UNIVERSITI PUTRA MALAYSIA

SELECTED HEAVY METAL IN TOYS AND ITS HEALTH RISK AMONG CHILDREN: A CASE STUDY IN SELANGOR, MALAYSIA

NURUL SYIFAA MOHAMAD

SELECTED HEAVY METAL IN TOYS AND ITS HEALTH RISK AMONG CHILDREN: A CASE STUDY IN SELANGOR, MALAYSIA

BY

NURUL SYIFAA MOHAMAD

Thesis submitted in fulfillment of the requirement for the degree of Bachelor Science (Environmental and Occupational Health) from the Faculty of Medicine and Health Sciences, Universiti Putra Malaysia

ACKNOWLEDGEMENTS

First of all, I would like to say Alhamdulillah, for giving me the strength and health to finish up my final year project report until it is done. Not forgotten to my family for providing everything, such as money, to buy anything that are related to this project and their advice, which is the most needed for this report. They also supported me and encouraged me to complete this task so that I will not procrastinate in doing it.

Then I would like to thank my Supervisor, Dr Sharifah Norkhadijah bt Syed Ismail and Dr. Karmegam a/l Karuppiah for their guidance, patience, financial and continuous support to complete my final year project.

I dedicate this thesis especially to myself, for not giving up on the obstacles faced to conduct this study and make it happen.

Last but not the least, my family and the one above all of us, the omnipresent God, for answering my prayers for giving me the strength to plod on despite my constitution wanting to give up and throw in the towel, thank you so much Dear Lord.

ABSTRACT

SELECTED HEAVY METAL IN TOYS AND ITS HEALTH RISK AMONG CHILDREN: A CASE STUDY IN SELANGOR, MALAYSIA

NURUL SYIFAA MOHAMAD

Introduction: Heavy metals in toys pose acute and chronic health risks to children. To date, there is no established data that indicates the safety status of toys in Malaysia market. **Objective:** This study determines the level of selected heavy metals in toys and its health risk to children. **Methodology:** Sampling was performed between February and March 2016 from various toy shops in Selangor. 42 toys were sampled by types (i.e. physical, intellectual, technical, creative, and social) and materials (i.e. paint, ink, polymer, textile, paper, paperboard, metallic, and plastic). The heavy metals concentration was determined by using High Definition X-ray Fluorescence (HDXRF®) technique (Model 800701-01). The health risk was calculated by using USEPA method. **Results:** Fifteen (15) heavy metals were detected in this study. Ba (11492.27±36753.53 mg/kg), Mn (467.06±738.22 mg/kg) and Cu (184.16±293.43 mg/kg) were significantly higher in intellectual toys. The intellectual toys were made from metallic materials which contain significantly higher concentration of Cr (432.90±553.69 mg/kg), Pb (171.67±83.11 mg/kg), and Ni (67922.67±117423.01 mg/kg). The intellectual toys were also made from printing inks and paint which contain significantly higher concentration of Cu (574.85±773.79 mg/kg) and Mn (589.83±666.33 mg/kg) respectively. There were positive correlation between Ba and Cr ($r = 0.80$), Cu ($r = 0.56$), Hg ($r = 0.41$), and Mn ($r = 0.71$). Mn has moderate correlation with Cu ($r = 0.61$), while Ni has moderate correlation with Cd ($r = 0.42$). Pb also has moderate correlation with Mn ($r = 0.44$). Only Ni (1.62×10^{-2}) and Co (3.61×10^{-3}) pose carcinogenic health risk through inhalation while Sb (HQ=4.03) and Sn (HQ=7.78) pose non-carcinogenic health risk through accidental ingestion. Other concentration of heavy metals detected in toy samples does not pose carcinogenic and non-carcinogenic health risk among children (As, Ba, Cd, Cr, Cu, Pb, Mn, Hg, Se, Sr, and Zn). **Conclusion:** The concentration of heavy metals in toys varied and strongly influenced one another. Children are at high health risk to expose to heavy metals via toys.

Keywords: heavy metals, toys, health risk, children

ABSTRAK

LOGAM BERAT TERPILIH DALAM MAINAN DAN RISIKO KESIHATAN DALAM KALANGAN KANAK-KANAK: SATU KAJIAN KES DI SELANGOR, MALAYSIA

NURUL SYIFAA MOHAMAD

Pengenalan: Logam berat dalam mainan menimbulkan risiko kesihatan yang akut dan kronik kepada kanak-kanak. Setakat ini, tidak ada data yang menunjukkan status keselamatan mainan di pasaran Malaysia. **Objektif:** Kajian ini menentukan tahap logam berat dalam mainan dan risiko kesihatan kepada kanak-kanak. **Metodologi:** Persampelan dilakukan antara Februari dan Mac 2016 dari pelbagai kedai mainan di Selangor. 42 mainan telah disampel berdasarkan jenis (iaitu fizikal, intelektual, teknikal, kreatif, dan sosial) dan bahan-bahan (iaitu cat, dakwat, polimer, tekstil, kertas, papan kertas, logam, dan plastik). Kepekatan logam berat telah ditentukan dengan menggunakan definisi tinggi X-ray pendarfluor (HDXRF®) teknik (Model 800.701-01). Risiko kesihatan telah dikira menggunakan kaedah USEPA. **Keputusan:** Lima belas (15) logam berat telah dikesan dalam kajian ini. Kepekatan Ba (11492.27 ± 36753.53 mg/kg), Mn (467.06 ± 738.22 mg/kg) dan Cu (184.16 ± 293.43 mg/kg) adalah tinggi dalam permainan jenis intelektual. Mainan intelek dibuat daripada bahan-bahan logam yang mengandungi kepekatan tinggi Cr (432.90 ± 553.69 mg/kg), Pb (171.67 ± 83.11 mg/kg), dan Ni (67922.67 ± 117423.01 mg/kg). Mainan intelek juga diperbuat daripada dakwat cetakan dan cat yang mengandungi kepekatan tinggi Cu (574.85±773.79 mg/kg) dan Mn (589.83±666.33 mg/kg). Terdapat korelasi positif antara Ba dan Cr ($r = 0.80$), Cu ($r = 0.56$), Hg ($r = 0.41$), dan Mn ($r = 0.71$). Mn mempunyai hubungkait sederhana dengan Cu ($r = 0.61$), manakala Ni mempunyai hubungan yang sederhana dengan Cd ($r = 0.42$). Pb juga mempunyai hubungan yang sederhana dengan Mn ($r = 0.44$). Hanya Ni ($1.62x\ 10^{-2}$) dan Co ($3.61x10^{-3}$) menimbulkan risiko kesihatan karsinogenik melalui inhalasi manakala Sb (HQ = 4.03) dan Sn (HQ = 7.78) menimbulkan risiko kesihatan bukan karsinogenik melalui oral. Kepekatan logam berat lain dalam sampel mainan tidak menimbulkan risiko karsinogenik atau bukan karsinogenik kepada kanak-kanak (As, Ba, Cd, Cr, Cu, Pb, Mn, Hg, Se, Sr, and Zn). **Kesimpulan:** Kepekatan logam berat dalam mainan adalah pelbagai dan dipengaruhi antara satu sama lain. Kanak-kanak yang terdedah kepada logam berat melaui permainan mempunyai risiko kesihatan yang tinggi.

Kata Kunci: logam berat, mainan, risiko kesihatan, kanak-kanak

TABLE OF CONTENTS

CHAPTER 4: RESULTS AND DISCUSSION

CHAPTER 5: CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

USEPA	United States Environmental Protection Agency
ADD	Average Daily Dose
HQ	Hazard Quotient
RFD	Reference Dose
RAIS	The Risk Assessment Information System
LADD	Lifetime Average Daily Dose
CSF	Cancer Slope Factor
CAIEPA	California Environmental Protection Agency
ISO	International Standard Organization
EU	European Union
Sb	Antimony
As	Arsenic
Cd	Cadmium
Cr	Chromium
Co	Cobalt
Cu	Copper
Pb	Lead
Mn	Manganese
Hg	Mercury
Ni	Nickel
Se	Selenium

Sr	Strontium
Sn	Tin
Zn	Zinc
UNEP	United Nations Environmental Program
ICP/MS	Inductively Coupled Plasma Mass Spectrometry

CHAPTER 1

INTRODUCTION

1.1 Background

Toys play an important role in the development of children in their young age. Toys not only bring fun, joy and entertainment to children but it also play a vital role in the development of physical, intelligence, and social abilities in children. , Under the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009, a "toy'' is defined as any goods designed or intended for the use in play by children below fourteen years of age. According to International Standard Organization 8124-1: 2014 Safety of Toys Part 1, a toy is any product or material that is designed intended for the use in play by children under fourteen years of age.

Toys should be made up and composed of material that is safe, not harmful, and not toxic for the children. For example, manufacturers supposedly not using the paint for building and furniture to paint toys as this type of paint are inorganic pigment that contains heavy metals. Materials and paints are loosely bound to the surface and can leach easily (Omolaoye et al., 2010). Manufacturers should consider the intellectual ability of children that is developing and most toys are misused by children with the act of licking, sucking, and biting of toys happens. Besides, a child spends most of their time holding and surrounding themselves with toys which can expose them to heavy metal toxicity by means of inhalation, ingestion, or dermal.

Heavy metal toxicity has become a serious health issue. This is because it has a density of at least 5 times that of water and it cannot be metabolized by the body. This can lead to the accumulation of heavy metals in body. Heavy metal poisoning can disrupt the functions of our mental, energy, nervous system, kidneys, lungs and other organs (Duffus, 2002).

According to Australian/ New Zealand Standard Safety of Toys Part 3: Migration of certain elements AS/NZS ISO 8124.3:2003, heavy metals that is a concern are antimony, arsenic, barium, cadmium, chromium, lead, mercury, and selenium. In the environment, heavy metals have been closely related to chronic diseases that are on the rise which includes breast cancer, learning and developmental disabilities, and reproductive health problems (Al-qutob et al., 2014). Children are vulnerable population to heavy metals toxicity.

In Malaysia, in exercise of the powers conferred by Section 19 of the Consumer Protection Act 1999 [Act599], the Minister had made the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009. This regulation had come into force on 30 January 2010. This regulation had imposed the safety standards for toys according to 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 this Act (Consumer Protection Act). 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 2010. This guideline functions to assist the businesses that supply toys in Malaysia to ensure compliance with the legislation and requirements by the Ministry. This guideline helps the industry to understand the main features of the legislations and requirements to comply the safety standards.

1.2 Problem Statement

The news of toys contaminated with the traces of heavy metals and other dangerous substances that are in dangerously high concentration continue to make alarming headlines to public. Heavy metals including lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), selenium (Se), mercury (Hg), barium (Ba), and zinc (Zn) are presence in these toys (Al-Qutob et al., 2014). These elements were added to enhance certain characteristics of toys such as softness, brightness, and flexibility to make the toys more attractive and fun for the children to play. The concern of this issue is that these heavy metals can migrate from toys when they are chewed or sucked by children, thus posed chronic effects in the long term (Al-Qutob et al., 2014). Not only that, the heavy metals may pose threat to children through inhalation (breathing) (Gordon, 2002).

Cases of heavy metal poisoning were widely reported all around the globe. A study conducted by Abdullah (2015) found out that inexpensive and yellow colour toys showed high lead concentration, and exceeded the permissible level. In North America, childhood lead poisoning linked to the ingestion of old paint chips (Adal et al., 2015). Another case of heavy metal poisoning that happened in Fengxiang County, China, 174 children from three villages were diagnosed with lead toxicity. The diagnose showed that 851 out of 1,016 children tested found to have abnormally high levels of lead in their body (Jia, 2009). Heavy metals have been linked to chronic diseases such as breast cancer, learning disabilities, and reproductive health problems (Al-qutob et al., 2014). For instance, exposure to high levels of lead can damage the brain, kidney, and eventually cause death (Martin & Griswold, 2009). Another example is cadmium. Cadmium is released as fine airborne particles which react with oxygen to form respirable cadmium oxide which may cause disruption in the nephritic system (Kumar & Pastore, 2007).

Over the past decade, regulations regarding potential hazardous elements and compounds such as heavy metals have increased in scope and decreased in permissible limits. Researchers are focusing more on children's consumer products due to its ability in exposing infants and children to these dangerous elements. In Malaysia, in exercise of the powers conferred by Section 19 of the Consumer Protection Act 1999 [Act 599], the Minister had made the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009. This regulation had come into force on 30 January 2010. This regulation had imposed the safety standards for toys according to 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 this Act (Consumer Protection Act 1999).

Ever since the enforcement dates of the amended local regulation, 30 January 2010, there is no established data that indicates the safety status of toys in the Malaysia market. There is a need to check whether the business manufacturers are complying the safety standard and level that is provided by the regulation. It is important to ensure that the concentration of heavy metals present in toys is safe as what is addressed by the law. This is because the amount of heavy metals in toys poses an acute and chronic health risks to the children. 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. Therefore, it is important to assess the concentration of heavy metals in toys by types of toys (i.e physical, intellectual, technical, social, and creative) and by toy materials (i.e paint coated toys, ink printed, polymer, textile, paper, paperboard, metallic, and plastic).

1.3 Research Justification

Toys should be made up from materials that are not harmful and toxic. The act of licking, sucking, and biting among children exposed them to harmful heavy metals in dangerous concentration. The concentration of heavy metals permitted by a child on a daily basis is estimated at 8 mg per day (Coelho, 2015). Heavy metals can migrate from toys when they are chewed and sucked by children, thus pose chronic effects in the long term. Furthermore, these toys are kept from one generation to another (for next child) making it more exposed to the heavy metals in the environment and surroundings.

A study shows that age differences affect the susceptibility to lead poisoning in children. In children, alimentary absorption is about 50% and, together with their greater volume of air inhaled in relation to body size, total lead absorption from the environment is around three times higher than for adults. Besides, iron deficiency and low dietary calcium promote the chance to increase rate of lead absorption (Gordon, 2002).

Children will be exposed to potentially harmful levels of heavy metals that may cause deleterious health effects. Thus, the health risks posed by these toys can be determined and used to improve the safety and quality of the toys in the Malaysia market. The result of this study can also be used as baseline information to monitor the quality of toys manufactured in this country so that the safety and health of toys users which is children can be taken care of at same time.

Next, the results obtained can be used to increase and improve the standard of local product so that more people will support the local manufactured toys instead of imported products. In addition, the results obtained can be used to set a new standard for toys manufacturers as the current regulation is using the standards provided by ISO 8124: 2010 Safety of Toys. The findings of this study can also be used to educate parents to be more considerate about the potentially harmful effects from the toys. Last but not least, this study can act as an evidence-based to create preventive action to ensure toy safety in Malaysia.

1.4 Conceptual Framework

There are two independent variables in this study; types of toys and materials that made up the toys. There are five types of toys; toys that are used for physical, intellectual, technical, creative activities and toys that are used to establish social relationship (ISO 8124, 2010). There are different types of toy materials as well. Toys that are paint coated, ink printed, made from polymer, textile, paper, paperboard, metallic, and plastic (ISO 8124, 2010). Toy samples for this study are below RM 10.00 which is considered as inexpensive toys and widely available in the market. The dependent variable for this study would be the concentration of heavy metals and the health risk assessment.

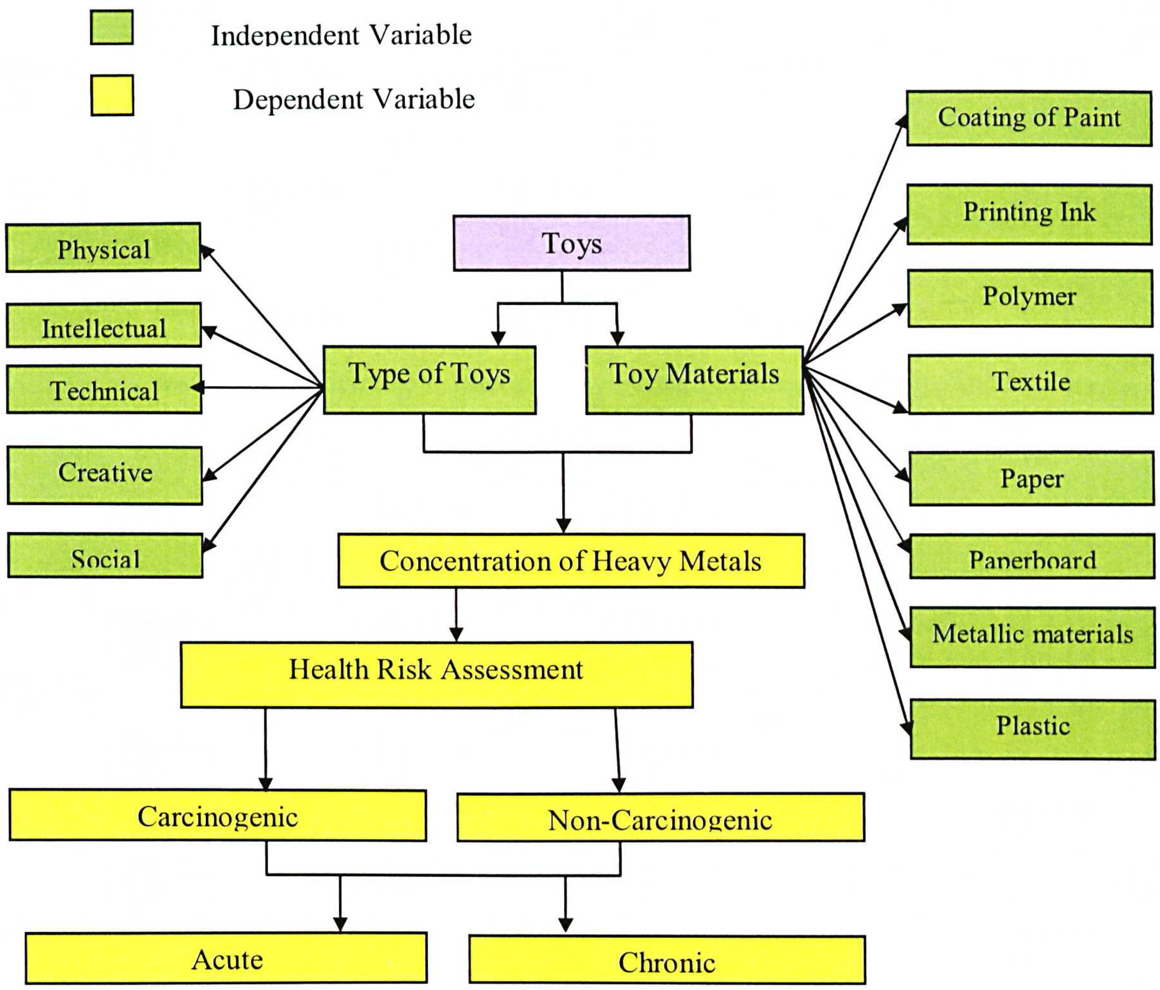

Figure 1.4: Conceptual framework of the study.

1.5 Definition of Terms

1.5.1 Conceptual Definition

i. Heavy Metals

Heavy metals are naturally occurring elements that have a high atomic weight and a density at least 5 times greater than that of water. The toxicity depends on the dose, route of exposure, chemical species and the health status of exposed individuals. Arsenic, cadmium, chromium, lead, and mercury are classified into high degree toxicity and are a concern to impact public health. These elements are believed to cause multiple organ damage, even at low concentration (Tchounwou et al., 2014).

i. Toys

Under the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009, a "toy" is defined as any goods designed or intended for the use in play by children below fourteen years of age.

ii. Health Risk Assessment

A multi-step procedure that comprise of data collection (gathering and analyzing human health data), exposure assessment (estimation of the magnitude of actual and/or potential human exposures), dose response (determination of adverse health effects associated with exposure to different chemicals), and risk characterization (summarizes and combines the results of the calculation of exposure and toxicity assessments whether carcinogenic or non-carcinogenic) (Ćrzetic & Ghariani, 2008).

1.5.2 Operational Definition

i. Heavy Metals

Heavy metals were detected using x ray fluorescent method.

ii. Toys

Any goods designed or intended for the use in play by children below fourteen years of age with price below RM 10.00.

iii. Health Risk Assessment

Health risk of toys among children is calculated by using USEPA (2009) method.

1.6 Objective

1.6.1 General Objective

To assess heavy metals concentration in toys and its health risk to children.

1.6.2 Specific Objectives

1. To compare the concentration of heavy metals in toys samples with the allowable concentration of heavy metals according to ISO 8124 Safety of Toys and European Union (EU) New Toy Safety Directive EN 71-3: 2013.
2. To compare the concentration of heavy metals by types of toys (i.e. physical, intellectual, technical, creative, and social).
3. To compare the concentration of heavy metals by toy materials (i.e. paint coatings, polymers, plastic, paper, paperboard, paper, and others).
4. To determine the association between concentrations of heavy metals in toys.
5. To determine the health risk from heavy metals exposure via toys among children.

1.7 Hypothesis

1. There is a significant difference in the concentration of heavy metals in five types of toys.

   (i.e. Physical, intellectual, technical, creative, and social).

2. There is a significant difference in the concentration of heavy metals in different types of toy materials. (i.e. paint coatings, ink printed, polymers, textile, paper, paperboard, metallic, and plastic).
3. There is a significant association between concentrations of heavy metals in toys.
4. There is a significant health risk from heavy metals exposure via toys among children.

CHAPTER 2

LITERATURE REVIEW

2.1 Toys

Toys must be safe. Toys cannot be toxicant to children. Under the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009, a "toy" is defined as any goods designed or intended for the use in play by children below fourteen years of age. Toys play an important role in the development of children in their young age. Toys not only bring fun, joy and entertainment to children but it also play a vital role in the development of physical, intelligence, and social abilities in children. According to International Standard Organization 8124-1: 2010 Safety of Toys Part 1, a toy is any product or material that is designed intended for the use in play by children under fourteen years of age.

Chemical exposure to children, especially from toys, is an emerging concern. The toys have paint which rubs off from time to time. Metals in materials and paints are loosely bound to the surface and can leach easily. Toys should be made up and composed of material that is safe, not harmful, and not toxic for the children. For example, manufacturers are not using the paint for building and furniture to paint toys as this type of paint is inorganic pigment. This kind of paint contains heavy metals in it. Materials and paints are loosely bound to the surface and can leach easily (Omolaoye et al., 2010). Manufacturers should consider the intellectual ability of children that is developing and children will not play the toys correctly. Therefore, most toys are misused by children whereby the act of licking, sucking, and biting of toys happens. Besides, a child spends most of their time holding and surrounding themselves with toys which can expose them to heavy metal toxicity by means of inhalation, ingestion, or dermal. Lead poisoning from toys causes learning disabilities, kidney failure, anemia and irreversible brain damage in children (WorldNet Daily, 2009).

2.1.1 Toy Types

Based on ISO 8124: 2010 Safety of Toys Part 8: Age Determination Guidelines, there are seven general types of toys that children use that comprise the play category. The categories are according to its own unique purposes and functions. The first category is toys used for sensorimotor activities. This type of toys is usually given to children at their first age to develop their sensorimotor. Second category is toys used for physical activities. Third type is toys used for intellectual activities. Another type is toys that reproduce children the technical world followed by toys that are played to develop feelings and empathy. Sixth, toys for creative activities. Last but not least, toys that are given to children to establish social relationship.

2.1.2 Toy Materials

ISO 8124: 2010 Safety of Toys Part 3: Migration of Certain Elements addresses maximum acceptable levels which are specified for the migration of the elements (Sb, As, Ba, Cd, Cr, Pb, Hg, and Se) that can be present in the following toy materials. The first material is coating of paints, varnishes, lacquers, printing inks, polymers, and similar coatings. Secondly, polymeric and similar materials which include laminates, whether it is textile-reinforced or not, but excluding other textiles and non-woven textiles. Thirdly, paper and paperboard, up to a maximum mass per unit area of 400 g/m^2. Fourth, natural, artificial, or synthetic textiles. Fifth, glass/ ceramic/ metallic, excepting lead solder when used for electrical connections. Sixth, other materials, whether mass-colored or not such as wood, fiberboard, hardboard, bone, and leather. Seventh, materials intended to leave a trace. Eighth, pliable modeling materials which include modeling clays and gels. Last but not least, paints to be used as such in the toy which includes finger paints, varnishes, lacquers, glazing powders, and similar materials in solid and liquid form.

2.2 Heavy Metals in Toys

Toys can made from many types of materials. One of the materials is Polyvinyl Chloride (PVC). PVC is one of the most used plastic materials (Mulder & Knot, 2001). Toys that are made of PVC are a potential source of heavy metal poisoning among children. This is because heavy metal such as lead and cadmium are added to PVC as stabilizers. Stabilizers function to prevent free chlorine radicals in PVC from reacting with hydrogen radicals to form hydrochloric acid. Hydrochloric acid weakens the structure of PVC (Tuczai & Cortolano, 1992).

When lead is banned under certain regulations, zinc is used to replace the function of lead to preserve the structure of PVC or other toy materials (Kumar & Pastore, 2007). Most manufacturers of PVC around the world use natural gas or petroleum as the raw materials. However, in China, where most plastic toys are made uses a different process that starts with coal as its raw material. In the coal-based process, mercury is used as a catalyst to enhance the chemical reaction among ingredients to manufacture the toys. Some of the mercury catalyst is lost during this industrial process. It is poorly understood where the lost mercury ends up, but part of it must have been mixed in the processed material (UNEP, 2008).

Selenium is used as coloring agents for several toy materials such as for paints, ceramics, plastics, and glazes. Selenium produces color that changes from deep red to light orange. It can also be used to decolorize glass and brings a scarlet red color to glass, glazes, and enamels (Al-Qutob et al., 2014).

2.3 Heavy Metals Toxicity

Generally, humans are exposed to heavy metals by ingestion (drinking or eating) or inhalation (breathing) (Martin & Griswold, 2009). Heavy metal toxicity has become a serious health issue in Malaysia. It has been an increasing reported case in other parts of the world, including Malaysia. This major health issue had been a serious concern since the industrial revolution. Heavy metals are worrying concern because it has a density of at least 5 times that of water and cannot be metabolized by the body. This can lead to the accumulation of heavy metals in body. Heavy metal poisoning can disrupt the functions of our mental, energy, nervous system, kidneys, lungs and other organs (Duffus, 2002).

According to Australian/ New Zealand Standard Safety of Toys Part 3: Migration of certain elements AS/NZS ISO 8124.3:2003, the heavy metals include are antimony, arsenic, barium, cadmium, chromium, lead, mercury, and selenium. In the environment, heavy metals have been closely related to chronic diseases that are on the rise which includes breast cancer, learning and developmental disabilities, and reproductive health problems (Al-qutob et al., 2014). Children are vulnerable population to heavy metals toxicity. Heavy metal such as cadmium appears to not cause any vital biological function. However, excessive exposure can damage liver and kidney (Friberg et al., 1974). As for lead, previous studies show that no level of lead in blood is safe or normal (National Referral Centre, 2009). Although, numerous investigations have been carried out on health impacts of lead and other heavy metals on humans, little has been done to ascertain their source in children environment in Nigeria (Needleman, 2007). Heavy metals have mostly been studied in soil, water, paints, and food (Sharma et al., 2005). Toys which are intimately linked to children's environment have not really been investigated as one of the sources of lead, cadmium and other heavy metals.

2.3.1 Adverse Effects of Heavy Metals

Arsenic is odorless and tasteless. Inorganic arsenic 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 arsenic can cause darkening of skin and the appearance of small warts on the palms, soles, and torso. Barium which present 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 barium will cause high blood pressure, changes in heart rhythm, paralysis, and possibly death. Cadmium is human carcinogens than can cause severe damage to lungs though high levels of breathing. Accidental ingesting of very high levels of cadmium irritates the stomach which leads to vomit and diarrhea. Chronic exposure to cadmium causes buildup in the kidneys and possible kidney disease, lung damage, and fragile bones. Chromium 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 lead levels can severely damage the brain and kidneys which eventually cause death (Martin & Griswold, 2009).

2.3.2 Heavy Metals Toxicity Mechanism

Children and pregnant women are particularly susceptible to lead poisoning. The digestive system of children absorbs up to 50% of the lead ingest (National Referral Centre, 2009). Disruption of brain function is one of the adverse effects of lead poisoning. Lead is able to mimic or in certain cases inhibit the action of calcium as a regulator of cell function. At a neuronal stage, exposure to lead can alter the release of neurotransmitter from pre-synaptic nerve endings. The alteration will cause the enhancement in the spontaneous release and inhibition in the evoked release. This will cause disruption of neuronal brain activity, which in turn alter and inhibit the development processes of synapse formation and result in a less efficient brain with cognitive deficits (Bressler & Goldstein, 1991).

Another carcinogenic element is cadmium. Cadmium normally attacks the vital organ kidney. Cadmium accumulates in the kidney as a result of its preferential uptake by receptor-mediated endocytosis of freely filtered and metallothionein bound cadmium (Cd-MT) in the renal proximal tubule. The Cd-MT is then degraded in endosomes and lysosomes which will release free Cd^{2+} into the cytosol. Here, reactive oxygen species (ROS) is generated and cell death pathways are activated. Early cadmium renal toxicity manifestation is impaired reabsorption of low molecular weight proteins which indicates a proximal tubular damage. Continued exposure to cadmium can progress to clinical renal Fanconi syndrome, and eventually renal failure (Johri et al., 2010).

2.4 Laws and Regulation

In Malaysia, in exercise of the powers conferred by Section 19 of the Consumer Protection Act 1999 [Act 599], the Minister had made the Regulation of the Consumer Protection (Safety Standards for Toys) Regulation 2009. This regulation had come into force on 30 January 2010. This regulation had imposed the safety standards for toys according to 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 this Act. (Consumer Protection Act). 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 2010. This guideline functions to assist the businesses that supply toys in Malaysia to ensure compliance with the legislation and requirements by the Ministry.

This guideline helps the industry to understand the main features of the legislations and requirements to comply the safety standards. For the purpose of this guideline, the relevant legislations are as follows:-

(i) Consumer Protection Act 1999 [Act 599];

(ii) Consumer Protection (Safety Standards for Toys) Regulations 2009 [Toys Regulations];

(iii) Consumer Protection (Certificate of Approval and Conformity Mark of Safety Standards) Regulations 2009 [Safety Regulations]; and

(iv) Customs (Prohibition of Import) Order.

2.5 Standard Limit of Heavy Metals in Toys

The concentration of heavy metals in toys can be compared to two standards, namely ISO 8124: 2010 Safety of Toys Part 3: Migration of Certain Elements and European Union New Toy Safety Directive EN 71-3: 2013. ISO 8124: 2010 Safety of Toys Part 3: Migration of Certain Elements is the standard that is used in the local regulation for safety of toys in Malaysia. The ISO standard functions to provide toxicity requirements for toys and toxicity labeling requirements for certain materials used in toys. There are eight permissible levels of heavy metals in toys addressed in the ISO standard. The requirements of this part of ISO 8124 are based on the bioavailability of certain elements which result from the use of toys and should not, as an objective, exceed the following level per day.

Table 2.5.1: Permissible levels of elements addressed in ISO 8124: 2010.

Element	Permissible level (μg)
Sb	0.2
As	0.1
Ba	25.0
Cd	0.6
Cr	0.3
Pb	0.7
Hg	0.5
Se	5.0

The European Union New Toy Safety Directive EN 71-3: 2013 is a new harmonized standard EN 71-3: 2013 under Directive 2009/ 48/ EC, which came into force on July 20, 2013. This standard specifies requirements and test methods for the migration of nineteen elements from materials and parts of toys. Besides, this standard also contains different requirements for the migration of certain elements from the following categories of toy materials. The categories are Category I: Dry, brittle, powder like or pliable materials, category II: liquid or sticky materials, and category III: scrapped –materials.

Table 2.5.2: The heavy metal permissible concentration in EU New Toy Safety Directive EN 71-3: 2013.

Element	EN 71-3: 2013 (Category III) (mg/kg)
Al	70.00
Sb	560.00
As	47.00
Ba	18.75
Bo	15.00
Cd	17.00
Cr	-
Cr (III)	460.00
Cr (VI)	0.20
Co	130.00
Cu	7.70
Pb	160.00
Mn	15.00
Hg	94.00
Ni	930.00
Se	460.00
Sr	56.00
Sn	180.00
Zn	46.00

2.6 Health Risk Assessment

Health risk assessment is a multi-step procedure that comprise of data collection (gathering and analyzing human health data), exposure assessment (estimation of the magnitude of actual and/or potential human exposures), dose response (determination of adverse health effects associated with exposure to different chemicals), and risk characterization (summarizes and combines the results of the calculation of exposure and toxicity assessments whether carcinogenic or non-carcinogenic) (Ćrzetic & Ghariani, 2008).

Previous study has stated that, the estimate value for one day bite for frequent mouthing activity is 8mg (RIVM, 2001). Also, age differences play huge impact to absorption of these heavy metals. The main route of absorption in adults is the respiratory tract where 30–70% of inhaled lead finds its way into the circulation. In children, alimentary absorption is about 50% and, together with their greater volume of air inhaled in relation to body size, total lead absorption from the environment is around three times higher than for adults (Gordon et al., 2015).

Health risk assessment models were developed in Europe (European Chemicals Bureau, 2003). The health risk assessment models were also developed in the United States (USEPA, 1991). However, the European model is under development and not as clear as the American model. Thus, it was decided to apply the American model developed by USEPA. 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.

CHAPTER 3

METHODOLOGY

3.1 Sample Collection

This is a cross-sectional study conducted to assess the health risk of heavy metals exposure in toys among children. Toy samples were grabbed purposively based on five types and eight materials as what were addressed in the regulation to ensure a homogenized samples. The types of toys and materials that were successfully sampled were toys that are used for physical, intellectual, technical, creative, and social activities. There toy samples were toys that were made from various materials such as paint coated toys, ink printed, polymer, textile, paper, paperboard, metallic, and plastic. In total, 42 toy samples were analyzed in this study.

All toy samples were below MYR 10 because the toys were made from China (Holmes, 2011). The toy samples were taken and chosen from two convenience shops that sell toys at Seri Kembangan, Selangor. The toy samples were taken here due to the availability of different and various types and toy materials.

Figure 3.1: Some of the toy samples collected in this study.

3.2 Sample Analysis

Heavy metals concentration in the toys was analyzed using High Definition X-ray Fluorescene (HDXRF®) HDRocksand uses a XOS's (Model 800701-01).XRF technology is a rapid and convenient high technology tool that enables instant detection of all the 8 heavy metals in the Australian Toy Standard AS/NZS ISO 8124.3:2003, which allows many toys to be tested compared to the time and cost that a lab would take to test toys. Savings are significant in both time and money. Furthermore, XRF determines the total amount of presence of heavy metals in a toy sample, which is not the same as the leachable or migratory properties of a heavy metal (Li, Brien, Advice, & Glass, 2011). The toy samples was kept in the original packaging before undergo screening by HD Rocksand to avoid any contamination from the environment. Besides, before the screening process, toy samples are wiped with alcohol swab to avoid contamination that may affect the results.

3.3 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.

3.4 Health Risk Assessment

Both equations for non-carcinogenic and carcinogenic health risk are adapted from USEPA, 2009.

3.4.1 Non-carcinogenic health risk

Average Daily Dose (ADD) is used in the calculation of non-carcinogenic risk. ADD was calculated as follows:

$$ADD = \frac{C \times IR \times EF \times ED}{BW \times AT}$$

Where,

Subject	Value	Unit	Source
C = Average concentration of heavy metal	Depend on results of this research	mg/kg	Based on results of this research
IR = Ingestion rate (child	0.0002	kg/day	Grzetic&Ghariani (2008)
EF = Exposure frequency	350	days/year	USEPA 2009
ED = Exposure duration (child)	6	years	Grzetic&Ghariani (2008)
BW = Body weight	15	Kg	Grzetic&Ghariani (2008)
AT = Average period of exposure (ED*365)	2190	days	Grzetic&Ghariani (2008)

US EPA = United State Environmental Protection Agency (2009)

For non-carcinogenic risk, HQ>1 indicates significant risk while HQ<1 indicates no significant risk. HQ was calculated as follow:

$$HQ = \frac{ADD}{RfD}$$

The Reference Dose (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 from 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. On the other hand, doses more than the RfD are likely to be associated with adverse health risks which is a concern (USEPA, 2015).

Table 3.4.1: Oral Reference Dose (RfD) for non-carcinogenic risk

Heavy metals	**Reference dose, RfD (mg/kg-d)**	**Source**
Sb	$4.0x10^{-4}$	RAIS
Ba	$6.0x10^{-1}$	RAIS
Cu	$4.0x10^{-2}$	RAIS
Mn	$1.4x10^{-1}$	RAIS
Hg	$1.0x10^{-4}$	RAIS
Se	$5.0x10^{-3}$	RAIS
Sr	$6.0x10^{-1}$	RAIS
Sn	$3.0x10^{-4}$	RAIS
Zn	$3.0x10^{-1}$	RAIS

US EPA = United State Environmental Protection Agency (2009)
RAIS= The Risk Assessment Information System (2013)

3.4.2 Carcinogenic health risk

Lifetime Average Daily Dose (LADD) is used in the calculation of carcinogenic risk. LADD were calculated as follows:

$$\text{LADD} = \frac{\text{C x IR x EF x ED}}{\text{BW x AT}}$$

Subject	Value	Unit	Source
C = Average concentration of heavy metal	Depend on results of this research	mg/kg	Based on results of this research
IR = Ingestion rate	0.0002	kg/day	Grzetic&Ghariani (2008)
EF = Exposure frequency	350	days/year	Grzetic&Ghariani (2008)
ED = Exposure duration	6	years	Grzetic&Ghariani (2008)
BW = Body weight	15	kg	Grzetic&Ghariani (2008)
AT = Average period of exposure (70*365)	25550	days	Grzetic&Ghariani (2008)

US EPA = United State Environmental Protection Agency (2009)

Risk between 1 x $10^{-6}$and 1 x $10^{-4}$is considered as acceptable risk. There is a significant risk if higher than1 x 10^{-4}. The carcinogenic risk were calculated as follow:

$$\text{Risk} = \text{LADD} \times \text{CSF}$$

Cancer Slope Factor (CSF) is used to estimate the risk of cancer which is associated with the exposure to a carcinogenic or potentially carcinogenic substance. CSF is expressed in units of proportion of a population affected per milligrams per kilogram of bodyweight per day (mg/kg/day) (USEPA, 2015).

Table 3.4.2: Oral Cancer Slope Factor (CSF)

Heavy metals	**Cancer slope factor, CSF (mg/kg-d)**	**Source**
As	1.50	RAIS
Cd	$3.8\text{x}10^{-1}$	Waalkes & Rehm
Cr	$5.00\text{x}10^{-1}$	RAIS
Co	3.00[a]	RAIS
Ni	$2.6\text{x}10^{-1}$ [a]	CalEPA
Pb	$8.50\text{x}10^{-3}$	RAIS

RAIS= The Risk Assessment Information System (2013)
CalEPA=California Environmental Protection Agency (1997)
Walkes & Rehm (1994)
[a] Inhalation Cancer Slope Factor

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Toy Samples

There were 42 toys sample involved in this study (Table 4.1). The samples were segregated into five types which are physical activity toys (N=10, 23.8%), intellectual (N=11, 26.2%), technical (N=5, 11.9%), creative (N=9, 21.4%), and social (N=7, 16.7%). The toys were also classified into eight materials, namely paint coated (N=4, 9.5%), ink printed (N=2, 4.8%), polymer (N=3, 7.1%), textile (N=2, 4.8%), paper (N=2, 4.8%), paperboard (N=5, 11.9%), metallic (N=3, 7.1%), and plastic (N= 21, 50.0%).

Table 4.1: Toy Samples (N=42)

Types	Frequency (%)	Toy Materials	Frequency (%)
Physical	10 (23.8)	Coating of Paint	4 (9.5)
Intellectual	11 (26.2)	Printing Ink	2 (4.8)
Technical	5 (11.9)	Polymer	3 (7.1)
Creative	9 (21.4)	Textile	2 (4.8)
Social	7 (16.7)	Paper	2 (4.8)
Total	42 (100)	Paperboard	5 (11.9)
		Metallic	3 (7.1)
		Plastic	21 (50.0)
		Total	42 (100)

4.2 Concentration of heavy metals in toys samples comparable to ISO 8124: 2010 Part 3 and EN 71-3: 2013 Category III.

The concentration of heavy metals in toys was compared to two standards, namely ISO 8124: 2010 Part 3 and European Union (EU) New Toy Safety Directive EN 71-3: 2013. ISO 8124: 2010 Part 3 is the standard that is used in the local regulation for safety of toys in Malaysia. All heavy metals in this study were exceeded the permissible concentration in ISO 8124: 2010. Meanwhile, six of the elements exceed the EU New Toy Safety Directive EN 71-3: 2013. There were Cr, Cu, Mn, Ni ,Sr, and Sn .

Table 4.2: Concentration of heavy metals in toys. Results expressed as range of values, average, standard deviation (SD), as well as their ISO 8124-3 and EN 71-3 2013 limits. (N=42)

Metal	Mean±SD (mg/kg)	Range	ISO 8124: 2010 Part 3 (mg/day)	EN 71-3: 2013 Category III (mg/kg)	No. of Samples exceed ISO 8124: 2010 Part 3 limit	No. of Samples exceed EN 71-3: 2013 Category III limit
Sb	126.03±521.46	56.50-2746.00	0.0002	560.00	5	2
As	36.80±214.37	1.00-1391.00	0.0001	47.00	10	3
Ba	4181.66±19195.32	95.70-122300.00	0.025	18.75	28	27
Cd	2.81±9.58	5.00-53.20	0.0006	17.00	6	3
Cr	81.37±194.03	9.30-1065.00	0.0003	0.20	28	25
Co	94.29±611.04	3960.00-3960.00	-	130.00	1	1
Cu	103.44±235.17	6.10-1122.00	-	7.70	-	27
Pb	46.81±116.58	1.50-639.00	0.0007	160.00	23	3
Mn	161.01±446.03	8.80-2246.00	-	15.00	-	17
Hg	2.77±10.65	2.30-66.90	0.0005	94.00	9	0
Ni	4875.72±31397.77	2.80-203511.00	-	930.00	-	1
Se	1.65±6.68	3.00-33.40	0.005	460.00	4	0
Sr	613.79±3701.74	2.00-24031.00	-	56.00	-	11
Sn	182.45±1102.28	33.00-6978.00	-	180.00	-	1
Zn	476.88±1583.19	3.90-10114.00	-	46.00	-	31

4.3 Comparison of heavy metal concentration by types of toys and materials.

The highest element detected in physical type of toys were Ba (11492.27±36753.53 mg/kg), followed by Sn (705.09±2204.13 mg/kg) and Zn (321.11±477.78 mg/kg). Elements that being a concern in human health also were highly detected in this toy types such as Cd (5.93±16.72 mg/kg), Pb (79.40±198.99 mg/kg), Cu (49.52±59.11mg/kg), Hg (7.98±20.96 mg/kg), Ni (27.76±75.69 mg/kg) and As (145.39±437.95 mg/kg). Similar trend was observed for intellectual toy types where Ba was the highest (11492.27±36753.53 mg/kg) followed by Sr (2218.33±7234.47 mg/kg), and Mn (467.06±738.22 mg/kg). Cu(184.16±293.43 mg/kg), Pb (58.99±101.31 mg/kg), Cd (0.45±1.51 mg/kg) and Ni (85.60±140.68mg/kg) also were detected. Hg (1.63±4.69 mg/kg) and As (2.50±6.65 mg/kg) were detected at low level for this toy type. As for the technical toy type, highest element detected was Ni (40703.08±91012.39 mg/kg), followed by Co (792.00±1770.97 mg/kg), and Zn (698.02±969.85 mg/kg). Other elements present in this toy type were Sb (405.80±907.40 mg/kg), Mn (249.96±514.95 mg/kg), Cu (92.16±170.20 mg/kg), Pb (72.22±112.45 mg/kg), and Cr (54.32±83.39 mg/kg).

In creative type toy, the concentration of Zn (1225.91±3336.94 mg/kg), was the highest, followed by Ba (38.00±84.97 mg/kg), Sb (305.11±915.33 mg/kg) and Sr (28.54±20.21 mg/kg). Other elements detected were Sn (26.16±46.36 mg/kg) and Cr (15.27±30.70 mg/kg). Social types of toys shows the highest concentration of Ba (880.81±894.96 mg/kg), followed by Cu (181.61±415.38 mg/kg), Zn (95.16±91.10

mg/kg), and Cr (27.53 ±38.98 mg/kg). Other elements of concern detected were Sr (23.89±26.64 mg/kg), Sb (10.70±28.31 mg/kg), Pb (8.24±19.61 mg/kg), and Ni (3.97±2.28 mg/kg).

There was a significant difference between concentrations of Ba(X^2 =0.57,ρ=<0.01) and Mn (X^2 =12.95,ρ=<0.01) by types of toys.

As for materials, the highest element was detected in paint coated toys is Ba (7556.75±14318.58 mg/kg), followed by Mn (589.83±666.33 mg/kg), and Ni (236.60±154.15 mg/kg). Other elements detected were Cr (215.48±160.93 mg/kg) and Cu (210.70±230.31 mg/kg). In ink printed toys, the highest heavy metal found was Ba (1698.00±758.02 mg/kg), followed by Cu (574.85±773.79 mg/kg), Zn (62.15±64.84 mg/kg), Sn (56.50±79.90 mg/kg), and Sr (34.30±38.89 mg/kg). Elements of concern were highly detected in polymer as well in which Zn occupies the highest concentration (5079.90±7119.29 mg/kg), followed by Ba (1127.50±1594.53 mg/kg), Sr (33.70±13.01 mg/kg), and Pb (1.80±2.55 mg/kg).

For textile based toys, the heavy metals present were Ba (642.00±907.93 mg/kg), Cr (62.35±70.22 mg/kg), Zn (38.55±13.22 mg/kg), and Sb (37.45±52.96 mg/kg). There were no heavy metals detected for toys made of papers. However, Ba (177.40±115.20 mg/kg), Zn (139.80±112.38 mg/kg), and Mn (110.62±69.20 mg/kg) were detected in paperboard toys. Low concentration of elements was detected in paperboard which was Cu (61.94±34.88 mg/kg), Se (49.86±16.72 mg/kg), and Cr (41.32±20.57 mg/kg).

For metallic toys, high concentration of Ni (67922.67±117423.01 mg/kg), Ba (40830.00±70555.15 mg/kg), Sr (8016.93±13868.59 mg/kg), Sb (676.33±1171.44 mg/kg), and Zn (465.67±446.58 mg/kg) were detected. Toys made of plastic were high with Ba (718.60±904.20 mg/kg), Sn (339.10±1521.27 mg/kg), Zn (327.13±567.79 mg/kg), and Sb (151.88±600.38 mg/kg). Cr (432.90±553.69 mg/kg), Pb (171.67±83.11mg/kg), and Ni (67922.67±117423.01 mg/kg) were significantly higher in metallic toys and Mn significantly higher in paint coated toys (589.83±666.33 mg/kg).

Table 4.3.1: Mean (±SD) heavy metal concentration (mg/kg) in toy samples between five types of toys. (N=42)

	Types of Toys						
Metal	Physical	Intellectual	Technical	Creative	Social	X^2	p-value
Sb	38.70±122.38	5.14±17.04	**405.80±907.40**	305.11±915.33	10.70±28.31	0.57	0.96
As	**145.39±437.95**	2.50±6.65	ND	7.14±17.42	ND	7.09	0.13
Ba	**3954.50±8867.17**	**11492.27±36753.53**	38.00±84.97	368.22±771.45	880.81±894.96	13.21	**<0.01**
Cd	**5.93±16.72**	0.45±1.51	5.66±12.66	2.83±6.41	ND	2.43	0.66
Cr	85.11±181..14	**178.60±321.93**	54.32±83.39	15.27±30.70	27.53±38.98	4.86	0.30
Co	ND	ND	**792.00±1770.97**	ND	ND	7.40	0.12
Cu	49.52±59.11	**184.16±293.43**	92.16±170.20	10.14±13.17	**181.61±415.38**	8.54	0.07
Pb	**79.40±198.99**	58.99±101.31	72.22±112.45	11.57±32.03	8.24±19.61	5.61	0.23
Mn	26.26±30.33	**467.06±738.22**	**249.96±514.95**	12.50±29.02	ND	12.95	**<0.01**
Hg	**7.98±20.96**	1.63±4.69	1.84±4.11	1.04±2.26	ND	2.55	0.63
Ni	27.76±75.69	85.60±140.68	**40703.08± 91012.39**	1.98±3.25	3.97±2.28	7.07	0.13
Se	0.44±1.39	ND	**6.68±14.94**	3.14±9.43	0.43±1.13	2.07	0.72
Sr	77.29±68.60	**2218.33±7234.47**	36.10±41.15	28.54±20.21	23.89±26.64	3.11	0.54
Sn	**705.09±2204.13**	5.83±19.33	ND	26.16±46.36	ND	5.10	0.27
Zn	321.11±477.78	148.05±85.15	698.02±969.85	**1225.91± 3336.94**	95.16±91.10	2.95	0.56

ND-Not Detected
*p-value is significant at the 0.05 level (2-tailed)
*Kruskal-Wallis test

Table 4.3.2: Mean (±SD) heavy metal concentration (mg/kg) in toy samples between different toy materials. (N=42)

Metals	ToyMaterials									
	Paint Coatings	Printing Ink	Polymer	Textile	Paper	Paperboard	Metallic	Plastic	X^2	p-value
Sb	ND	ND	ND	37.45±52.96	ND	ND	**676.33± 1171.44**	151.88± 600.38	6.04	0.53
As	8.13±10.64	ND	ND	ND	ND	1.02±1.01	ND	**71.81± 302.66**	7.42	0.38
Ba	7556.75± 14318.58	1698.00±758.02	1127.50± 1594.53	642.00±907.93	ND	177.40±115.20	**40830.00±70555.15**	718.60± 904.20	5.752	0.56
Cd	ND	3.30±4.67	ND	ND	ND	1.00±2.24	**9.43±16.33**	3.72± 12.11	4.80	0.68
Cr	215.48± 160.93	15.30±21.64	ND	62.35±70.22	ND	41.32±20.57	**432.90± 553.69**	40.46± 127.18	17.36	**0.02**
Co	ND	ND	ND	ND	ND	ND	**1320.00± 2286.31**	ND	13.00	0.07
Cu	210.70±230.31	**574.85±773.79**	ND	23.35±17.04	ND	61.94±34.88	**449.57± 460.00**	23.46± 40.03	20.22	**<0.01**
Pb	**127.45± 2.84**	ND	1.80±2.55	0.75±1.06	ND	11.44±6.00	**171.67± 83.11**	41.85± 38.89	18.07	**<0.01**
Mn	589.83± 666.33	ND	ND	4.40±6.22	ND	110.62±69.20	**1138.13± 1123.31**	12.80± 27.04	20.69	**<0.01**
Hg	**16.73±33.45**	1.45±2.05	ND	ND	ND	0.50±1.12	5.20±9.01	1.35± 3.18	3.41	0.84
Ni	236.60± 154.15	ND	ND	4.80±0.42	ND	5.92±4.51	**67922.67± 117423.01**	3.56± 2.84	17.32	**<0.01**
Se	ND	ND	ND	ND	ND	ND	**11.13± 19.28**	1.70± 6.20	4.63	0.70
Sr	43.73± 84.87	34.30±38.89	33.70±13.01	29.70±37.34	ND	49.86±16.72	**8016.93± 13868.59**	46.53± 48.86	4.15	0.76
Sn	16.03±32.05	56.50±79.90	ND	ND	ND	ND	ND	**339.10± 1521.27**	4.99	0.66
Zn	157.58±65.21	62.15±64.84	**5079.90± 7119.29**	38.55±13.22	ND	139.80±112.38	465.67± 446.58	327.13± 567.79	6.30	0.50

ND-Not Detected
*p-value is significant at the 0.05 level (2-tailed)
*Kruskal-Wallis test

4.4 Association between concentration of heavy metals in toys based on their types and materials.

Findings show low positive correlation between Co and Cd (r = 0.42). Ba was positively correlated with Cr (r = 0.80), Cu (r = 0.56), Hg (r = 0.41), and Mn (r = 0.71). Mn was moderately correlated with Cu (r = 0.61) while Ni was low correlated with Cd (r=0.42) and strongly correlated with Co (r=1.00). Pb has low correlation with Mn (r = 0.44) and moderate correlation with Cr (r = 0.66). In addition, Se and Sb (r = 0.97), Sn and Cd (r = 0.84), Sr and Ba (r = 0.98) were strongly correlated to each other. This result indicates a significant influence of one heavy metal to another and suggests the possibility of the metals coming from the same source.

Table 4.4: Correlation among heavy metals in toys (N=42)

	As	Ba	Cd	Co	Cr	Cu	Hg	Mn	Ni	Pb	Sb	Se	Sn	Sr	Zn
As	1														
Ba	-0.01	1													
Cd	-0.04	-0.06	1												
Co	-0.03	-0.03	0.42**	1											
Cr	-0.05	0.80**	-0.01	0.10	1										
Cu	-0.05	0.56**	0.02	0.20	0.57**	1									
Hg	0.12	0.41**	-0.07	-0.04	0.20	0.11	1								
Mn	-0.03	0.71**	0.09	0.36*	0.84**	0.61**	0.10	1							
Ni	-0.03	-0.03	0.42**	1.00**	0.10	0.20	-0.04	0.36*	1						
Pb	0.07	0.17	0.08	0.29	0.66**	0.19	0.06	0.44**	0.29	1					
Sb	0.11	-0.04	0.18	-0.04	-0.02	-0.08	-0.05	-0.08	-0.04	0.10	1				
Se	0.09	-0.04	0.13	-0.04	-0.04	-0.08	-0.05	-0.08	-0.04	0.10	0.97**	1			
Sn	-0.02	-0.03	0.84**	-0.03	-0.04	-0.07	-0.04	-0.06	-0.03	-0.06	-0.03	-0.04	1		
Sr	-0.02	0.98**	-0.05	-0.03	0.80**	0.56**	0.20	0.74**	-0.03	0.16	-0.04	-0.04	-0.02	1	
Zn	0.11	-0.04	-0.06	-0.03	-0.08	-0.09	-0.04	-0.06	-0.03	-0.05	0.04	0.05	-0.05	-0.03	1

*Correlation is significant at the 0.05 level (2-tailed)

** Correlation is significant at the 0.01 level (2-tailed)

4.5 Health Risk of Heavy Metals in Toy Samples

The carcinogenic and non-carcinogenic health risk due to the exposure to selected heavy metals was estimated using Lifetime Cancer Risk (LCR) and Hazard Quotient (HQ) in this study were highlighted in in Table 4.5. According to The Risk Assessment Information System database (2013), As, Cd, Co, Cr, Ni, and Pb are known to be human carcinogens whereas other elements (i.e. Sb, Ba, Cu, Mn, Hg, Se, Sr, Sn, Zn) are non-carcinogens to human. Levels of 10^{-6} to 10^{-4} be given as a range of "generally acceptable risk" (Kelly, 1991) for LCR.

In this study, heavy metals concentration in the toy samples did not posed significant carcinogenic risks to children's health except for Co ($3.61 x 10^{-3}$) and Ni ($1.62 x 10^{-2}$) as the value exceeded levels of 10^{-6} to 10^{-4} which is the acceptable risk range. The non-carcinogenic elements in this study does not pose health risk among children except for Sb (HQ=4.03) and Sn (HQ=7.78). Hazard quotient of other elements were lower than 1.

Table 4.5: Non-carcinogenic hazard quotient and carcinogenic lifetime risk for individual heavy metals.

Heavy Metal	Minimum Concentration (mg/kg)	Maximum Concentration (mg/kg)	Mean Concentration± SD (mg/kg)	LADD (mg/kg-day)	Carcinogenic Risk	ADD (mg/kg-day)	Non-carcinogenic Risk
Sb	**56.50**	**2746.00**	**126.03±521.46**	**-**	**-**	**<0.01**	**4.03**
As	1.00	1391.00	36.80± 214.37	<0.01	7.06×10^{-4}	-	-
Ba	95.70	122300.00	4181.66±19195.32	-	-	0.05	0.09
Cd	5.00	53.20	2.81±9.58	<0.01	1.37×10^{-5}	-	-
Cr	9.30	1065.00	81.37±194.03	<0.01	5.20×10^{-4}	-	-
Co	**3960.00**	**3960.00**	**94.29±611.04**	**<0.01**	$\mathbf{3.61 \times 10^{-3}}$	-	-
Cu	6.10	1122.00	103.44±235.17	-	-	<0.01	0.03
Pb	1.50	639.00	46.81±116.58	<0.01	5.09×10^{-6}	-	-
Mn	8.80	2246.00	161.01±446.03	-	-	<0.01	0.01
Hg	2.30	66.90	2.77±10.65	-	-	<0.01	0.01
Ni	**2.80**	**203511.00**	**4875.72±31397.77**	**0.06**	$\mathbf{1.62 \times 10^{-2}}$	-	-
Se	3.00	33.40	1.65±6.68	-	-	<0.01	0.01
Sr	2.00	24031.00	613.79±3701.74	-	-	<0.01	0.01
Sn	**33.00**	**6978.00**	**182.45±1102.28**	**-**	**-**	**<0.01**	**7.78**
Zn	3.90	10114.00	476.88±1583.19	-	-	<0.01	0.02

4.6 Discussion of Findings

The presence of heavy metals in toys could be due to various reasons. One of it was due to elements that were added to increase the durability, attractiveness, and give colour to toys (Al-Qutob et al., 2014). Findings in this study have obtained high Ba, Mn, and Cu in intellectual toys. According to Miller et al., (2015), toys may contain barium sulfate which is white and often mixed with other colors to vary the shade. Toys that are made from metals often came as silver, grey, and white color that may contain high heavy metals. In this study, toys made of metal were highly detected with Cr, Pb and Ni compared to other materials. Samples with Pb concentration often contained Cr. Both Cr and Pb are used as stabilizer in toys to enhance its material properties and also to reduce cost on plastics (Al-Qutob et al, 2014). Based on the correlation test, there is a strong positive correlation between concentration of Ba with Cr and Pb. These metals are combined to be used as colorants or pigments (Al-Qutob et al, 2014). Rangos (2003) indicates that lead chromate ($PbCr0_4$) was a standard colorant for plastic toys even lead sulfate ($PbS0_4$) and lead oxide (PbO) were mixed with the chromate to produce light yellow colour to red.

Mn was significantly higher in intellectual and paint-coated toys in this study. To date, there is no study conducted in determining the concentration of Mn in toys. However, a study to determine the concentration in paint was conducted by Ben Ali & Salah, 2010. The study aims to determine the percentages of Mn in driers which are used in paints. According to the study, drier is the main additive used in paints which contain heavy metals such as Co and Mn. Based on Grzetic & Ghariani (2008), Mn can bring variety and serious toxic responses for prolonged exposure to high concentration, either through ingestion and inhalation. Further monitoring of Mn is necessary to improve this study. However, there is no study yet that had proven the significance of Mn in toys.

High concentration of As and Hg was observed in paint-coated toys in this study. There were strong positive correlation between Ba, Cr, Cu, Hg, and Mn. Hg is used as a catalyst to enhance chemical reaction among ingredients (UNEP, 2008). There could be a possibility that both Mn and Hg were used as stabilizer or colorant for toys. However, 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 shows high concentration of Pb, Cr, and Cd. Cu is highly 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 (Obene & Clark, 2013).There is a positive strong correlation between Cu and Mn.

As for health risk in this study, only Ni and Co poses carcinogenic health risk among children via inhalation. Therefore, children in Selangor are exposed to the adverse health effect of Ni which is allergic reaction (RAIS, 2007). Adverse health effects caused by Co exposure through inhalation may cause asthma, alveolitis, and occasionally, fibrosis (Raffn et al., 1988). As for non-carcinogenic health risks, 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 (Oliver, 1916). A study conducted by Kimbrough (1976) has shown ingestion of tin can had caused skin irritation in rabbits. Although other heavy metals determined in this study did not show significant health risk as the HQ is less than 1 for non-carcinogenic risk or the carcinogenic risk did not exceed the acceptable risk range, the elements are still possible to create adverse health effects among children. This is because bioaccumulation of heavy metals from toys could still happen. Bioaccumulation is the accumulation of substances or chemicals in an organism. Children spent long hours playing with toys. Half of their childhood is surrounded by toys. Bioaccumulation can also happen when the children are exposed to plants or soil that are contain high levels of heavy metals. When these plants are harvested for use and ingest, the heavy metals present in the plant are ingested by children. This can cause accumulation in the childrens' body. Bioaccumulation is not only exposed through ingestion, but through inhalation and dermal contact as well (Martin & Griswold, 2009).

4.7 Study Limitation

There were several limitations experienced while conducting this study. Due to financial constraint, this study was only performed to small samples size which cannot be generalized for the whole toys in the market. The sample size were also not homogenized as this study did not measure the concentration of heavy metal in all types and materials as what is addressed in the ISO 8124: 2010 Part 3 Safety of Toys. Besides, the toy samples were taken purposively from Selangor area and market only. Another limitation would be the health risk calculation which was calculated based on secondary data and study (Grzetic & Ghariani, 2008). Furthermore, the instrument used in this study (High Definition X-ray Fluorescence (HDXRF®) technique (Model 800701-01) has low detection limit which unable to detect trace element at very low concentration.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH

5.1 Conclusion

In conclusion, all heavy metals in this study were exceeded the permissible concentration in ISO 8124: 2010. Meanwhile, six of the elements exceed the EU New Toy Safety Directive EN 71-3: 2013. There were Cr, Cu, Mn, Ni ,Sr, and Sn . There is a significant difference in the concentration of heavy metals in five types of toys. (i.e. physical, intellectual, technical, creative, and social). The concentration of Ba (11492.27±36753.53 mg/kg), Mn (467.06±738.22 mg/kg), and Cu (184.16±293.43 mg/kg) are significantly higher in toys that are used for intellectual activities. The concentrations of heavy metals are strongly associated between one another. There is a significant difference in the concentration of heavy metals in different types of toy materials. (i.e. paint coatings, polymers, plastic, paper, paperboard, paper, and others). The concentration of Cr (432.90±553.69 mg/kg), Pb (171.67±83.11 mg/kg), and Ni (67922.67±117423.01 mg/kg) are significantly higher in toys that are made from metals. In addition, Mn (589.83±666.33 mg/kg) and Cu (574.85±773.79 mg/kg) are significantly higher in paint-coated toys and ink printed toys respectively. There is a significant association between concentrations of heavy metals in toys. Based on the findings, there is low positive correlation between Co and Cd ($r = 0.42$). Ba was positively correlated with Cr ($r = 0.80$), Cu ($r = 0.56$), Hg ($r = 0.41$), and Mn ($r = 0.71$). Mn was moderately correlated with Cu ($r = 0.61$) while Ni was low correlated with Cd ($r=0.42$) and high correlated with Co ($r=1.00$). Pb has low correlation with Mn (r = 0.44) and moderate correlation with Cr (r = 0.66). In addition, Se and Sb (r = 0.97), Sn and Cd (r = 0.84), Sr and Ba (r = 0.98) were strongly correlated to each other. This result indicates a significant influence of one heavy metal to another and suggests the possibility of the metals coming from the same source. The concentration of Co (3.61×10^{-3}) and Ni (1.62×10^{-2}) are of worrying concern to be played by children because it pose carcinogenic health risk through inhalation whereas the concentration of Sb (HQ=4.03) and Sn (HQ=7.78) are dangerous for children that are exposed to the toys sold in Selangor because it pose non-carcinogenic health risks through ingestion. Other concentration of heavy metals detected in toy samples does not pose carcinogenic and non-carcinogenic health risk among children (As, Ba, Cd, Cr, Cu, Pb, Mn, Hg, Se, Sr, and Zn).

5.2 Recommendation

Based on the findings obtained, parents should avoid purchasing toys that are made from metallic materials. This is because high number of heavy metals concentration was found to be present in metallic toys. Second, the findings of this study can act as preliminary data and evidence to recommend the Ministry of Domestic Trade and Consumer Affairs to revise and regulate new standards that addresses the permissible concentration of heavy metals in toys. The revision is necessary because currently, the regulation is using the international standards form International Standard Organization (ISO).

For future study, the study should be conducted with bigger sample size so that the result can be generalized to the community. Besides, all types and materials should be taken account as what is addressed in ISO 8124: 2010 Part 3 Safety of Toys to ensure all samples are homogenized. The samples should also be taken from all district in Malaysia and not limited to one district or location only. This is very important to ensure compliance from the business manufacturers in other districts towards local regulation Consumer Protection (Safety Standards for Toys) Regulation 2009 where the permissible concentration of heavy metals in toys is addressed. The heavy metals included are Sb, As, Ba, Cr, Cd, Pb, Hg, and Se. Since the local regulation only addressed eight heavy metals permissible concentration, future studies should be conducted to identify the health risks concerning other types of heavy metals that are present in toys such Mn, Cu, and Sn. In the future, the health risk calculation should be calculated by using primary data and not secondary data. The primary data can be collected through 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.

REFERENCES

Abdullah, Z. (2015). Health Risk Assessment of Lead in Various of Toys and Parental Awareness On Children Exposure. Unpubliched Manuscript, Universiti Putra Malaysia, Selangor, Malaysia.

Adal, A., Wiener S. W., VanDeVoort, J.T., Benitez, J.G. & Tarabar, A. (2015). Heavy Metal Toxicity-Medscape (2015). Retrieved from 2016, March 7, from http://emedicine.medscape.com/article/814960-overview.

Al-qutob, M., Asafra, A., Nashashibi, T., & Qutob, A. A. (2014). Determination of Different Trace Heavy Metals in Children ' s Plastic Toys Imported to the West Bank / Palestine by ICP /MS-Environmental and Health Aspects. *Journal of Environemntal Protection*. 5:1104-1110.

Agarwal, R. Toxics link. Toying with Toxics. Retrieved 2016, March 7, from www.toxiclink.org.

Application of FISCHER products-Detecting Heavy Metals in Toys using X-Ray Fluorescene. Retrieved 2016, May 10, from http://www.fischer-technology.com/fileadmin/user_upload/default/Application-Notes/en/AN027en_heavy_metals_in__toys.pdf.

Ali, B. & Salah, M. (2010). Determination of Cobalt and Manganese percentages in paint driers by complex formation titration. *Al-Nawah*. 9(13): 29-42.

Bressler, J.P. & Goldstein, G.W. (1991). Mechanisms of lead neurotoxicity. *Biochemical Pharmacology*. 41(4): 479-484.

California Environmental Protection Agency (CalEPA). *Technical Support Document for the Determination of Noncancer Chronic Reference Exposure Levels. Draft for Public Comment.* Office of Environmental Health Hazard Assessment, Berkeley, CA. 1997.

Ćrzetic, I., & Ghariani, R. H. A. (2008). Potential health risk assessment for soil heavy metal contamination in the central zone of Belgrade (Serbia). *J. Serb. Chem. Soc.* 73 (8-9): 923-934.

Consumer Protection Act 1999 Consumer Protection (Certificate of approval and Conformity Mark of safety standards) regulations 2009. Ministry of Domestic Trade, Co-operatives and Consumerism.

Davidowski, L., Grosser, Z. & Thnompson, L. (2007). Heavy Metals Analysis for the Toy Industry by Inductively Coupled Plasma Optical Emission Spectroscopy. ICp-Optical Emission Spectroscopy.

European Chemical Safety for Toys. Underwriters Laboratories (UL). (2013). Retrieved 2016, May 10, from http://ul.com/toys.

ECB, European Chemicals Bureau (ECB), European Commission – Joint Reasearch Centre, (2003) EUR 20418 EN/1.

Gordon. J.N., Taylor. A. & Bennet, P. N. (2002). Lead Poisoning: case studies. *Br J Clin Pharmacol.* 53 (5): 451-459.

Heavy Metal Poisoning- NORD (National Organization for Rare Disorder). (2016). Retrieved 2016, May 10, from http://rarediseases.org/rare-diseases/heavy-metal-poisoning/.

Holmes, T.J., McGrattan, E.R. & Prescott, E.C. (2011). Technology Capital Transfer. Federal Reserve Bank of Minneapolis.

International Toy Safety Standards Expected. (2012, December 4th).Toy Industries of Europe.

ISO/IEC IS 8124:2010: Safety of Toys Part 3: Migration of Certain Elements, 2016 May 10th, International Organization for Standardization, Geneva, Switzerland. http://www.iso.org/iso/catalogue_detail?csnumber=43471/ 2016-May-10th

ISO/IEC IS 8124:2010: Safety of Toys Part 8: Age Determination Guidelines, 2016 May 10th, International Organization for Standardization, Geneva, Switzerland. http://www.iso.org/iso/catalogue_detail?csnumber=43471/ 2016-May-10th.

Jia., H. (2009). Heavy Metal Poisoning Sparks Protests in China. *Royal Society of Chemistry.* Retrieved 2016, March 7, from http://www.rsc.org/chemistryworld/News/2009/August/25080902.asp

Johri, N., Jacquillet, G. & Unwin, R. (2010). Heavy Metal Poisoning: The Effects of Cadmium on the Kidney. *BioMetals.* 23(5): 783-792.

Kelly, K. E. (1985). Neurotoxicity: Toxic Effects in the Nervous System. In : Williams, P.L. and Burson, J. L. (eds.). *Industrial toxicology.* New York : Van Nostrand Reinhold.

Kelley, M., Watson, p., Thorton, D. & Halpin, T.J. (1993). Lead intoxication associated with chewing plastic wire coating. *Morbidity Mortality Wkly Rep.* 42: 465-467.

Kimbrough, R.D. (1976). Toxicity and Health Effects of Selected Organotin Compounds: A review. *Environ Health Prospect.* 14: 51-56.

Kumar, A., & Pastore, P. (2007). Lead and cadmium in soft plastic toys. *Current Science*, 93(6), 818–822.

Li, E., & O'Brien, E. (2011). A fact sheet for Australian toy importers and traders. Retrieved 2016, March 7, from https://www.lead.org.au/fs/Fact_sheet_for_Australian_toy_importers_and_trad ers.pdf.

Martin, S. & Griswold, W. (2009). Human Health Effects of Heavy Metals. Environmental Science and Technology Briefs for Citizens. Retrieved 2016, February 18, from http://www.engg.ksu.edu/chsr/files/chsr/outreach-resources/15HumanHealthEffectsofHeavyMetals.pdf.

Miller, G. Z., & Harris, Z. E. (2015).Hazardous Metals in Vintage Plastic Toys Measured by a Handheld X-ray Fluorescene Spectrometer. *Journal of Environmental Health.* 77(6): 8-13.

Mulder, K. & Knot, M. (2001). PVC Plastic: A History of Systems Development and Entrenchment.

National Referral Centre for Lead Poisoning in Indian, (n.d). Retrieved 2016, March 7, from http://www.tgfwotld.org/lead.htm

Needleman, H. L. & Bellinger, D. (2007). The health effects of low level exposure to Lead. *Ann. Rev. Public Health.* 12: 111-140.

Obene, P., & Clark, I. (2013). High Resolution Copper Ink Printing for Electronic Interconnects. *Electronic Engineering Times Europe.* 20-22.

Oliver, T. Diseases of Occupation: from the legislative, social, and medical points of view. (1908). 3rd ed. Methuen, London: 1916.

Omolaoye, J. A., Uzairu, A., & Gimba, C.E. (2010).Heavy Metal Assesment of Some Soft Plastic Toys Imported into Nigeria from China. *Journal of Environmental Chemistry and Ecotoxicology.* 2(8): 126-130.

RAIS, The Risk Assessment Information System. (2007). Retrieved 2016, January 15 from http://rais.ornl.gov/tox/rap_toxp.shtml.

Raffn, E., Mikkelsen, S., Altman, D. G., Christensen, J. M. & Groth, S. (1988). Health Effects due to Occupational Exposure to Cobalt Blue Dye among Plate Painters in a Porcelain Factory in Denmark. *Scandinavian Journal of Work.* 14(6): 378-384.

Rangos, G. (2003). Inorganic colored pigments. In R.A. Charvat (Ed.), *Coloring of plastics*: Fundamentals (pp. 134–136). New York: Wiley Interscience.

Sharma, M., Maheshwari, M., & Morisawa, S. (2005). Dietary and Inhalation Intake of Lead and Estimation of Blood Lead Levels in Adults and Children in Kanpur, India. *Risk Anal.,* 25(6): 1573-1588.

Tamaddon, F. & Hogland, W. (1993). Review of Cadmium in Plastic Waste in Sweden. Waste Management & research. 11: 287-295.

Tchounwou, P.B., Yedjou, C.G., Patlolla, A.K.,. & Sutton, D. J. (2014). Heavy Metals Toxcity and the Environment. *Molecular, Environmental and Clinical Toxicology*. 101: 133-164.

United Nations Environmental Program (UNEP). (2008). Expanding the Number and Scope of Partnership Areas within the United Nations Environment Program Global Mercury Partnership. Retrieved March 7, 2016, from http://www.chem.unep.ch/MERCURY/UGMP/K0820334_%20Partnership%20-%201-3.doc.

USEPA, United States Environmental Protection Agency, *Risk*, 1991, OSWER Directive 9285.7-01B.

U.S. EPA. Exposure Factors Handbook - 2009 Update (External Review Draft) . U.S. Environmental Protection Agency, Washington, DC, EPA/600/R-09/052A, 2009.

Waalkes, M.P. & Rehm, S. (1994). Cadmium and prostate cancer. *J. Toxicol. Environ. Health*, 43, 251–269.

Worldnet Daily News. China Exports Lead Poisoning. (2009). Retrieved 2016, March 7, from http://www.worldnetdaily.com/news/article.

Zeitoun, M. M. & Mehana, E.E. (2014). Impact of Water Pollution with Heavy Metals on Fish Health. *Global Veterinaria*. 12(2): 2019-231.