].This causes high Pb and Cd in plastic toys. Cr and Pb also are used as stabilizer in toys to enhance its material properties and also to reduce cost on plastics [3, 37, 38]. Cr is widely used as a valuable dye and as a coloring for PVC material, as well as heat stabilizer. Samples with high Pb concentrations also often contained Cr. This could be attributed to that both elements were used as dyes or pigments rather than stabilizers and the combination of these elements was lead chromate.

When Pb is banned under certain regulations, Zn is used to replace the function of Pb to preserve the structure of PVC [4]. This explain why Zn presence in plastic toys. Zinc could be used in toys in the form of zinc borate or zinc omadine (zinc 2-pyridinethiol-1-oxide). Zinc borate is used alone or in combination with other halogen synergists, such as antimony trioxide as a fire retardant additive for PVC products and as an anticorrosive pigment in coatings [39]. Zinc omadine is used as antimicrobial agent in PVC plastics [40]. Due to its low solubility in water (8 mg·L-1 at neutral pH), its decomposition by ultraviolet light is slow, providing years of protection even against direct sunlight.

Arsenic (As) was detected in paint coated material, paperboard and plastic. It was detected in 23% (N = 10) of the sample in this study. This possibly related to some pigments that may contain as or some of its compounds used. For example, despite of its toxicity, Paris green (copper acetoarsenite) was used as a blue colorant pigment for fireworks [26].

Hg was detected in pain coated material in this study. Hg is used as a catalyst to enhance chemical reaction among ingredients [12]. There could be a possibility that both Mn and Hg were used as stabilizer or colorant for toys. Most manufacturers of PVC around the world use natural gas or petroleum as the feedstock or raw material from which the plastic is manufactured. However, most plastic manufacturing in China uses a different process that starts with coal as the feedstock. In that coal-based process, Hg is used as a catalyst to spark the chemical reaction among ingredients [12].

As for health risk assessment in this study, only Ni and Co poses carcinogenic health risk among children via inhalation. Children may be exposed to the allergic reaction cause by the effect of Ni [24]. Co may cause asthma, alveolitis, and occasionally, fibrosis [41]. As for non-carcinogenic health risks, Hg, Sb and Sn were the elements that produce significant health risk. Accidental ingestion of Sb can lead to headache, abdominal pain, constipation, colic, and loss of appetite among children [42]. A study conducted by Kimbrough (1976) has shown ingestion of Sn can had caused skin irritation in rabbits. Exposure to Hg is linked with neurodevelopment problems and possible autistic disorders in children [44].

Although other heavy metals determined in this study did not show significant health risk as these elements are still possible to create adverse health effects among children. This is because bioaccumulations of heavy metals from toys still happen. Children spent long hours playing with toys. Half of their childhood is surrounded by toys. This can cause metals accumulation in the children's body. Bioaccumulation is not only exposed through ingestion, but through inhalation and dermal contact as well [16].

There were several limitations experienced while conducting this study. Due to financial constraint, this study was only performed to small samples of toys which cannot be generalized for the whole toys in Malaysian market. Besides, the toy samples were taken purposively from Selangor area only which may not represent the

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whole country. Another limitation would be the health risk calculation was based on the secondary default data in Grzetic&Ghariani, (2008). The default data used was children average body weight (15 kg), expected exposure duration, average period of exposure etc. These default values may not represent the children in Malaysia accurately. However, the health risk results are still valuable as preliminary baseline information for the state. Another limitation in this study is the used of High Definition X-ray Fluorescence (HDXRF®) technique. This machine has low detection limit which unable to detect trace element at very low concentration and its measure the total content of heavy metals in toys. Even though this information is important, but metals in a form of ion is best to represent the bioavailability in toys and its exposure to children via chewing or licking.

Thus, based on these limitations, we would like to recommend for future study to consider a bigger sample size to be generalized for the whole community. This is very important to ensure compliance from of the manufacturers and importers towards local regulation Consumer Protection (Safety Standards for Toys) Regulation 2009 where the permissible concentration of heavy metals in toys is addressed. Future studies may also include other heavy metals in the assessment to identify the health risks concerning in toys. The health risk calculation should be calculated based on primary data of children characteristics in the study area. Data such as body weight, amount of toys played, duration of exposure, period of exposure etc can be collected through a survey. In terms of the instrumentation, a high detection limit instrument should be used such as ICP/MS. A high detection limit instrument can produce a more reliable and accurate data for the study. Future study also should consider determining the metal availability instead of the total content of metals as studied in this article.

CONCLUSIONS

In conclusion, most of the samples in this study were detected with Zn (97%, N = 41), Sr (90%, N = 38) and Cu (79%, N = 33) and half of the samples were detected with Ni (64%, N = 27), Cr (59%, N=25), Ba (57%, N = 24) and Pb (50%, N = 21). One third of the samples were detected with Mn (43%, N=18) and As (24%, N = 10) while other metals such as Cd, Hg, and Sn also detected in the sample. Only Cd and Pb exceed the EN 71-3: 2013 Category III limit in 3 samples while Co exceeds the limit in 1 sample. Physical toy was highly detected with metals (14 elements) versus only 9 elements were detected in social toy category.

The highest element detected in all toy categories was Ba which is in creative, physical and social toy categories. Ba also was high in most of the toy material such as polymer, printed ink, plastic and textile. Mn and Zn also among the highest metals detected in this study. These elements were high in metal, paint coated, plastic and paperboard material. Other element such as Co, Pb, Cu and Cr also were among the highest detected in the sample.This study revealed that plastic and metal toys available in Malaysian markets contained 14 and 13 metals elements respectively. The least material detected with metals was paper with only 5 elements. Most of the reason of this was possibly due to colour pigment and stabilizer agents in plastic material.

Co and Ni are of worrying concern in this study as it poses carcinogenic health risk whereas Hg, Sb and Sn pose non-carcinogenic health risks. Other elements were within an acceptable range of carcinogenic and non-carcinogenic health risk among children. Further study is needed to assess metals present in toys. Purchasing toys that are made from plastic and metal material seems to be high risk with high metals contaminant.

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