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Multi-technique Quantitative Analysis and Socioeconomic Considerations of Lead, Cadmium, and Arsenic in Children's Toys and Toy Jewelry

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Abstract

A wide spectrum and large number of children's toys and toy jewelry items were purchased from both bargain and retail vendors and analyzed for arsenic, cadmium, and lead metal content using multiple analytical techniques, including flame and furnace atomic absorption spectroscopy as well as x-ray fluorescence spectroscopy. Particularly dangerous for young children, metal concentrations in toys/toy jewelry were assessed for compliance with current Consumer Safety Product Commission (CPSC) regulations (F963-11). A conservative metric involving multiple analytical techniques was used to categorize compliance: one technique confirmation of metal in excess of CPSC limits indicated a "suspect" item while confirmation on two different techniques warranted a non-compliant designation. Sample matrix-based standard addition provided additional confirmation of non-compliant and suspect products. Results suggest that origin of purchase, rather than cost, is a significant factor in the risk assessment of these materials with 57% of toys/toy jewelry items from bargain stores non-compliant or suspect compared to only 15% from retail outlets and 13% if only low cost items from the retail stores are compared. While jewelry was found to be the most problematic product (73% of non-compliant/suspect samples), lead (45%) and arsenic (76%) were the most dominant toxins found in non-compliant/suspect samples. Using the greater Richmond area as a model, the discrepancy between bargain and retail children's products, along with growing numbers of bargain stores in low-income and urban areas, exemplifies an emerging socioeconomic public health issue.

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46 **1. Introduction:**

47 Exposure to metals such as arsenic (As), cadmium (Cd), and lead (Pb) represents a
48 significant threat to children's health and their behavioral/intellectual development.¹² High
49 concentrations of these metals have been linked to hindered brain/sensory motor development
50 (As, Cd, Pb), decreased kidney function (As, Cd), gastrointestinal complications (Cd), bone
51 softening (Cd), and cancer.³⁴ At a critical developmental time, children under six are particularly
52 vulnerable to the harsh effects of these toxins.⁵ The risk of significant exposure from
53 contaminated toys/toy jewelry is elevated because of mouthing and threat of ingestion common
54 with children this age. The Intergovernmental Forum on Chemical Safety has identified a "risk
55 triangle" with metal-tainted children's toys, accessibility of the toys to children, and their
56 vulnerability to exposure.⁶ Even with well-known dangers and increasing regulations,
57 contamination of children's toys/toy jewelry continues to be a serious concern. In 2006, a four
58 year old died after ingesting a metallic charm containing lead, prompting the Consumer Product
59 Safety Commission (CPSC) to re-evaluate its lead regulations.⁷

60 Lead, cadmium, and arsenic may be present in products such as children's toys/toy
61 jewelry for a variety of reasons. Lead is often used as a stabilizer in certain plastics, a paint color
62 enhancer, or an anti-corrosion agent.⁸ As restrictions on lead have increased, cadmium has been
63 increasingly substituted.⁹ Similar to lead, cadmium is used to brighten paint color and stabilize
64 plastic, preventing hydrochloric acid formation that subsequently degrades the polymer.⁹ In
65 children's jewelry, cadmium can create a lustrous appearance and add mass to make the product
66 more realistic.¹⁰ The reason for the inclusion of arsenic in children's toys is unclear but may be
67 related to certain color dyes.¹¹

68 Government agencies have enacted restrictions on heavy metal concentration in food,
69 paint, toys, and many other children's products. The CPSC has issued numerous toy/toy jewelry
70 recalls including 79 product recalls in 2007 and 48 in 2008, which affected 14.5 million and 4
71 million toys, respectively.¹² Imported toys represent approximately 87% of the total toy market
72 with 74% imported from China. Of the 127 aforementioned recalls, 113 were for Chinese
73 imports.¹³ Demand for low cost products presumably drives Chinese manufacturers toward cheap
74 and often toxic materials, including inexpensive lead-based paints or metals leftover from China's
75 depreciating lead/nickel-cadmium battery industry and prevalent metallic electronic waste.^{14,15} With
76 the CPSC's 2012 endorsement of the American Society for Testing and Materials newest
77 regulation (F963-11), the United States has one of the most comprehensive standards for toys¹⁶ -

78 in line with legislation from the European Union (EN 71-3) and International Standards
79 Organization (ISO 8124).^{16,19}

80 Increasing regulation of these metals in consumer products has elicited an effort to
81 evaluate screening effectiveness and assessment of regulatory limits. Several reviews are
82 available, including Becker et al. (2010) and Zagury and coworkers (2012), that address heavy
83 metal contamination of toys/toy jewelry as well as summarize the legislative/regulatory/business
84 issues and challenges.^{20,22} Studies by Weidenhamer and coworkers^{14,23} represent important
85 contributions here, including a 2006 report where atomic absorption spectroscopy (AAS) was
86 used to determine lead in low-cost, purely metallic jewelry items (e.g., charms, bracelets) from
87 retail stores in different geographic locations - a study finding nearly half of 139 jewelry items
88 were over 80% lead by weight with an average lead content around 44% (wt.).²⁴ Their results
89 reiterated findings of a larger, 2005 study by Maas et. al which used flame AAS to identify 285
90 jewelry items with lead greater than 30% (wt.).²⁵ Using flame AAS, Weidenhamer et. al also
91 investigated lead contamination in the coating of inexpensive plastic jewelry, showing that many
92 products exceeded the then current U.S. regulatory limit.¹⁴ Using x-ray fluorescence
93 spectroscopy (XFS) and AAS, Weidenhamer expanded to cadmium screening of inexpensive
94 jewelry, finding non-compliant metal content (2011 CPSC standards; U.S. 2010e).²³
95 Overshadowed by lead, arsenic contamination was found using XFS in ~10% of jewelry items
96 studied by Saunders et al.²⁶ In terms of the bioaccessibility of metal contaminants in children's
97 products, Weidenhamer studied cadmium leached under simulated mouthing and digestion
98 conditions,²³ reporting a linear trend of extractable metal over time, dangerous if swallowed.
99 Zagury et al. reported the bioaccessibility of six metals under simulated gastrointestinal
100 conditions, concluding that lead and cadmium were particularly prone to high leaching and
101 exceeded EU regulation for items tested.²⁸

102 In this paper, a variety of nearly 100 children's toys and toy jewelry items were
103 purchased from either bargain or retail chain stores and were analyzed for arsenic, cadmium, and
104 lead with multiple analytical techniques, including different types of AAS and XFS, to determine
105 if the products violate current regulation and assess potential socioeconomic consequences related
106 to the contaminated products. To our knowledge, a larger, more comprehensive multi-metal,
107 multi-technique study of children's toys/toy jewelry items with different composition and from a
108 range of store-types was lacking from the literature. During the course of our research, Zagury et
109 al. published a seminal report examining metal content of ten metals, including those targeted
110 here, in a large number of North American marketed children's toys/jewelry.²⁸ Our effort builds

111 on these studies, adding multi-technique analysis and socioeconomic considerations by analyzing
112 and comparing the metal content of children's toys/toy jewelry from bargain and retail stores.

113

114 **2. Experimental:**

115 *2.1 Samples/Digestion.* Children's toys/toy jewelry samples labeled "Made in China" were
116 randomly selected/purchased from nationally recognized bargain and retail chain stores in the
117 Richmond metropolitan area. Multiples of each toy or jewelry item were attained including low
118 cost items (<\$5.00) purchased from both store types and high cost items (>\$5.00), not available at
119 bargain stores, only purchased from retail stores. Separate analysis of individual samples
120 representing multiples of the same toy were reported as an average (Supporting Information or
121 SI). Toys/jewelry, as received or cut into smaller pieces, were digested with concentrated HNO₃
122 (Standard Method 3030). To aid removal of coatings, certain samples were stirred and/or heated
123 during digestion. Acid digestions involving metallic products/components can be violent and
124 were covered to protect against analyte loss. Sample digests were subsequently gravity-filtered
125 (Whatman #43), diluted in volumetric glassware (18 MΩ water), and stored in plastic bottles until
126 analysis. Post-digestion solids were oven dried 24-48 before massing.

127

128 *2.2 Atomic Absorption Spectroscopy Analysis.* Sample digests were screened for cadmium and
129 lead on a Varian AA240FS flame atomic absorption spectrophotometer (FL-AAS) whereas
130 samples were tested for arsenic, cadmium, and lead on a Varian GTA120AA240Z graphite
131 furnace atomic absorption spectrophotometer (GF-AAS). A stabilizing matrix modifier,
132 Ni(NO₃)₆•6H₂O, was added (50 ppm) to samples/standards tested for arsenic. Standard calibration
133 curve analysis (SI, Figs. SM-1 and SM-2) was performed on digested samples. As a secondary
134 confirmation, standard addition (SA) analysis was run on specific samples, the range of SA
135 increasing the sample signal 1.5-3.0 times the original value (SI, Fig. SM-3). Standardized
136 aqueous metal solutions provided routine quality control of calibration curve effectiveness (GF-
137 AAS; FL-AAS) with discrepancies quantifying the standard solution resulting in the generation of
138 a new curve.

139

140 *2.3 X-ray Fluorescence Spectroscopy Analysis.* A Niton XL3t-700 X-Ray Fluorescence Analyzer
141 (Thermo) was operated in multiple modes within the "Consumer Goods" function of the device as
142 per manufacturer recommendation. The exact mode (e.g., painted product, metals/ceramics,

143 plastics) used was matched with the assessed sample composition (60 second scans). Reference
144 samples (Thermo) were routinely analyzed for quality control/instrument functionality checks.

145

146 *2.4 Bioavailability Studies.* Select non-compliant samples (BLCJP-01, BLCJP-02, BLCJP-06)
147 were analyzed to determine bioavailability of metal contaminants via established protocol,
148 relative bioavailability leaching procedure (RBALP), designed to simulate ingestion without
149 certain digestion enzymes and other physiological conditions deemed negligible with regard to
150 bioavailability.³⁰ Briefly, toys/toy jewelry were subjected to a 96 hour end-over-end (~ 10 rpm)
151 wash in a dilute HCl (0.4 M) in glycine solution (pH 1.53; 37°C). Wash aliquots taken between 6
152 and 96 hours were analyzed using GF-AAS. A matrix matched standard (HCl-glycine)
153 calibration curve was used with results additively compounded after certain intervals for total
154 metal leaching over time.

155

156 **3. Results and Discussion**

157 Children's toys/toy jewelry items, selected at random and purchased from both bargain
158 and retail stores in the greater Richmond metropolitan area, were analyzed for arsenic, lead, and
159 cadmium content. Analysis for these metals was performed with multiple analytical techniques
160 including flame (FL) and graphite furnace (GF) atomic absorption spectroscopy (AAS) and/or x-
161 ray fluorescence spectroscopy (XFS). Metal concentrations determined from these techniques
162 were compared to the United States Consumer Product Safety Commission (CPSC) regulation
163 levels (CPSC, F963-11).

164

165 *3.1 Analysis of Toys/Toy Jewelry From Bargain Stores*

166 **Table 1** summarizes, by analytical technique, the average and range of detected
167 concentrations for arsenic, cadmium, and lead in jewelry/toys purchased from bargain stores-all
168 considered "low cost" (<\$5.00). A sample was labeled non-compliant if the metal concentration
169 of arsenic, cadmium, or lead concentration exceeded the CPSC limit of 25, 75, and 90 ppm,
170 respectively, with additional confirmation by a secondary technique. Non-compliant toys/toy
171 jewelry measurements are shaded black. If only one technique showed metal concentrations
172 exceeding CPSC limits, the sample was classified as "suspect" and shaded grey. A third
173 classification, identified by bolded outline, signifies "borderline" products with metal
174 concentrations below regulation but still high enough to elicit concern due to cumulative metal
175 exposure over time.

176 Table 1 shows 16 of 46 (35%) bargain samples were found to be non-compliant for at
177 least one of the metals as confirmed by two different analytical techniques. Of the remaining toys
178 (30), an additional 10 toys/toy jewelry items were classified as suspect for one or more of the
179 metals because they exceeded limits on one analytical technique. Thus, out of the 46 toys/toy
180 jewelry tested from bargain stores, nearly 60% (26 of 46) were determined to be contaminated at
181 some level by at least one metal. This result (i.e., 60%) is similar to previous reports employing
182 only one analytical technique to investigate toxic metal content in low cost children's
183 toys/jewelry.^{24,28} The similarity of percent non-compliance from one technique studies suggests
184 that a conservative, multi-technique approach, may more effectively indicate actual non-
185 compliance. Single technique analysis have disadvantages that can affect results, including
186 digestion ineffectiveness (AAS) and inherent XFS variability stemming from analyte depth,
187 proper use, and the user's operational mode selection.

188 Of the 16 items identified as non-compliant, all but two (BLCTP-03, BLCTM-04) were
189 classified as "jewelry" for children, with the bulk composition of these items split between plastic
190 (6) and metal (8). More than half of the non-compliant products (56% or 9 of 16) were found to
191 exceed CPSC regulations for lead with one also having excessive cadmium content (BLCJP-02).
192 The other non-compliant metal items (7 of 16) were found to exceed the acceptable arsenic limit.
193 While multiples of each item were tested and the results averaged, a significant level of
194 variability, similar to other reports of this nature,^{8,12,14,28} persisted in the measurements. As in those
195 studies,^{12,28} the variability is partly attributed to analyzing toys/toy jewelry from different
196 production periods (i.e., the same toy purchased from different locations at different times). Prior
197 reports suggest variability may stem from opportunistic use of contaminated raw materials at the
198 manufacturing stage.¹⁵

199 The matrices of some of the digested toys/toy jewelry required for AAS analysis are
200 likely complex, eliciting concern over matrix effects on analyte signal. To compensate, standard
201 addition (SA) methodology, utilizing the sample's own matrix, was employed. Most of the
202 sixteen non-compliant products (Table 1) identified in initial screening (i.e., two or more
203 techniques (FL-AAS, GF-AAS, or XFS) measured concentrations over CPSC limits) were
204 subsequently confirmed with SA (SI, Table SM-1). Some samples (BLCJM-03, BLCJM-07,
205 BLCJM-11, BLCJP-04, BLCJP-005, BLCJP-09) initially measuring just below CPSC limits were
206 eventually labeled non-compliant (Table 1) upon additional SA analysis (GF-AAS). Collectively,
207 SA analysis with GF-AAS, confirmed the non-compliance claim of all 16 samples. SA analysis
208 (GF-AAS) was also used to check the "suspect" status bargain toys that initially exceeded CPSC

209 limits when tested with GF-AAS. In all cases with samples originally deemed “suspect,” the
210 classification was sustained by SA analysis (SI, Table SM-2).

211

212 *3.2 Analysis of Toys/Toy Jewelry From Retail Chain Stores*

213 A primary objective was comparing children’s toys/toy jewelry purchased from bargain
214 versus large retail stores to determine if place of purchase correlated impacted non-compliance.
215 To facilitate this comparison another 46 children’s toys/toy jewelry items were purchased from
216 retail chain stores in the same Richmond (VA) area and surveyed for As, Cd, and Pb with XFS, a
217 surface analysis technique. Retail toys/toy jewelry were further categorized into low cost (LC),
218 <\$5.00, (n=23) or high cost (HC), >\$5.00, (n=23) items. Because many of the bargain toy/toy
219 jewelry samples found to be non-compliant (9 of 16, 56%, Table 1) were acid-digested to remove
220 only the outer coating (rather than the entire product), retail store samples were only surveyed
221 with XFS to assess CPSC compliance. XFS, the industry standard for commercial testing, allows
222 for non-destructive (i.e., no acid digestion) and high-throughput screening of products. Of the 46
223 products, only 7 (15%) were found to contain unsafe levels of one or more of the three analytes
224 (**Table 2**) and were designated as “suspect” based on the use of a single technique (XFS). As in
225 the case of bargain products, children’s jewelry at retail stores was the product of greatest concern
226 with 71% of the items deemed suspect. The overwhelming majority of retail store regulatory
227 violations (86%) were from arsenic. While our study focused on arsenic, cadmium, and lead XFS
228 detected non-compliant levels (>60 mg/kg, F963-11) of mercury in 19 of 92 items total (20%),
229 including 12 bargain products and 7 retail products.

230 In comparing collective product analysis of bargain stores versus retail stores, several key
231 findings emerge. First, the rate of non-compliant/suspect toys/toy jewelry from bargain stores
232 (57%) is nearly four times that of items purchased at large retail stores (15%). This finding
233 indicates that the type of store from which these products are purchased may be a significant
234 factor in public exposure. Secondly, both store types showed similar patterns in which of the
235 three targeted metals products were found. Of the non-compliant/suspect bargain store samples,
236 54% (14 of 26) and 73% (19 of 26) were in violation of CPSC limitation for lead and arsenic,
237 respectively, whereas only 14% and 86% of suspect retail items were high in lead and arsenic
238 (SI, Fig. SM-4). The high arsenic levels in both types of samples may be a symptom of the
239 attention lead contamination has received over the years compared to lesser known toxins as well
240 as new policies of retail chains aimed at banning lead from children’s toys.^{31,32} Lastly, it is evident
241 from the results that jewelry items, as opposed to toys, are much more likely to have metal

242 concentrations that exceed CSPC limits, and therefore represent a greater threat to children who
243 accidentally ingest them. Nearly 73% (24 of 33) of the total number of samples found to exceed
244 limits with one or more techniques, regardless of purchase origin, were classified as children's
245 jewelry (SI, Fig. SM-5). Metallic-based jewelry (16 of 24, 67%) was more problematic than
246 plastic jewelry (8 of 24, 33%), a result consistent with the Zagury/Guney²⁸ report on low cost
247 metallic jewelry versus Weidenhamer's examination¹⁴ of low cost plastic jewelry (45% vs. ~10%
248 high in lead, respectively).

249 In the current study, analysis of only low cost (LC) items, regardless of the place of
250 purchase, resulted in a suspect/non-compliance rate of 42% (29 of 69 LC items), a rate similar to
251 previous studies dedicated to LC items. If one examines only data for low cost items, the
252 discrepancy between retail and bargain is slightly larger - a 57% non-compliant/suspect rate for
253 bargain stores versus a 13% rate at retail stores. A 2005 report by Maas and coworkers studying
254 lead content of inexpensive jewelry items, reported that nearly 60% of the 285 items tested had
255 unacceptable levels of the metal.²⁵ Weidenhamer et al. focused on lead analysis in low cost,
256 metallic jewelry,¹⁵ and later in low-cost plastic jewelry,¹⁴ reported non-compliance rates of 42%
257 and 9%, respectively. A more recent study (2010) by Green, found only 4% of jewelry exceeded
258 the lead limit allowable by California law.³³ Collective results of this and prior studies suggest a
259 trend toward lower rates of lead non-compliance in low cost, children's items. However, results
260 from the current study, suggest that price may not be as significant of a factor as the origin of
261 purchase (i.e., type of store). Out of the 7 suspect items from the retail stores, there was a nearly
262 even split between low cost (n=3) and high cost (n=4) products. If XFS determined "suspect"
263 samples are assumed to actually be non-compliant, the difference between bargain vs. retail
264 stores, while slightly less pronounced, remains significant (SI, Figs. SM-6 and SM-7).

265 The finding that low cost items from retail stores have a smaller likelihood of unsafe
266 arsenic, cadmium or lead content suggests, as in other studies,^{24,28} that the selection of supplier may
267 be critical. One explanation postulated in the literature is that manufacturers are being
268 opportunistic in the production of the toys/jewelry, using cheaper, contaminated raw materials to
269 lower costs.¹⁵ Alternatively, some national retail chains have, as of 2007, taken pre-emptive steps,
270 self-imposed standards, testing, and enforcement, including requiring suppliers take back shipped
271 merchandise failing to meet those standards.^{31,32}

272

273 *3.3 Bioavailability Studies*

274 While total metal content of a completely digested sample or the outer coating of a
275 toy/toy jewelry item is significant, an important aspect of overall toxicity is the bioaccessibility of
276 the metals in contaminated children's products - that significant ingestion of toxins can occur
277 either through ingestion or mouthing behaviors with the toys/jewelry. While ingestion may
278 induce more acute illness or death, the latter mode is of particular concern with young children
279 because of the frequency of the behavior (i.e., mouthing, licking, chewing, sucking), their critical
280 developmental stage of life, and the cumulative health effects of continuous exposure.²¹ Studies
281 show a median of 39 and 9 contacts per hour for toy mouthing in children under two and over two
282 years old, respectively.²¹ Lead, for example, has a half-life of 35 days in erythrocytes, two years in
283 the brain, and decades in the bone, with evidence of greater adsorption into the gastrointestinal
284 tract and brain of children compared to adults.²⁷ With almost 90% of our non-complaint samples
285 being small pieces of jewelry, bioavailability through mouthing and/or ingestion is of significant
286 concern.

287 To assess bioavailability, a number of the 16 bargain store non-compliant items with a
288 removable coating were selected. GF-AAS analysis of extracts from the bioavailability digests
289 over time revealed a significant amount (e.g., ~0.04 µg Pb/hr) of lead leached from the items (SI,
290 Fig. SM-9), albeit below the CPSC regulation for migratable metal (e.g., 6 µg Pb/6 hours). While
291 our levels of migratable metal were lower than other reports,^{23,28} those studies were often limited to
292 bulk metallic items with much higher starting total metal concentrations (e.g., 10,000-100,000
293 ppm Cd).^{23,28} Extrapolation of our results, suggests the leachable lead content of items tested
294 would exceed CPSC regulation after ~100 hours of exposure. Weidenhamer reports²³ similar
295 findings to our study: leeching was linear with time and accelerated if the item was purposefully
296 damaged prior to exposure to simulate wear-and-tear. Given the cumulative nature of these
297 toxins in the body, extended exposure over time remains a significant concern even with items
298 yielding migratable amounts well below government standards.

299

300 *3.4 Socioeconomic Considerations*

301 Our results suggest that for inexpensive toys/toy jewelry (<\$5.00), manufactured in
302 China, the origin of purchase (i.e., type of store) may be one of the most significant factors in
303 assessing potential health risks to child consumers. Coupled with the increasing number and
304 success of bargain stores in the current sluggish economy, our findings may serve as a harbinger
305 of an important emerging socioeconomic public health issue.³⁴ Bargain stores have seen their net
306 retail sales as well as the number of stores drastically increase between 2005 and 2011 while

307 retail chain stores continue to recover from recession (2008).³⁴ A 2011 national review noted that
308 the store count of the four major dollar store chains (21,500 locations) exceeded the number of
309 three major national drugstore chains (19,700 locations).³⁴ If the increasing popularity, numbers,
310 and profit margins of these bargain stores are considered in concert with our results, a greater
311 number of children could be at risk for both acute (ingestion) and chronic (mouthing behaviors)
312 exposure to contaminated toys/jewelry.

313 Bargain stores have achieved their recent success with a strategy built around offering the
314 consumer convenience and low prices by stocking an inventory with larger percentages of
315 inexpensive imports predominantly from China.^{33,34} Studies suggest, however, that their success is
316 partly a function of strategically locating their stores to target specific consumer demographics:
317 low-income families (low prices), urban center populations (convenience), and rural populations
318 with limited access to retail chains (convenience).³⁴ Dollar General, for example, is the dominant
319 rural bargain store, with over 70% of its stores servicing communities of less than 20,000.³⁴
320 Family Dollar stores, on the other hand, tend to purposefully locate in less desirable urban
321 markets. A 2012 study of bargain store locations in the country showed a disproportionate
322 number in low income states such as West Virginia, Mississippi, Alabama, and Louisiana.³⁵ The
323 study indicated high correlation of bargain store density with areas populated with mostly blue
324 collar working families, lower levels of education, and low median income.³⁵

325 In the context of this current report, consumer demographics in the aforementioned
326 studies identify those most likely to purchase these items and have greater risk for acute or
327 chronic metal exposure – a group also more likely to not have adequate health insurance coverage
328 and suffer health issues. In 2009, The Urban Institute (TUI) showed low-income families (i.e.,
329 average incomes below twice the federal poverty level) had lower rates of health insurance
330 coverage and were more likely to be uninsured, including an estimated 15% of children (2005).^{36,37}
331 The Institute for Research on Poverty, examining the Affordable Care Act's effect on low income
332 families, reported that this demographic is particularly vulnerable, suffering from poorer health
333 outcomes, lower life expectation, higher chronic illness rates, and more unaddressed health needs
334 compared to middle or upper class families.³⁸ In short, those most vulnerable to exposure may also
335 be the least likely to recognize and treat health consequences from toys/toy jewelry with unsafe
336 metal levels, creating a socioeconomic driven public health hazard.

337 Using the greater Richmond, Virginia metropolitan area as a model, locations of bargain
338 versus large retail chain stores reflect our investigations' suggested socioeconomic implications.
339 Defined by the I-295/SR-288 barrier, **Figure 1** shows locations of Richmond's three major chains

340 of bargain stores (60 locations) versus retail stores (19 locations). Interestingly, the bargain stores
341 are more concentrated within the city limits (i.e., urban center) and to the east and south of the
342 region. Retail stores, on the other hand are more prevalent in the north and west of the greater
343 Richmond area. An overlay and examination of average income per region indicates a close
344 correlation.³⁹ Bargain stores are more concentrated in low income and urban areas where large
345 retail stores are much less plentiful. Store locations are likely the result of store assessment, with
346 large retail stores selling children's products at a higher cost not wanting to directly compete with
347 the convenience and low prices of the bargain stores in these areas.⁴⁰ We note that 12 of the 19
348 (>60%) retail stores are found in Richmond's affluent "West End" (i.e., highest median
349 household income)³⁹ along with notably fewer bargain stores. This model system may be
350 representative of similar metropolitan areas across the country, suggesting that this issue may be a
351 developing national concern.

352

353 **4. Conclusion**

354 Multi-technique, multi-metal spectroscopic analysis of a number of randomly selected
355 children's toys/toy jewelry items indicates a significant difference exists in the toxic metal
356 content between products purchased at bargain versus retail stores, establishing origin of purchase
357 as a major factor in assessing the safety of these particular products. The discrepancy between
358 bargain and retail products is likely to be even more pronounced if sample selection were
359 conducted with bias from these findings or prior studies.^{9,10,12,14,23-25,28} With a nearly four-fold higher rate
360 of violation for low cost children's toys/toy jewelry compared to retail stores, bargain stores that
361 seem to also employ marketing strategies targeting low-income and urban areas, may be creating
362 an socioeconomic public health concern. Healthcare-centered reports indicate consumers most
363 likely to suffer even low levels of poisoning from these metals in these products are also more
364 likely to suffer from poor health or not have health insurance. This study suggests that stores can
365 make decisions to limit exposure risk to consumers by screening their own merchandise, more
366 careful selection of import manufacturers, and/or decreasing their percentage of imported
367 products. This particular issue will experience another variable with the advent CPSC
368 certification tags (June 2013) for toys 3rd party independently tested for metal contaminants,
369 though voluntary and cumbersome requirements for certification may dampen participation and
370 effectiveness.⁴¹

371

372

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379

380 **Supporting Information (SI)**

381 Additional experimental details; Representative FL-AAS and GF-AAS calibration curves, SA
382 analysis (GF-AAS); Data tables: SA analysis (GF-AAS) for non-compliant and "suspect"
383 toys/jewelry; Analysis summary as a function of metal; bioavailability GF-AAS analysis of
384 selected non-compliant bargain toys/jewelry.

385

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Figure 1. GoogleMaps© of greater Richmond, Virginia metropolitan area showing locations of (A) three national chain bargain and (B) three national retail stores. Regions reflecting highest median household income between 2005-2010⁹⁹ is shaded yellow (B).