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1 **Rare earth elements in plastics**

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25

26 **Abstract**

27 Because of their unique properties, rare earth elements (REEs), comprising the lanthanide elements
28 plus Sc and Y, have a variety of integral applications in modern electronic equipment. Consequently,
29 it has been suggested that REEs may act as contaminants of and tracers for recycled electrical and
30 electronic plastics in consumer goods. In this study, REEs have been determined in a range of
31 consumer plastics of different polymeric makeup ($n = 31$), and purchased new and in societal
32 circulation, by inductively coupled plasma-mass spectrometry following acid digestion. Samples
33 were also screened by X-ray fluorescence spectrometry for Br and Sb as markers of brominated
34 flame retardants and the retardants synergist, Sb_2O_3 , respectively. One or more REE was detected in
35 24 samples, with four samples returning detectable concentrations of all REEs analysed and with
36 total REE concentrations up to 8 mg kg^{-1} . REEs were most commonly observed in samples containing
37 Br and Sb at levels insufficient to effect flame retardancy and, therefore, likely derived from recycled
38 electronic plastic, but were not detectable in new electrical plastics. Various REEs were also present
39 in plastics with no detectable Br and Sb, however, and where unregulated recycling is prohibited
40 (e.g. food packaging). This observation, and correlations between pairs of REEs for all samples
41 considered, suggests a more generic source of these elements in consumer plastics in addition to the
42 recycling of electrical and electronic waste. REEs reported in the literature for beached marine
43 plastics were characterised by similar concentrations and inter-element correlations, suggesting that
44 REEs are ubiquitous and pervasive contaminants of both contemporary and historical consumer and
45 environmental plastics.

46

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

48

49 **Introduction**

50 From manufacturing, health and environmental perspectives there has been an increasing interest in
51 the nature, amount and mobility of additives and residues in plastics (Kwon et al., 2017; Liu et al.,
52 2017; Massos and Turner, 2017; Mercia et al., 2018; Schmidt et al., 2019; Filella, 2020). Functional
53 additives are generally added to improve the processibility, performance, durability and safety of
54 plastic, while residues may be present as contaminants from recycling or as remains of, for example,
55 polymerisation catalysts (Tolinski, 2015; Turner, 2018; Wagner and Schlummer, 2020). When lost to
56 the environment, plastics may also be contaminated by chemicals and materials acquired from their

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

59 Most of the literature dealing with additives and contaminants, and exemplified above, has focussed
60 on organic chemicals (e.g. brominated flame retardants, phthalates and bisphenols), metals (e.g. Cd,
61 Cr, Pb) or metalloids (e.g. As, Sb), with guidelines or regulations based on concentrations or
62 migratabilities of these substances in new products (e.g. European Parliament and Council of the EU,
63 2009; Commission Regulation, 2011; European Parliament and Council, 2011; US Food & Drug
64 Administration, 2014). Recycling, however, may convert a product that is compliant for its original
65 use into a product that is non-compliant with regards to its new use. This is a problem for electrical
66 and electronic plastic waste that is recycled illegally or through poor management practices into
67 food contact articles or children's toys (Guzzonato et al., 2017; Fatunsin et al., 2020).

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

80 As far as we are aware, the only quantitative data on REEs in consumer plastics are reported by
81 Puype et al. (2015). Here, and using inductively coupled plasma-optical emission spectroscopy (ICP-
82 OES) following sample digestion in acid, six REEs (Y, La, Ce, Pr, Nd, Dy) were detected in four out of
83 seven recycled food-contact plastics purchased in Europe and that contained brominated flame
84 retardants. In the present study, we aim to further our understanding of the occurrence and
85 signature of REEs in plastics by using a more sensitive technique (ICP-mass spectrometry) to analyse
86 a broader suite of REEs in a wide range of consumer plastics, including samples that are
87 contaminated by Br (as a proxy for brominated flame retardants) and those that are free of
88 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 μ A and 20 s at 50 kV and 40 μ A, 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 ($\sim 50\ mg$) sub-sample was cut from the

120 original material and was inserted into a quartz capillary tube with quartz wool plugs. The capillary
121 tube was then loaded into the pyroprobe and heated to 600 °C for 90 s. The temperatures of the GC
122 injection port (split ratio of 100:1) and transfer line were set at 300 °C. Separation was performed on
123 a capillary column (30 m x 0.25 mm x 0.25 µm film thickness; Restek Rtx-5MS) with a helium carrier
124 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
125 min before being heated at a rate of 10 °C min⁻¹ to 300 °C, where it was held for a further 10 min.
126 The mass spectrometer was programmed to collect data over a mass to charge ion (m/z) range from
127 35 to 350 Daltons. Blanks were analysed between each sample to check for carry-over and were run
128 continuously until initial baselines were achieved.

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

134 *REE analysis*

135 Plastics were brought into solution via a microwave digestion procedure. Thus, sub-samples (0.15 g –
136 0.78 g) were digested with 8 mL HNO₃, 2 mL HCl and 2 mL H₂O₂ using a Milestone Ethos system with
137 the following digestion program: step 1, 30 min @ 80 °C; step 2, 45 min @ 180 °C; with resulting
138 digestates subsequently diluted to a final volume of 50 mL with deionised water. In addition to the
139 samples, digestion blanks and digestion matrix spikes were processed with each batch of samples for
140 quality control purposes.

141 Scandium, Y and the lanthanides (except Pm) were analysed in triplicate with a Thermo Elemental
142 PQ EXcell inductively coupled plasma-mass spectrometer (ICP-MS) fitted with a Meinhard nebuliser.
143 RF forward Power was 1350 W and coolant, auxiliary and nebuliser gas flows were 15 L min⁻¹, 0.60 L
144 min⁻¹ and 0.70 L min⁻¹, respectively. Tellurium, gold and uranium at concentrations of 10 ng mL⁻¹
145 were used as internal standards and the instrument was externally calibrated with standards
146 procured from SPEX Certiprep. Analysis of reagent and digest blanks demonstrated that the methods
147 and procedures were free from contamination, and independent check standards prepared from an
148 independent SPEX Certiprep stock and analysed after every ten samples revealed precisions of +/-
149 20%. Method detection limits, based on instrument detection limits and limits of quantification,
150 varied depending on the element and sample mass digested, with specific values reported in Table 2
151 ranging from 0.002 to 0.3 mg kg⁻¹. Average recoveries derived from the analysis of the matrix spikes

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

154

155 **Results**

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

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

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

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

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

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

193

194 **Discussion**

195 *Sources of REEs in consumer plastics*

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

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

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

226 *REEs in environmental plastics*

227 Given the occurrence REEs in a wide range of plastics, we would also expect to encounter them in
228 plastics lost to the environment. Many studies have reported metals in environmental plastics as
229 additives or adsorbed to the surface from the surrounding aqueous medium (Ashton et al., 2010;
230 Nakashima et al., 2012; Massos and Turner, 2017; Filella and Turner, 2018; Munier and Bendell,
231 2018) but REEs have not, explicitly, been considered. However, in a recent study of marine
232 microplastics retrieved from the Colombian coast of the Caribbean, Acosta-Coley et al. (2019)
233 included nine REEs amongst a range of metals analysed. Here, fourteen polyethylene and
234 polypropylene pellets and secondary fragments were dissolved in HF and aqua regia and the digests
235 analysed by ICP-MS. The results, summarised in Table 4, reveal frequencies of detection, median
236 concentrations and concentration ranges that are similar to the corresponding values reported for
237 the consumer plastics in Table 3. Moreover, correlation analyses of the beached microplastic data
238 revealed inter-element associations with values of $r > 0.9$ in all cases.

239 *REEs in consumer plastics versus REEs in environmental plastics*

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

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

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

263 *Contamination of the plastisphere and human exposure*

264 REEs are emerging contaminants that have been reported from anthropogenic sources (e.g. mining,
265 electronic waste, fossil fuel combustion, agricultural applications) in a variety of environments,
266 including ground water (Ayedun et al., 2017), surface water (Hatje et al., 2016), soils (Hu et al.,
267 2006), sediments (Brito et al., 2018), biota (Bosco-Santos et al., 2018) and the atmosphere (Wang
268 and Liang, 2014). However, the present study is significant in that it has demonstrated the wide REE
269 contamination of the “plastisphere” that does not appear to be related to a single source or activity
270 (e.g. electronic waste recycling).

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

279

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284

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399 Wagner, S., Schlummer, M., 2020. Legacy additives in a circular economy of plastics: Current
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402 Table 1: Descriptions, polymeric makeup and Br and Sb concentrations in the 31 consumer plastic
 403 samples. Respective detection limits for Br and Sb were in the range 5 to 15 mg kg⁻¹ and 30 to 65 mg
 404 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

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416 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-
 417 specific method detection limits.

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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

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

	<i>n</i>	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
423 sum	24	1.85	2.48	0.65	0.0055	7.89	

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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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	<i>n</i>	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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446 Figure 1: Correlation matrix highlighting significant paired relationships in REE concentrations in the
 447 consumer plastics. The shading denotes the level of significance of the correlation coefficient and
 448 the number in parentheses is the number of sample pairs.

REE	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb
Y															
La		0.754 (9)													
Ce		0.702 (14)	0.728 (9)												
Pr		0.829 (14)	0.737 (11)	0.909 (14)											
Nd		0.760 (14)	0.710 (9)	0.985 (14)	0.964 (14)										
Sm															
Eu		0.630 (12)		0.641 (12)	0.581 (12)	0.649 (12)									
Gd				0.780 (8)	0.990 (8)	0.888 (8)	0.910 (5)								
Tb				0.830 (7)	0.868 (7)	0.856 (7)			0.904 (7)						
Dy					0.964 (6)	0.863 (7)			0.987 (6)	0.855 (6)					
Ho		0.819 (6)			0.816 (6)		0.907 (5)		0.876 (6)		0.868 (6)				
Er												0.980 (4)			
Tm												0.974 (4)	0.999 (4)		
Yb												0.976 (4)	0.997 (4)	0.999 (4)	
Lu												0.956 (4)	0.990 (4)	0.996 (4)	0.997 (4)

Legend

$p = 0.01-0.05$

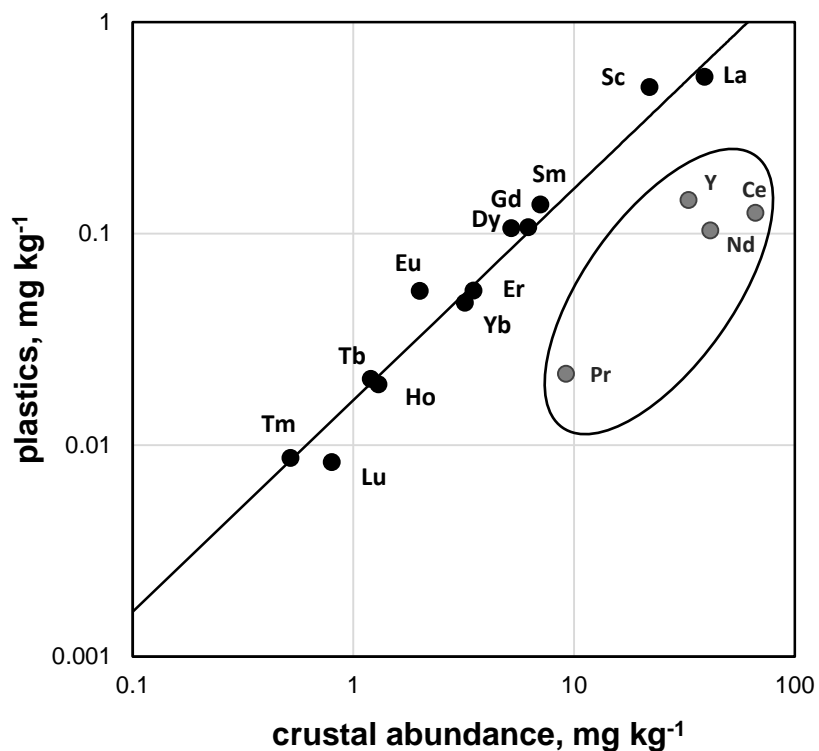
$p = 0.001-0.01$

$p < 0.001$

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

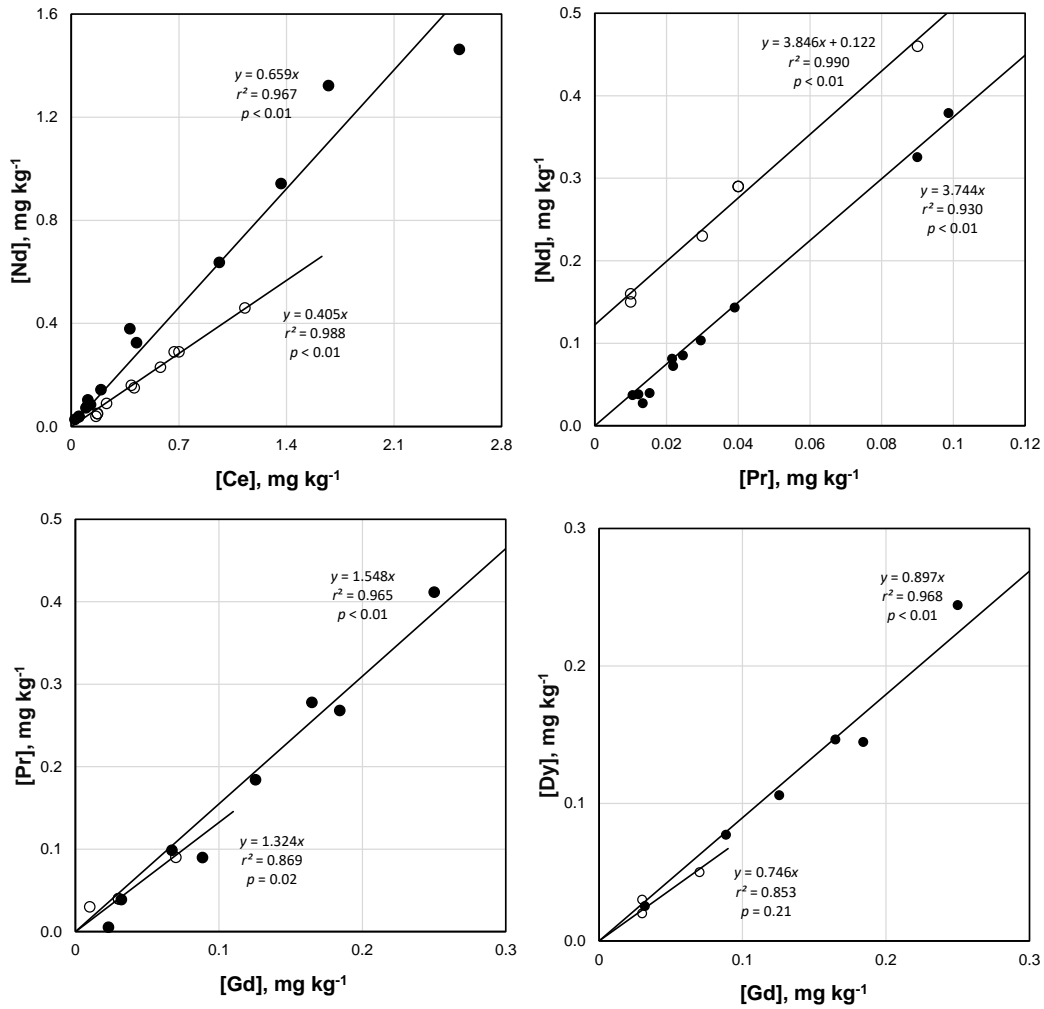
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470 Figure 3: Scatter plots of concentrations of pairs of individual REEs in consumer plastics (●) and
 471 beached microplastics (○; Acosta-Coley et al., 2019). Annotated are statistical results of linear
 472 regression analysis and the best-fit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



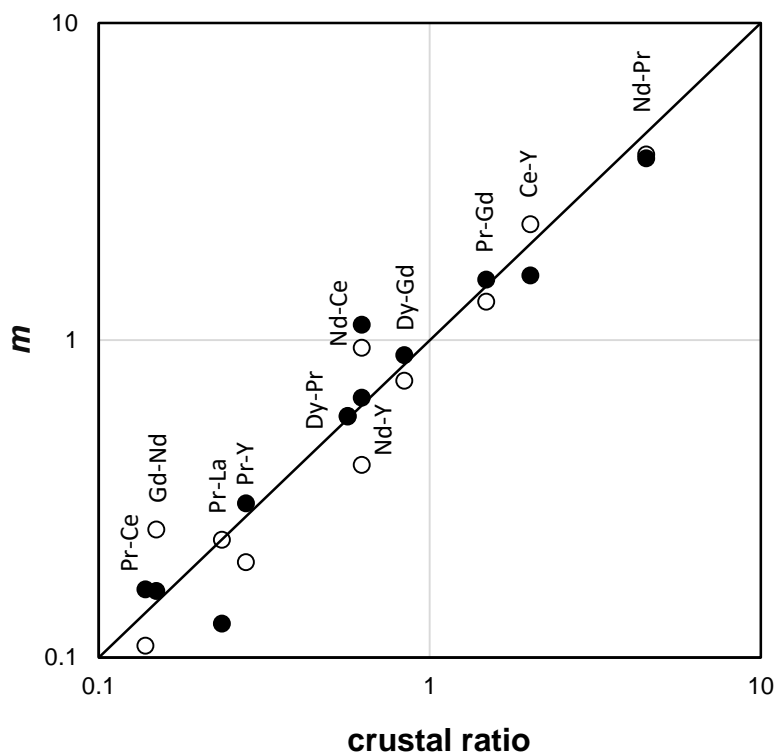
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478 Figure 4: Gradients, m , defining inter-element relationships in consumer plastics (●) and beached
 479 microplastics (○; Acosta-Coley et al., 2019) versus crustal abundance ratios (Lide, 1997). Values of m
 480 were derived from linear regression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



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