Metal Contamination in Low-Cost Jewelry and Toys in Cambodia

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Background. The existence of lead-contaminated consumer products is a global issue. Toys and low-cost jewelry may contain toxic metals and Cambodia is known to have consumer products with toxic metals.
Objectives. It is important to inform Cambodians about sources of toxic metals so that they can reduce their exposure risk, particularly for children.
Methods. Student volunteers purchased, or brought from home, low-cost jewelry and toys to either the University of Health Science or a Don Bosco Institute in Phnom Penh, Cambodia, where they were analyzed using X-ray fluorescence (XRF). The initial analysis was performed in 2011. A subset of the 2011 samples was re-analyzed in 2015 using new preparation techniques and a new x-ray fluorescence (XRF) unit.
Discussion. The analysis of low-cost jewelry in Phnom Penh in 2015 indicated that lead in jewelry clasps is a more serious health concern than was first perceived in 2011. Mercury, nickel, cadmium and copper were also found in toys, and occasionally these toys had been produced by well-known companies. Sources of jewelry production of samples in the present study are unknown. Lead in clasps in low-cost jewelry appeared to be the greatest risk to children in our sampling.
Conclusion. One-third of toys and low-cost jewelry exceeded the United States and European Union guidelines for heavy metals. XRF analysis allows for rapid screening of lead and other toxic metals and could be used to reduce the sales of low-cost jewelry and toys containing toxic metals.
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Introduction

Cambodia imports most of its consumer products from nearby countries and the rate of economic growth of Cambodia has produced a high demand for consumer products.[1] This influx of consumer products requires a more rapid, effective system of quality control. Consumer products that are imported and sold in Cambodia can contain chemical contaminants that pose serious adverse health effects to consumers. About one-third of the skin creams analyzed in Phnom Penh contained mercury (Hg) in amounts higher than the Association of Southeast Asian Nations (ASEAN) recommended guideline of 1 $\mu g/g$.[2,3] Ninety percent of the enamel paints sampled in Cambodia exceeded a voluntary standard of 100 $\mu g/g$ lead (Pb) of the producing country, Thailand.[4] The problem is not limited to Cambodia, as studies in the US, India, and China have found elevated Pb in children's toys, low-cost jewelry, and paints.[5,6,7]

Elemental Pb is added to paints and plastic toys as a coloring agent and to prevent free radicals from reacting to form hydrochloric acid.[7,8] Sources of metals used to make low-cost jewelry include recycling facilities for lead acid batteries and electronic wastes.[5,9,10] Such recycled metals lower the cost of manufacturing and imitate shiny, better quality jewelry.

Common effects of Pb in children include reduced intellectual capacity, anemia, kidney damage and a suppressed immune system.[11,12,13,14,15] Blood lead levels (BLL) as low as 3 $\mu g/dl$ were associated with neurobehavioral deficits in visual motor integration, attention, reaction time and off-task behaviors.[16] Bellinger[17] found that adverse outcomes such as reduced IQ performance and academic deficits occurred at BLL <10 μg/dl, and at prolonged exposure there was association with attention deficit hyperactivity disorder (ADHD). Similarly, children aged 2 through 5 years had an 18-point increase in the Total Behavior Problem Score (TBPS) if their BLLs were higher than >15 μg/dL.[18]

Abbreviations			
As	Arsenic	*Ni*	Nickel
Cd	Cadmium	*Pb*	Lead
Cr	Chromium	*Sb*	Antimony
Cu	Copper	*Sn*	Tin
EU	European Union	*XRF*	X-ray fluorescence
Hg	Mercury	*Zn*	Zinc

Lead in Inexpensive Jewelry

In the United States, there are at least 4 million households with children who have been exposed to Pb, and approximately half a million US children one to five years old with BLLs above 5 μg/dL.[19] Recognizing that Pb exposure can affect every system in the body, the United States (US) Centers for Disease Control and Prevention (CDC) initiated a Childhood Lead Poisoning Prevention Program which is committed to eliminating BLLs > 10 μg/dL by 2020.[19] However, despite efforts to eliminate Pb exposure in children, there have been numerous cases reported in which workers and children have been the victims of Pb exposure. One of the lead exposure pathways is low-cost jewelry items, which are imported into the US from developing countries such as China.[6]

A study conducted by Weidenhamer and Clement[6] found that 77 out of 130 inexpensive jewelry samples imported from China to the US exceeded the former United States Consumer Product Safety Commission (USCPSC) guideline of 0.06% (600 μg/g).[20] Moreover, out of 311 inexpensive jewelry samples purchased, Maas *et al.*[21] found more than 50% of samples contained more than 3.0% Pb in at least one portion of the jewelry piece, while 39.5% of the samples contained more than 50% Pb and 29.2% contained more than 75% Pb.

The above findings were highlighted by the death of a four-year old child in Minnesota after ingesting a charm composed of 99.0% Pb and stress the need to improve monitoring and regulation of imported Pb-containing products in the US.[22] In 2011, the Consumer Product Safety Commission (CSPC)[23] issued a recall of 150 million pieces of metallic toy jewelry due to concerns over Pb levels.

A similar, serious incident was reported in 2009 when an American-born child of Cambodia-born parents was found to have been wearing an amulet obtained from a monk in Cambodia that contained more than 45% Pb. The level of Pb in his blood was 20 μg/dL.[24] Subsequently, the CDC[24] advised parents who had traveled to foreign countries and may have had their children wearing amulets or inexpensive jewelry items obtained from these countries to register for a BLL test out of concern over potential Pb contamination.

A recent analysis of low-cost jewelry in China found items with 65% Pb, 71% copper (Cu) and 37% cadmium (Cd), and 3-7 other samples had high levels of metals.[25] A survey in Seattle of children's jewelry in 2015 indicated that lead was still present in 23% of samples; the highest Pb concentration of 50,100 μg/g was lower than found in some earlier surveys, probably reflecting improved monitoring and management.[26]

Lead in Plastic Toys

In plastic toys, Pb is used as a pigment for color or as a stabilizer to provide rigidity and high heat stability.[9] Toys are an integral of part of a child's development.[10] Children with their hand-to-mouth habits can chip off parts of the surface of the contaminated toy, or leach the toxic element in their mouths. Because toys play an important role in a child's development during the hand-to-mouth stage, toys contaminated with Pb can obstruct a child's neurological development.[27]

Similar to inexpensive jewelry, over the past few years there have been multiple recalls of toys because of chemical safety hazards.[28] Table 1 reviews selected recalls on children's toys containing excessive Pb concentrations between 2004 and 2016. In India, Kumar and Pastore[10] tested 111 non-branded toy samples and found a level of Pb ranging from 0.65 – 2104 *μg*/g with the Mumbai average having the highest level (278.3 *μg*/g). Twenty percent of the painted toy samples analyzed exceeded the US Environmental Protection Agency (USEPA) guideline at the time of 600 *μg*/g. In Turkey, Aliyev *et al.*[29] collected 50 toys, 19 of which were manufactured in Turkey and 31 were imported from China. The results from the atomic absorption spectrometry analysis indicated that the mean Pb on the surface of the toys imported from China was 85.3 *μg*/g, compared

Distributor	N	Description	Substrate	Origin	Year
Four firms	1500000	Toy jewelry	Paint	India	2004[23]
Fisher-Price	967000	Toys	Paint	China	2007[35]
Mattel Inc.	253000	Toy cars	Paint	China	2007[36]
Greenbrier	300000	Beads & cars	Paint	China	2008[37]
Toys "R" Us	16000	Military figures	Paint	China	2008[38]
S.U. Wholesale	5000	Toys	Paint	China	2008[39]
Oriental Trading Co.	220000	Toy banks	Paint	China	2008[40]
Sportime	1000	Sports balls	Paint	China	2010[41]
Playmates toys	252000	Jewelry	Metallic	China	2010[42]
Blip toys	15000	Toys	Paint	China	2010[43]
Jide Trading	2100	Military toys	Paint	China	2010[44]
S&S Worldwide	1000	Wooden beads	Paint	China	2010[45]
LM Import & Export	1900	Toy cars	Paint	China	2011[46]
Build a Bear Workshop	28900	Toys	Paint	China	2011[47]
Cost Plus Inc.	1000	Toy drum	Paint	China	2011[48]
G.A. Gertmenian	600	Toy bowling	Paint	China	2011[49]
Dillon Importing	6970	Toy guns	Paint	China	2012[50]
Lee Carter Co.	7000	Wrestling figures	Paint	China	2012[51]
Discount School Supply	3700	Educational game	Paint	China	2014[52]
Minga Fair Trade Imports	135	Toys	Paint	Peru	2014[53]
Things Remembered	10000	Jewelry	Metallic	China	2014[54]
GSI Outdoors	6700	Water bottles	Lead solder	China	2016[55]
Far East Brokers	6000	Children's furniture	Paint	China	2016[56]
LaRose Industries	170000	Jewelry	Metallic	China	2016[57]
KHS America	150	Musical toy	Paint	Israel	2016[58]

Table 1—Toys Recalled Due to Lead Contamination—US Consumer Safety Product Commission

2004-2008 are examples of recalls, whereas 2010-2016 are inclusive of recalls by CPPC

Color*	Standard value	LPA-1 XRF	XRF 3t
Yellow	1.0 mg/cm^{-2}	8765	8389
Green	1.9 mg/cm^{-2}	16450	15555

Table 2— Calibration of XRF XL3t and LPA-1 XRF with a Certified Reference Material

Note: Values are in μg/g

*NIST Certified Reference Material, RMD Inc. Colors refer to paint reference materials.

with 41.4 μg/g for the toys made in Turkey. The difference in Pb content between these two groups of toys was significant (P<0.05).

Despite several federal recalls on toys contaminated with high levels of Pb, the following study continued to find excessive Pb content in children's plastic toys. Greenway and Gerstenberger[7] analyzed 535 toys collected from 10 different daycare centers in Las Vegas, Nevada using an XRF analyzer (XLt 797 2W) and found 29 toy samples (5.4%) contained a Pb concentration higher than the USCPSC standard in 2008 of 600 μg/g. There have been several reviews of an acceptable Pb level for toy surface coatings and paints, and the current US Pb guideline has been reduced to 90 μg/g.[30] The European Union (EU) has three categories for heavy metals in toys with the scraped-off category (160 μg/g for Pb) *(Table 3)* being the most relevant to toys in our study. Although significant, the concentrations of Pb in plastic toys in the literature are generally much less than that found in low-cost jewelry.

The purpose of this study was to use a handheld X-ray fluorescence analyzer (XRF) to evaluate the lead content in inexpensive jewelry and plastic toys that are sold in markets in Phnom Penh, Cambodia and compare the findings with the European Standard EU EN 71-3:2013 for scraped-off materials.[31]

Methods

XRF Calibration

The XRF analyzer (Niton Thermo Fisher, Billerica, MA; model XL3t) used in the initial analysis in 2011 was calibrated in the Thermo Fisher Scientific Laboratories (Billerica, Massachusetts, USA), verified several times in Phnom Penh with certified reference materials supplied by Thermo Fisher Scientific, and validated by comparison with another independent portable analytical tool, LPA-1 XRF, provided by the Environmental Health and Safety Office of Buffalo State, State University of New York. The LPA-1 XRF had been compared to other analytical procedures.[32] Consumer products were measured using either a general metals or plastic mode setting. The validation results are shown in Table 2.

In 2015, a new Niton XL3T970 handheld XRF analyzer was used to re-evaluate the clasps of jewelry collected in 2011. In 2015, we developed a simple technique to increase the size

		As	Cd	Cr	Cu	Hg	Ni	Pb	Sb	Sn	Zn
LOW-COST JEWELRY	Max detected	1704	3786	290	827366	677	477694	706510	66890	987706	21267
	EU Guideline	47	17	460	7700	94	930	160	560	180000	46000
	% EU Guide	4.5	12.5	0	18.2	3	13.6	19.1	15.9	1.1	0
	% 100x EU	0	1.1	0	2.3	0	0	11.2	1.1	0	0
TOYS	Max detected	4160	107	434	495469	204479	95551	3520	590	12239	90685
	EU Guideline	47	17	460	7700	94	930	160	560	180000	46000
	%EU Guide	6.3	4.8	0	6.3	26	7.9	4.2	1.8	0	31.8
	% 100x EU	0	0	0	0	6	1.5	1.4	0	0	0

Table 3—Heavy Metal Content of Low-Cost Jewelry and Toys

Max detected is the maximum detected concentration of metal.

% EU Guide is the percentage of samples that exceeded the EU guideline for scraped-off metals.

% 100x EU is the percentage of samples that exceeded the EU guideline for scraped-off metals by 100x.

of the clasps. The new XRF analyzer was calibrated in the Thermo Fisher Scientific laboratories, Hong Kong, and verified several times in Phnom Penh with certified reference materials supplied by Thermo Fisher Scientific. Furthermore, in 2015 a new Niton XL3T970 XRF analyzer with "small spot" capability was used at Cleverich Inc., Bangkok, to assess the added value of this instrument enhancement for this type of analysis.

Lead Content in Low-Cost Jewelry

Inexpensive jewelry pieces were purchased by student volunteers randomly from three markets in Phnom Penh, while other jewelry owned by female students attending a vocational training institute at the time (Don Bosco Vocational Training Institute) was analyzed in 2011. In total, there were 89 jewelry pieces, most of which cost less than US $5 each. Although the origin of these jewelry pieces could not be geographically determined, it is likely that they were imported from neighboring countries and China, but some might have been made in Cambodia.

Prior to and after the analysis, jewelry pieces were kept in plastic bags to prevent cross contamination. Because the jewelry pieces were different from one another in terms of length, composition and style, the jewelry part analyzed varied from one sample to another. Because we were concerned the jewelry clasps might be richer in lead (soft and dense) than the XRF results indicated, the 2011 clasps were stored for further analysis. In 2015, prior to XRF analysis, the clasps were flattened with a hammer into disks 7 mm to 10 mm in diameter. Subsequently, they were analyzed using the new model XRF in Cambodia and in Bangkok, using the "small spot" unit.

Lead Concentration in Toys

Seventy-one children's toys were purchased by students from local markets, and some by the senior author in Thailand. After the toys were tested on Plastic or General Metals mode for 30 seconds, they were kept in plastic bags in the laboratory or returned to the donor. All were collected in 2011.

Results

Lead Concentration in Low-cost Jewelry

Of the 89 jewelry pieces tested, 35% failed the EU guidelines for heavy metals. Of these, 18% were found with more than 160 μg/g Pb, the EU guideline for lead in scraped-off materials. Of concern is that 11% of these 88 samples contained 100 times the EU guidelines for lead.[31] These results, presented in Table 3, illustrate the lead content as measured directly with the Niton XL3T970 in 2015. In 2011, the concentration of lead measured with a XRF XL3t in the same 10 jewelry clasps was significantly less (*Figure 1*). It appears that the older analyzer was not as accurate in measuring small pieces, i.e. 4 mm in diameter or 10 mm by 4 mm. Furthermore, the analysis showed higher Pb levels in the clasps when they were flattened into a thin sheet and measured with the Niton XL3T970 in 2015 (*Figure 1*). The mean Pb concentration of the 10 clasps as measured in 2011was 8.9±9.6%; 40.2±22.2% in 2015 (unflattened); and 76.7±12.2% in 2015 (flattened). These latter clasps exceeded the EU guidelines for Pb in scraped-off materials of 160 μg/g Pb by 5000-fold. Although the increase in the diameter of the flattened clasp might appear to produce this effect, the details of the analysis suggest that the increased signal reflects a surface coating of copper and nickel (Ni) over a relatively pure lead core with minor impurities, especially antimony. In Figure 2, a relatively strong relationship between the lead and copper content of the clasps can be seen. The three outlying data points in Figure 2 are samples with about 40% nickel, which is considerably higher than the other clasps. These three outlying data points pull the line down from the rest of the data set. Excluding these outliers is one option, but an equivalent response is formed when the copper and nickel content are combined and plotted against the lead content, showing a strong relationship (r^2=.994)

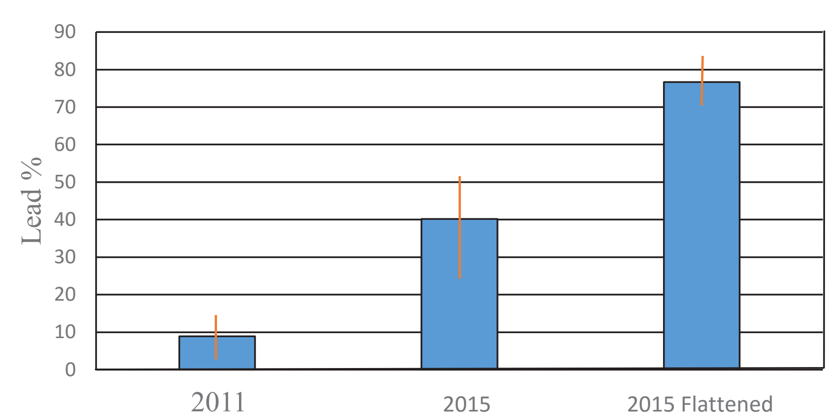

Figure 1 — Lead in clasps

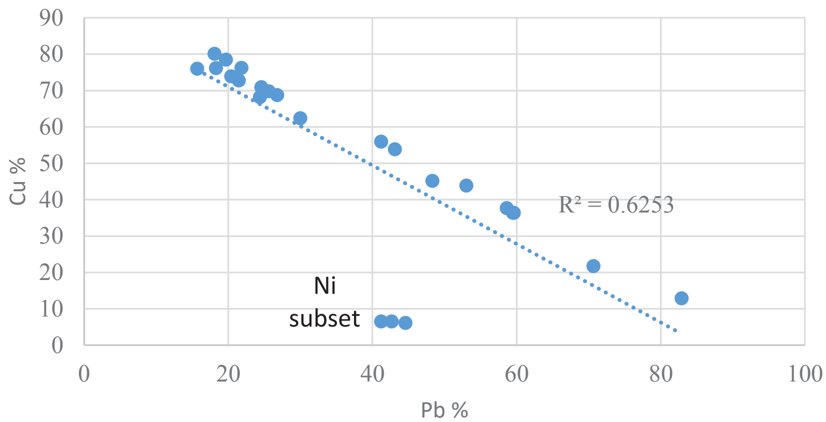

Figure 2—Lead clasps plated by copper

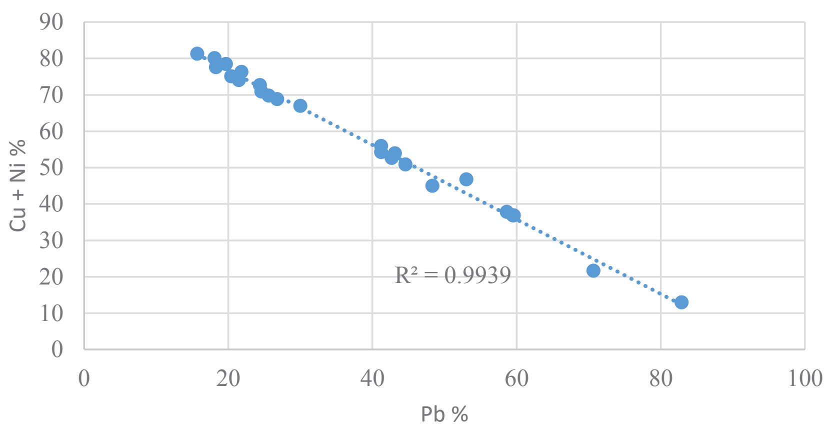

Figure 3—Lead clasps plated by copper and nickel

Figure 4—Toy car with paint chipped off

(*Figure 3*). When the clasps were flattened, sometimes the sides of the clasp looked quite different, with one side being much richer in lead and containing less copper and nickel. One flattened clasp was very different, with the lead content of one side at 39.3% and the other at 83%. Because of the high metal content, the analytical error was small. Prior to flattening, the 10 clasps processed in 2015 in triplicate had an average relative standard deviation of 10.2%. Further analysis supported this interpretation. We used another Niton XRF analyzer with "small spot" capability that can measure pieces 3 mm in diameter. For three jewelry clasps, in the "small spot" analysis, the lead content of the initial analysis and the flattened clasp was 61.9±26.9% and 84.0±5.3%, respectively. It is not possible to compare the results of the earlier analysis in 2015 to the "small spot" analysis, as flattening the clasp is a one-way destructive process. Furthermore, flattening removes any uncertainty about small size weakening the analysis.

Jewelry clasps of necklaces contained the highest levels of Pb, but other jewelry pieces commonly contained some lead: hairclips, 88,224 μg/g; earrings, 72,070 μg/g; necklaces, 64,544 μg/g; bracelets, 14,030 μg/g; rings, 3,523 μg/g; and anklets, 1,632 μg/g.

Lead most commonly seriously exceeded the EU guidelines, but Ni, Cu, Cd and antimony (Sb) also commonly exceeded the EU guidelines (13% to 18% of samples). Only zinc and chromium did not exceed the guidelines.

Metal Contamination in Toys

Out of 71 toy samples tested, four toys (6%) contained more than 100-fold the EU Hg guideline, i.e. >1% Hg, and 26% of toys failed the EU guidelines due to mercury. In two toy cars, the mercury-containing paint (1-2%) readily chipped off (*Figure 4*). Nickel and lead were also present in concentrations 100 times greater than the EU guidelines. Overall, 38% of toys failed the EU guidelines.

Discussion

The greatest health risk in low-cost jewelry is associated with lead, particularly in the clasps. Newer model XRF analyzers are very effective for screening lead, even with small pieces. At times, a surface coating can partially mask the actual lead content, but by flattening the item, a better estimate of the total lead content can be made. These coating interferences have been reviewed by Maas et al.[21] Our limited analysis of the small spot feature indicated that this feature would not resolve an underestimation of lead, as often there are surface coatings. It is possible that higher energy XRF analyzers using radioactive sources of X-rays, such as is used to measure leaded paint under layers of paint, might do a better job of detecting high levels of lead under surface coatings. In university environments, particularly when the XRF analyzer is used in educational clinics, the use of more powerful XRF analyzers might create restrictions. For example, licensing of more powerful XRF analyzers is more difficult.

Students can learn a lot about toxic metals in educational clinics.[33] Students can also educate their families and friends, and thus reduce the general exposure to toxic metals. This is a very effective way to get important information to the people most at risk. Moreover, having students collect items for analysis can remove the bias associated with purchasing of consumer goods; vendors often detect professionals and avoid selling bootleg items.

Handheld XRF analyzers with x-ray tubes and lower energy x-rays are sufficient for educational clinics and adequate for any review of toxic metals in such products. There is a great deal of variation in the coating on the clasps, including relatively pure lead, lead coated with a mixture of lead and copper, and primarily lead plated with nickel. It is possible that the nickel and copper might reduce the extraction of lead in gastric solutions, and this question should be resolved, but this would not make these clasps safe. There is still a high lead content on the surface of many clasps. Jewelry vendors at times admit to selling jewelry that can initiate skin irritations, and point out better quality jewelry that is a lot less likely to cause skin irritations. About 10-15% of women are known to be allergic to nickel[34], and individuals can commonly be sensitive to cadmium, copper, etc. However, some jewelry vendors are not so honest. In educational clinics at universities in Phnom Penh, we processed four "gold" rings that were sold as high quality gold, but were mostly copper. In our analysis of jewelry bought by students, we had very little opportunity to process expensive jewelry. Two attempts to work with owners of jewelry stores failed for different reasons. Initially, one vendor agreed that we could analyze his gold, but upon reflection, he changed his mind. We would not have been able to be onsite often to check the gold that he was buying, but we could likely have found counterfeit gold in his shop. Another vendor protested that all jewelry was the same, and if we found toxic metals in jewelry in her shop, she would lose many of her customers.

The problem of lead in jewelry requires further evaluation. The major source of the low-cost jewelry is not clear, but likely represents both local production in Cambodia and other countries in the region, including China. This significant issue has not been resolved due to lack of resources. The case of the boy poisoned by a lead amulet in the US that was bought in Cambodia[24] probably represents a much bigger problem in Cambodia. Many men and boys wear religious amulets, usually around their waist, but at times around their neck. These amulets are made of lead and are inscribed by monks for good luck. Sometimes amulets around the waist are wrapped in plastic, but sometimes they are worn directly on the skin. Very little study has been done on dermal absorption of lead, but it is generally believed to be lower than oral absorption. The greatest risk is when an amulet is worn around the neck of a child and the child (or adult) sucks on the amulet. It would be a useful exercise to evaluate the lead content in the blood of people wearing such amulets to assess their potential health risk. Handheld XRF analysis lacks the sensitivity to measure lead in blood and an alternative technique such as atomic absorption spectrometry is required.

In our study of toys, the greatest health risk was associated with mercury used in paints to produce shiny surfaces. As in other studies *(Table 1)*, we found toys with toxic metals produced by name brands/stores such as Mattel and Toys "R" Us with headquarters in the US. Bootlegging of consumer goods is widespread in Southeast Asia, so the producer is not always as labelled. It is also possible that retailers in Southeast Asia are selling old stock that was initially intended to be exported to the United States/EU and most likely would be detected in the developed world. However, a survey of jewelry in Seattle in 2015 found that lead was still present in 23% of samples.[26] It is likely that toxic toys continue to be present in the US, but hopefully as with the jewelry study in Seattle, at lower concentrations than found in earlier surveys.

Four toys (6%) contained more than 1% Hg and 26% of toys failed the EU guidelines for toys because of mercury. Overall, 38% of toys failed the EU guidelines. Technically, it is very easy to monitor mercury and other metals in paint, but the producing countries are not doing this very well. Future analysis in Southeast Asia should include assessment of mercury in paints used for other purposes. By far the most common cause for a CSPC recall of lead in the toy in the United States was associated with lead in paint *(Table 1)*. Since much of the developing world still uses enamel paint with lead,[4] painted toys in such countries should be expected to be occasionally contaminated with lead or even mercury. Although the four largest recalls of toys in the USA because of lead occurred between 2004 and 2008, the problem of lead contamination remains serious (*Table 1*).

No amount of Pb is considered safe for children, and there is a possibility they might chew and ingest the surface of a toy coated with Pb, contributing to elevated BLLs. Maas *et al.*[21] cited two studies that showed handling of Pb-contaminated items with lower Pb weight percent than the jewelry pieces in this study can result in significant transfer of Pb to the skin. Additionally, up to 4 μg of Pb could be transferred from the skin to the surface of food items after a single handling of items contaminated with Pb.[20]

Like the jewelry items, the Pb content of toys could not be determined by visual inspection. Different parts of the toy may or may not possess Pb or may contain uneven concentrations. The toys appeared in different colors and designs to which children are attracted. Only after using an XRF analyzer could the metals concentrations be determined quickly. Thus, XRF analyzers are reliable, robust tools for screening environmental samples and consumable goods.

Conclusion

Because there is a lack of treatment facilities and experience with Pb poisoning in Cambodia, it is important to implement prevention measures and make the public aware of the health risks. Processing consumer goods in educational clinics in schools can be an effective method of education. Labeling of certified consumer goods could help inform parents whether toys or jewelry are suitable for children. Increased monitoring in the US, the EU and in the developing world with XRF analyzers could increase consumer protection.

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