Hazardous Metals in Vintage Plastic Toys Measured by a Handheld X-ray Fluorescence Spectrometer

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Abstract **Over 100 plastic toys from the 1970s and 1980s, both polyvinyl chloride ("vinyl") and nonvinyl, were analyzed in the study described here using a handheld X-ray fluorescence spectrometer to quantify hazardous metal content. A sampling of recent vinyl toys was also tested. The majority of nonvinyl samples were Fisher Price brand toys. The vinyl toys consisted largely of Barbie dolls and other dolls. Overall, lead or cadmium was found in 67% of vintage plastic toys, frequently at concentrations exceeding current U.S. and European limits. Arsenic was detected at levels of concern in 16% of the samples. In the nonvinyl toys, heavy metal content was found to correlate with certain colors of plastic. The likely sources of the detected metals are discussed. None of the contemporary vinyl toys contained detectable cadmium, lead, or arsenic. Given that vintage toys remain in widespread use by children in homes and other locations, the results illuminate a potential source of heavy metal exposure for children.**

Introduction

Hazardous Metals in Contemporary Toys

Recent years have seen a sharp increase in revelations of so-called toxic toys and other everyday products contaminated with hazardous metals in the U.S. The reports have come from a number of sources: academic research studies, nonprofit centers, and occasionally, news outlets (Gregory & Roe, 2007; Guney & Zagury, 2012; HealthyStuff.org, 2008; Kumar & Pastore, 2007; Pritchard, 2012). Researchers have found handheld X-ray fluorescence (XRF) spectrometers to be convenient and accurate for rapidly testing consumer products (Reames & Charlton, 2013).

Prior to 2009, when the Consumer Product Safety Improvement Act (CPSIA) went into effect, the U.S. had no laws restricting heavy metals in consumer products, including toys. The CPSIA restricted total lead in children's products to 600 ppm in 2009 and reduced the limit to 100 ppm in 2011. (Note that ppm, parts per million, is equivalent to mg/kg.) No mandatory limits exist on cadmium or other heavy metals in toys, although voluntary guidelines borrowed from Europe were adopted.

Table 1 lists current U.S. and EU mandatory limits for five heavy metals in children's toys (Guney & Zagury, 2012). Some European countries have their own restrictions; Denmark's are listed as an example.

Usage of Vintage Toys

An Internet search revealed a robust trade in pre-1990 vintage toys, including toys identical to those tested in our study. Some buyers are collectors; some are parents seeking remembered toys to give to their own children. The market for old toys, together with personal observation, suggests that vintage plastic toys are in widespread use in homes, daycares, church nurseries, and waiting rooms. A search of the academic literature shows a lack of research on old toys. A handheld XRF study of daycare center toys carried out in the Las Vegas area found a high rate of lead- and cadmium-containing toys, both vinyl and nonvinyl, in 10 different child care centers (Greenway & Gerstenberger, 2010). In that study, however, the age and origin of the toys were not known.

Health Risks

The developing brains and bodies of infants and young children are especially vulnerable to toxic exposures. This is because 1) they absorb and retain lead more efficiently than do adults (Ziegler, Edward, Jensen, Mahaffey, & Fomon, 1978); 2) they are exposed to contaminated dust particles by playing close to the floor; 3) they chew on, mouth, and occasionally swallow items; and 4) they handle many toys and frequently put their fingers into their mouths (Guney & Zagury, 2012).

No safe blood level of lead has been established; even very low amounts in a child's body are linked to reduced intelligence (Canfield et al., 2003; Lanphear et al., 2005). The long-term consequences of lead exposure, which affects virtually every organ system

TABLE **1**

Governmental Restrictions on Heavy Metals in Toys

Metal	Limits in the U.S.	Limits in the EU
Cadmium	No mandatory limit*	Migratable cadmium <75 ppm[a] (EU); total cadmium <75 ppm (Denmark)
Lead	Total content <100 ppm	Migratable lead <90 ppm (EU); total lead <100 ppm (Denmark)
Arsenic	No mandatory limit	Migratable arsenic <25 ppm (EU)
Mercury	No mandatory limit	Migratable mercury <60 ppm (EU); total mercury <100 ppm (Denmark)
Barium	No mandatory limit	Migratable barium <1000 ppm (EU)

[a]ppm = parts per million.
*Although no limit is defined for toys, in children's jewelry, cadmium cannot exceed 300 ppm.

TABLE **2**

Examples of Toys Tested in Each Category

1970s–1980s non-PVC[a]	1970s–1980s PVC	2010–2013 PVC
Fisher Price Little People and accessories	Katie Kangaroo by Fisher Price (1976–77)	Animal figures
Fisher Price and Little Tikes stoves and cookware	My Little Pony and accessories	"Rubber" bath ducks
Barbie doll torsos	Barbie and other doll heads and legs	Barbie and other doll heads
Colored foods, brand unknown	American Girl doll (1988)	

[a]PVC = polyvinyl chloride.

in the body, are reviewed in Rosin (2009). Tests of bioavailability of lead and cadmium from toys and jewelry have been reviewed in Gunery and Zagury (2012). Cadmium, while less well studied than lead, appears to impact brain development (Kippler et al., 2012). Cadmium is also known to damage renal function, may contribute to osteoporosis, builds up in the placenta, and may increase cancer risk (Järup & Akesson, 2009).

Mercury, arsenic, and barium are additional chemicals of concern in children's products. Profiles of the toxicological effects of those metals can be found in the Agency for Toxic Substances and Disease Registry (ATSDR, 2011).

One concern with old toys is degradation of the materials. Plastic objects will degrade over time, releasing small plastic particles as well as embedded metals or metal compounds. Thus, vintage toys may pose a greater exposure hazard than new toys.

Methods

A handheld XRF spectrometer, model Niton XL3t-980 GOLDD+, was rented from Thermo Scientific. The XRF analyzer was calibrated by the manufacturer to allow quantitative elemental analysis for several material classes, including plastics, metals, and ceramics. Plastics mode was chosen for all samples in our study. Within plastics mode, the XRF analyzer identifies polyvinyl chloride (PVC) plastic based on chlorine content, but cannot distinguish between other types of plastic. In this article, "PVC" and "vinyl" are used interchangeably.

A calibration check was performed at the beginning and end of every test session and approximately every two hours in between. For plastics mode, the certified calibration standard used was a plastic disc impregnated with ppm levels of multiple metals.

Ninety-one plastic toys purchased in the 1970s and 1980s, 14 toys purchased between 2010 and 2013, and one doll from 2005 were collected from the homes of families in the Midwest. Examples of these toys are given in Table 2.

The toy surfaces were wiped with Kimwipes. The 77 nonvinyl toys were subject to a total of 125 XRF analyses on different solid-color parts. Likewise, the 26 vintage vinyl toys were subject to 61 scans and the 14 contemporary vinyl toys to 24 scans. Most toy parts were tested once, but a few were scanned twice, in some cases days apart, to verify repeatability. Each XRF scan consisted of a 30-second exposure to the X-ray beam while the analyzer collected and averaged spectra multiple times. Error ranges calculated by the XRF instrument for each element concentration are reported here as two standard deviations, 2σ, which is considered the 95% confidence interval (Piorek, 2009).

Toys were held steady in a clamp or placed on a high-density polyethylene block. The polyethylene block was chosen because it contained no elements that would confound the results from the test samples. Care was taken to position toys to avoid any signal from the clamp.

A thickness correction was programmed into the XRF analyzer before scanning toy parts that were thinner than 10 mm. A caliper was used to measure thickness for most samples. A few toy thicknesses were estimated by visual inspection because their shapes prevented caliper use.

Toys with curved surfaces presented a challenge. For all such toys, the flattest possible location was chosen for analysis to minimize air gaps and loss of quantitative accuracy. Also, since the XRF beam window is recessed, spherical surfaces such as the Little People heads could be covered by the window opening without air gaps.

An Omnic Fourier Transform Infrared (FTIR) spectrometer was used in transmission mode to help identify the plastic type of nonvinyl toys. Thin strips of plastic were shaved from four Fisher Price toys of different colors and placed in the path of the infrared beam in air. The resulting spectra were examined after subtracting a background spectrum.

Results and Discussion

Table 3 summarizes the measurements. A majority (69% and 66% of PVC and non-PVC, respectively) of vintage plastic toys contained either cadmium or lead or occasionally both metals.

The minimum concentrations counted in Table 3 were chosen as follows. 1) Cadmium and lead: readings of 90 ppm would be considered noncompliant (Piorek, 2009), or very close, because the upper error limit is close to the 100 ppm legal limit. 2) Arsenic: only readings above 65 ppm had error ranges of ±20% or less and thus could be considered quantitative (Piorek, 2009). 3) Barium: values >250 ppm showed unambiguous barium peaks in the spectra, whereas lower readings had high error ranges ($2\sigma > 30\%$).

The three toy categories—vintage nonvinyl, vintage vinyl, and recent vinyl—are discussed in separate sections below.

TABLE **3**

Summary of Hazardous Metal Measurements in Three Toy Categories

Measurement	Vintage Non-PVC[a] Toys	Vintage PVC Toys	Recent PVC Toys
# of toys tested	77	26	14
Detectable cadmium or lead	51 (66%)	18 (69%)	0
Lead >90 ppm[b]	26 (34%)	8 (31%)	0
Lead >1000 ppm	18 (23%)	3 (12%)	0
Cadmium >90 ppm	23 (30%)	10 (38%)	0
Cadmium >1000 ppm	9 (12%)	1 (4%)	0
Arsenic >65 ppm	14 (18%)	2 (8%)	0
Mercury >10 ppm	12 (16%)	1 (4%)	0
Barium >250 ppm	17 (22%)	18 (69%)	5 (36%)

[a]PVC = polyvinyl chloride.
[b]ppm = parts per million.

Vintage Nonvinyl Toys

Out of 91 old plastic toys collected for our study, 77 were nonvinyl or, in the case of the Barbie dolls, partly nonvinyl. Barbie dolls were found by XRF to have PVC heads and legs and non-PVC backs or torsos. Twelve doll torsos were therefore included in the nonvinyl toy category. (The heads and legs of the same dolls were included in the vinyl category.)

As described in the Methods section, plastic from four nonvinyl toys (excluding Barbie doll torsos) of different colors were analyzed by FTIR. The resulting spectra from the four samples showed similar spectral profiles. The spectra are consistent with polyethylene or polypropylene that has undergone partial oxidation, forming C=O bonds, as suggested by a peak at 1,739 cm^{-1} (Socrates, 2001). A peak due to vinyl CH_2 groups also appears at 908 cm^{-1}, which is consistent with commercial polyethylene but not polypropylene (Socrates, 2001).

The plastic strips were observed to float in water and to sink in 70% isopropyl alcohol. This places the density of the plastic between 0.88 and 1.0 g/cm^3, which is consistent with polyethylene but not polypropylene. We concluded that the toys (excluding Barbie torsos) are likely made of polyethylene, and that the chemical structure of the polymer has degraded to some degree since manufacture.

The Barbie torso material was not identified other than as a nonvinyl plastic containing no halogens. This plastic was difficult to scrape or shave off and was not analyzed by FTIR or by the density test.

Table 3 shows that 34% of the 77 nonvinyl toys would violate the current U.S. and Denmark limits on lead. Thirty percent would violate the limits on cadmium. Furthermore, 23% of the toys contained more than 10 times the lead limit, and 12% contained more than 10 times the cadmium limit. Of the cadmium- and lead-containing samples, most had one metal or the other, but not both.

Arsenic was unambiguously detected in 18% of nonvinyl toys (Table 3). Barium was measured >250 ppm in 22% of samples. Some of these toys might exceed EU limits for arsenic and barium (Table 1), but direct comparison is not possible because the EU standard is a migration, or soluble, limit and migration of metals from toys was not studied in this work.

Figure 1 shows lead and cadmium content as measured by XRF of individual nonvinyl samples sorted by color. Within each color, the results are shown in order of increasing lead content. Overall, the highest concentrations of both cadmium and lead were found in yellow toy parts. Most of the leaded yellow toys contained anywhere from 10 times to nearly 70 times the current limit of 100 ppm.

Green, orange, and brown nonvinyl samples were similarly likely to contain high lead or cadmium concentrations, including a few >1,000 ppm. Red and peach colors frequently contained lead or cadmium well over 100 ppm. Peach plastic was especially likely to contain excessive cadmium. In white, magenta, turquoise, black, and gray samples, cadmium and lead were either nondetectable or very minimal, although we note that only a small number of samples was available for those colors.

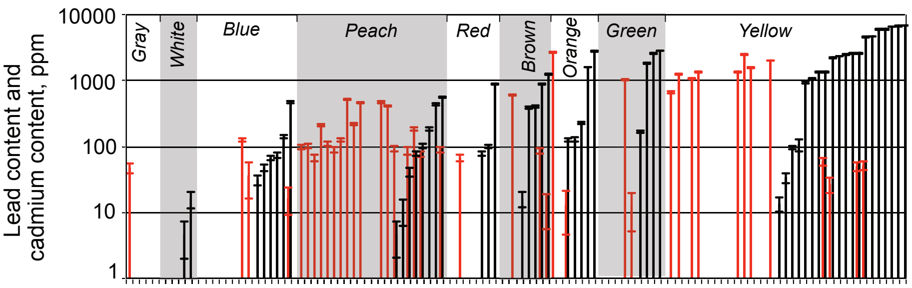
Twelve of the nonvinyl toys (16%) contained mercury below regulatory limits, ranging from 13.6±3.9 ppm to 46.4±5.1 ppm. Ten of these toys were the 10 light peach colored heads of Little People figures. Mercury was not detected in the 12 Barbie torsos of similar color.

All of the 14 toys containing arsenic (Table 3) also contained lead. Not all lead-containing samples contained arsenic, however. Arsenic measurement with XRF requires caution because the arsenic K peak overlaps with the lead L peak. The Niton XRF instrument was calibrated to differentiate between lead and arsenic. Failing to include a thickness correction for a sample under 10 mm, however, increases the chance of incorrect separation of the arsenic and lead peaks. In our study, thickness corrections were used when needed and the calibration checks gave reasonably accurate results for a plastic sample containing known amounts of both arsenic and lead. Hence, we take the arsenic measurements in our study to be real.

Why are hazardous metals present in so many old nonvinyl toys? The metals are almost certainly colorants or pigments. Lead chromate, $PbCrO_4$, also known as chrome yellow, was a standard plastic colorant in the era in which these toys were made. Lead sulfate ($PbSO_4$) and

FIGURE **1**

Lead (Black Bars) and Cadmium (Red Bars) Measured in Nonvinyl Vintage Toys Sorted by Color



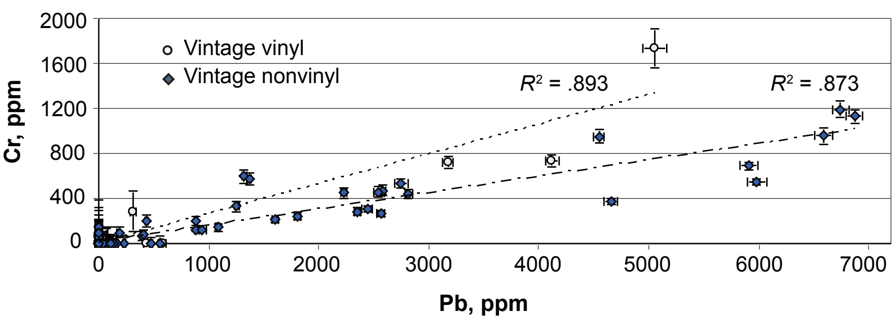
Each tick mark on lower x axis represents one toy part. Color of plastic is given at top of chart.

Ppm = parts per million.

Within each color, samples are arranged in order of increasing lead content. Black, magenta, and turquoise toys are not included because they contained no significant lead or cadmium. A log scale is used to display the wide range of measured concentrations. Error bars show ±2σ. Upper and lower error bars are shown for every measurement, but appear as one bar when the gap is small.

FIGURE **2**

Chromium (Cr) Content as a Function of Lead (Pb) Content in Nonvinyl (Shaded Diamonds) and Vinyl (Open Circles) Vintage Samples



Error bars represent ±2σ. Linear regression results are shown. Ppm = parts per million.

lead oxide (PbO) were mixed with the chromate in varying amounts to produce a color palette from light yellow to red (Rangos, 2003).

Similarly, cadmium compounds such as cadmium sulfide and cadmium selenide were—and still are—used to produce yellow and orange hues in plastics (Rangos, 2003; Vonkeman, Thornton, & Makuch, 2001). Mercury-cadmium sulfides were also used to produce colors from orange to maroon (Rangos, 2003).

In the 1980s and early 1990s, European countries restricted the use of these chemicals due to concerns about toxicity and environmental contamination, both during manufacture and disposal. As a result, the usage of lead-, cadmium-, and mercury-based colorants has been greatly reduced, and less toxic substitutes have been developed (Rangos, 2003; Vonkeman et al., 2001).

Although XRF cannot determine the chemical form of the elements in the tested samples, the data do provide evidence suggesting that lead, cadmium, and mercury are pigment constituents. Figure 2 shows measured chromium content as a function of lead content for the complete nonvinyl sample set (all colors). If lead is in the form of lead chromate, chromium content should increase linearly with lead content. Figure 2 shows a roughly linear trend. The ratio of lead to chromium in pure lead chromate would be approximately 4:1 by weight. In our measurements, most samples showed a lead:chromium weight ratio of around four or greater, even as high as 12. Ratios higher than four may reflect the presence of additional lead compounds added for color tuning such as lead oxide and lead sulfate (Rangos, 2003).

The six blue samples with detectable lead did not show detectable chromium. This may be because all but one of the blue leaded samples had lower lead (31±5 to 140±8 ppm), and if chromium is present in those, its concentration is likely below the limit of detection in XRF. Judging from the 2σ values, below 100 ppm the chromium readings cannot be considered quantitative. It is worth noting that all 23 samples containing at least 100 ppm chromium also contained significant lead.

The presence of cadmium did not show a trend with color as clearly as lead did, except for the peach samples (faces and heads of light-skinned Little People as well as Barbie torsos). All 11 of the peach Little People samples contained cadmium ranging from 68±7 to 523±13 ppm. Of the doll torsos, cadmium was found in 7 of the 12, ranging from 85±11 to 611±14 ppm. Since sulfur was not measured in the XRF mode used here, it is not possible to see if cadmium follows sulfur content, which might indicate cadmium sulfide pigments. Selenium was not detected in any samples, ruling out the presence of cadmium selenide.

Cadmium content was linearly correlated with barium content. A linear regression of cadmium versus barium gave a slope of 1.07 with an R^2 value of .93 (graph not shown). This relationship suggests a common source of both cadmium and barium in the majority of cadmium-containing toys. It is possible these toys contain a yellow or orange cadmium-based pigment, plus barium sulfate, which is white and often mixed with other colors to vary the shade.

Vintage Vinyl Toys

Twenty-six toys were vinyl or partially vinyl (e.g., doll heads and legs). Most of the vintage vinyl toys were dolls with peach or tan skin. Of the cadmium- and lead-containing samples, most had one metal or the other, but not both. Arsenic was clearly detected in two of the 26 toys. Barium was found in 69% (Table 3).

Since lead and cadmium were the most frequent toxic metals found aside from barium (Table 3), we focus on those. In general, both lead and cadmium in various chemical forms may be added to PVC for three purposes: as pigments, as stabilizers against degradation, and, less commonly, as plasticizers. Lead-based stabilizers such as tribasic lead sulfate and lead stearates continue to be widely used in PVC products (Jennings & Starnes, 2005). Cadmium-barium-based stabilizers have also been used.

In the 1970s and 1980s vinyl toys tested, lead and cadmium are mostly likely part of pigment compounds added for color. The evidence is as follows. First, the concentrations of lead and cadmium, while high relative to current acceptable limits in toys, are lower than would be expected for stabilizer compounds. If lead- or cadmium-based stabilizers were present, metal content should be on the order of 1%–3% (Vonkeman et al., 2001). The highest weight concentrations detected in vinyl toys here were ~0.1% cadmium and ~0.5% lead.

Second, the darker colored toys in our sample set had the highest levels of lead or cadmium, consistent with more pigment for a more intense shade. Although the majority of vintage PVC samples were light peach colored, a few were darker: a dark tan Barbie, a brown kangaroo, and a dark brown doll shoe. These contained, respectively, 1077±26 ppm cadmium in the cheek, 4123±57 ppm lead in the body, and 5052±106 ppm lead in the shoe bottom.

As with the nonvinyl toys, leaded vintage PVC toys may contain the pigment lead chromate. Figure 2 shows chromium content in vintage vinyl (open circles) as a function of lead content. The two metals are moderately linearly correlated. No significant correlations were seen between cadmium and barium (ruling out cadmium-barium stabilizers), cadmium and selenium, cadmium and chromium, or lead and barium.

Contemporary Vinyl Toys

As shown in Table 2, of the 14 recent PVC toys tested, none contained detectable cadmium, lead, arsenic, or mercury. Roughly one-third contained clearly detectable barium. The toys were a variety of colors.

Concerns about toxic metals in pigments and PVC stabilizers has spurred manufacturers to find substitutes (Jennings & Starnes, 2005). The sample set of recent PVC toys in our study was small, but the absence of detectable hazardous metals (with the exception of relatively small amounts of barium) likely reflects their overall decrease in toys sold in the U.S.

We caution, however, that hazardous heavy metals have been found in a significant number of new toys as recently as 2008 (HealthyStuff.org, 2008). Also, federal regulators have been criticized for not enforcing existing restrictions (Pritchard, 2012). Since new U.S. legislation took effect in 2009, it is possible that the frequency of contamination in toys has continued to decrease, but a larger sample set of new toys is needed to assess this.

Potential Exposure From Plastic Toys

As noted in the Introduction, solid polymers degrade over time as they are exposed to light, heat, oxidizing agents, and handling. Indeed, the vintage nonvinyl samples in our study are visibly rough and worn away along the edges. FTIR confirmed the presence of oxidized bonds. The heavy-metal pigments discussed above and likely contained in many of the vintage toys are not chemically bound to the polymers. Instead, pigments are tiny particles embedded within the polymer. Toxic particles may therefore be released due to chemical breakdown of plastic toys or by mouthing or handling (Guney & Zagury, 2012).

Limitations

Our study was limited to measuring hazardous metals present in vintage and new toys. Additional research is needed to determine how much heavy metal from a given toy will migrate onto a child's skin or be ingested during mouthing. Future work should therefore include tests that simulate handling and mouthing of the tested toys.

Conclusion

Vintage plastic toys frequently contain toxic heavy metals, particularly lead or cadmium, at concentrations exceeding current restrictions. Pigments appear to be a major source of these metals. Old toys are still in frequent use and thus present an exposure pathway that may be overlooked for children, especially children already burdened with toxic exposures from other sources.

Acknowledgements: The authors wish to thank the St. Ambrose University Summer Research Institute for funding. Special thanks to Dr. Jodi Prosise, Dr. Barret Ferm, and the Department of Chemistry. We received technical assistance from James Collins, Gary McEachern, and Ritch Vernon, all of Thermo Scientific. Finally, we acknowledge assistance from St. Ambrose students Barb Anderson, Kialee Bowles, and Alex Ducray.

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