Lead in plastics Recycling of legacy material and appropriateness of current regulations

Turner, Andrew

http://hdl.handle.net/10026.1/16664

10.1016/j.jhazmat.2020.124131
Journal of Hazardous Materials
Elsevier

Lead in plastics – recycling of legacy material and appropriateness of current regulations

Andrew Turner
School of Geography, Earth and Environmental Sciences, Plymouth University, Drake Circus, Plymouth PL4 8AA, UK

Montserrat Filella*
Department F.-A. Forel, University of Geneva, Boulevard Carl-Vogt 66, CH-1205 Geneva, Switzerland; email: montserrat.filella@unige.ch

https://doi.org/10.1016/j.jhazmat.2020.124131

Accepted: 26 September 2020

Abstract

X-ray fluorescence spectrometry has been employed to measure Pb in a wide range of consumer and environmental plastics, including food-packaging material, household goods, electronic casings, beach litter and agricultural waste. Results reveal high concentrations of Pb (> 1000 mg kg^{-1}) in historical items that are still in use or circulation (e.g. toys, construction plastics, wiring insulation) and variable, but generally lower concentrations in more recently manufactured articles. Analysis of Br, Cl and Cr, proxies for brominated flame retardants, polyvinyl chloride (PVC) and chromate pigments, respectively, suggests that as historical material is recycled, Pb from electronic plastics and pigments, but not PVC, is dispersed into a variety of newer products. Although most cases in the consumer sector comply with relevant EU Directives, some products that are non-compliant highlight shortfalls in regulations where recycling is involved and potential problems arising from the direct fashioning of industrial plastics into new consumer goods through attempts to be environmentally positive. The uncontrolled loss of historical and recycled plastics has also resulted in Pb contamination of the environment. Here, it is proposed that litter can be classified as hazardous depending on its Pb content and according to existing regulations that embrace consumer plastics.

Keywords: XRF; historical plastic; consumer goods; recycling; contamination; environmental litter; EU directives

1. Introduction

Current consensus in the scientific community is that there is no safe level of exposure to lead (Pb), and in particular for young children (Lanphear, 2017; Spungen, 2019). Thus, cumulative childhood exposure can result in damage to the brain and nervous system, slowed growth, anaemia, hearing loss, and behavioural and learning problems, while in adults exposure can increase blood pressure and incidence of hypertension, decrease kidney function and reduce fertility (Agency for Toxic Substances and Disease Registry, 2007).

The widespread use of leaded gasoline caused the dispersion of large quantities of airborne Pb throughout the environment in the 20th century, resulting in serious exposure for both humans and ecosystems (Caprino and Togna, 1998; Kristensen, 2015). Despite the phasing out of Pb as an antiknock agent, dusts and soils with high concentrations of legacy automotive Pb are still present in cities and close to major roads (Mielke et al., 2010; Filella and Bonet, 2017). Other legacy sources of Pb in the environment and/or in the household include old paints that are deteriorating or disturbed, leaded plumbing and industrial and mining waste (Clark et al., 2004; Howard et al., 2015; Shu et al., 2015; Ruckart et al.,, 2019).

An additional source of legacy Pb that has received less attention is historical plastic or plastic that has been recycled from historical plastic. Lead was commonly used in a range of plastics as a series of chromate pigments and in polyvinyl chloride as a heat and UV stabiliser (Hansen et al., 2013). However, strict regulations on Pb concentrations in electrical plastics (Restriction of Hazardous Substances – RoHS – Directive; Commission Delegated Directive, 2015), toys (Toy Safety Directive 2009/48/EC; European Parliament and Council of the EU, 2009), packaging (Packaging and Packaging Waste Directive 94/62/EC; European Parliament and Council of the EU, 1994) and food contact material (EC Directive 2002/72/EC; Commission Directive, 2002), coupled with the voluntary phasing out of Pb by the PVC industry (VinylPlus, 2014), have effectively eliminated the intentional introduction of the metal into new products on the European market. In the US, the Consumer Products Safety Improvement Act now limits the amount of Pb in products intended for children under 12 years, including plastics (Consumer Product Safety Commission, 2008), while the Institute of Electrical and Electronics Engineers Standard 1680 regarding personal computer products adopts the RoHS and packaging and Packaging Waste directives and refers to an optional limit of intentionally added Pb in plastic computer components (IEEE, 2006). Despite these regulations, evidence for the dispersion of Pb at lower levels in contemporary consumer plastics that result from the legal and illegal recycling of historical plastics has recently emerged (Turner and Filella 2017; Turner, 2018).

In this study, we use a rapid, non-destructive X-ray fluorescence (XRF) technique to determine the concentrations of Pb and various other elements serving as proxies for the origin of Pb in both contemporary and historical consumer plastics and in material lost to the environment. The results provide a valuable insight into the extent of Pb contamination in plastics in circulation and that pervade in the environment, and allow us to assess whether current regulations are being met or, in many circumstances, are entirely appropriate, and in particular where material is recycled.

2. Materials and methods

2.1. Materials

About 1500 samples were considered here that had been analysed as part of previous research programmes (Turner and Solman, 2016; Turner and Filella, 2017; Filella and Turner, 2018) or had been acquired specifically for the present study. Samples constitute hard plastics (i.e. excluding rubbers and foams) and, while textiles have not been included, we consider constructions of coarser and longer fibres like rope and twine. Table 1 categorises and quantifies the plastics according to use or source and provides general examples for each category. Thus, agriculture and beached refer to plastics lost in nature through agricultural and aquatic-maritime activities and from littering and municipal (and industrial) waste and embraces primary objects and secondary fragments (including microplastics of < 5 mm in diameter). Agricultural samples were collected from the edges of fields in Luxembourg and Spain during spring and summer of 2018 and beached samples were retrieved from the strandlines of sandy shores of southwest England in mid-2015 and the gravel shores of Lake Geneva in March 2016.

Single-use food defines plastics used for the packaging of food and the containment or takeaway of fast food and drinks that had been acquired since 2016. Consumer goods refers to products commonly encountered in the household, office or workplace and includes items purchased in the UK within the last five years and in Switzerland in 2018 as well as older articles (up to 45 years) that are in common circulation or use because of their durability or their function (e.g. structural and plumbing). Electronic plastics are casings and housings of electronic and electrical equipment and insulation for wiring (excluding rubbers). Note that in Table 1, and based on signage or estimated age, consumer and electronic plastics are subdivided (by number) according to whether articles had been purchased, manufactured or installed before or since the original RoHS Directive (2002/95/EC; European Parliament and Council, 2003) came into effect in July 2006.

Table 1: Quantities (*n*) and categories of plastic considered in the present study, along with general examples and the number of PVC-based samples (*n*-PVC) in each category. Shown in parentheses are the numbers of samples estimated or known to be manufactured pre-RoHS and post-RoHS.

Category	*n*	*n*- PVC	Examples
Agriculture	55	2	film, gauze-mesh, packaging, potting, tree protection, twine, tarpaulin
Beached (lacustrine)	584	32	primary and secondary plastics
Beached (marine)	217	2	primary and secondary plastics and microplastics (< 5 mm)
Consumer goods	353 (193/160)	58 (31/27)	toys, storage, stationery, apparel, sports gear, plumbing, construction, tools, decor
Electronic	193 (115/78)	18 (16/2)	phones, chargers, wiring, laptops, white goods, appliances, sockets-switches, remotes
Single-use food	95	2	packaging, trays, cutlery, cups, bottles, lids, stirrers
Total	1497	114	

2.2. XRF analysis

Samples were analysed by energy-dispersive FP-XRF using a Niton XL3t 950 He GOLDD+ operated in a standardless 'plastics' mode (Turner and Solman, 2016). The majority of samples were analysed in the laboratory in an accessory stand and by remote activation of the instrument, with a thickness correction algorithm applied between 50 μm and 12 mm. Thickness was determined through the measurement surface using Allendale digital callipers or, where inaccessible, was estimated from the thickness of samples of similar construction. Samples were analysed for a suite of elements, of which the present focus was on Pb, Br (a proxy for brominated flame retardants), Cl (a proxy for PVC above a concentration of 15%; Turner and Filella, 2020) and Cr (whose association with Pb may indicate the presence of lead chromate pigments). Counting was undertaken for periods ranging from 30 to 180 s, depending on sample thickness, that were distributed equally or in a 1:2 ratio between a low energy range (20 kV and 100 μA) and main energy range (50 kV and 40 μA). Spectra were quantified by fundamental parameter coefficients to yield concentrations on a dry weight basis (in mg kg^{-1}) and with a counting error of 2σ (95% confidence). For samples too large to be contained by the accessory stand or that were permanent fixtures in the household setting the instrument was used handheld and with a backscatter shield under the conditions described above.

As a performance check, polyethylene reference discs Niton PN 180-619 (Cr = 101 $\pm$ 10 mg kg^{-1}; Pb = 150 $\pm$ 12 mg kg^{-1}) and Niton PN 180-554 (Br = 495 + 20 mg kg^{-1}; Cr = 995 $\pm$ 40 mg kg^{-1}; Pb = 1002 + 40 mg kg^{-1}) were analysed throughout each measurement session, with the instrument returning concentrations that were consistently within 15% of certified values. Detection limits varied depending on counting time, sample size and thickness and whether the instrument was deployed in a stand or activated handheld but indicative values based on the lowest counting errors returned throughout the study were about 6 mg kg^{-1} for Br and Pb and 12 mg kg^{-1} for Cl and Cr. Precision, defined as the relative standard deviation arising from quintuplicate measurements of selected samples, was better than 10% in most cases but approached 20% for small or thin samples or where concentrations were close to detection limits.

3. Results

The number of cases in which Pb was detected and summary statistics for concentrations of the metal are shown for each plastic sample category in Table 2. Note that the data for the beached samples differ slightly to those published previously because here we have focused on hard plastics and have neglected foams, paints and rubbers (Turner and Solman, 2016; Filella and Turner, 2018). Detection occurred across all categories and was most frequent (on a percentage basis) among beached samples and electronic plastics and was lowest in the single-use food category. Overall, Pb concentrations were variable, spanning four orders of magnitude and ranging from < 10 mg kg^{-1} to about 3.4% by weight, and >20% of Pb-positive samples in each category with the exception of single-use food exceed the RoHS limit for Pb of 1000 mg kg^{-1}.

Samples of PVC, defined as returning a Cl content greater than 15% by the XRF, were encountered in all categories but were most abundant (on a percentage basis) among consumer goods and least abundant in beached marine plastics (Table 1). Associations of Pb with PVC were most frequent in consumer goods and electronic plastics while associations with Br and

Cr were most frequent in beached litter; in contrast, no associations of Pb with PVC or Cr and just one association with Br were observed in the single-use food category (Table 2).

Table 3 shows the number of cases in which Pb was detected and summary statistics for concentrations of the metal for electronic and consumer plastics categorised according to whether they were estimated or known (from signage) to have been manufactured, sold or installed pre-RoHS or post-RoHS. Thus, about a third of electronic articles manufactured before the directive came into effect (in 2006) contained detectable Pb, with an exceedance of the RoHS limit of 1000 mg kg^{-1} in 19 cases and an association with PVC in 14 samples. In contrast, only four post-RoHS electronic samples contained detectable Pb with no exceedance of the RoHS limit or association with PVC. Despite these differences, however, a Mann-Whitney U test undertaken in Minitab *v*19 revealed no significant difference (p = 0.119) in median concentrations between the two groups. With respect to consumer plastics, Pb detection rate was similar among products manufactured pre-RoHS and post-RoHS, and although mean, median and maximum concentrations were greater in pre-RoHS consumer articles than post-RoHS items, a Mann-Whitney U test indicated no significant difference (p = 0.131) in median concentrations.

Table 2: Number and percentage of samples in which Pb was detected (n (%)) and summary statistics defining Pb concentrations (in mg kg^{-1}) in each category. Also shown are the number of samples that exceed the RoHS limit of 1000 mg kg^{-1} (n > RoHS), and the number of cases where Pb was detected in PVC (n-PVC) and with Br (n-Br) or Cr (n-Cr).

	Agriculture	Beached (lacustrine)	Beached (marine)	Consumer goods	Electronic	Single-use food
n (%)	6 (10.9)	134 (22.9)	47 (21.7)	42 (11.9)	41 (21.2)	4 (4.2)
mean	4500	2150	765	3300	5570	114
sd	8060	4010	2010	5560	8080	152
median	390	433	142	573	512	43.2
min	62.1	5.9	6.3	3.9	17.3	26.9
max	20400	23500	13200	21700	34100	342
Q1	302	41.0	30.8	137	104	32.8
Q3	4230	2210	704	3720	10000	124
n > RoHS	2	55	9	18	19	0
n -PVC	0	16	0	14	14	0
n -Br	0	43	20	11	19	1
n -Cr	5	88	32	12	4	0

Table 3: Number and percentage of pre-RoHS and post-RoHS electrical plastics and consumer goods in which Pb was detected (*n* (%)) and summary statistics defining Pb concentrations (in mg kg^{-1}) in each category. Also shown are the number of samples that exceed the RoHS limit of 1000 mg kg^{-1} (*n* > RoHS), and the number of cases where Pb was detected in PVC (*n*-PVC) and with Br (*n*-Br) or Cr (*n*-Cr).

	Pre-RoHS	Post-RoHS	Pre-RoHS	Post-RoHS
	Electronic		Consumer	
n (%)	37 (32.2)	4 (5.1)	23 (11.9)	19 (11.9)
mean	6160	149	5070	1160
sd	8300	91.0	6870	1970
median	1070	149	1190	302
min	17.3	48	3.87	9.64
max	34100	251	21700	7240
Q1	158	89.7	282	68.0
Q3	10500	209	9000	1040
n > RoHS	19	0	13	5
n -PVC	14	0	10	4
n -Br	16	3	5	6
n -Cr	4	0	6	6

4. Discussion

4.1. Legacy Pb in plastics

Evidently, Pb is widely and heterogeneously distributed in plastics that are in circulation and production as well as lost in nature. The presence of Pb in older plastics is expected because the metal chromate was used in a number of coloured pigments in a range of plastics and various leaded compounds acted as heat and UV stabilisers in PVC until they were restricted or phased out according to a series of international regulations and agreements (Hansen et al., 2013). Thus, in many older consumer and electronic plastics, high concentrations of Pb were encountered in unplasticised PVC (e.g. window and door frames) and plasticised PVC (e.g. electrical wire insulation, garden hosing and inflatable toys) and in a range of brightly coloured articles. These traits also characterise many (presumably older) articles and fragments retrieved from the environment, and in particular from beaches, where decadal-old plastics derived from the municipal waste stream, littering or loss (e.g. at sea) may be washed up or exposed (Watts et al., 2017; Turner et al., 2020).

4.2. Legacy use and recycling of lead chromate

The presence and pervasiveness of lead chromate is evident from the association of Pb and Cr in consumer goods and environmental plastics shown in Figure 1. Thus, the highest concentrations ([Pb] > 1000 mg kg^{-1}) generally arise from older samples coloured with chrome yellow ($PbCrO_4$), chrome green ($PbCrO_4$ mixed with Fe-based Prussian blue) and chrome orange-red ($PbCrO_4 \cdot PbO$) (Oldring, 2001), with the majority of data close to the line defining the mass ratio of [Pb] to [Cr] in pure $PbCrO_4$ (~ 4). Samples lying close to the line but having Pb and Cr concentrations too low to act as a colourant (e.g. [Pb] ~ 100 to 1000 mg kg^{-1}) were encountered in each category shown and for both contemporary and historical articles. Here, presumably, chromate-based pigments are widely encountered as contaminants of the mechanical recycling of coloured plastics. For Pb concentrations below about 100 mg kg^{-1}, data points in Figure 1 are more heterogeneously dispersed and generally lie well below the slope defining the composition of $PbCrO_4$. This may be attributed to the more general Pb contamination of recycled plastic (including electronic-based waste; see below) and the use of additional Cr pigments that are free of Pb.

The general observations above are consistent with the phasing out of Pb chromate pigments in Europe and, effectively, since an EU court overruling authorisation for production and export by a Canadian company in March 2019, an outright ban (EVISA, 2019). However, XRF results returned for some samples analysed suggest that these pigments are still circulating as colourants in a limited number contemporary products. Specifically, Pb above a concentration of 1000 mg kg^{-1} was found in association with Cr above a concentration of a few hundred mg kg^{-1} in a green clothes peg, red and yellow "environmentally sustainable" shoulder bags that had been fashioned from PVC truck tarp, and pieces of yellow and green agricultural packaging that appeared to have been discarded recently.

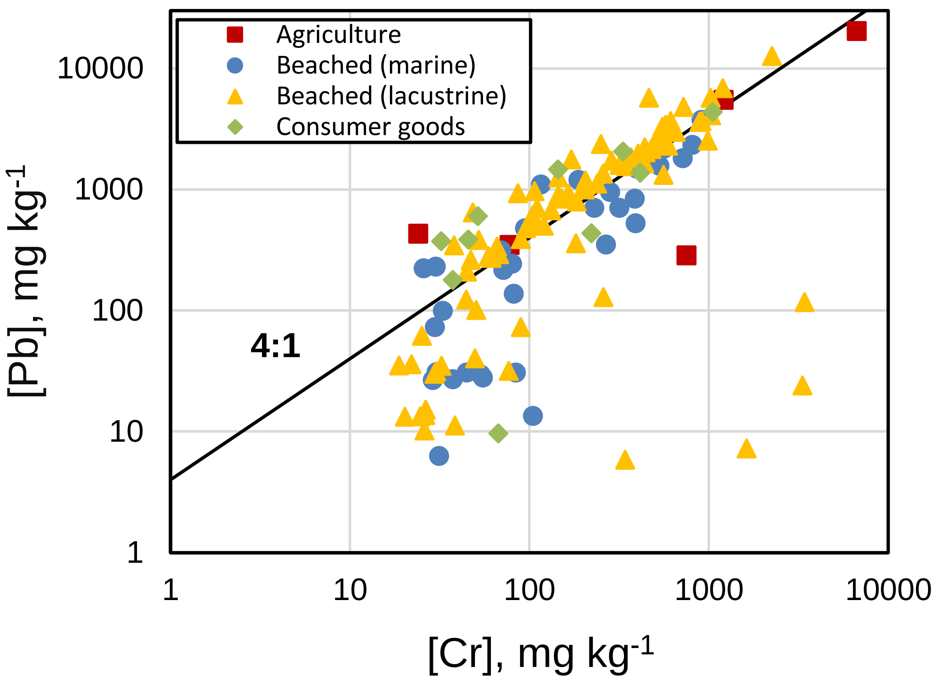

Figure 1: Concentration of Pb versus concentration of Cr in consumer goods and plastics lost to the environment. Note that samples of PVC employing Pb-based stabilisers are not included. The line of slope 4:1 defines the mass ratio of Pb to Cr in pure lead chromate.

4.3. Lead contamination from electrical and electronic plastic recycling

Although Br is a constituent of the halogenated copper phthalocyanine pigments (Ranta-Korpi et al., 2014), the principal use of brominated compounds in plastics is as flame retardants in electronic components, casings and insulation (Papazoglou, 2004). Thus, an association of Pb with Br provides an upper estimate of the number of samples in each category where the metal is derived from the poorly managed and often illegal use or recycling of contaminated electronic and electrical waste (Turner, 2018). The highest percentage of Br-Pb associations among Pb-positive samples occurs in the electronic category (Table 2) and associations are observed in both pre- and post-RoHS articles (Table 3), presumably reflecting the use of recycled electronic and electrical plastic in what is intended to be a regulated, circular economy. Significantly, there were no associations of Br-Pb-Cr in this category, suggesting that Pb in electronic and electrical plastic is contaminated by additional sources other than lead chromate pigments (e.g. PVC and soldering residues). Associations of Br-Pb were also observed in several consumer products, a single-use food item (cocktail stirrer) and various plastic items and fragments retrieved from coastal and lacustrine beaches. Moreover, in these categories Cr was also detected in the presence of both Br and Pb in many cases. This suggests that the recycling of electronic waste is not constrained to the electrical and electronic industries but that some material has been (and continues to be) exported for use in a broader array of plastic products that may or may not be contaminated by residues of lead chromate pigments.

4.4. Lead in PVC

The frequency distributions of Pb concentrations amongst the samples of PVC in each category are shown in Table 4. Overall and within each category Pb concentrations display a distinctly bimodal distribution; that is, out of 114 PVC-based samples Pb concentrations are focussed above 1000 mg kg^{-1} (n = 39) and below the detection limit (n = 69). PVC samples containing

[Pb] > 1000 mg kg^{-1} were dominated by older consumer products, plastics associated with pre-RoHS electrical and electronic items (and occasionally containing traces of Br) and articles and fragments of beached lacustrine litter, while Pb-free PVC samples comprised newer consumer goods, post-WEEE electrical plastics, single-use food articles and various plastics lost to the environment (Tables 2 and 3). Presumably, this observation reflects the historical use of Pb-based heat and UV stabilisers in PVC in various sectors and the gradual and voluntary phasing out and replacement of Pb in the more recent manufacture of PVC (VinylPlus, 2014). Unlike the case for Pb chromate, however, there is no evidence for the widespread contamination of newer PVC products by Pb-based stabilisers. (The only notable exception is the pair of PVC shoulder bags described above, but here Pb appears to be related to the more general use of lead chromate in colour pigments rather than the presence of Pb-based stabilisers.) These observations suggest one or more of the following: the mechanical recycling of PVC has been more targeted at and successful in eliminating older Pb-based materials; Pb-based PVC is recycled for more specific, industrial or professional purposes; the recycling of PVC in general has been reduced in order to avoid product contamination.

Table 4: Frequency distribution of Pb concentrations (mg kg^{-1}) in samples of PVC from each category. < LOD = below the detection limit.

Category	< LOD	< 100	100-1000	1000-10,000	>10,000
Agriculture	2				
Beached (lacustrine)	15	1	1	10	4
Beached (marine)	2				
Consumer goods	44		4	6	4
Electronic	4			4	11
Single-use food	2				

4.5. Compliance with and appropriateness of current regulations

There are several cases in the present study that highlight regulations which target certain products but neglect the life cycle of the material. For example, the current RoHS limit for Pb in any component of electrical and electronic equipment is 1000 mg kg^{-1} (Commission Delegated Directive, 2015), and while this is only exceeded among electrical plastics which pre-date the 2006 implementation of the original Directive (European Parliament and Council, 2003), it is exceeded in several newly purchased consumer plastics, some of which are likely to have been manufactured from recycled (and pre-RoHS) electronic plastic. That is, a directive that is specific to electrical plastic does not apply to products that are repurposed from regulated material.

Adding to this complexity, and although not electrical in origin, PVC truck tarp that appears to be free of leaded stabilisers but that is coloured by Pb chromate pigments would be non-compliant according to the RoHS. However, this material has been fashioned directly (without mechanical recycling) into shoulder bags produced in Switzerland that are currently on sale in the EU. Subsequent acquisition and XRF analyses of a wider range of bags (n = 9) revealed the more general presence of Pb chromate pigments in such products. This is an example of what is designed to be an environmentally positive process that transfers a hazardous plastic from the industrial (transportation) sector to consumer products and one that evades the various regulations on Pb that are currently in place.

The dispersion of Pb into plastics more widely through recycling can also result in the non-compliance or potential non-compliance of specific types of consumer plastic. For example, a recent amendment to the latest iteration of the Toy Safety Directive stipulates a migration limit (in dilute HCl) of Pb from material that can be scraped off, including plastic, of 23 mg kg^{-1} (The Council of the European Union, 2017). This means that, in theory, any toy contaminated with Pb above the concentration limit (and ascertained by XRF) could be subject to migration testing. Article II of the Packaging and Packaging Waste Directive (European Parliament and Council of the EU, 1994) states that the sum of concentrations of Pb, Cd, Hg and Cr(VI) present in packaging or packaging components shall not exceed 100 mg kg^{-1}. Since the directive also includes industrial packaging, it would appear that at least two fragments of agricultural wrapping waste greatly exceed the limit value with respect to Pb alone or with respect to Pb combined with Cr(VI). Directive 2002/72/EC relating to plastics intended to come into contact with foodstuffs (Commission Directive, 2002) stipulates an upper limit of 2 mg kg^{-1} of Pb in the raw material prior to granulation. On this basis, therefore, four single-use food contact items reported here (three drinks stirrers and a coffee cup lid; Pb = 27 to 342 mg kg^{-1}) are non-compliant.

One of the key objectives of many of the directives above was to limit noxious metals in plastics because of their environmental impacts, and in particular, to reduce their presence in emissions, ash or leachate arising from controlled disposal. Specific regulations are, however, neither feasible nor appropriate for metal-rich plastics that have accumulated in the environment from a multitude of historical and, likely, transboundary sources. That said, existing regulations or limit values could be used as a framework to define whether plastic litter, including microplastics, is chemically hazardous or not and whether it poses a risk to wildlife or the environment.

5. Conclusions

This study has revealed the wide distribution of Pb in plastics that are in circulation, in production and lost in nature. Observations are attributed to the historical use of the metal as a pigment and additive in plastics (including PVC) and the contamination of contemporary products through mechanical material recycling. Consequently, some currently manufactured products are non-compliant with respect to various directives aimed at protecting human health and the environment. Although plastics lost in nature are not embraced by any specific regulation, limit values could be used as an aid to assess potential impacts in the environment.

Funding: This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

References

Agency for Toxic Substances and Disease Registry, 2007. Toxicological Profile for Lead. US Department of Health & Human Services, Atlanta, GA.

Caprino, L., Togna, G.I., 1998. Potential health effects of gasoline and its constituents: A review of current literature (1990-1997) on toxicological data. Environmental Health Perspectives 106, 115-125.

Clark, S., Grote, J., Wilson, J., Succop, P., Chen, M., Galke, W., McLaine, P., 2004. Occurrence and determinants of increases in blood lead levels in children shortly after lead hazard control activities. Environmental Research 96, 196-205.

Commission Delegated Directive, 2015. EU Directive 2015/863 amending Annex II to Directive 2011/65/EU of the European Parliament and of the Council as regards the list of restricted substances. Official Journal of the European Union L137/10.

Commission Directive, 2002. EC Directive 2002/72/EC of 6 August 2002 relating to plastic materials and articles intended to come into contact with foodstuffs. Official Journal of the European Communities L220/18.

Consumer Product Safety Commission, 2008. The Consumer Product Safety Improvement Act (CPSIA) https://www.cpsc.gov/Regulations-Laws--Standards/Statutes/The-Consumer-Product-Safety-Improvement-Act

European Parliament and Council, 2003. Directive 2002/95/EC on the restriction of the use of certain hazardous substances in electrical and electronic equipment. Official Journal of the European Union L37/19.

European Parliament and Council of the EU, 1994. Directive 94/62/EC of 20 December 1994 on packaging and packaging waste. Official Journal of the European Union L365.

European Parliament and Council of the EU, 2009. Directive 2009/48/EC of the European Parliament and of the Council of 18 June 2009 on the safety of toys. Official Journal of the European Union L170/1.

EVISA, 2019. EU court rules commission authorisation of lead chromate pigments was illegal. http://www.speciation.net/News/EU-court-rules-Commission-authorisation-of-lead-chromate-pigments-was-illegal-;~/2019/03/12/9006.html, accessed 6/20.

Filella, M., Bonet, J., 2017. Environmental impact of alkyl lead(IV) derivatives: perspective after their phase-out. Metal Ions in Life Sciences 17, 471–490.

Filella, M., Turner, A., 2018. Observational study unveils the extensive presence of hazardous elements in beached plastics from Lake Geneva. Frontiers in Environmental Science https://doi.org/10.3389/fenvs.2018.00001

Hansen, E., Nilsson, N.H., Lithner, D., Lassen, C., 2013. Hazardous Substances in Plastic Materials. Oslo: COWI and the Danish Technological Institute on behalf of The Norwegian Climate and Pollution Agency, 150 pp.

Howard, A.J., Kincey, M., Carey, C., 2015. Preserving the Legacy of Historic Metal-Mining Industries in Light of the Water Framework Directive and Future Environmental Change in Mainland Britain: Challenges for the Heritage Community. Historic Environment – Policy & Practice 6, 3-15.

IEEE, 2006. Standard for environmental assessment of personal computer products, including laptop personal computers, desktop personal computers, and personal computer monitors. The Institute of Electrical and Electronics Engineers Inc, New York, 32pp.

Kristensen, L.J., 2015. Quantification of atmospheric lead emissions from 70 years of leaded petrol consumption in Australia. Atmospheric Environment 111, 195-201.

Lanphear, B.P., 2017. Low-level toxicity of chemicals: No acceptable levels? PLOS Biology 15, e2003066.

Mielke, H.W., Laidlaw, M.A.S., Gonzales, C., 2010. Lead (Pb) legacy from vehicle traffic in eight California urbanized areas: Continuing influence of lead dust on children's health. Science of the Total Environment 408, 3965-3975.

Oldring, P.K.T., 2001. Coatings, colorants, and paints. In: Encyclopedia of Physical Science and Technology, 3rd Edition, ed. R.A. Meyers. Elsevier, London, pp.175-190.

Papazoglou, E.S., 2004. Flame retardants for plastics, in: C.A. Harper (ed.), Handbook of Building Materials for Fire Protection, McGraw-Hill, New York, pp4.1-4.88.

Ranta-Korpi, M., Konttinen, J., Saarimaa, A., Rodriguez, M., 2014. Ash forming elements in plastics and rubbers. VTT Technical Research Centre of Finland, Espoo, Finland, 131pp.

Ruckart, P.Z., Ettinger, A.S., Hanna-Attisha, M., Jones, N., Davis, S.I., Breysse, P.N., 2019. The Flint water crisis: A coordinated public health emergency response and recovery initiative. Journal of Public Health Management and Practice, 25(1 Supp), S84–S90.

Shu, Z., Axe, L., Jahan, K., Ramanujachary, K.V., 2015. Metal leaching from the bridge paint waste in the presence of steel grit. Chemosphere 119, 1105-1112.

Spungen, J.H., 2019. Children's exposures to lead and cadmium: FDA total diet study 2014-16. Food Additives and Contaminants Part A: Chemistry Analysis Control Exposure & Risk Assessment 36, 893-903.

The Council of the European Union, 2017. Council Directive (EU) 2017/738 of 27 March 2017 amending, for the purpose of adapting to technical progress, Annex II to Directive 2009/48/EC of the European Parliament and of the Council on the safety of toys, as regards lead. Official Journal of the European Union L110/6.

Turner, A., 2018. Black plastics: linear and circular economies, hazardous additives and marine pollution. Environment International 117, 308-318.

Turner, A., Filella, M., 2017. Bromine in plastic consumer products – Evidence for the widespread recycling of electronic waste. Science of the Total Environment 601-602, 374-379.

Turner, A., Filella, M., 2020. The influence of additives on the fate of plastics in the marine environment, exemplified with barium sulphate. Marine Pollution Bulletin 158, 111352.

Turner, A., Solman, K.R., 2016. Analysis of the elemental composition of marine litter by field-portable-XRF. Talanta 159, 262-271.

Turner, A., Arnold, R., Williams, T., 2020. Weathering and persistence of plastic in the marine environment: Lessons from LEGO. Environmental Pollution 262, 114299.

VinylPlus, 2014. The European PVC industry’s experience in replacing lead and cadmium-based stabilisers https://www.stabilisers.eu/wp-content/uploads/2015/11/VinylPlus_Contribution-Cefic_Eu-Industry.pdf. Accessed 6/20.

Watts, A.J.R., Porter, A., Hembrow, N., Sharpe, J., Galloway, T.S., Lewis, C., 2017. Through the sands of time: Beach litter trends from nine cleaned north Cornish beaches. Environmental Pollution 228, 416-424.