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Abstract

Because of their unique properties, rare earth elements (REEs), comprising the lanthanide elements plus Sc and Y, have a variety of integral applications in modern electronic equipment. Consequently, it has been suggested that REEs may act as contaminants of and tracers for recycled electrical and electronic plastics in consumer goods. In this study, REEs have been determined in a range of consumer plastics of different polymeric makeup (*n* = 31), and purchased new and in societal circulation, by inductively coupled plasma-mass spectrometry following acid digestion. Samples were also screened by X-ray fluorescence spectrometry for Br and Sb as markers of brominated flame retardants and the retardants synergist, Sb_2O_3, respectively. One or more REE was detected in 24 samples, with four samples returning detectable concentrations of all REEs analysed and with total REE concentrations up to 8 mg kg^{-1}. REEs were most commonly observed in samples containing Br and Sb at levels insufficient to effect flame retardancy and, therefore, likely derived from recycled electronic plastic, but were not detectable in new electrical plastics. Various REEs were also present in plastics with no detectable Br and Sb, however, and where unregulated recycling is prohibited (e.g. food packaging). This observation, and correlations between pairs of REEs for all samples considered, suggests a more generic source of these elements in consumer plastics in addition to the recycling of electrical and electronic waste. REEs reported in the literature for beached marine plastics were characterised by similar concentrations and inter-element correlations, suggesting that REEs are ubiquitous and pervasive contaminants of both contemporary and historical consumer and environmental plastics.

Keywords: rare earth elements; plastics; bromine; ICP-MS; recycling; contamination

Introduction

From manufacturing, health and environmental perspectives there has been an increasing interest in the nature, amount and mobility of additives and residues in plastics (Kwon et al., 2017; Liu et al., 2017; Massos and Turner, 2017; Mercia et al., 2018; Schmidt et al., 2019; Filella, 2020). Functional additives are generally added to improve the processibility, performance, durability and safety of plastic, while residues may be present as contaminants from recycling or as remains of, for example, polymerisation catalysts (Tolinski, 2015; Turner, 2018; Wagner and Schlummer, 2020). When lost to the environment, plastics may also be contaminated by chemicals and materials acquired from their

surroundings by adherence, (ad)sorption and precipitation (Ashton et al., 2010; Huffer et al., 2019; Prunier et al., 2019; Steinman et al., 2020).

Most of the literature dealing with additives and contaminants, and exemplified above, has focussed on organic chemicals (e.g. brominated flame retardants, phthalates and bisphenols), metals (e.g. Cd, Cr, Pb) or metalloids (e.g. As, Sb), with guidelines or regulations based on concentrations or migratabilities of these substances in new products (e.g. European Parliament and Council of the EU, 2009; Commission Regulation, 2011; European Parliament and Council, 2011; US Food & Drug Administration, 2014). Recycling, however, may convert a product that is compliant for its original use into a product that is non-compliant with regards to its new use. This is a problem for electrical and electronic plastic waste that is recycled illegally or through poor management practices into food contact articles or children's toys (Guzzonato et al., 2017; Fatunsin et al., 2020).

The most common markers of electrical and electronic plastic that are often evident in recycled products are various brominated flame retardants, with or without the flame suppression synergist, Sb_2O_3 (Turner, 2018; Filella et al., 2020). However, Puype and co-workers (Puype et al., 2015; Puype et al., 2017; Guzzonato et al., 2019) have proposed that rare earth elements (REEs) may also be used as a proxy for recycled electrical and electronic plastic and for more precise information on the origin of the original product. REEs, comprising the fifteen lanthanide elements (La through to Lu), plus Sc and Y, have a variety of critical applications in modern electronic equipment because of their magnetic, phosphorescent and electrochemical properties (Balaram, 2019). Unlike brominated compounds and Sb_2O_3, REEs are not deliberately added to plastic to serve any function; rather, they are suspected of incidentally contaminating plastic during the mechanical separation and processing of recoverable components containing REEs (e.g. magnets, compact lamps, precision glass, electrodes, light emitting diodes).

As far as we are aware, the only quantitative data on REEs in consumer plastics are reported by Puype et al. (2015). Here, and using inductively coupled plasma-optical emission spectroscopy (ICP-OES) following sample digestion in acid, six REEs (Y, La, Ce, Pr, Nd, Dy) were detected in four out of seven recycled food-contact plastics purchased in Europe and that contained brominated flame retardants. In the present study, we aim to further our understanding of the occurrence and signature of REEs in plastics by using a more sensitive technique (ICP-mass spectrometry) to analyse a broader suite of REEs in a wide range of consumer plastics, including samples that are contaminated by Br (as a proxy for brominated flame retardants) and those that are free of detectable Br. We also review the literature that reports measurements of REEs in environmental

89 microplastics in order compare the concentrations and signatures of weathered and unweathered
90 plastics.

91

92 **Methods**

93 *Samples*

94 A range of consumer plastics that were purchased new or that were in societal circulation (and up to
95 20 years old) were analysed in the present study. Samples are described and coded in Table 1 and
96 include single-use food contact articles, toys, electronic housings, office equipment, cosmetic
97 containers, automotive parts, plumbing accessories and decorations that were acquired in the EU (*n*
98 = 14) and the US (*n* = 17).

99 *XRF screening*

100 Because REEs are believed to be contaminants in recycled electronic plastic, samples were initially
101 screened for Br and Sb as indicators of brominated flame retardants and the retardant synergist,
102 Sb_2O_3, respectively, by portable X-ray fluorescence (XRF) spectrometry using a Niton XL3t 950 He
103 GOLDD+. The instrument was operated remotely in a laboratory accessory stand and in a
104 standardless 'plastics' mode with thickness correction after the thickness of the measurement
105 surface had been determined with digital callipers. Counting was undertaken for 40 s at 20 kV and
106 100 μA and 20 s at 50 kV and 40 μA, and spectra were quantified by fundamental parameter
107 coefficients to yield concentrations on a dry weight basis (in mg kg^{-1}) and with a counting error of 2σ
108 (95% confidence). As a performance check, polyethylene reference discs Niton PN 180-619 (Sb = 96
109 $\pm$ 10 mg kg^{-1}) and Niton PN 180-554 (Br = 495 $\pm$ 20 mg kg^{-1}) were analysed after every ten samples.
110 Detection limits varied depending on sample thickness and composition but were generally about 40
111 mg kg^{-1} for Sb and 4 mg kg^{-1} for Br.

112

113 *Pyrolysis Gas Chromatography Mass Spectrometry*

114 Material identification was performed by pyrolysis-gas chromatography mass spectrometry (py-
115 GCMS) using a CDS Analytical 5200 Series pyroprobe coupled to a Shimadzu QP2010 SE gas
116 chromatograph mass spectrometer. This technique involves heating the material to a high
117 temperature and monitoring the products formed during decomposition. Identification of the
118 original material is achieved by investigating the sum of all components detected (average spectra
119 and by identification of the major products formed). A small (~ 50 mg) sub-sample was cut from the

original material and was inserted into a quartz capillary tube with quartz wool plugs. The capillary tube was then loaded into the pyroprobe and heated to 600 °C for 90 s. The temperatures of the GC injection port (split ratio of 100:1) and transfer line were set at 300 °C. Separation was performed on a capillary column (30 m x 0.25 mm x 0.25 μm film thickness; Restek Rtx-5MS) with a helium carrier gas set for a flow rate of 1.0 mL min^{-1}. At the initiation of pyrolysis, the oven was held at 40 °C for 2 min before being heated at a rate of 10 °C min^{-1} to 300 °C, where it was held for a further 10 min. The mass spectrometer was programmed to collect data over a mass to charge ion (m/z) range from 35 to 350 Daltons. Blanks were analysed between each sample to check for carry-over and were run continuously until initial baselines were achieved.

Mass spectra were averaged over the entire chromatogram and were matched with spectra in CDS Analytical and in-house pyrogram libraries. In addition, chromatograms were manually evaluated peak-by-peak to confirm materials identified in the libraries or to identify those not contained therein. Average spectra and major pyrolysis products were also verified by consulting published reference data (Wampler, 2006; Tsuge et al., 2011).

REE analysis

Plastics were brought into solution via a microwave digestion procedure. Thus, sub-samples (0.15 g – 0.78 g) were digested with 8 mL HNO_3, 2 mL HCl and 2 mL H_2O_2 using a Milestone Ethos system with the following digestion program: step 1, 30 min @ 80 °C; step 2, 45 min @ 180 °C; with resulting digestates subsequently diluted to a final volume of 50 mL with deionised water. In addition to the samples, digestion blanks and digestion matrix spikes were processed with each batch of samples for quality control purposes.

Scandium, Y and the lanthanides (except Pm) were analysed in triplicate with a Thermo Elemental PQ EXcell inductively coupled plasma-mass spectrometer (ICP-MS) fitted with a Meinhard nebuliser. RF forward Power was 1350 W and coolant, auxiliary and nebuliser gas flows were 15 L min^{-1}, 0.60 L min^{-1} and 0.70 L min^{-1}, respectively. Tellurium, gold and uranium at concentrations of 10 ng mL^{-1} were used as internal standards and the instrument was externally calibrated with standards procured from SPEX Certipep. Analysis of reagent and digest blanks demonstrated that the methods and procedures were free from contamination, and independent check standards prepared from an independent SPEX Certiprep stock and analysed after every ten samples revealed precisions of +/- 20%. Method detection limits, based on instrument detection limits and limits of quantification, varied depending on the element and sample mass digested, with specific values reported in Table 2 ranging from 0.002 to 0.3 mg kg^{-1}. Average recoveries derived from the analysis of the matrix spikes

(taken through the entire sample processing) ranged from about 104% for La, Ho and Tm to 110% for Sc, Nd and Sm.

Results

The polymeric compositions of the 31 consumer plastics are shown in Table 1, along with concentrations of Br and Sb determined by portable XRF. Bromine is likely present as a range of unidentified brominated flame retardants in a variety of polymers, and while Sb may be present as a flame retardant synergist in association with Br, it may also be present as catalytic residue and without measurable Br in PET (Puype et al., 2015). Bromine concentrations above about 5% generally indicate the deliberate addition of brominated compounds for fire suppression in electrical and electronic plastics (e.g. sample #8) while concentrations below this level in plastics that are normally coloured black suggest the presence (and dilution or blending) of recycled electrical and electronic plastics (Turner, 2018).

Table 2 shows the concentrations of each REE in the consumer plastics and Table 3 provides a statistical summary of the data. Each element was detected in at least four cases, with La, Ce, Pr and Nd detected in fifteen or more samples. Overall, 24 plastics contained at least one detectable REE, with all elements detected in four samples (#s 6, 9, 10, 14) and no elements detected in seven cases (#s 8, 15, 21, 24, 25, 26, 30). Also shown in Table 3 is the rank order of REE abundance in the plastic samples based on median concentrations. Thus, La and Sc are most abundant with median concentrations above 0.45 mg kg^{-1} and Tm and Lu are least abundant with median concentrations below 0.01 mg kg^{-1}.

A closer inspection of the data reveal that REEs are least abundant in relatively new electronic plastics (e.g. #s 8, 15, 30), including one sample containing percentage concentrations of Br and Sb, and most prevalent in samples that have concentrations of Br insufficient to effect flame retardancy and that are mainly coloured black (#s 1-5, 7, 8, 10, 14). However, REEs are encountered in a wider range of samples, including contemporary plastics used to package or store food (for example, a clear PET fruit container, #28; two polyethylene bottle tops, #s 20, 27; and a polymethylpentene candy container, #19).

Figure 1 presents a correlation matrix (showing Pearson product-moment correlation coefficients) for the concentrations of individual REEs detected in the plastic samples, with significant relationships highlighted and shaded according to the magnitude of p. Out of 120 elemental pairs tested, 41 were statistically significant ($p < 0.05$), with Sc the only REE not significantly correlated

with another element and Pr exhibiting significant correlations with nine other elements (and as exemplified in Figure 3).

Overall, there was no clear relationship between concentrations of REEs in consumer plastics (as median values) and crustal concentrations given in Lide (1997). However, amongst the data there were two groups that revealed distinctly different distributions, as shown in Figure 2. Thus, for Y, Ce, Pr and Nd, four of lightest and most crustally abundant REEs, there was no significant association with median concentrations in plastic ($p = 0.276$), but for the remaining REEs (Sc, La, and Sm through to Lu) there was a striking and significant linear relationship ($p < 0.01$) between crustal and consumer plastic concentrations.

Discussion

Sources of REEs in consumer plastics

This study is the first to systematically investigate the full suite of REEs in a broad range of consumer plastics. A limited number of measurements of selected REEs have been previously reported for ten black food contact utensils and containers constructed of various polymers but suspected as being derived from recycled electrical and electronic plastics based on the presence of Br and Sb (Puype et al, 2015). Specifically, Ce, Dy, Er, La, Nd, Pr and Y were targeted because of their industrial demand and abundance in waste electrical and electronic equipment, and were analysed by ICP-OES following sample digestion. Cerium and Nd were detected in three samples, Y in two sample, and Dy, La and Pr one case each. Single or median concentrations reported were similar to or greater than the highest concentrations reported in Table 3 but we note that the detection limits cited for several elements were greater than our corresponding median concentrations.

The presence of REEs in recycled electrical and electronic plastic (but evidently not in new electronic plastic) is likely to arise through contamination of the material from traces of non-plastic components as shreds and dust (Marra et al., 2018). Many REEs are particularly concentrated in small devices such hard drives, phones and iPods (Buechler et al., 2020), with specific uses including Ce, Dy, La and Y in LEDs, Ce, La and Y in fluorescent powders, Ce, La, Nd and Pr in rechargeable batteries and Nd and Pr in magnets (Puype et al., 2017). In contrast, Er and Tm appear to be encountered least in e-waste, with analysis of a range of pulverised and digested electronic devices and components failing to return detectable or quantifiable concentrations of these REEs (Buechler et al., 2020).

These observations are qualitatively consistent with our results for REEs in consumer plastics in that Y, La, Ce, Pr and Nd were among the elements most commonly detected and that exhibited many of the strongest inter-element correlations, while Er and Tm (along with Yb and Lu) were detected in the fewest cases. However, lack of correlation of between individual REE concentrations and Br content and the detection of REEs in a range of consumer plastics that do not have an e-waste signature, including food contact articles in which the mixing of recycled materials is restricted or illegal (FDA, 2006; Commission Regulation, 2011) and in PET that is contained in a controlled closed-loop recycling process (Guzzonato et al., 2017), suggests additional or alternative more general sources of contamination. These sources could be related to the occurrence of REEs in crude oils (Akinlua et al., 2008) or their use in or contamination of catalysts used in petrochemical cracking (Jha et al., 2016; Akah, 2017) or plastic manufacturing (Ferron and Henry, 2015).

REEs in environmental plastics

Given the occurrence REEs in a wide range of plastics, we would also expect to encounter them in plastics lost to the environment. Many studies have reported metals in environmental plastics as additives or adsorbed to the surface from the surrounding aqueous medium (Ashton et al., 2010; Nakashima et al., 2012; Massos and Turner, 2017; Filella and Turner, 2018; Munier and Bendell, 2018) but REEs have not, explicitly, been considered. However, in a recent study of marine microplastics retrieved from the Colombian coast of the Caribbean, Acosta-Coley et al. (2019) included nine REEs amongst a range of metals analysed. Here, fourteen polyethylene and polypropylene pellets and secondary fragments were dissolved in HF and aqua regia and the digests analysed by ICP-MS. The results, summarised in Table 4, reveal frequencies of detection, median concentrations and concentration ranges that are similar to the corresponding values reported for the consumer plastics in Table 3. Moreover, correlation analyses of the beached microplastic data revealed inter-element associations with values of $r > 0.9$ in all cases.

REEs in consumer plastics versus REEs in environmental plastics

The relationships between pairs of REEs in consumer plastics and beached microplastics are exemplified as scatter plots in Figure 3, with linear regression analysis also applied and best-fit lines annotated ($y = mx$ or $y = mx + c$, where m is the gradient and c is the y-axis intercept). The data illustrate significant relationships ($p < 0.05$) for both types of plastic and between elements that were either related or unrelated in Figure 2, but with different gradients (Nd-Ce), the same gradient but different intercepts (Nd-Pr), or similar gradients (Pr-Gd and Dy-Gd).

Differences in the gradients or intercepts observed in some cases could be related to different digests employed (of different REE recoveries from the plastic matrix), contamination of beached material by particulates captured from the environment or the selective mobilisation of specific REEs from microplastics on weathering. Nevertheless, the broad similarities in REE signatures amongst environmental pellets and fragments of unknown age and origin and consumer goods of variety of ages, functions and polymer construction are remarkable and highlight the pervasiveness and ubiquity of these elements and their signatures in both contemporary and historical plastics.

For the eleven cases where the most significant inter-element correlations ($p < 0.01$) were observed in the consumer plastics (Figure 1) and where these metals were also measured in marine microplastics, the gradients arising from linear regression analysis are plotted against corresponding crustal abundance ratios in Figure 4. Most data points for both groups of plastics lie close to the line of unit slope, with respective Pearson product-moment correlation coefficients of 0.984 ($p < 0.01$) and 0.983 ($p < 0.01$) defining the consumer plastic and marine microplastic data. This observation, coupled with lack of a relationship between gradients and ratios based on industrial demand in 2015 and as given in Roskill (2016), provides further evidence of the widespread, general contamination of plastics by REEs rather than contamination by REEs restricted to or more critical to the electronics sector.

Contamination of the plastisphere and human exposure

REEs are emerging contaminants that have been reported from anthropogenic sources (e.g. mining, electronic waste, fossil fuel combustion, agricultural applications) in a variety of environments, including ground water (Ayedun et al., 2017), surface water (Hatje et al., 2016), soils (Hu et al., 2006), sediments (Brito et al., 2018), biota (Bosco-Santos et al., 2018) and the atmosphere (Wang and Liang, 2014). However, the present study is significant in that is has demonstrated the wide REE contamination of the “plastisphere” that does not appear to be related to a single source or activity (e.g. electronic waste recycling).

The occurrence REEs in consumer plastics, and in particular, food contact articles, presents a potential route of human exposure through migration from the polymeric matrix. Although the health impacts arising from chronic exposure to small quantities of these metals, and their more general mechanisms of human and environmental toxicity, are unknown (Gwenzi et al., 2018), when REEs in food and tap water and certain medicines are considered (Manousi et al., 2020), plastics are unlikely to represent a significant vector of exposure to the general population. Nevertheless, contamination of plastics by REEs needs to be considered amongst the range of other, more widely known and better-studied chemical additives and residues.

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Table 1: Descriptions, polymeric makeup and Br and Sb concentrations in the 31 consumer plastic samples. Respective detection limits for Br and Sb were in the range 5 to 15 mg kg^{-1} and 30 to 65 mg kg^{-1} (nd = not detected).

sample	description	polymeric composition	Br, mg kg^{-1}	Sb, mg kg^{-1}
1	board game	styrene-divinylbenzene copolymer	341	nd
2	coat hanger	polystyrene	7580	3825
3	tyre cap	ethylene-propylene copolymer	37	nd
4	poster tube cap	acrylonitrile-styrene copolymer	2690	1520
5	makeup brush handle	styrene-propylene	261	385
6	makeup brush bristles	polypropylene	300	nd
7	bag handle	acrylonitrile-butadiene-styrene copolymer	3100	2100
8	timer switch casing	acrylonitrile-butadiene-styrene copolymer	57100	36800
9	torch casing	diallyl phthalate	165	2940
10	magnifying glass handle	polypropylene	nd	nd
11	games counter	polystyrene	862	nd
12	water pipe adaptor	poly(vinyl chloride)	141	nd
13	cocktail stirrer	acrylonitrile-butadiene-styrene copolymer	3430	1240
14	xmas beads	styrene-methyl methacrylate	10900	13900
15	ink cartridge	styrene-polybutadiene Copolymer	nd	nd
16	CD case (backing)	polystyrene	3640	1350
17	CD case (cover)	polystyrene	nd	nd
18	desk corner cover	styrene-divinylbenzene copolymer	nd	nd
19	xmas candy container	poly(4-methyl-1-pentene) or related	nd	nd
20	bottle top	polyethylene	nd	nd
21	bottle top	polypropylene	nd	nd
22	take out container	acrylonitrile-styrene copolymer	nd	nd
23	remote control	acrylonitrile-butadiene-styrene copolymer	nd	nd
24	water bottle	poly(ethylene terephthalate)	nd	84
25	water bottle lid	polyethylene	nd	121
26	yoghurt pot	polypropylene	nd	nd
27	bottle top	polyethylene	nd	nd
28	fruit container	poly(ethylene terephthalate)	nd	263
29	ink pen	acrylonitrile-styrene copolymer	nd	nd
30	blackberry case	MDI-polyactone polyurethane	nd	nd
31	computer mouse	acrylonitrile-styrene copolymer	67	108

Table 2: Individual and summed concentrations of REEs in each consumer plastic (in mg kg^{-1}). Note that numbers preceded by "<" are sample- and element-specific method detection limits.

sample	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Sum
1	0.31	0.28	0.32	0.19	0.039	0.14	<0.05	0.024	0.032	0.006	0.025	0.006	<0.02	<0.005	<0.02	<0.005	1.38
2	0.33	0.21	0.50	0.38	0.099	0.38	<0.05	0.096	0.067	0.005	<0.02	<0.005	<0.02	<0.005	<0.02	<0.005	2.07
3	0.84	0.69	1.49	1.37	0.27	0.94	0.21	0.076	0.18	0.028	0.14	0.028	<0.3	<0.02	<0.3	<0.02	6.27
4	<0.2	0.06	0.31	0.13	0.025	0.085	<0.05	0.028	<0.02	<0.005	<0.02	<0.005	<0.02	<0.005	<0.02	<0.005	0.64
5	0.51	0.80	0.83	0.42	0.090	0.33	0.094	0.33	0.088	0.015	0.077	0.017	0.048	0.008	0.044	0.008	3.71
6	<0.3	<0.03	1.04	<0.03	<0.005	<0.03	<0.05	<0.005	<0.07	<0.015	<0.07	<0.005	<0.3	<0.005	<0.3	<0.005	1.04
7	<0.2	0.051	<0.2	0.095	0.022	0.072	<0.05	0.068	<0.02	<0.005	0.068	<0.005	<0.02	<0.005	<0.02	<0.005	0.38
8	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.04	<0.004	<0.02	<0.004	<0.02	<0.004	<0.02	<0.004	<0.02	<0.004	<0.60
9	0.54	0.91	2.60	0.96	0.18	0.64	0.14	0.054	0.13	0.021	0.11	0.022	0.060	0.009	0.051	0.009	6.43
10	0.30	0.96	1.49	1.67	0.41	1.32	0.23	0.24	0.25	0.027	0.24	0.038	0.085	0.014	0.077	0.012	7.37
11	0.75	<0.02	<0.2	<0.02	<0.002	<0.02	<0.03	<0.003	<0.02	<0.003	<0.02	<0.003	<0.02	<0.003	<0.02	<0.003	0.75
12	<0.2	0.078	<0.2	0.048	0.011	0.037	<0.04	<0.004	<0.02	<0.004	<0.02	<0.004	<0.02	<0.004	<0.02	<0.004	0.17
13	<0.2	0.055	<0.2	0.11	0.022	0.081	<0.05	0.026	<0.02	<0.005	<0.02	<0.005	<0.02	<0.005	<0.02	<0.005	0.29
14	0.33	0.530	1.92	2.53	0.28	1.46	0.13	0.30	0.16	0.026	0.15	0.013	0.032	0.005	0.025	0.005	7.89
15	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
16	0.48	0.027	<0.2	0.050	0.015	0.040	<0.05	0.037	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.64
17	0.25	<0.02	0.24	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.50
18	0.53	0.023	0.55	0.050	0.012	0.038	<0.05	0.006	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	1.21
19	0.60	<0.02	0.29	<0.02	0.005	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.90
20	<0.2	0.038	<0.2	0.11	0.030	0.10	<0.05	<0.003	0.023	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.30
21	<0.2	<0.02	<0.2	<0.02	<0.002	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
22	<0.2	<0.02	0.65	<0.02	0.005	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.66
23	<0.2	<0.02	0.47	<0.02	<0.002	<0.02	<0.05	0.005	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.48
24	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
25	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
26	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
27	<0.2	<0.02	0.48	<0.02	0.004	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.49
28	<0.2	<0.02	<0.2	<0.02	0.005	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.005
29	0.37	<0.02	<0.2	<0.02	0.004	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.37
30	<0.2	<0.02	<0.2	<0.02	<0.003	<0.02	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	<0.59
31	0.53	<0.02	<0.2	0.024	0.013	0.027	<0.05	<0.003	<0.002	<0.003	<0.02	<0.003	<0.02	<0.005	<0.02	<0.003	0.59

Table 3: Number of cases in which each REE was detected in the 31 consumer plastics (*n*) and summary statistics for their concentrations (in mg kg^{-1}). The rank order is based on median concentrations.

	n	mean	sd	median	min	max	rank
Sc	14	0.48	0.18	0.49	0.25	0.84	2
Y	14	0.34	0.36	0.14	0.023	0.96	3
La	15	0.88	0.70	0.55	0.24	2.60	1
Ce	15	0.54	0.75	0.13	0.024	2.53	6
Pr	20	0.08	0.12	0.022	0.0036	0.41	12
Nd	15	0.38	0.49	0.10	0.027	1.46	8
Sm	5	0.16	0.056	0.14	0.094	0.23	4
Eu	13	0.099	0.11	0.054	0.0050	0.33	9=
Gd	8	0.12	0.079	0.11	0.023	0.25	5
Tb	7	0.018	0.010	0.021	0.0050	0.028	13
Dy	7	0.12	0.071	0.11	0.025	0.24	7
Ho	6	0.021	0.012	0.019	0.0056	0.038	14
Er	4	0.056	0.023	0.054	0.032	0.085	9=
Tm	4	0.0090	0.0036	0.0087	0.0049	0.014	15
Yb	4	0.049	0.022	0.047	0.025	0.077	11
Lu	4	0.0084	0.0029	0.0083	0.0049	0.012	16
sum	24	1.85	2.48	0.65	0.0055	7.89	

438 Table 4: Number of cases in which REEs were detected in 14 beached microplastics (*n*) and summary
439 statistics for their concentrations (in mg kg^{-1}) (Acosta-Coley et al., 2019). The rank order is based on
440 median concentrations.

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	n	mean	sd	median	min	max	rank
Sc	9	0.084	0.11	0.030	0.010	0.34	6=
Y	10	0.19	0.15	0.15	0.040	0.47	4
La	10	0.18	0.15	0.16	0.030	0.49	2=
Ce	10	0.46	0.31	0.40	0.16	1.13	1
Pr	6	0.037	0.029	0.035	0.010	0.090	5
Nd	10	0.18	0.14	0.16	0.040	0.46	2=
Sm	4	0.038	0.022	0.030	0.020	0.070	6=
Gd	4	0.035	0.025	0.030	0.010	0.070	6=
Dy	3	0.033	0.015	0.030	0.020	0.050	6=

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Figure 1: Correlation matrix highlighting significant paired relationships in REE concentrations in the consumer plastics. The shading denotes the level of significance of the correlation coefficient and the number in parentheses is the number of sample pairs.

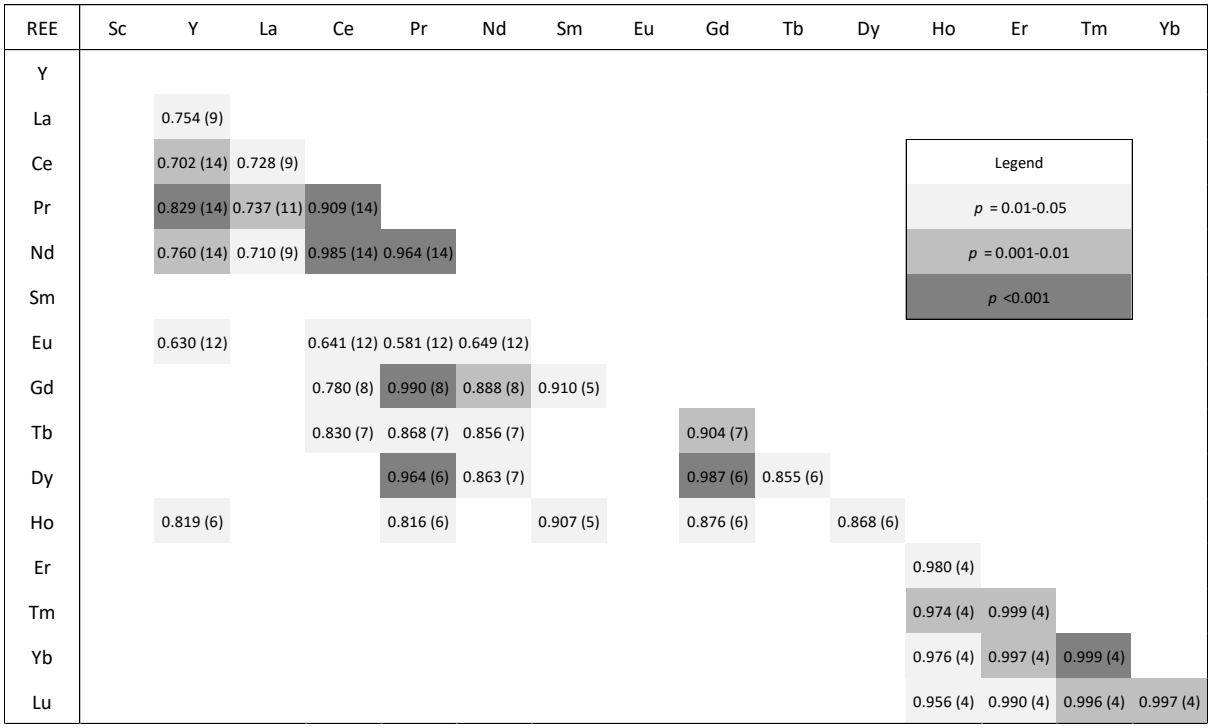

Figure 2: Median REE concentrations in consumer plastics versus corresponding crustal abundances. Elements in black are defined by the annotated line, derived from linear regression analysis, while those circled in grey exhibit no significant relationship.

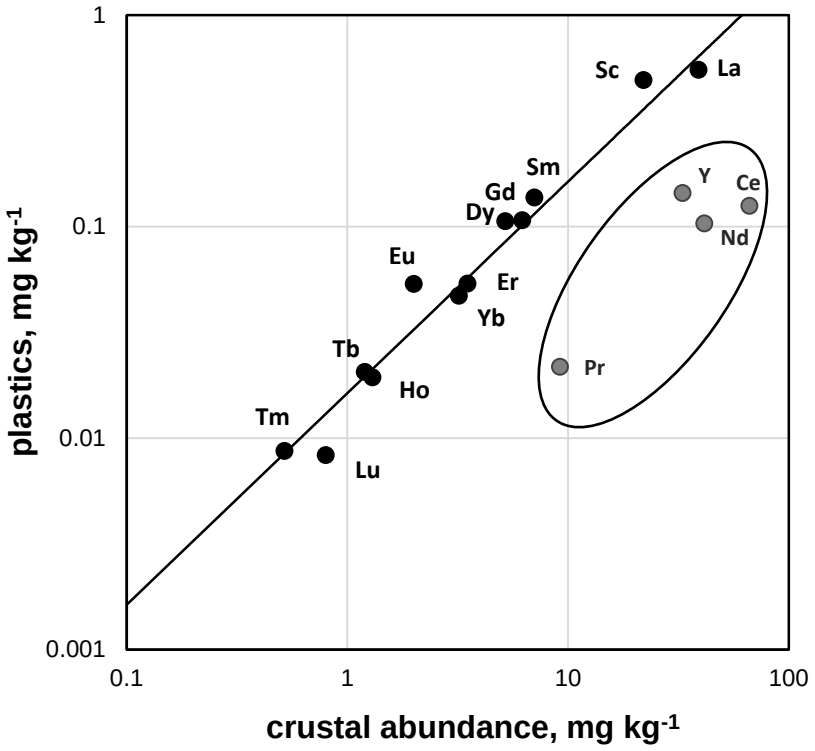

Figure 3: Scatter plots of concentrations of pairs of individual REEs in consumer plastics (●) and beached microplastics (o; Acosta-Coley et al., 2019). Annotated are statistical results of linear regression analysis and the best-fit lines forced through the origin and of the form $y = mx$ (except for Nd-Pr in beached microplastics; $y = mx + c$).

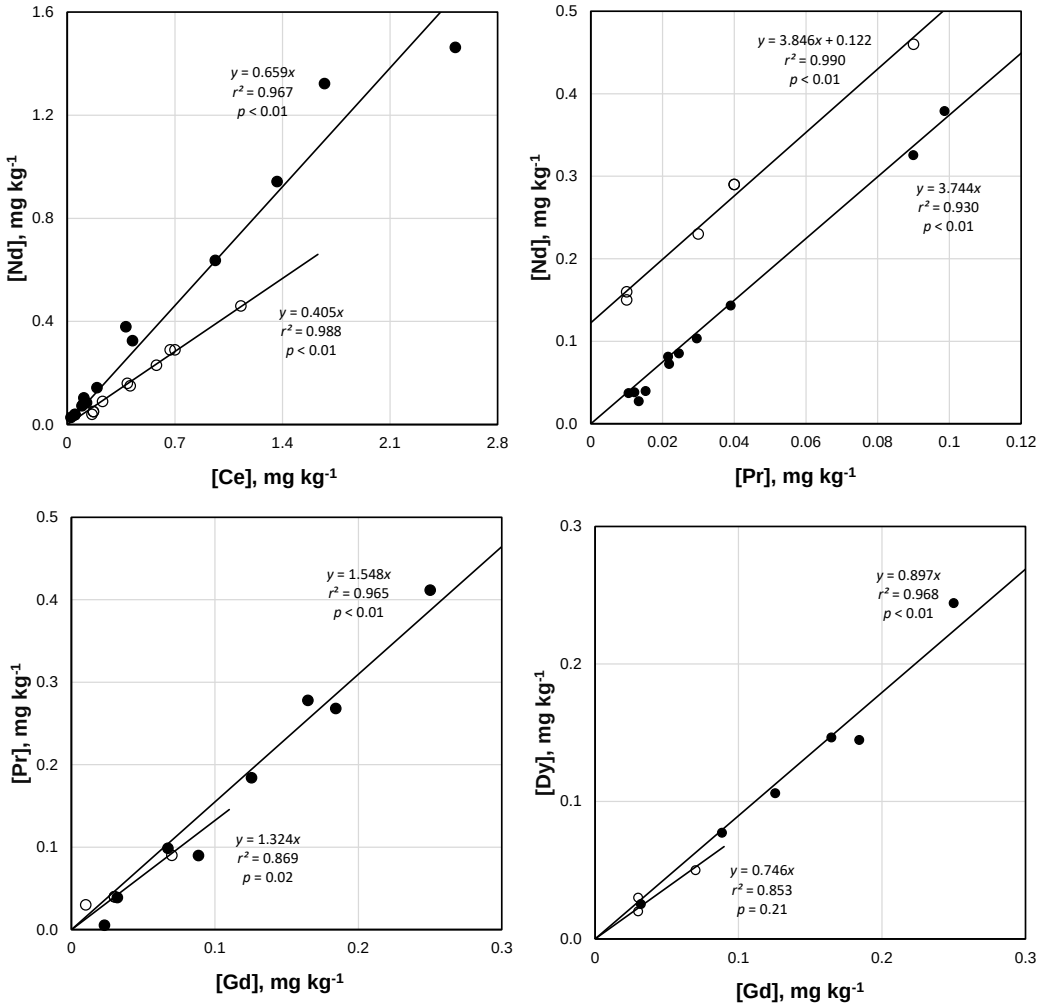

Figure 4: Gradients, *m*, defining inter-element relationships in consumer plastics (●) and beached microplastics (o; Acosta-Coley et al., 2019) versus crustal abundance ratios (Lide, 1997). Values of *m* were derived from linear regression analysis forced through the origin except for Nd-Pr, Pr-Y, Pr-La and Gd-Nd in beached microplastics (analysis involved a *y*-axis intercept). The solid line denotes unit slope.