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Determination of Lead in Candies and their Packaging Sold in Malaysia and its Potential Health Risk to Children

Nur Aziema Azreen Samsuddin, Siti Zulfa Zaidon, Yu Bin Ho*

Department of Environment and Occupational Health, Faculty of Medicine and Health Science, University Putra Malaysia 43400 UPM Serdang, Selangor, Malaysia

Corresponding author: Yu Bin Ho, <u>yubin@upm.edu.my</u>, Department of Environmental and Occupational Health, Faculty of Medicine and Health Sciences, Universiti Putra Malaysia, 43400 UPM

Serdang, Selangor, Malaysia

ABSTRACT

Objective: This study was done to determine the concentration of lead in 3 different types of candies (type I: sugar based candies, type II: milk based candies and type III: chocolates based candies) sold in local market in Malaysia and to assess their potential health risk to children.

Method: The candies were purchased from a local market and categorised according to their types. For each category, 15 samples from different brands were selected and the total samples were 45 samples. The sample was extracted using microwave acid digestion and maple furnace and analysed by using Graphite Furnace Atomic Absorption Spectrometry (GF-AAS). The concentration of lead found in different type of candies and their packaging were used to calculate the target hazard quotient (THQ) for health risk assessment.

Result: The concentration of lead in 3 different types of candies which were sugar based candies, milk based candies and chocolate based candies varied with the range $0.04-4.24 \ \mu g/kg$, $0.04-0.26 \ \mu g/kg$ and $0.05-0.47 \ \mu g/kg$ respectively, and packaging varies with the range of $0.01-0.08 \ \mu g/kg$, $0.02-0.08 \ \mu g/kg$ and $0.01-0.5 \ \mu g/kg$ respectively.

Conclusion: Generally, the concentrations of lead in candies and their packaging for the entire sample were not exceeded the permissible limit and the THQ were below 1 for all candies and their packaging, indicating that there was no significant non-carcinogenic health risk by consuming the candies by the children

Keywords: Lead (Pb), candies and packaging, maple furnace and microwave acid digester, GF-AAS, health risk assessment (HRA)

1. Introduction

There are many types of locally made candies sold in Malaysian market and most of them have food label on the packaging with the list of the ingredient and name. There are many ingredients that usually used in the production of candies and that ingredient may be the source of lead contamination (Dahiya, Karpe, Hegde, & Sharma, 2005). Children are the most sensitive and vulnerable age group to any kind of metal contamination in the food chain (Dahiya et al, 2005). This exposure may result in acute and longterm effects on health particularly during growth and development. Since growing children consume more food per body weight than their adult counterparts, they are at a higher risk of illness from exposure to chemical hazards in food (Iwegbue et al., 2014).

Many candies packages are poorly design which allow the migration of heavy metal components from the ink of the printed surface into the candies surface. Previous study by Kim et al. (2008) showed that heavy metals that were used in the painting of the candies packaging were detected in candies. Lead or hexavalent chromium are especially commonly as a based ink in food packaging.

Lead is a classic toxic chemical and it can cause damage to the kidney, cardiovascular, immune, hematopoietic, central nervous and reproductive system. If the surface of the candy is sticky, severe migration could be occurred and the high exposure to the lead can cause gastrointestinal distress, anaemia, encelophathy and also may cause death. The effect of exposure to the lead varies according to the dosage and age of the exposed person (ATSDR & Sciences, 2007).

Therefore, this study was aimed to detect the concentration of lead in local candies and their packaging and also the migration of lead into the candies. This allowed us to know the probability of packaging material that containing lead. Furthermore, there are no reported study on lead in candies and their packaging in Malaysia and there are also no published reports on the heavy metals such as lead in candies and its packaging sold locally. This study was important to investigate the level of heavy metals that was presented in our local candies and to raise the public awareness of food safety.

2. Materials and Method

2.1 Sample Collection

The candies were purchased at the local market and categorised according to their types (type I: sugar based candies, type II: milk based candies and type III chocolates based candies). For each category, 15 samples from different brand were selected and the total samples were 45 samples. The sample was extracted using microwave acid digestion and analysed by using Graphite Furnace Atomic Absorption Spectrometry GF-AAS.

2.2 Instrumentation

2.2.1 Microwave Acid Digester

The candy samples were extracted using microwave acid digester Multiwave 3000 (Anton Paar, Graz, Austria) according to the method adopted from Kim et al. (2008) and Faridah et al. (2008). Briefly, the procedure as follows:

approximately, 0.2 g of sample were weighted and then 4 mL of HNO₃ and 2mL of ultrapure water was added into the vessels. After that, the vessels were sealed with sealed cap and put into the microwave oven. The microwave program is summarized in Table 1. After the second step of the program, the vessels were cooled to 50 °C and vented for 10 minutes. The extracts were filtered by using 0.45 μ m filter paper and then diluted with ultrapure water to the final volume of 25mL.

2.2.2 Maple Furnace

For digestion of candy packaging, dry ash method was used according to the procedure from Duran et al. (2009). Approximately 1 g of sample was added into a ceramic crucible and then ashed for 6 hours at 500°C by using muffle furnace (CarboliteELF 11/6). Then the sample was diluted with 5mL of HNO₃ and heated slowly by using hot plate to dissolve the residue. The solution then filtered with 0.45 μ m filter paper and added the ultrapure water to make up to 25 mL of final volume.

Table 1: Microwave Acid Digester characteristic							
Step	Temperature	Power	Time	Fan			
		(watt)	(Min)	Level			
Power	-	600 W	10	1			
Ramp							
Power	-	700 W	10	1			
Hold							
Cooling	50°C	0 W	-	3			

2.2.3 Graphite Furnace Atomic Absorption Spectrometry (AAS) Instrument

For this study, Perkin Elmer analyst 600 atomic absorption spectrometry were used to analyse the candies and packaging of samples. The instrument was calibrated using six-point calibration curve (0 μ g/L, 1 μ g/L, 2 μ g/L, 3 μ g/L, 4 μ g/L and 6 μ g/L) prior to the analysis process.

2.3 Quality Control

The apparatus was cleaned by soaking the in 5-10% of hydrochloric acid, washed with detergent, rinsed with ultrapure water and air dried before every used. All the candies were stored in the zip block bag according to their type of candies to avoid any cross contamination occur during storage. The sample was stored in the refrigerator to ensure the candies not become sticky and affect the other sample. Furthermore, for every batch of sampling, the blank was prepared to check for possible contamination along the extraction procedure (Dahiya et al., 2005). Then the sixpoint calibration curves were prepared before the sample analysis. The over range samples were diluted in order to ensure the reading fall within the calibration curve.

Percent of recovery for extraction was carried by comparing the concentration of heavy metals spiked before the microwave digestion or dry ash digestion process (AI) with the concentration of heavy metals spiked after the extraction but prior to GF-AAS injection (A2) to ensure the efficiency of the extraction method. Eq. 1 was used to calculate the recovery percentage.

Recovery (%) =
$$\frac{A1 - blank(ppm)}{A2 - blank(ppm)} \times 100$$
 (Eq. 1)

The result of quality control was summarized in table 2.

Table2: Summary of quality control result						
Samples	\mathbf{R}^2					
	(Mean ± RSD) %, N=3	range (ppb)				
candies	82.6 %	0-6	0.9996			
packaging	80.6%					

2.4 Health Risk Assessment (HRA)

Health risk evaluation indicator such as estimated daily intake (EDI) and target hazard quotient (THQ) was calculated to assess the potential chronic risk according to Iwegbue et al. (2014) by using Eq. 2.

$$EDI = \frac{MI \times CM}{BW}$$
(Eq. 2)

where, EDI is estimated daily intake of lead which was obtained from this study, CM is concentration of pollutant in food (μ g/kg) which was obtained from this study, MI is mass of product ingested per day (20g/day) which was obtained from study by Dahiya et al. (2005) and BW is body weight (15kg), the value was obtained from Dahiya et al. (2005) and Iwegbue et al. (2014).

In order to assess the level of lead arising through consumption of candies, the THQ was calculated by using Eq. 3 from USEPA (1989) in which the measured concentrations of lead was used.

$$THQ = \frac{EF \times ED \times MI \times CM}{(Eqx B)^{-3}}$$

where, THQ is Target hazard quotient which was obtained from this study, EF is exposure frequency (365/day), this was obtained from Iwegbue et al. (2014), ED is exposure duration (74 years) which was obtained from Malaysia: Health Profile, 2012, AT is averaging time (days) (6 years or 2190 days) according to Iwegbue et al. (2014) and RfD is reference dose for lead ($4\mu g/kg/day$) which was obtained from Iwegbue et al. (2014).

2.5 Statistical Analysis

Kruskal Wallis test was used to determine the concentration of lead in 3 different types of candies and their packaging. Whereas, Pearson correlation was used to test the association between the concentration of lead and the type of candies and to test the association between the concentrations of lead and candies packaging.

3. Results

3.1 Concentration of lead in candies and their packaging

The concentration of lead in 3 different types of candies which were sugar based candies, milk based candies and chocolate based candies varied with the range of 0.04-4.24 μ g/kg, 0.04-0.26 μ g/kg, and 0.05-0.47 μ g/kg respectively.

The highest lead content was found in sugar based candies with the reading 4.24 μ g/kg. The concentrations of lead in different type of candies and their packaging are summarized in Table 4.

For the packaging, the concentration of lead in the 3 types of candies varies with the range 0.01-0.08 μ g/kg, 0.02-0.08 μ g/kg, and 0.01-0.5 μ g/kg respectively. The highest concentration of lead was reported in one of the milk based candies packaging with the reading 0.8 μ g/kg. However the highest concentration of lead in both samples were considered safe as it does not exceed the permissible limit that was list by Food and Drug Association (2006) which is 100 μ g/kg. Figure 1 illustrates the mean concentration of lead in candies and their packaging.



Figure 1: Mean concentration of Pb in candies and packaging

3.1 Association between the concentration of lead and the type of candies.

Table 3 shows that the value for the association between the lead concentration and the type of can-dies is 0.084 which is more than 0.05. This shows that the data is not significantly distributed and there is no association between the concentrations of lead with the type of candies. So no further investigation will be made.

Table 3: Correlation bet	ween lead	and types of	f candies
	m	3.4	CD

	ſ	I	(N=45)
Lead concentration and type of candies	1.00	0.084	0.35 ± 0.78

3.3 Association between the concentration of lead and type of packaging.

Table 5 shows that the value for association between the lead concentration and the type of candies packaging is 0.164 which is more than 0.05. This shows that the data is not significantly distributed. Therefore, there is no association between the concentrations of lead with the type of candies. So no further investigation will be made.

Table 5: Correlation between lead concentration and types of candies packaging

	r	t	Mean± SD (N=45)
Lead concentration and type of candy packaging	1.00	0.164	0.86 ± 0.14

3.4 Health risk assessment (HRA)

The estimated daily intake (EDI) for the consumption of contaminated candies and their packaging ranged from 0.04×103 to $5.65 \times 103 \ \mu g/kg$ bw/day and from 0.01×103 to $1.09 \times 103 \ \mu g/kg$ bw/day respectively. The value of THQ of candies and packaging ranged from $1.5 \times 10-4$ to $174.2 \times 10-4$ and $0.4 \times 10-4$ to $33.5 \times 10-4$ respectively. The interpretation THQ is binary, the THQ either >1 or 1<.

When the THQ is >1, it is indicate a reason for health concern. All of the THQ values are <1 for all candies and packaging. Table 4 summarizes the EDI and THQ values for all samples.

4. Discussion

A similar study was reported by Kim et al. (2008) where the concentration of lead in 3 different type of candies ranged from ND to 1.31 mg/kg. Another study from Dahiya et al. (2005) reported that lead level ranged from 49 to $8.04 \times 10^3 \mu g/kg$ with an average of $9.3 \times 10^{-2} \mu g/kg$. Sugarbased candies were found to have higher contents of heavy metals compared to chocolate-based and milk-based candies. In comparison, study from Duran et al. (2009) have found that the average lead concentration in chocolatebased samples is 1.347 $\mu g/g$ and average concentration of lead in sugar based samples is almost 77% of the chocolatebased samples. Another study from Jacobs et al. (2004) have found that two sample out of eleven sample were exceeded the regulatory limit for lead. The highest concentration of lead detected was at 0.890 mg/kg.

The study by Kim et al. (2008) has also detected high lead concentration in 10 out of 92 candy packaging with the concentrations ranged from 110.3 mg/kg to 6394.1 mg/kg. The outer cover of these candy packaging was green or yellow in colour which may contained lead chromate used as inorganic pigments in ink or paint. An assessment of heavy metals in packaging prepared by The Toxic In Packaging Clearinghouse found that, 25 sample are detected exceeding the permissible limit for packaging which is 100 mg/kg with the total mean 1740 with different types of product categories (Council, 2007).

Overall, there was no association between type of candies and the concentration of lead; and type of candy packaging and the concentration of lead. Hence, no migration of lead from the candy packaging to the candy was observed in this study.

Provisional Tolerable Weekly Intake (PTWI) of 25 μ g/kg bw/week (3.6 μ g/kg bw/day) was used as an indicative value for comparing the results of EDI of lead in candies and their packaging. EDI for all candies and their packaging were below the PTWI except for one of the sugar-based candies with the reading of 5.65 μ g/kg bw/day. THQ value for lead in candies are higher compared to their packaging which implies that the candies possess higher potential risk than their packaging. However, THQ value for all candies and their packaging is <1. This indicates that there is no significant non-carcinogenic health risk. The value of THQ do not provide the quantitative estimation on the probability on the exposed population, it only serves as

an indicator of the risk level due to metal exposure (Iwegbue et al., 2014).

5. Conclusion

The candy samples were collected from the local market in Malaysia, and the concentration of lead were quantified. Generally, the concentration of lead in candies and their packaging do not exceed the permissible limit that established by FDA which is 100 μ g/kg. The result in this study shows that there are no significant different of lead in 3 types of candies and packaging. There was no significant of chronic non-chronic health risk due to the exposure of lead through digestion. The THQ for the entire samples are below 1 which showed no significant of chronic non-carcinogenic health risk for the consumption of contaminated candies.

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CONFLICT OF INTEREST

There is no conflict of interest present.

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Type of candy	Sample	Characteristic	Main colour of		Candy			Packaging	
canay			Pacing	Mean (μg/kg) ± SD	EDI	THQ	Mean (μg/kg) ± SD	EDI	THQ
Type I:	CS1	hard candy	white	0.09±0.02	1.30E-07	9.38E-02	0.06±0.03	9.30E-08	2.88E-04
Sugar	CS2	hard candy	dark pink	0.04±0.01	6.50E-08	2.00E-04	0.04±0.02	5.50E-08	1.69E-04
Based	CS3	hard candy	soft green	0.05±0.02	7.80E-08	2.42E-04	0.05±0.02	6.80E-08	2.10E-04
Candy	CS4	hard candy	orange	0.18±0.15	2.40E-07	7.45E-04	0.03±0.02	4.50E-08	1.38E-04
	CS5	hard candy	pink	0.04±0.02	5.50E-08	1.70E-04	0.04±0.01	5.90E-08	1.82E-04
	CS6	soft candy	white	0.06±0.07	8.30E-08	2.57E-04	0.05±0.02	6.20E-08	1.90E-04
	CS7	hard candy	orange	0.04±0.03	6.50E-08	2.00E-04	0.03±0.03	4.40E-08	1.36E-04
	CS8	hard candy	red	0.04±0.01	6.30E-08	1.95E-04	0.02±0.02	3.00E-08	9.00E-05
	CS9	hard candy	orange	0.20±0.09	2.80E-07	8.53E-04	0.05±0.01	6.40E-08	1.96E-04
	CS10	hard candy	red	0.18±0.07	2.40E-07	7.45E-04	0.05±0.02	7.50E-08	2.30E-04
	CS11	hard candy	green	0.19±0.07	2.70E-07	8.17E-04	0.07±0.02	9.40E-08	2.89E-04
	CS12	hard candy	red	0.93±1.41	1.25E-06	3.84E-03	0.08±0.02	1.09E-07	3.36E-04
	CS13	soft candy	red	0.06±0.03	9.30E-08	2.88E-04	0.01±0.03	2.00E-08	5.00E-05
	CS14	soft candy	green	3.34±3.23	4.46E-06	1.38E-02	0.02±0.01	2.30E-08	7.00E-05
	CS15	hard candy	white	4.24±0.44	5.70E-05	1.74E-02	0.02±0.01	3.00E-08	9.25E-05
Type II:	CM1	hard candy	green	0.17±0.01	2.30E-07	7.19E-04	0.30±0.10	4.00E-07	1.12E-03
Milk	CM2	hard candy	orange	0.26±1.5	3.50E-07	1.09E-03	0.02±0.04	3.00E-08	8.00E-05
Based	CM3	hard candy	pink	0.04±0.08	6.50E-08	2.00E-04	0.09±0.12	1.00E-07	3.74E-04
Candy	CM4	soft candy	yellow	0.17±0.2	2.35E-07	7.25E-04	0.08±0.06	1.06E-07	3.27E-04
	CM5	soft candy	pink	0.14±0.09	2.00E-07	5.91E-04	0.04±1.15	7.00E-08	2.06E-04
	CM6	soft candy	white	0.11±0.01	1.50E-07	4.63E-04	0.03±0.05	4.00E-08	1.34E-04
	CM7	soft candy	bluish purple	0.13±0.07	1.80E-07	5.55E-04	0.04±0.03	6.00E-08	1.74E-04
	CM8	soft candy	pink	0.15±0.15	2.10E-07	6.48E-04	0.02±0.04	3.20E-08	1.00E-05
	CM9	hard candy	orange	0.07±0.05	1.00E-07	3.19E-04	0.04±0.15	6.00E-08	1.86E-04
	CM10	soft candy	black	0.07±0.09	9.50E-08	2.93E-04	0.08±0.15	1.00E-07	3.44E-04

Table 4: Characteristics of samples and concentrations of lead in candies and their packaging.

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Turnet	Pb								
candy	Sample ID	Characteristic	Main colour of packaging	Candy			Packaging		
				Mean (μg/kg) ± SD	EDI	тно	Mean (μg/kg) ± SD	EDI	тно
	CM11	sticky candy	gold and silver	0.07±0.05	9.50E-08	2.93E-04	0.05±0.03	7.40E-08	2.27E-04
Type II:	CM12	sticky candy	black	0.04±0.01	6.00E-08	1.85E-04	0.02±0.02	3.40E-08	1.04E-04
Milk	CM13	hard candy	purple	0.06±0.03	8.00E-08	2.52E-04	0.04±0.02	4.80E-08	1.48E-04
Based Candy	CM14	soft candy	black and white	0.09±0.04	6.00E-07	1.82E-03	0.05±0.03	7.00E-08	2.01E-04
	CM15	sticky candy	gold and silver	0.04±0.06	4.00E-07	1.23E-03	0.8±0.30	1.09E-06	3.35E-03
	CC1	soft candy	silver blue	0.05±0.06	5.40E-07	1.68E-03	0.01±0.02	2.00E-08	6.00E-05
	CC2	soft candy	red	0.18±0.08	6.20E-07	1.91E-03	0.01±0.02	2.00E-08	5.00E-05
	CC3	sticky candy	silver	0.07±0.08	8.30E-07	2.56E-03	0.20±0.01	2.74E-07	8.45E-04
	CC4	soft candy	silver purple	0.35±0.06	4.80E-07	1.46E-03	0.11±0.17	1.00E-07	4.55E-04
	CC5	soft candy	green	0.23±0.03	3.00E-07	9.51E-04	0.07±0.02	1.00E-07	3.00E-04
	CC6	soft candy	gold and purple	0.47±0.03	6.30E-07	1.94E-03	0.01±0.01	1.80E-08	5.55E-05
Type III:	CC7	soft candy	green	0.14±0.08	2.00E-07	6.17E-04	0.03±0.02	4.00E-08	1.09E-04
Chocolat	CC8	soft candy	silver	0.07±0.06	1.00E-07	2.98E-04	0.05±0.05	7.00E-08	2.20E-04
e Based	CC9	soft candy	gold	0.07±0.01	9.50E-08	2.93E-04	0.02±0.03	3.00E-08	9.00E-05
Candy	CC10	soft candy	green	0.06±0.03	8.30E-08	2.57E-04	0.02±0.01	3.00E-08	9.25E-05
	CC11	soft candy	silver	0.18±0.10	2.50E-07	7.71E-04	0.01±0.01	1.40E-08	4.00E-05
	CC12	soft candy	silver	0.20±0.10	2.80E-07	8.53E-04	0.28±0.11	3.80E-07	1.17E-03
	CC13	soft candy	green	0.18±0.08	2.50E-07	7.66E-04	0.05±0.00	6.00E-08	1.95E-04
	CC14	soft candy	red	0.30±0.02	4.00E-07	1.25E-03	0.08±0.02	1.05E-07	3.24E-04
	CC15	soft candy	gold and	0.05±0.04	7.30E-08	2.26E-04	0.5±30	7.00E-08	2.20E-04
			dark purple						