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Concentrations and migratabilities of hazardous elements in second-hand children's plastic toys

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Abstract

About 200 second-hand plastic toys sourced in the UK have been analysed by x-ray fluorescence spectrometry for hazardous elements (As, Ba, Cd, Cr, Hg, Pb, Sb, Se) and Br as a proxy for brominated flame retardants. Each element was detected in > 20 toys or components thereof with the exception of As, Hg and Se, with the frequent occurrence of Br, Cd and Pb and at maximum concentrations of about 16,000, 20,000 and 5000 μg g^{-1}, respectively, of greatest concern from a potential exposure perspective. Migration was evaluated on components of 26 toys under simulated stomach conditions (0.07 M HCl) with subsequent analysis by inductively coupled plasma spectrometry. In eight cases, Cd or Pb exceeded their migration limits as stipulated by the current EU Toy Safety Directive (17 and 23 μg g^{-1}, respectively), with Cd released from yellow and red Lego bricks exceeding its limit by an order of magnitude. Two further cases were potentially non-compliant based on migratable Cr, with one item also containing > 250 μg g^{-1} migratable Br. While there is no retroactive regulation on second-hand toys, consumers should be aware that old, mouthable, plastic items may present a source of hazardous element exposure to infants.

Introduction

Mounting evidence for their acute and chronic toxicities at relatively low doses has resulted in increasingly restricted use of many heavy metals and metalloids in consumer products over the past few decades (1,2). Young children are particularly susceptible to the health impacts of such elements because of their higher metabolic rate, greater surface area to weight ratio, more rapid growth of organs and tissues, and longer time period to develop diseases with latency periods compared with adults (3). Infants are also potentially exposed to greater quantities of metals and metalloids in consumer products through the mouthing of non-food objects as they seek oral stimulation and explore taste, texture and shape. Consequently, products designed to be used by (or in contact with) young children have received particularly stringent regulatory attention in terms of both the concentrations and migratabilities of toxic chemicals in accessible components.

The original European Council Directive 88/378/EEC on toy safety (4) stipulated migratable limits for eight hazardous elements, listed in Table 1, that are based on the ingestion of a small quantity of material and defined by two-hour extraction under simulated gastric conditions (dilute HCl at 37 ^{o}C) according to the European standard, EN 71-3 (5). An amended directive that applied to products placed on the market from July 2013 provided revised limits on migration in dilute HCl that depended on the matrix being tested (liquid or sticky; brittle, powder-like or pliable; material that can be scraped off) (see Table 1), as well as limits for an additional number of elements and different oxidation states of Cr (6). Precise concentration limits for some elements have since been revised downwards in line with new scientific data, and there is currently a proposal to substantially reduce the concentration limits of Pb because of a growing body of scientific evidence that suggests there is no lower threshold of safety for its levels in blood (7).

Table 1: Migratable limits (in $\mu g\ g^{-1}$) of eight hazardous elements in toys and as defined by the original and amended EC Toy Safety Directives. Note that different species of Cr are discriminated in the latter directive, and that limits for Ba, Cd and Pb have since undergone (or are currently being proposed for) further reduction, with revised values shown in parentheses.

	88/378/EEC	2009/48/EC		
		brittle, powder, pliable	liquid, sticky	scraped-off
As	25	3.8	0.9	47
Ba	1000	4500 (1500)	1125 (375)	56000 (18,750)
Cd	50	1.9 (1.3)	0.5 (0.3)	23 (17)
Cr	60			
Cr (III)		37.5	9.4	460
Cr(VI)		0.02	0.005	0.2
Hg	60	7.5	1.9	94
Pb	90	13.5 (2.0)	3.4 (0.5)	160 (23)
Sb	60	45	11.3	560
Se	500	37.5	9.4	460

While the current Toy Safety Directive applies to new products, there is no retroactive regulation on the recycling or re-sale of older toys. Second-hand toys are an attractive option because they can be inherited directly from relatives and friends or obtained cheaply and readily from charity stores, flea markets and the internet (8). Moreover, in the UK the re-use of working, old and outgrown toys is actively encouraged through the Toy Recycling in Your Community ('Tric') scheme (9). Whereas liquid or sticky toys have a limited shelf-life and parents are likely to be wary of second-hand toys that are brittle or have visibly flaking paint, old plastic products often appear to be in good condition, presumably because of the durability of synthetic polymers and the color-fastness of many pigments, and tend to be re-used more extensively.

Despite the market for second-hand plastic toys and their abundance in homes, waiting rooms, day-care centers and nurseries (8), there has been little scientific study of the presence of hazardous elements in such items. Specifically, two studies conducted in the US and employing a handheld x-ray fluorescence (XRF) spectrometer report total concentrations of Pb in a variety of vintage plastic toys from day centers and family homes that exceeded US limits of 100 $\mu g\ g^{-1}$, but migration from the matrix was not evaluated (8,10). Other metals and metalloids, including As, Ba, Cd and Cr, are mentioned in the latter study, but none of these elements have US mandatory limits in toys with the exception of Cd in jewelry.

The present study describes the first systematic investigation of the occurrence and migratability of hazardous elements in second-hand plastic toys in the UK. Portable XRF was used to determine the concentrations of the eight elements listed in the original toy Safety Directive (and defined in Table 1) in addition to Br as a proxy for brominated flame retardants (BFRs) and whose concentration is limited in electronic plastic waste according to EU Directive 2002/95/EC (11). Based on the XRF results, selected samples were subjected to extraction in dilute HCl in order to evaluate element migratability and potential for exposure through ingestion.

Experimental Section

Sample collection and categorization

About 200 toys that were designed for young children, containing parts that were small or accessible enough to be mouthed in part or in whole and that had been acquired second-hand or second-generation, were sourced from two pre-school nurseries, a primary school, various charity shops and five family homes within the city of Plymouth, south west England. Toys were constructed entirely or largely of synthetic, molded plastic (and not foam, rubber or textile) and excluded products that housed electrical parts or that had been painted. Products and distinct components thereof (e.g. different colored blocks, various body parts of figures, separate constituents of games and puzzles) are listed in Table S1 of the Supporting Information where they are categorized as follows: ‘activity’ (balls, marbles, yoyos, tools, letters), ‘cars and trains’ (and other toys with wheels, plus accessories), ‘construction’ (blocks and studded bricks), ‘food-related’ (bottle caps, cutlery, crockery, model food), ‘figures’ (animals, dinosaurs, dolls, characters), ‘games and puzzles’ (board games, shape sorters, numeracy toys), ‘jewelry’ (straps, beads, rings) ‘sound-generating’ (rattles, whistles, bells, musical instruments) and ‘water’ (mainly bath toys).

XRF analysis

Toys and components were analysed by energy-dispersive FP-XRF using a Niton XL3t 950 He GOLDD+ that was configured nose-upwards in the laboratory and in a 4000 cm^3 Thermo Scientific accessory stand. The instrument was operated in a low-density, ‘plastics’ mode with thickness correction down to 50 μm. A suite of elements may be determined in this mode but the present study focuses on the eight metals and metalloids defined in the original Toy Safety Directive (listed in Table 1) and that are generally regarded as most hazardous for children (12), as well as Cl and Br as indicators of polyvinyl chloride (PVC) BFRs, respectively.

Where possible, sample thickness was determined through the measurement surface using 300 mm Allendale digital calipers, while for hollow objects (mainly dolls, balls, model food and sound-generating items) thickness was estimated from accessible surfaces of objects of similar construction and rigidity. Samples were placed on the stainless steel base plate of the accessory stand with the measurement surface above the XRF detector window before analyses with appropriate thickness correction and collimation (3 mm or 8 mm beam width depending on the accessibility and homogeneity of the area to be probed) were activated remotely through a laptop for counting periods of 40 s at 50 kV/40 μA (main energy) and 20 s at 20 kV/100 μA (low energy). Spectra were quantified by fundamental parameters yielding elemental concentrations in parts per million ($\mu g\ g^{-1}$) and with a counting error of 2σ that were downloaded to the laptop via Niton data transfer (NDT) software.

Measurement detection limits (as 3σ) varied depending on sample thickness and composition, but median values based on counting errors arising from analyses that failed to deliver a concentration ranged from < 10 $\mu g\ g^{-1}$ for As, Br and Pb to about 300 $\mu g\ g^{-1}$ for Ba. (A complete list of detection limits can be found in Table S1.) Polyethylene and polyvinyl chloride (PVC) reference discs of 13-mm thickness and that had been impregnated with As, Ba, Cd, Cr, Hg, Pb, Sb and Se (PN 180-619, LOT#T-81), Cd, Cr, Br, Hg and Pb (PN 180-554, batch SN PE-071-N) or Br and Sb (PVC-4C80) were analysed at regular intervals throughout each measurement session and returned concentrations that were always within 10% of mean certified values. A detailed comparison of metal concentrations in plastics derived by portable XRF and, as an independent measure, by inductively coupled plasma (ICP) spectrometry following digestion in H_2SO_4, is given elsewhere (13).

Sample extraction

Based on the results of XRF analysis, and subject to sufficient accessible material and permission to sacrifice, 34 components of 26 plastic toys (and as described in Table S1) were extracted in dilute HCl according to EN 71-3 for scraped off material (5). Hard plastics were fractionated to < 2 mm through a stainless steel grater, with 100 to 300 mg of material collected on white A4 paper subsequently transferred to a series of pre-weighed 50 ml polypropylene centrifuge tubes with the aid of a Nylon brush; softer plastics (mainly PVC) or thin items were cut into small (< 5 mm) pieces using stainless steel scissors or a scalpel before being transferred to centrifuge tubes likewise. Ten ml of a solution of 0.07 M HCl, simulating the human gastric environment and prepared by dilution of Fisher Scientific Trace Analysis grade acid in Elga ultrapure water, was then pipetted into each tube and the screw-capped contents inverted twice before being placed in a water bath at 37 ^{o}C for 2 h (1 h under lateral agitation and 1 h without agitation). At the end of the incubations, 5 ml from each tube were filtered through Whatman 0.45 μm PES syringe-cartridge filters into 10 ml polypropylene centrifuge tubes and diluted to 10 ml with 0.07 M HCl.

Extract analysis

Within 24 h of preparation, extracts were analysed for As, Ba, Br, Cd, Cr, Hg, Pb, Sb and Se by ICP mass spectrometry using a Thermo X-series II (Thermo Elemental, Winsford UK) with a concentric glass nebulizer and conical spray chamber. The instrument was calibrated externally using five mixed standards prepared by serial dilution of mixed standard solutions (LabKings, Hilversum, NL) in 0.07 M HCl, and internally by the addition of 10 μg L^{-1} of ^{115}In and ^{193}Ir to all samples, standards and blanks. RF power was set at 1400 W and coolant, auxiliary, nebulizer and collision cell gas flows rates were 13 L Ar min^{-1}, 0.70 L Ar min^{-1}, 0.75 L Ar min^{-1} and 3.5 mL 7% H_2 in He min^{-1}, respectively. Data were acquired over a dwell period of 10 ms, with 50 sweeps per reading and three replicates. Limits of detection, based on three standard deviations about multiple measurements of blanks ranged from 0.05 μg L^{-1} for Cd and Pb to about 10 μg L^{-1} for Br. Analyses of a reference drinking water (EP-L-2; SPC Science, Quebec) after every ten samples revealed analyte concentrations that were within 10% of certified values with the exception of Sb (within 15%).

Results

Sample categorization and elemental concentrations

The number and categorization of plastic toys and components considered in the present study is summarized in Table 2. Thus, 285 XRF analyses were performed on 197 products, with multiple measurements undertaken on distinctly different components of various toys like the wheels, base and body of a car, different colored blocks in a set, and the constituent parts of a puzzle or game. Overall, 28 products were constructed of PVC (defined by the XRF as having a Cl content greater than 15% by weight; 14), with the majority of this polymer encountered in the figures and water categories. Also shown in Table 2 is the number of measurements in which each hazardous element was detected by XRF. Thus, Hg was not detected in the products analysed, and the sequence of decreasing number of cases detected among the remaining elements was: Ba > Br/Cr > Sb > Cd/Pb > As > Se; with Ba and Br encountered across all categories considered. Hazardous elements were most frequently detected among plastic figures, construction toys, games and puzzles, and jewelry, where, on average, more than one element was detected per measurement, and detection was least frequent among activity products, cars and trains and toys designed for use in water.

The total concentrations of each element, where detected by XRF, are illustrated in Figure 1 in ascending order, with PVC- and non-PVC-based materials discriminated and statistical summaries for each dataset annotated. Also shown is the number of cases in which total concentrations exceed the respective migratable limits defined by Directive 2009/48/EC and its subsequent amendments (see Table 1). (Note that the higher migratable limit for Cr in its lower oxidation state is shown and, while Br is not included in the Toy Safety Directive, the value presented is based on the Restriction of Hazardous Substances concentration limit for certain brominated flame retardants, but not total Br, in electronic plastics; 11.) While the two measures are not directly comparable, total concentrations that exceed migratable limits should act as a trigger for further investigation of a product. Thus, overall, there were 73 cases of exceedance, encompassing 49 measurements and 31 products (of which seven were PVC-based) from all categories with the exception of toys designed for water. Co-associations of multiple elements exceeding their respective limit values were encountered in two types of bead (Br-Cd-Pb-Sb), a number of Lego bricks (Cd-Se or Cr-Sb), a small games mat (Ba-Pb), various games counters (Cd-Se) and figures (Cd-Pb), and the plastic bowl of a bell (Cr-Sb).

Table 2: Quantity and categorization of toy samples, along with the number of analyses performed and the number of cases in which each hazardous element was detected.

	products (analyses)	PVC (analyses)	As	Ba	Br	Cd	Cr	Hg	Pb	Sb	Se
activity	38 (45)	4 (5)	1	12	7	0	2	0	2	0	0
cars and trains	25 (31)	1 (1)	1	7	3	1	1	0	2	2	0
construction	25 (46)	1 (1)	3	15	4	7	12	0	2	9	3
figures	39 (71)	11 (16)	5	24	18	10	13	0	15	10	1
food-related	21 (21)	2 (2)	0	5	3	0	7	0	0	0	0
games and puzzles	24 (41)	1 (1)	1	20	3	5	5	0	2	3	2
jewelry	8 (8)	0	1	5	4	2	3	0	4	3	0
sound-generating	7 (9)	0	0	2	3	0	1	0	0	1	0
water	10 (13)	8 (9)	0	3	3	0	0	0	0	0	0
total	197 (285)	28 (35)	12	93	48	25	44	0	27	28	6

Hazardous element migratability

Extraction tests were performed on 34 components of 26 toys that could be sacrificed and that were homogeneous, readily accessible and yielded sufficient quantities of plastic material on grating or slicing. These included many of the items where the XRF returned one or more hazardous element above its corresponding EU Directive migration limit (Table 1) in order to ascertain compliance/non-compliance, and samples of lower elemental concentrations but of a variety of color and type in order to explore possible controls on and variations in migratability. Significantly, and despite constraints on the amount of material required for extraction, a number of items tested (e.g. small blocks, counters and beads) were small enough to be ingested whole.

Table 3 presents results for all samples in which at least one element was detected in the extract by ICP (n = 30) in terms of both weight-normalized migratable concentration and percentage migration relative to total elemental content (i.e. a measure of bioaccessibility). Note that where the element was detected in the extract but not by XRF, a lower limit of bioaccessibility is given that is based on the detection limit returned by the Niton XL3t (and as shown in Table S1). Thus, while As and Se were never detected in the extracts, Ba and Cd were encountered in 30 and 11 cases, respectively, and in a variety of toys. Migratable concentrations ranged from < 1 μg g^{-1} for Cd in two components to > 100 μg g^{-1} for Ba, Br, Cd, Pb and Sb in at least one case each, and bioaccessibility ranged from below 1% for Ba, Cd and Cr in a number of products to over 10% for Cr in a molded food toy and Pb in a black bead. Among the elements considered, and despite variations in percentage bioaccessibility, statistically significant correlations between migratable concentrations and total concentrations were exhibited by Cd (n = 11, r = 0.825, p = 0.002) and Pb (n = 7, r = 0.972, p < 0.001).

With respect to the current EU Directive, non-compliance occurred for Cd in four yellow or red building bricks from two Lego sets, and for Pb in two body parts of a PVC-based model dinosaur and on both surfaces of a child's PVC tape measure. Although concentrations of extractable Cr were compliant in respect of total migration, it is suspected that a number of brightly colored items or parts (a building block, model dinosaur and tape measure) and a black bead were non-compliant in respect of Cr(VI) since the co-association of Cr and Pb suggests the presence of the pigment, lead chromate. Overall, therefore, various components from four toys were non-compliant, with a further two products potentially non-compliant.

Table 3: Migratable concentrations (in μg g^{-1}) and percentage bioaccessibilities (in parentheses and relative to total concentrations) of hazardous elements detected in the extracts of different toys or components thereof that are numbered and lettered

according to the identification given in Table S1 and that are classified as PVC- or non-PVC-based. Note that figures in bold denote non-compliance or potential non-compliance according to the current EU Toy Safety Directive and the proposed, revised limit for Pb.

sample/component, colour	category	Ba	Br	Cd	Cr	Pb	Sb
1a. tape measure, red	activity (PVC)	13.2 (0.28)			**0.34 (>0.50)**	**137 (3.4)**	
1b. tape measure, white	activity (PVC)	7.8 (>0.1)				**163 (3.6)**	
2. building brick, pink	construction (PVC)	6.9 (0.74)		0.18 (0.18)			
3. building brick, purple	construction	3.8 (>1.6)					
4. Sticklebrick, green	construction	6.6 (>4.7)					
5. megablock, yellow	construction	24.1 (5.1)			**3.2 (0.77)**	16.0 (1.2)	
6a. Lego brick, yellow	construction	62.5 (7.1)		**217 (3.1)**			
6b. Lego brick, grey	construction	10.1 (>2.3)		11.3 (0.87)			
6d. Lego brick, red	construction	394 (3.8)		**274 (1.4)**			
6e. Lego brick, red	construction	79.8 (3.5)		**18.0 (0.19)**			1.1 (>0.93)
7. building block, red	construction	58.2 (2.6)		7.9 (0.30)			
8. Lego brick, yellow	construction	10.2 (2.6)		**105 (1.6)**			
10a. dinosaur model, red	figure (PVC)	68.7 (8.1)		8.6 (8.9)	**4.4 (3.5)**	**41.1 (4.0)**	6.3 (1.5)
10b. dinosaur model, grey	figure (PVC)	71.3 (4.9)		8.8 (7.3)	**4.5 (2.8)**	**43.7 (3.9)**	
11. farm animal, white	figure (PVC)	3.4 (>1.4)					
12. dinosaur model, brown	figure (PVC)	3.6 (1.0)			0.59 (>2.8)		
13. helmet, grey	figure	2.5 (>2.4)					
14. molded food, brown-red	food-related (PVC)	3.1 (1.0)			18.6 (1.8)		
15. molded food, yellow	food-related (PVC)	6.4 (3.8)					
16. molded food, red	food-related	2.6 (>1.9)			0.76 (0.26)		
17. plate, yellow	food-related	4.3 (>2.0)			1.8 (2.6)		
18. bowl, yellow	food-related	3.3 (>1.7)			1.6 (5.6)		
19. spoon, green	food-related	6.6 (3.9)					
20. molded food, brown	food-related	2.1 (1.4)					
22a. cup, orange	games and puzzles	1.2 (0.11)		0.83 (0.03)			
22b. mat, brown	games and puzzles	23.9 (0.02)				6.6 (1.1)	
23. counter, red	games and puzzles	14.3 (0.19)		6.6 (0.27)			
24. cylinder, blue	games and puzzles	2.1 (>1.9)					
25. bead, black	jewellery	50.5 (4.1)	257 (1.8)		**4.5 (20.4)**	18.2 (10.8)	104 (1.2)
26. bell, orange	sound-generating	7.9 (>8.5)			3.65 (0.32)		

Discussion

The results of the present study reveal high concentrations of many elements listed by the original EU 88/3781/EEC Toy Safety Directive (4) in products that remain in circulation, being handed-down by parents, recycled via charity shops, and donated to or purchased (historically) by nurseries, hospitals and schools. Both the frequencies of detection and median elemental concentrations are greater than corresponding values for new plastic toys sourced from major retailers (15,16) but are more in line with measurements reported for new, low cost items purchased from bargain stores and road-side vendors (16,17,18). Regarding older plastic toys, similar conclusions to the present study have been reached for Pb in toys in day care centers in Nevada (10) and for several hazardous metals in products purchased in the 1970s and 1980s in the USA but whose precise source was not specified (8). The latter studies, however, implicitly relate exposure and health hazard to total elemental concentrations in plastic, in line with current US restrictions, rather than migratable levels. The results shown in Table 3 clearly indicate a wide variation in bioaccessibility on a percentage basis for any given element, with correlations between total and migratable concentrations significant only for Cd and Pb. Presumably, this reflects variations in product composition, age and usage, but suggests that total concentration is not, necessarily, a good proxy for exposure though ingestion.

Elemental concentrations extracted by dilute HCl in this study that exceed current or proposed EU migratable limits include Cd and Pb (and potentially Cr) in various building blocks and bricks, and in PVC figures. In these products, Pb appears to have been employed in compounds used as stabilizers in PVC or, in association with Cr(VI), as the brightly colored yellow pigment, $PbCrO_4$, whose precise hue may be varied through to red by addition of $PbMoO_4$ or $PbSO_4$ (19). Cadmium was evident as a stabilizer in one PVC-based component (the black caterpillar track from an excavating vehicle) but was more generally encountered in a variety of brightly-colored toys. Here, the compound CdS, or a mixture of CdS and ZnS, is likely to have been employed as a yellow pigment, with successively darker hues of orange and red effected by the progressive replacement of S by Se (19). Consistent with this assertion, Se was absent from all yellow toys that were Cd-positive, while the mass ratio of Cd:Se increased from about 4 in an orange games cup to about 7.3 in a dark red counter.

Cadmium and Pb-based pigments found widespread use in plastics because of their pure, brilliant shades, opacity, light-fastness and weather-resistance, and, at least when new, high chemical resistance and little tendency to migrate (20). Accordingly, such pigments had a key role in manufacture of colorful toys before health and environmental concerns resulted in their restriction and replacement by safer organic and inorganic alternatives. Of the toys analysed in the present study, the highest levels of total and migratable Cd were encountered in some, but not all, red and yellow studded Lego bricks. Specifically, those in sets that appear to have been purchased in the 1970s yielded migratable Cd concentrations that sometimes exceeded the EU migration limit by an order of magnitude, while those purchased in the 1990s, and that were visually indistinguishable from the older bricks, contained no detectable Cd. The introduction of high quantities of Cd in Lego bricks is likely to have coincided with the introduction of acrylonitrile butadiene styrene (ABS) as a replacement for cellulose acetate as the material of construction in the 1960s (21) since Cd sulphoselenides were favorable colorants for styrenic-based polymers at the time (22); when, precisely, Cd-based chemicals in Lego were subsequently replaced by safer pigments, however, is unclear. Given their popularity, durability, collectability and compatibility with newer products, older, ABS-based Lego sets, and in particular those containing brightly-colored pieces, should be treated with caution.

One element not embraced by the original or amended Toy Safety Directive but of concern from a health perspective is Br. Although the halogen is found in the organic pigment, copper phthalocyanine green 36, that has limited use in plastics (23), its most important application in synthetic polymers is as a constituent of free radical-scavenging BFRs. These chemicals represent a wide variety of organic compounds designed to increase resistance to ignition and slow down developing fires in heat-generating materials like electronic casings and construction materials. Because many commonly-employed BFRs are persistent, bioaccumulative and toxic, their use in new and recycled electrical products is restricted according to EU Directive 2002/95/EC (11). Accordingly, and given the practical and analytical difficulties in discriminating BFRs (including those that are unregulated; 24), Br should be absent from non-electrical plastics that are not pigmented with copper phthalocyanine (14). That nearly 40 non-green (and mainly neutrally-colored) products or components thereof were Br-positive in the present study, and usually at levels well below those required for flame retardancy (between about 3 and 8% by weight; 24), suggests many children's toys have been manufactured, directly or indirectly, from recycled waste electrical casings. This issue has been highlighted more generally in new consumer products, including toys (25,26,27), but the presence of Br in older toys raises the possibility that residues of the more hazardous BFRs that were banned 15 years ago and are subject to more stringent regulation, like polybrominated diphenyl ethers and polybrominated biphenyls, remain in plastics that are available to young children. Because of the relatively small molecular weight of most BFRs, they also have a greater propensity to migrate compared with many other organic additives (28). This is evident from the extraction of considerable quantities of Br from an item of jewelry presented in Table 3.

With the introduction and refinement of the Toy Safety Directive, the design and development of safer and more sustainable products, and the publicity generated by new goods violating chemical standards, there is clearly good reason for the plastic manufacturing industry to eliminate hazardous elements from new toys. The latest statistics provided by the US Consumer Product Safety Commission (29) confirm a steady decline in the number of recalls based on the presence of hazardous substances, and in particular Pb. However, the attraction of second-hand products to consumers in terms of cost, convenience and recyclability is acting as a conduit for exposing 'legacy' chemicals to the current generation of young children. For consumer products more generally, some authorities have advocated a 'right to know' policy, whereby goods are labelled should they contain any toxic constituents (3, 30). Because of the plethora and variety of old toys on sale or passed down whose precise ages and origins are unknown, this approach would be difficult to implement in the current context. However, a specific recommendation of this investigation is that consumers should be aware of the potential risks associated with small, mouthable, and brightly colored (and in particular red and yellow) old plastic toys or components. The present study has also provided evidence for the occurrence of historical BFRs in some second-hand toys that are neutrally colored; this finding is part of a broader and more complex issue concerning the recycling of electronic plastic waste and one that warrants further study (14,31).

Declaration

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Associated content

Supporting information available

A brief description of each toy and component, XRF results with counting errors, and cases of potential non-compliance are provided in Table S1.

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Figure 1: Total concentrations of the hazardous elements detected in the toys and components thereof by XRF. Concentrations are shown in ascending order and open bars denote samples composed of PVC. Dashed lines represent the current or proposed (Pb) EC Toy Safety Directive migration limits, and shown inset are summary statistics for each element (n_e = number of samples exceeding the corresponding migration limit).