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Chemical characteristics of artificial plastic plants and the presence of hazardous elements from the recycling of electrical and electronic waste

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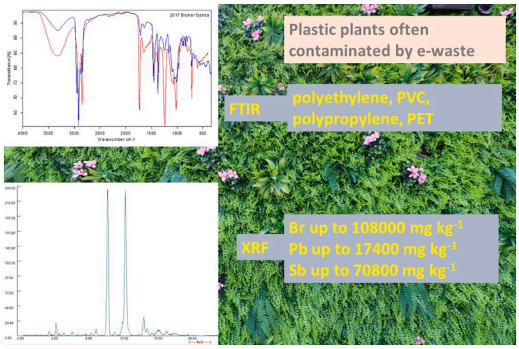
HIGHLIGHTS

- Elemental and polymeric composition of 175 artificial plastic plants determined.
- Most plants consist of moulded structure (mainly polyethylene-polypropylene).
- Many plants have fabric appendages (e.g., leaves) constructed of polyethylene terephthalate.
- Moulded components often contain traces or high levels of Br, Pb and Sb.
- Elemental signatures suggest material is often derived from recycled electronic waste.

GRAPHICAL ABSTRACT



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ABSTRACT

Because of their convenience, the demand for decorative plastic plants has been increasing over recent years. However, no information exists on the origin or nature of the polymers employed or the type of additives used in order to understand potential environmental impacts and inform safe and sustainable disposal or recycling practices. In this study, 203 parts or offcuts from 175 plastic plants acquired from European shops and venues have been analysed by X-ray fluorescence spectrometry to determine elemental content, while a selection has been analysed by infrared spectrometry to establish polymer type. The (usually green) moulded components ($n = 159$) were commonly constructed of polyethylene or polypropylene, while leaves and colourful petals ($n = 40$) were generally made of polyethylene terephthalate fabric that had been glued to the moulded component. However, both components also exhibited evidence of being coated with a resin or adhesive for support, protection or appearance. Barium, Fe, Ti and Zn-based additives were commonly encountered but more important from an environmental and health perspective were variable concentrations of potentially hazardous elements in the moulded parts: namely, Br (6.1 to 108,000 mg kg^{-1}; $n = 78$), Pb (7.6 to 17,400 mg kg^{-1}; $n = 53$) and Sb (58.6 to 70,800 mg kg^{-1}; $n = 17$). These observations suggest that many of the moulded components are derived from recyclates that are contaminated by waste electronic and electrical plastic, introducing brominated flame retardants, the flame retardant synergist, Sb_2O_3, and Pb into the final product. There are no standards for these chemicals in plastic plants, but regulations for electronic plastic, toy safety and packaging are frequently

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exceeded or potentially exceeded. Widespread contamination of plastic plants may impose constraints on their recycling and disposal.

1. Introduction

Artificial plants (or faux plants), including trees, foliage and flowering plants, wall panels, wreaths and stem flowers, are an increasingly popular form of decorative or functional product used in the domestic setting and in public places, offices, hotels, airports, theme parks and hospitality venues. Demand for artificial plants has surged rapidly over the past few years, with the market predicted to grow at a rate of over 4 % per year until 2030 when its value is estimated to be almost $1bn (Market Research Future, 2023). This growth may be largely attributed to the benefits and convenience of artificial plants. Thus, they are cheaper and more durable than natural plants, need little maintenance, have no light or temperature requirements, are not subject to seasonal change and do not act as a source of potential allergens. Artificial plants are not, however, able to clean indoor air, can lack authenticity and may fade over time when deployed outdoors. Moreover, while artificial plants may offer some physiological (relaxation) and psychological benefits as indoor visual stimuli, recent research suggests that they are not as significant as those engendered by real plants (Oh et al., 2019; Jeong and Park, 2021).

Although plastic is one of the most common materials used in artificial plants, very little information exists on the sources of the material or the polymers used. For example, a review of products sold by major online retailers usually refer to "plastic" or "foam", with occasional mention of the polymers "polyethylene", "PVC", "polystyrene", "polyester" or "rayon". Some retailers also make reference to specific properties that imply the incorporation of plastic additives, like "antibacterial" or "ultraviolet light protection". Metal- and organic-based additives in artificial turf have been the subject of recent research based on health concerns although the turf fibres themselves (i.e., the plastic component) appear to be of low risk (Pavilonis et al., 2014).

In order to improve our understanding of the sources, recyclability and potential environmental impacts of plastic plants, we have analysed a variety of samples obtained from retail outlets and public places for their polymeric and elemental compositions. More generally, information of this kind is becoming increasingly important as the United Nations seeks to end plastic pollution with an international legally binding agreement by 2024 (United Nations Environment Programme, 2022).

2. Methods

2.1. Sampling

A total of 175 plastic plants were sampled from different European countries (Belgium, Germany, Luxembourg, Portugal, Spain, Switzerland, UK). Whole plants were purchased from DIY stores, homeware shops, supermarkets, gift shops and second-hand stores, while detached parts were taken from a variety of internal and external public venues (shopping malls, public houses, libraries, railway stations, airports). Plants were labelled with a permanent marker pen and stored in a series of polyethylene boxes.

2.2. FTIR analysis

A selection of plant parts (e.g., stems, leaves, blades, petals, fibres, berries; $n = 128$) from 112 plants was analysed by attenuated total reflectance Fourier-transform-infrared (ATR-FTIR) spectrometry using a Bruker Vertex 70 or Perkin-Elmer Spectrum Two N FT-IR spectrometer. Thin samples (<0.5 mm), small, surficial offcuts of thicker samples, or inner slices of spherical berries were clamped against the diamond crystal before spectra were acquired with four to sixteen scans in the region 4000 to 600 cm^{-1} and at a resolution of 4 cm^{-1}. Sample spectra were compared with polymer spectra in various libraries, and a positive identification was defined with a hit rate of >65 %.

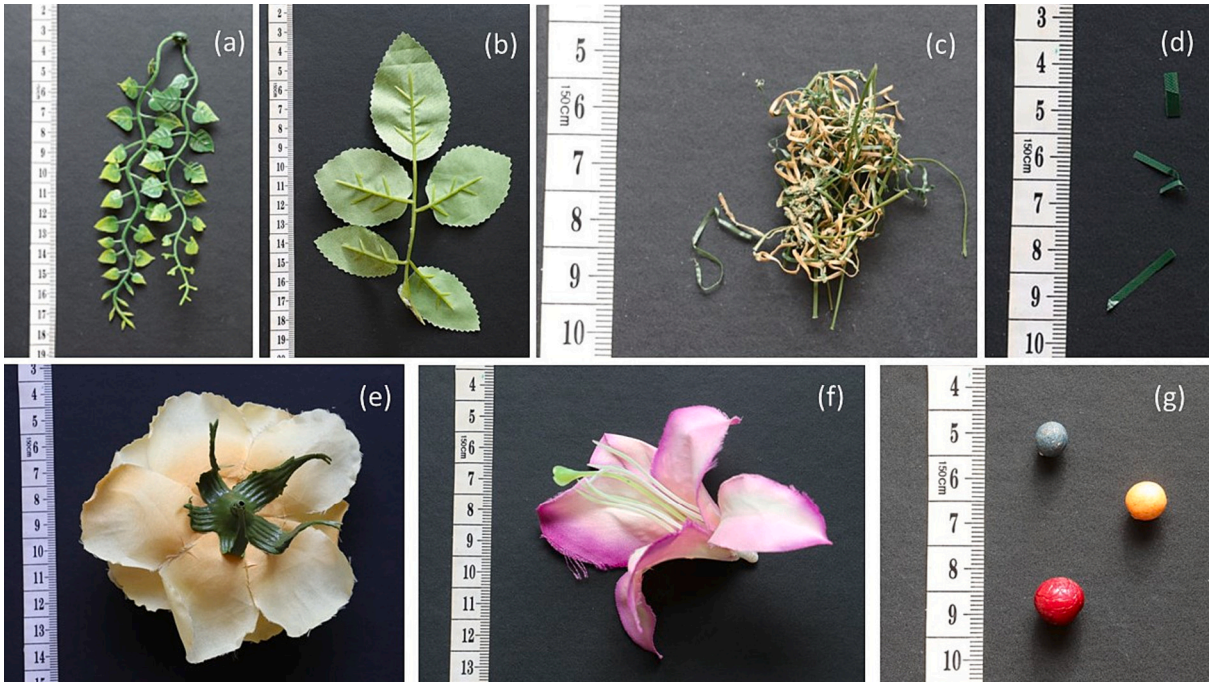

Fig. 1. A selection of plastic plant parts considered in the present study. (a) Part of a moulded, hanging plant; (b) fabric leaves glues to a moulded stem; (c) a clump of artificial turf; (d) fibres from various Christmas trees; (e) the yellow petals and moulded sepal of a rose; (f) purple fabric petals and moulded carpel and stamens of a lily; (g) coloured (painted), lightweight spheres from various floral arrangements.

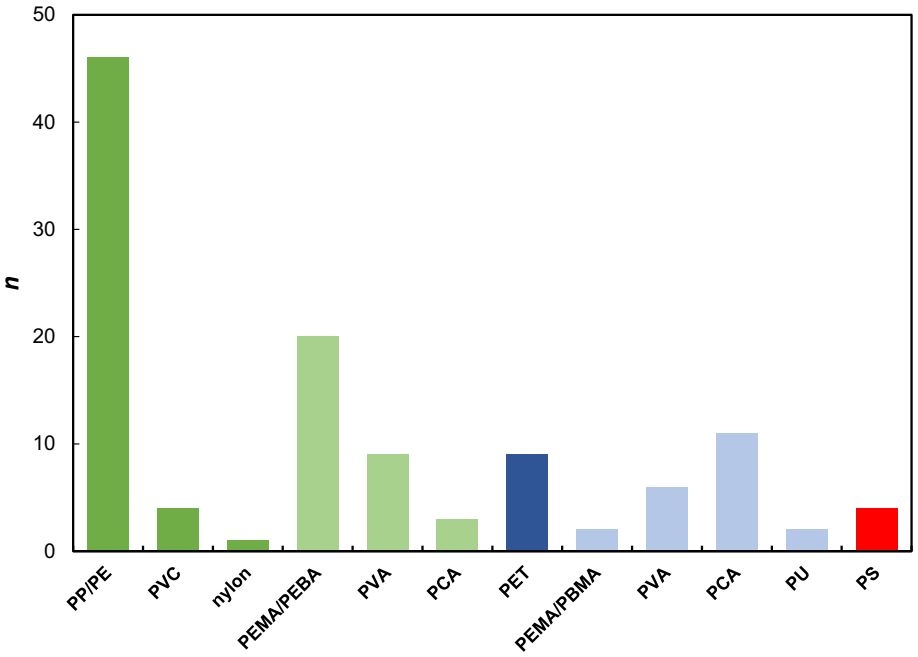

Fig. 2. Polymer frequency (*n*) among the 117 plastic sample components that returned a positive identification by FTIR (dark green = moulded components; light green = moulded component adhesives and coatings; dark blue = fabric leaf or flower components; light blue = fabric component adhesives and coatings; red = painted spheres). PP = polypropylene; PE = polyethylene, including chlorinated polyethylene; PVC = polyvinyl chloride; PEMA = poly (methyl methacrylate); PEBA = poly(butyl methacrylate); PVA = polyvinyl acetate; PCA = polycarprolactone; PET = polyethylene terephthalate; PU = polyurethane; PS = polystyrene.

2.3. XRF analysis

The elemental constituents ($Z > 17$, and $n = 18$) of all plant parts ($n = 203$) were determined by energy-dispersive X-ray fluorescence (XRF) spectrometry using a Niton XL3t 950 He GOLDD+ configured in a laboratory test stand. The instrument was operated in a standardless 'plastics' mode and with a thickness correction algorithm applied between 50 μm and 12 mm after sample thickness had been determined through the measurement area using Allendale digital callipers. The measured area was usually the central or thickest part of a component and was carefully selected to exclude any permanent pen marking. Counting was performed for 60 s in total (30 s each in low and main energy modes) and 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).

As a performance check, polyethylene reference discs Niton PN 180–619 (As = 51 ± 7 mg kg^{-1}; Ba = 688 ± 45 mg kg^{-1}; Cd = 292 ± 20 mg kg^{-1}; Cr = 101 ± 10 mg kg^{-1}; Hg = 101 ± 10 mg kg^{-1}; Pb = 150 ± 12 mg kg^{-1}; Sb = 96 ± 10 mg kg^{-1}; Se = 200 ± 15 mg kg^{-1}) and Niton PN 180–554 (Br = 495 ± 20 mg kg^{-1}; Cd = 150 ± 6 mg kg^{-1}; Cr = 995 ± 40 mg kg^{-1}; Hg = 1000 ± 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 sample size and thickness, but indicative values based on the lowest counting errors returned throughout the study were <10 mg kg^{-1} for all elements except Ba, Sb and Sn (about 50 mg kg^{-1}). Precision, defined as the relative standard deviation arising from quintuplicate measurements of selected samples, was better than 10 % in most cases but approached 25 % for the smallest or thinnest samples or where concentrations were close to detection limits.

3. Results and discussion

3.1. Visual characteristics

A total of 203 samples were obtained from 175 plants, with a selection shown in Fig. 1. The majority of samples consisted of a single,

Table 1
Number of detects and summary statistics for different elements in moulded components (including a wired stem, turf and Christmas tree fibres), fabrics and painted spheres of the plastic plants. Concentrations are in mg kg^{-1}.

	As	Ba	Br	Cr	Cu	Fe	Pb	Sb	Sn	Ti	Zn
Moulded (n = *159)*											
n	20	110	78	109	72	81	53	17	11	151	64
Median	28.1	1040	124	43.7	54.3	65.0	103	305	421	477	46.1
Min	10.8	331	6.1	17.2	7.6	13.5	7.6	58.6	103	14.5	9.1
Max	177	59,500	108,000	5770	9100	3080	17,400	70,800	2310	54,500	3350
Fabric (n = *40)*											
n	1	34	9	34	21	10	2	19	0	38	12
Median	13.4	2090	65.9	51.0	173	96.1	144	277		961	51.2
Min		720	15.5	24.7	19.6	29.3	137	110		35.7	10.7
Max		232,000	196	550	512	953	152	406		8410	624
Spheres (n = *4)*											
n	0	2	3	3	1	0	1	0	0	4	1
Median		1060	60.7	693	16,000		42.5			172	1740
Min		620	13.2	42.6						12.7	
Max		1500	1240	3160						47,800	

moulded plastic or constructions of multiple pieces of the same or similar plastic ($n = 159$). These were usually green and occasionally partly painted or glittered for decoration, and included one stem that contained metal wire and five fibres from Christmas trees and artificial turf. The leaves or more colourful petals of some plants were composed of fabric ($n = 40$) that had been glued on to the plastic stems or blades (presenting two distinct types of plant component), while brightly painted spherical and lightweight foam berries were loosely bound to some wreaths and arrangements ($n = 4$).

3.2. Polymeric composition

Out of a total of 128 plastic parts analysed by FTIR, 117 returned positive results (hit rate > 65 %) which are summarised in Fig. 2. For the (usually green) moulded sections ($n = 83$), polypropylene or polyethylene (including chlorinated polyethylene in one case) was the most abundant polymer, with contributions from polyvinyl chloride (PVC) and nylon. We also detected poly(methyl methacrylate) (PMMA), poly (butyl methacrylate) (PBMA), polyvinyl acetate (PVA) and the biodegradable polyester, polycarprolactone (PCA), that we suspect have been used as adhesives or as coatings for protection, moisture resistance or a smooth, life-like appearance (Blesius, 2018; Save et al., 2002). Presumably, the coatings in these cases were sufficiently thick to block an absorption signal from the underlying plastic matrix. Regarding the fabric-type leaves and petals ($n = 30$), polyethylene terephthalate (PET) was detected in nine cases and was assumed to be the dominant construction polymer present (and in the form of polyester fibre). However, we also detected PMMA, PBMA, PVA, PCA and polyurethane (PU) that we suspect are present as adhesives or coatings (as above) or act to effect some structural rigidity. All sections of the small, lightweight and painted spheres analysed were constructed of white, expanded polystyrene.

3.3. Elemental composition

Table 1 provides a summary of the elemental concentrations in the different components of the plastic plants. Note that Cd, Hg and Ni were detected as contaminants in three or less cases of the moulded components at concentrations below 100 mg kg^{-1} and data are not shown. Barium, Br, Cr, Cu, Fe, Pb, Sb, Ti and Zn were detected in at least 50 (or 30 % of) moulded components and at concentrations spanning at least two orders of magnitude. In the leaf and petal fabrics, detection rate was similar to or lower than that in the moulded components with the exception of Sb (greater in the fabric), and concentration ranges were smaller. The difference between median concentrations in the moulded and fabric components was greatest for Ba, Br, Cu and Ti but, according to a series of Mann Whitney tests performed in Minitab *v*19, these differences were not significant ($p > 0.05$). In the lightweight, painted spheres, Br, Cr and Ti were most frequently detected while As, Fe, Sb and Sn never returned a positive result.

A closer inspection of the data, coupled with visual observations, allow the following inferences to be made. The presence of Ba, Fe, Ti and Zn reflects the addition of common, inorganic pigments and fillers, such as barium sulphate, various Fe oxides, titanium oxide and zinc oxide, to reduce costs and improve performance or appearance (Dufton, 1998; Pritchard, 1998). Concentrations of Cr above 1000 mg kg^{-1} are restricted to samples containing high concentrations of Pb or that are painted red, suggesting the presence of lead chromate and red chrome oxide, respectively, and requiring Cr to be in its higher (VI) oxidation state (Lennartson, 2014). High concentrations of Sn (>500 mg kg^{-1}) in the samples identified as PVC suggest the presence of organo-tin heat stabilisers used to prevent thermo-oxidative degradation (Turner and Filella, 2021a).

The wide occurrence of Cu at relatively low concentrations (<300 mg kg^{-1}) suggests pigmentation of the samples, and in particular the moulded components, by the synthetic halogenated organic pigment, phthalocyanine green 7 (Christie and Abel, 2021). Because the 16-ring hydrogen atoms of the phthalocyanine molecule are replaced virtually completely by chlorine, the compound has a much lower Cu content (about 5 %) than more traditional, inorganic Cu-based pigments. Its bluish-green may also be modified to yellowish-green by the partial replacement of Cl by Br (phthalocyanine green 36). By contrast, concentrations of Cu above 1000 mg kg^{-1} are restricted to three samples that are painted grey, a colour that can be created by mixing complementary green (including Cu-based) and red pigments.

3.4. Concentrations of hazardous elements

The wide occurrence and variable concentrations of Br, Pb and Sb, coupled with their known or potential health and environmental impacts in plastics, merit particular attention. Fig. 4 shows all cases where these elements were detected and in ascending order and by sample component type.

The presence of Br at relatively low concentrations may partly reflect its incorporation in phthalocyanine green 36 where moulded plastics have a yellowish hue. However, and more generally, the heterogeneous distribution of Br concentrations among the samples (except those known to be constructed of PVC) likely reflects the occurrence of historical and contemporary brominated flame retardants in the polymer. In most cases, Br is present at concentrations insufficient to engender flame retardancy (typically at least 3 to 4 % by weight for polypropylene, for example; Papazoglou, 2004), suggesting that the material has been manufactured from recyclate that is derived, at least partly, from waste electrical and electronic (WEE) plastic (Gallen et al., 2014; Puype et al., 2017). However, in four moulded polyethlene-polypropylene components the Br content exceeds 4 %, requiring material to be sourced entirely from WEE plastic.

The presence of rather invariant concentrations of Sb in the fabric samples is consistent with its use as a catalyst (mainly as Sb_2O_3) in the manufacture of PET and its consequent contamination of the polymer (Filella, 2020). However, a more variable concentration in the moulded plastics suggests its use as a synergist (as Sb_2O_3) with certain brominated flame retardants (including tetrabromobisphenol A, TBBPA, and commercial mixtures of polybrominated diphenyl ethers, PBDEs; Papazoglou, 2004). In support of this assertion, Fig. 3 shows a highly significant relationship between the concentrations of Br and Sb in the moulded components when one sample with an exceptionally high Sb content (whose use or origin is unclear) is excluded. Note, the presence of Br but absence of Sb in the painted (polystyrene) spheres in Table 1 is also consistent with material recycling. Thus, expanded polystyrene used in the construction and electronic sectors has commonly been flame retarded with hexabromocyclododecane (HBCDD), a brominated compound that is highly effective at low concentrations and without the requirement of Sb_2O_3.

Lead is also an indicator of plastic contamination, and in particular contamination of recyclate by materials produced when restrictions on the metal were not in place. These materials include WEE plastic, PVC containing lead-based stabilisers and plastics coloured by leaded pigments such as lead chromates (Ren et al., 2021; Turner and Filella, 2021b). Results for our plastic plants indicate heterogeneous contamination by Pb is largely restricted to the moulded components and suggest its presence arises from all aforementioned sources. For example, Pb

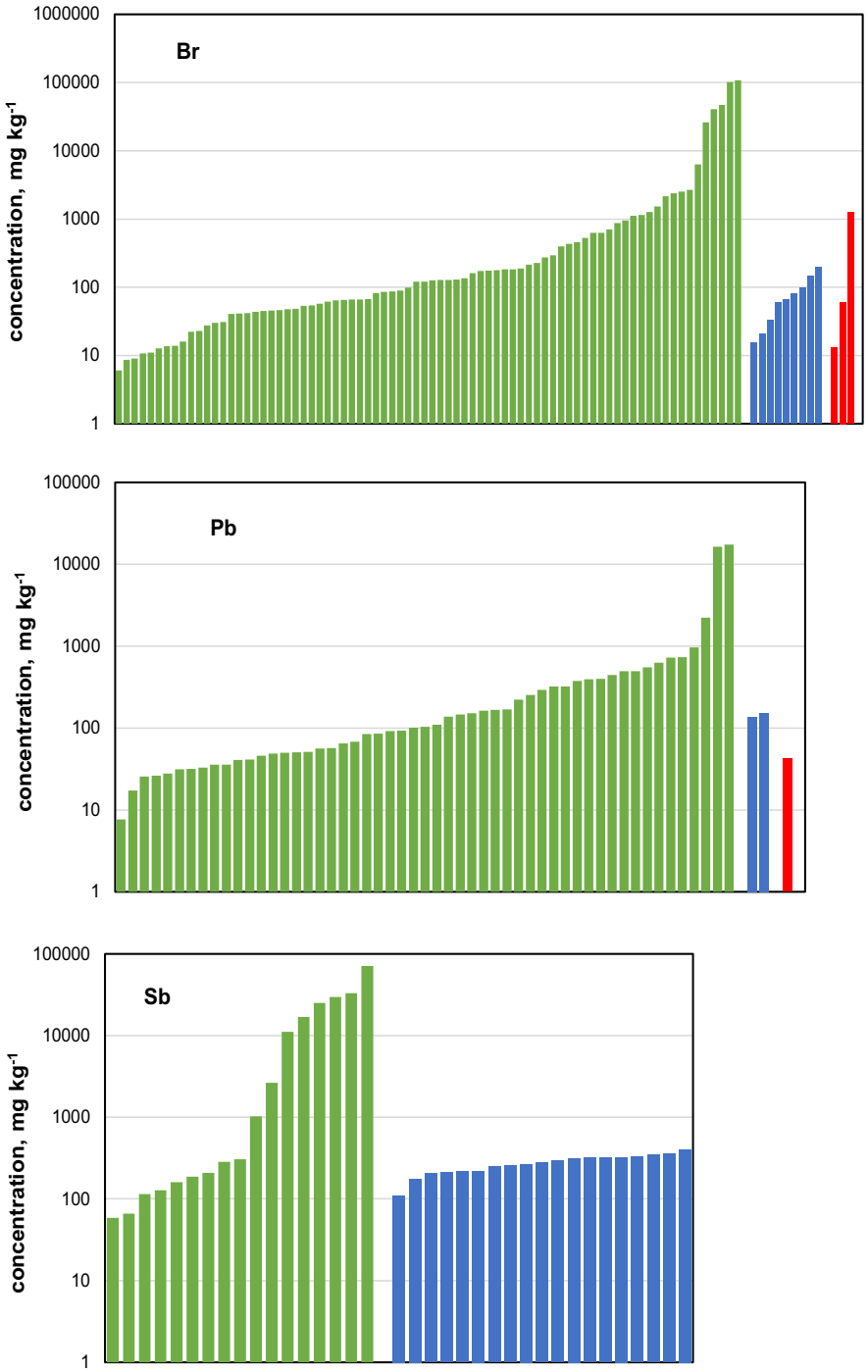

Fig. 3. Concentrations of Br, Pb and Sb, where detected by XRF and in ascending order, in the moulded components (green), fabric leaves or flowers (blue) and coloured spheres (red).

is encountered in two samples confirmed as PVC, and the two cases where Pb exceeded concentrations of 10,000 mg kg^{-1} (in polypropylene and chlorinated polyethylene) were associated with Cr concentrations above 1000 mg kg^{-1}.

3.5. Restrictions and regulations on hazardous substances

Many of the commonly employed (historical) brominated flame retardants are classified as persistent organic pollutants (POPs) and have been subject to regulation in both new products and plastic waste. In the EU, PBDEs and HBCDD are regulated, with TBBPA currently under review (ECHA, 2023). Specifically, PBDEs and HBCDD at concentrations above 1000 mg kg^{-1} cannot be recycled until the retardants have been destroyed or irreversibly transformed, with a new limit value of 500 mg kg^{-1} recently proposed (European Commission, 2019; ECHA, 2023). The EU has also introduced an Unintentional Trace Contaminant (UTC) limit, or the level of a substance that is incidentally present in a minimal amount, of 100 mg kg^{-1} for HBCDD and 10 mg kg^{-1} for PBDEs (European Commission, 2019).

Although the results of the present study report the content of total Br rather than the concentrations of specific brominated compounds, the percentage of Br in TBBPA and PBDEs employed in electrical and

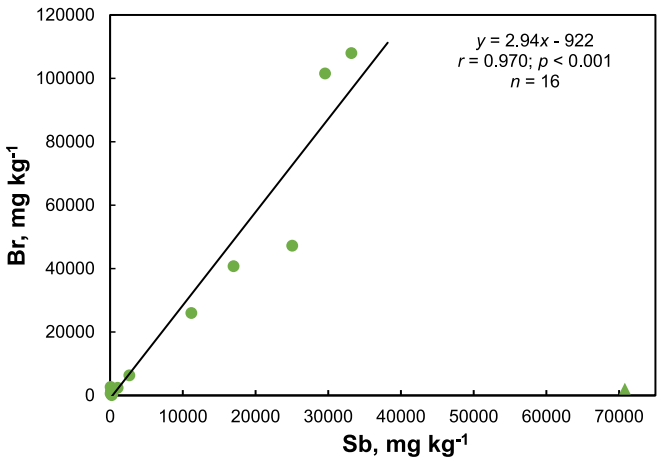

Fig. 4. Concentrations of Br versus concentrations of Sb in moulded plastic components. The equation of best fit and correlation coefficient define all data shown as circles (an outlier with Sb > 70,000 mg kg^{-1} and Br ~ 2000 mg kg^{-1} is shown as a triangle).

electronic plastics is between about 60 and 80, while that in HBCDD used in construction foam is around 80. This suggests that the moulded plastic plant components and the expanded polystyrene spheres are "potentially" non-compliant at Br concentrations above 6 to 8 mg kg^{-1} or 60 to 80 mg kg^{-1} (UTC), 300 to 400 mg kg^{-1} (proposed POP regulations), or 600 to 800 mg kg^{-1} (current POP regulations) if they contain restricted brominated compounds. On this basis, therefore, and according to the data shown in Fig. 4, as many as 78 moulded components, nine fabric components and one coloured sphere have brominated compound concentrations that are potentially above UTC limits, while 19 moulded components are potentially non-compliant under existing POP regulations.

Regarding Pb, the XRF data are, in principle, more straightforward to interpret with regard to regulations as total lead content is generally stipulated. However, there are no specific regulations that apply to interior or exterior decorative plastics. According to the current Restriction of Hazardous Substances (RoHS) Directive, Pb in any component of electrical and electronic equipment must not exceed 1000 mg kg^{-1} (Commission Delegated Directive, 2015), meaning that three moulded plant components would be non-compliant if they were electrical or electronic. 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), while 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}. Under these regulations, the number of plastic plant components that are potentially non-compliant is about 50 (toy safety, and depending on Pb migratability from the plastic matrix) and at least 30 (packaging; with some samples containing Pb below 100 mg kg^{-1} potentially non-compliant because of the additional presence of Cr(VI)).

Despite its co-association with Br, Sb is not included in the RoHS Directive, although it is subject to possible future regulation (Baron et al., 2021). Limits exist for the migration of Sb from plastic into bottled water and packaged food, but concentrations are on a weight-volume basis or per kg of contaminated food (Filella et al., 2020). Regarding the Toy Safety Directive, the maximum migratable concentration of Sb in materials that can be scraped off is 560 mg kg^{-1} (European Parliament and the Council of the EU, 2009) and on this basis eight moulded plastic plant samples would be potentially non-compliant.

3.6. General discussion

This study has provided the first chemical data on plastic plants that are widely encountered in the domestic, commercial and hospitality settings. Thus, the (mainly green) moulded components are most commonly constructed of polyethylene or polypropylene, with additional fabric components, like petals and leaves, usually constructed of PET and glued to the moulded section. However, many components are also coated with adhesives and resins in order to effect a smooth appearance or provide some protection and rigidity. XRF analysis of the moulded plastic reveals heterogeneous contamination by Br, Pb and Sb, regardless of the country of acquisition and plant design, although we note that samples obtained from two outlets of an international houseware store (n = 13) were persistently free of these contaminants. In many cases, therefore, the moulded material appears to be sourced from post-consumer recyclates that are contaminated, to varying degrees, by WEE plastic, a practise that is unwanted and, depending on the chemicals and their concentrations present, potentially illegal (Puype et al., 2015). In previous studies, consumer products derived more directly from this source have been reported to be mainly black, reflecting the principal colour of the plastic casings of small to medium-sized electrical and electronic devices and the general technological constraints on recycling of black plastics from consumer recyclate (Kuang et al., 2018; Turner, 2018; Fatunsin et al., 2020). Although we note that polypropylene is often used to encase larger, white electrical goods (Khanna et al., 2014), we suspect that in the present context, mixed recyclates (WEE plastic and non-WEE plastic) with resulting neutral colours (shades of grey) are used because they are, presumably, readily sourced and can be re-coloured with a suitable masterbatch.

Retailers are keen to publicise the recyclability of plastic plants, but given the wide contamination by traces or non-compliant concentrations of hazardous or potentially chemicals, recycling could be problematic. Regardless of their precise origin, the promotion and increasing popularity of plastic plants is clearly at odds with the goals of the proposed United Nations "End Plastic Pollution" resolution (United Nations Environment Programme, 2022).

CRediT authorship contribution statement

Andrew Turner: Conceptualization, Investigation, Formal analysis, Writing – original draft, Writing – review & editing. **Montserrat Filella:** Conceptualization, Investigation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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