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A LARGE-SCALE INVESTIGATION OF MICROPLASTIC CONTAMINATION: ABUNDANCE AND CHARACTERISTICS OF MICROPLASTICS IN EUROPEAN BEACH SEDIMENT

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Abstract

Here we present the large-scale distribution of microplastic contamination in beach sediment across Europe. Sediment samples were collected from 23 locations across 13 countries by citizen scientists, and analysed using a standard operating procedure. We found significant variability in the concentrations of microplastics, ranging from 72±24 to 1512±187 microplastics per kg of dry sediment, with high variability within sampling locations. Three hotspots of microplastic accumulation (>700 microplastics per kg of dry sediment) were found. There was limited variability in the physico-chemical characteristics of the plastics across sampling locations. The majority of the microplastics were fibrous, less than 1 mm in size, and blue/black in colour. In addition, using Raman spectrometry we identified particles as polyester, polyethylene, and polypropylene. Our research is the first large spatial-scale analysis of microplastics on European beaches giving insights into the nature and extent of the microplastic challenge.

Key words: Citizen Science; Microplastics; Beach Sediment; Europe; Plastic Pollution

1. Introduction

Since the first commercial manufacture of plastics in the 1940s, plastic production and consumption have increased rapidly (Cole et al. 2011), with approximately 322 million tonnes (Mt) of plastic produced in 2015 (PlasticsEurope 2016). Approximately 5 to 13 Mt of plastic waste entered the ocean in 2010 (Jambeck et al. 2015), where it will persist and accumulate (Barnes et al. 2009). One subgroup of plastic that has raised particular concern are microplastics (MPs), commonly defined as pieces of plastic smaller than 5 mm (Thompson 2004; Arthur et al. 2009; Cole et al. 2011). MPs are now ubiquitous in the marine environment (Eriksen et al. 2014): their presence has been recorded near densely-populated areas, remote regions, and in different types of marine environments, such as beaches (e.g. Besley et al. 2017), estuaries (e.g. Leslie et al. 2013), surface water (e.g. Lusher et al. 2015) and deep sea sediment (e.g. Van Cauwenberghe et al. 2015).

A distinction is commonly made between primary and secondary MPs. Primary MPs are manufactured to be of microscopic size and are often purposefully added to products (Derraik 2002; Napper et al. 2015) or can be used as raw material in industry. These MPs likely enter the environment via wastewater treatment plants and industrial drainage systems (Derraik 2002; Napper et al. 2015). Secondary MPs are the result of the gradual weathering or abrasion of larger plastics, mainly through prolonged exposure to solar UV radiation resulting in photo-degradation, or mechanical abrasion (Barnes et al. 2009; Andrady 2011; GESAMP 2015). Weathering is particularly evident on beaches, where temperatures and oxygen concentrations are higher than in water (Andrady 2011; GESAMP 2015).

As fragmentation and weathering decreases the size of plastics, their potential to be ingested by marine biota increases (Browne et al. 2008). The bioavailability of MPs in the marine environment has been demonstrated in different studies. MPs have been found in mussels (Santana et al. 2016), demersal and pelagic fish species (Bellas et al. 2016; Rummel et al. 2016), worms and seabirds (Cole et al. 2013). The direct effects of MP ingestion include reduced feeding, blocking of the intestinal tract leading to starvation and impaired bodily functioning, and translocation to the circulatory system (Browne et al. 2008; Cole et al. 2013; Wright et al. 2013). Furthermore, a limited number of studies have demonstrateding the trophic transfer of MPs have raised concerns about MPs and their possible negative impact on the health of marine food webs and humans (Farrell and Nelson 2013; Setälä et al. 2014; Van Cauwenberghe and Janssen 2014; Rochman et al. 2015).

Numerous studies have quantified the abundance of MPs in marine sediment in locations in Europe and other continents. There is a wide range in concentrations of MPs recorded in Europe: from less than 1 MP/kg dry weight (d.w.) (Friere et al. 2017), to over 2000 MP/kg d.w. (Vaniello et al. 2013; Popa et al. 2014; Leslie et al. 2017). Part of this variation can be attributed to the different methodologies employed for extraction, as well as different size definitions of MPs (Cole et al. 2011; Besley et al. 2017). For example, there were differences in the way in which samples were obtained, how the MPs were separated from the sediment, and how MPs were subsequently identified across the literature (Besley et al. 2017). Additionally, the identification of MPs can be performed using different instruments with varying degrees of accuracy (Song et al. 2015; Käppler et al. 2016; Qiu et al. 2016). These differences can limit the comparability of the reported abundances, making it difficult to gain an understanding of the broader spatial distribution of MP abundance (Cole et al. 2011; Besley et al. 2017).

Besley et al. (2017) investigated the major sources of variation in sampling and extraction procedures. The main source of variation resulted from the extraction procedure, and not the sampling technique. Based on these outcomes we developed a citizen science project where samples were collected by non-professional volunteers (Bosker et al. 2017). Recently, researchers have begun to realise the value of these volunteers regarding the significant resources that they can provide in terms of labour, skills, and even finance (Silvertown 2009). Citizen science is particularly valuable to large-scale projects that require extensive data collection (Silvertown 2009; Dickinson et al. 2010). There are a variety of ways citizen scientists can participate in research, ranging from sample collection (as in the current study), to helping analysing and processing data (Kobori et al. 2015). In return, the citizen scientist actively contributes to increasing the scientific understanding of microplastics, a topic which has received considerable public attention and many feel concerned about. Citizen scientists have participated in previous research on marine litter, but Thiel and Hidalgo-Ruz (2015) noted that in the current literature on marine litter, citizen science studies do not tend to focus on MPs. This is because advanced techniques are needed to adequately identify small MPs (Hidalgo-Ruz and Thiel 2013; Zettler et al. 2017). Therefore, the two studies in wich citizen scientists participated in the quantification of MP contamination had to use a lower size limit of 1 mm (Hidalgo-Ruz and Thiel 2013; Davis and Murphy 2015). In the current study, the citizen scientists followed a protocol to collect bulk sediment samples and then to send them to our laboratory. This allowed for smaller MPs to be properly identified and for the continent-wide, spatial distribution of MPs to be examined with increased accuracy. The aim of this study was first to quantify MP contamination of European beach sediment, allowing examination of MP distributions, and secondly to characterise MPs in terms of their physical properties and polymer type.

2. Methodology

2.1 Sampling, extraction and identification procedure

Sample collection – Five samples per beach were collected between June 2015 and January 2017. Beach sediment was collected from 23 different locations across 13 different countries (Table S1). Samples from Israel and Turkey were also included, because they adjoin the Mediterranean Sea, which is a specific area of interest due to possible trapping of MPs. Participation in sample collection for this study was volunteer-based, with recruiting predominantly via social media. Within Leiden University, participants were also recruited via personal emails. The participants were provided with 6 re-sealable plastic bags and a link to the sampling instructions. The only other materials needed to obtain the samples were a metal spoon and a smartphone to take a picture of the sampling location, and note the GPS coordinates. For details on the sample collection protocol see: www.lucmicroplastic.wordpress.com. Participants were first asked to look for the high tide line, described as the line of deposition, take a picture and note the GPS coordinates if possible. Five replicate samples were obtained from a 40 m stretch of beach at the high tide line. Every 10 m, approximated by 10 large steps, a zip-lock bag was filled with roughly 100 g of sand of the top 5 cm of the beach using the metal spoon.

Extraction – All samples were sent by mail or transported in person back to Leiden University for extraction. A standardised, density separation method of extraction was used to extract the MPs from the sediment (Besley et al. 2017). A total of 100 g of the sediment was weighed, put into a glass dish and dried for 48 hours at 60 °C. The dried sediment was sieved through a 5-mm sieve. Next, a 250 mL flask was filled with 50 g of dry sediment and 200 mL of a fully-saturated, filtered salt solution (358.9 g of NaCl in 1 L of demineralized water; water density of 9,043 kg/m3 at 20 ºC). Finally, it was sealed with Parafilm. If <50 g of sand was provided by the participants all of the available sediment was used, and the final abundance was adjusted accordingly. The mixture was then stirred at 900 RPM for 2 minutes, after which it was left to settle. After a minimum of 8 hours, approximately 75-100 mL of the supernatant was poured off the surface and filtered through a vacuum pump covered with 47 mm Millipore, 0.45 μm filter paper (Fisher scientific, the Netherlands). The filter paper was transferred to a covered petri dish to avoid contamination and left to dry at room temperature. This extraction process was repeated three times for each sample to increase the recovery rate (Besley et al. 2017).

Visual identification -- The filter papers were examined under a stereo-microscope (Motic Classmag 41, Motic, Germany); at up to 40x magnification and MPs counted. This process allowed for quantification of MPs in the range of 0.3 – 5 mm (NOAA 2015). This was done systematically by dividing the filter paper up into four quarts with the top clearly marked. The approximate location on the filter paper, the colour and shape (fibre, film or particle) were noted for all MPs. Colours were then grouped in the categories ‘blue/black’ and ‘red’, as these were the most abundant, with all other colours grouped within the category ‘other’. The visual identification was partially guided by a set of rules reported by Hidalgo-Ruz et al. (2012). They mention three important characteristics of MPs: i) there should be no cells or organic structures visible, ii) fibres should be equally thick throughout their entire length, and iii) they should exhibit clear and homogenous colour throughout. However, there are exceptions to these rules. For example, biofouling and bleaching can change the colour and apparent thickness of a fibre (Marine & Environmental Research Institute 2015). Therefore, the identification was additionally guided by a visual comparison to pictures of MPs from other publications (Leslie et al. 2013), and the observed colour (perceived as bright or unusual, as depicted in Dekiff et al. 2014).

For every sampling location, 10 MPs were selected randomly to measure the length of the MPs (DinoCapture software, version 2.0, Dino-Lite Europe, the Netherlands). The fibres were measured by tracing their length (mean length ± standard error [mm]). For particles and films, the largest cross-section was measured. Only in 2.6% of measurements did the fibre length exceed 5 mm (due to coiling it is difficult to visually ensure that fibres are below 5 mm); for transparency they were included in the analysis.

Contamination -- To avoid contamination, all equipment used during the extraction process was rinsed with distilled water before usage. All Petri dishes for storage of samples were wiped (Kimberly Clark cellulose wipe, Fisher Scientific, the Netherlands). During the extraction process, all equipment and vessels were covered when they were not in use. Additionally, the complete extraction process for one sampling location was repeated without beach sediment to quantify the procedural contamination. An analysis using a procedural blank was performed, finding an average of 3 MPs per 5 replicates, or less than one MP per replicate. The maximum level of procedural contamination among replicates was 4 MPs.

2.2 Polymer identification

A total of 221 MPs were analysed to determine their chemical composition. Raman spectroscopy was used to determine the chemical composition of the visually identified MPs (HR800UV, Jobin Yvon Horiba, Japan, with an integrated Olympus BX21 microscope, Japan). The method used here was similar to the method described by Horton et al. (2017). A near-infrared laser (785 nm) was used to obtain the spectra to achieve an optimum balance between high signal intensity and limited fluorescence (which can override the readable spectrum) (Löder and Gerdts 2015**.** Acquisition time was 40 s and accumulation was set at 2x, with the range set to acquire between

200 - 1800 cm^{-1}. For each item analysed, laser intensity was adjusted using an inbuilt filter, as dark-coloured items can be damaged by the laser.

The spectra were analysed using the Bio-Rad KnowItAll® Informatics System – Raman ID Expert (2015) software (Bio-Rad Laboratories, California, USA). The software matches the sample spectra to several potential spectra from a database of known compounds, and it ranks and rates these matches (for a more detailed description see Horton et al. 2017). Given a selection of possible matches, the most suitable match was selected based on peak position. The version of the software used provided limited spectrum editing capabilities, therefore most spectra were manipulated with the spectrum acquisition software LabSpec 6.0 (Horiba, Japan) before they were analysed with the BioRad KnowItAll® matching software. These manipulations consisted of baseline corrections and truncating the spectrum to eliminate noise that may interfere with the interpretation.

2.3 Data analysis

Classification of zones and subzones -- To examine large-scale trends, data was aggregated into zones, similar to Hidalgo-Ruz and Thiel (2013). In the study by Hidalgo-Ruz and Thiel (2013) zones were classified according to climate and water regime. Similarly, we classified our samples into 3 zones: Zone I covers all beaches bordering the Mediterranean; Zone II covers the beaches adjacent to the Atlantic Ocean and North Sea; and, Zone III those adjacent to the Baltic Sea (see Table S2 for the coastal attributes of these zones). These zones have different characteristics. For example, the Atlantic coast has the highest average wind speed, waves and annual precipitation, while the surface water temperature is highest along the Mediterranean coast, which is also most densely populated (Gazeau et al. 2004; Table S2). Furthermore, the Mediterranean Sea has been shown to contain particularly high concentrations of plastic due to its semi-enclosed structure and large plastic input (Cózar et al. 2015). The Baltic Sea is similarly semi-enclosed. The Mediterranean Sea is commonly divided into an eastern and western basin that are divided near the Tunisian and Sicilian coast (International Hydrographic Organization 1953). The hydrological characteristics of these basins can lead to different behaviours of plastic in the marine environment. In our study we also make a distinction between the eastern and western Mediterranean coasts. The Atlantic zone was similarly divided into the North Sea and Atlantic, the former of which is boreal whereas the Atlantic is warm-temperate (Dauvin 2008). The main European ports are situated in the southern North Sea and maritime traffic in the northern English Channel is the busiest in the world (Dauvin 2008). As a result, MP abundance will therefore be examined within 3 zones and 5 subzones.

Some locations are situated in transition regions between zones (one) and subzones (two). The Drøbak location is situated on the border of the North Sea and the Baltic Sea, near the Skagerrak strait. We follow Gazeau et al. (2004) who considered Skagerrak to be a part of the Atlantic zone. Two sample locations from Normandy were included in the North Sea subzone, as they are also partially closed from the Atlantic current. A map showing the level of MP contamination was made using ArcGIS (version 10.2) (Figure 1).

Statistical analysis – MP concentrations for sampling locations were reported as mean ± SEM of the 5 replicates expressed in MPs per kg of dry weight sediment. We conducted an analysis of variance (ANOVA) (using R version 0.98) on the 23 sampling locations (with 5 replicate samples per location) with significance set at $\alpha < 0.05$. A nested ANOVA with the same significance level was performed on the aforementioned zones and subzones. The data was checked for normality and homogeneity of variance using Shapiro-Wilk’s W-test and Levene’s test respectively. Although ANOVAs are robust for the violation of these assumptions, if they are violated, results need to be interpreted with caution when p-values are close to α, which was noted in the results section where applicable. If significant differences were observed, a Tukey's post-hoc test was conducted.

3. Results

3.1 Microplastics abundance

The distribution of sampling locations and their relative contamination were shown in Figure 1, with Table 1 reporting the average abundance of MPs per sampling location. The average abundance ranged from 72 ± 24 MPs kg^{-1} d.w. in Tromsø, Norway, to 1512 ± 187 MPs kg^{-1} d.w. in Lido di Dante, Italy. The majority of locations had abundances below 248 MPs kg^{-1} d.w. (Figure 1). Zone I and III, the Mediterranean zone and the Baltic zone, were on average the most polluted sites with means of 291 and 270 MPs kg^{-1} d.w., respectively (see Table 2 for more details). The Atlantic zone was the least polluted with a mean of 190 MPs kg^{-1} d.w. These differences were not statistically significant (nested ANOVA, $F_{2,20} = 0.21$, $p = 0.809$).

Within Zone I, the western Mediterranean subzone was found to be less contaminated than the eastern subzone, showing average abundances of 147 and 387 MPs kg^{-1} d.w., respectively (Table 2). The levels of microplastics in the western subzone were relatively low and homogenously distributed. In the eastern subzone, the sample locations in Greece and Turkey showed relatively high levels of contamination (Table 1 and 2). Within Zone II, the North Sea and Atlantic Ocean had respective average abundances of 131 and 238 MPs kg^{-1} d.w. respectively. These differences were not statistically significant (nested ANOVA, $F_{4,18} = 0.44$, $p = 0.778$). However, within Figure 1 it was shown that mainland Europe gave comparable levels of moderate contamination, whereas other locations in the Atlantic zone showed low contamination. The location in Iceland was an exception to this.

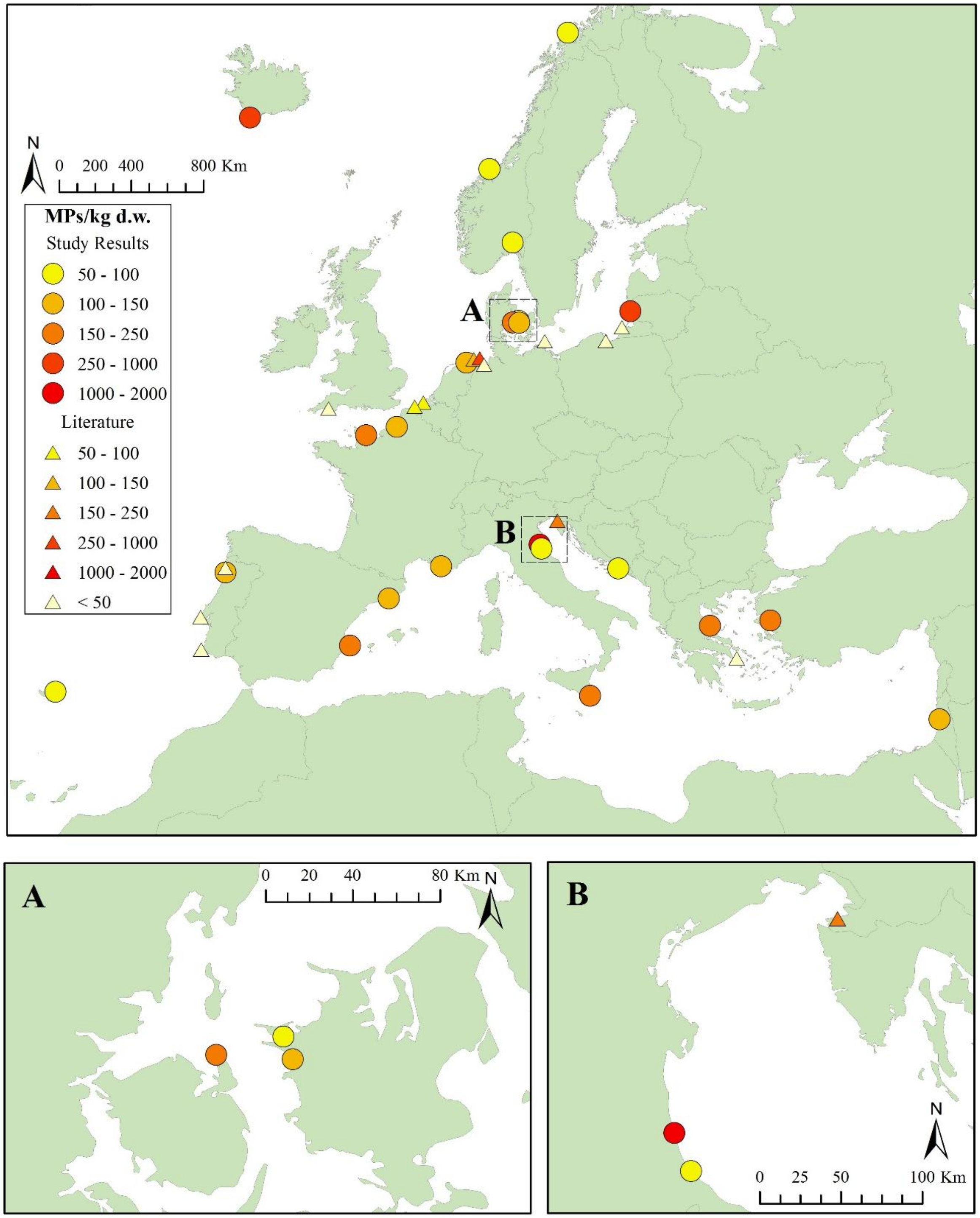

Figure 1. A map showing the contamination levels across Europe [O: locations from current study; Δ: data obtained from literature (Table S3)]. Contamination is reported in number of microplastics per kg of dry sediment. (A) Map of sampling locations in Denmark. (B) Map of sampling locations in Italy, Adriatic coast.

Table 1. Abundance, length, and colour are presented per location. Abundance is expressed as the average number of plastics from 5 replicates per kg of dry sediment (± SEM). The statistical significance is indicated. Length is based on a sample of n = 10 per beach and is expressed in mm. Error margins are expressed in standard error. Colours are expressed as a percentage of the total count.

	Group		Abundance		Length		Colour (%)[b]		
Location	Zone	Subzone[a]	(MPs/kg d.w.)		(mm)		Blue/black	Red	Other
Sicily, IT	I	W	160 ± 31	c	1,32 ± 0,30	a	70,0	20,0	10,0
Denia, ES	I	W	156 ± 29	c	1,96 ± 0,71	a	79,5	12,8	7,7
Barcelona, ES	I	W	148 ± 23	c	1,13 ± 0,36	a	81,1	8,1	10,8
Cassis, FR	I	W	124 ± 36	c	1,28 ± 0,32	a	87,1	9,7	3,2
Lido di Dante, IT	I	E	1512 ± 187	a	1,38 ± 0,37	a	72.0 *	11,2 *	16,8 *
Dikili, TR	I	E	248 ± 47	c	1,01 ± 0,17	a	62,9	14,5	22,6
Pilion, GR	I	E	232 ± 93	c	0,93 ± 0,48	a	77,6	10,3	12,1
Tel Aviv, IL	I	E	168 ± 16	c	0,94 ± 0,31	a	81,0	9,5	9,5
San Mauro, IT	I	E	84 ± 12	c	1,42 ± 0,58	a	90,5	9,5	0
Bosnia	I	E	76 ± 13	c	1,54 ± 0,33	a	73,7	26,3	0
Vik, IS	II	A	792 ± 128	b	1,80 ± 0,33	a	84,8	8,1	7,1
Porto, PT	II	A	140 ± 26	c	1,34 ± 0,32	a	74,3	8,6	17,1
Smøla, NO	II	A	92 ± 21	c	0,96 ± 0,24	a	78,3	8,7	13,0
Madeira, PT	II	A	92 ± 15	c	1,98 ± 0,73	a	91,3	4,3	4,3
Tromsø, NO	II	A	72 ± 24	c	1,60 ± 0,48	a	77,8	16,7	5,6
Normandy, FR	II	NS	156 ± 29	c	0,91 ± 0,28	a	92,3	5,1	2,6
Normandy, FR	II	NS	143 ± 13	c	1,36 ± 0,42	a	78,8	12,1	9,1
Rottumeroog, NL	II	NS	124 ± 27	c	1,28 ± 0,54	a	80,6	16,1	3,2
Drøbak, NO	II	NS	100 ± 21	c	1,50 ± 0,36	a	80,0	12,0	8,0
Klaipéda, LT	III	B	700 ± 296	b	1,42 ± 0,29	a	75.0 *	14,4 *	10,6 *
Fyns Hoved, DK	III	B	164 ± 21	c	1,26 ± 0,44	a	82,9	9,8	7,3
Bjergje Nord, DK	III	B	128 ± 31	c	1,34 ± 0,44	a	84,4	12,5	3,1
Kalundburg, DK	III	B	88 ± 33	c	1,55 ± 0,45	a	81,8	13,6	4,5

[a] E = Mediterranean-East, W = Mediterranean-West, A = Atlantic Ocean, NS = North Sea and B = Baltic Sea.

[b] and * indicates a subsample was taken due to high MP abundance.

Table 2. A summary of the mean abundance (± SEM), mean length (± SEM), and colour per zone and subzone (see Table 1). No significant differences were found between locations.

Zone/Subzone	Abundance (#/kg d.w.)	Length (mm)	Colour (%)		
			Blue/black	Red	Other
I: Mediterranean	291 ± 62	1.29 ± 0.13	77.5	13.2	9.3
West	147 ± 14	1.43 ± 0.22	79.4	12.7	7.9
East	387 ± 100	1.20 ± 0.16	76.3	13.6	10.2
II: Atlantic	190 ± 35	1.41 ± 0.14	82.0	10.2	7.8
North Sea	131 ± 12	1.26 ± 0.20	82.9	11.3	5.7
Atlantic	238 ± 62	1.54 ± 0.20	81.3	9.3	9.4
III: Baltic	270 ± 90	1.39 ± 0.20	81.0	12.6	6.4

Individual sampling locations across all zones showed significantly different MP abundances (ANOVA, $F_{22,92} = 15.58$, $p < 0.001$). Lido di Dante, Italy, was the most polluted site. With a mean abundance of 1512 MPs kg^{-1} d.w., it was significantly more polluted than all other sites (Table 1). The concentrations found for Vik, Iceland, and Klaipéda, Lithuania, were also significantly different from the other locations with means of 792 and 700 MPs kg^{-1} d.w., respectively.

3.2 Microplastics characterization

Physical characteristics – The majority of MPs identified in this study were fibrous (98.7 %). Other types of MPs found were films (5 items, 0.35 %) and particles (13 items, 0.91 %). Only one particle was identified as a potential primary MP because of its spherical shape (Figure S1a). Other particles were more angular and irregularly shaped (Figure S1b), suggesting they resulted from breakdown of larger plastics. As a proportion of MPs, blue/black MPs were 77.5-82.9%, red MPs was 9.3-13.6% (Table 1). Other colours that were identified were green, orange, purple, grey, white, and multi-coloured (photographic examples fibres identified were shown in Figure S1c-g). The average length of the MPs ranged from 0.91 mm in Normandy to 1.97 mm in Madeira (Table 1). These results were not statistically significant (ANOVA, $F_{22,207} = 0.51$, $p = 0.967$). Among different zones, the average length ranged from 1.26-1.54 mm (Table 2). Zones and subzones showed no statistically significant differences (nested ANOVA, $F_{sub,\ 2,20} = 0.22$, $p = 0.719$, $F_{zone,\ 4,18} = 0.52$, $p = 0.801$). The majority of the MPs measured (54.8%) were < 1 mm in size. The distribution of MPs within size categories was shown in Figure 2, and follows an exponentially decreasing number of MPs with increasing size.

Chemical composition -- Of the 221 visually confirmed MPs analysed using Raman spectrometry, 92 (42%) did not have discernible peaks in their spectra, even after several trails.

Of the remaining 129 visually confirmed MPs, only 10 (4.5%) were matched to a specific polymer type. The three types of polymer that were identified are polyester (7 items), polypropylene (2 items) and polyethylene (1 item). Additionally, 10 MPs were matched to several types of dyes, such as mortoperm blue (3 items), hostaperm blue (2 items) and neozapon blue FLA (2 items). The remaining 3 fibres were matched to Drimaren navy blue, Drimaren brilliant green, and cobalt phthalocyanine. Mortoperm blue, hostaperm blue, neozapon blue, and cobalt phthalocyanine are all phthalocyanine dyes. Several times a reoccurring spectrum was noticed that did not match any compounds from the database. Additionally, two fibres were matched to the dye Indigo. These fibres were part of a group of 29 fibres which were visually grouped together based on peak position.

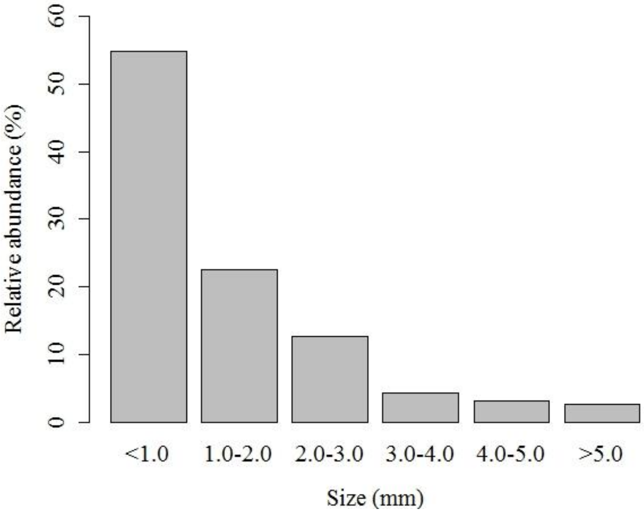

Figure 2. The distribution of microplastics (%) in different size fractions based on a subsample of n = 10 per sampling location. Size classification adapted from Laglbauer et al. (2014).

4. Discussion

Here we present data from a large-scale MP investigation using citizen science and robust lab techniques. Our findings were summarised into three main themes: the MP abundance and spatial distribution across Europe; characterization of MP types; and, efficacy of citizen science as a tool for MP research.

4.1 Microplastics abundance and spatial distribution

Using a standardised sampling and extraction protocol, our results confirmed that MP pollution on European beaches is ubiquitous. All 23 sampling locations in the current study were found to have substantial levels of MP contamination. Our results suggested that the Mediterranean zone, and particularly the eastern subzone is the most contaminated, showing the highest average abundance of MPs. This could be due to the partial geographic trapping of MPs, combined with high coastal population density and waste input (Table S2).

Within the Baltic Sea, one sampling location in Lithuania showed much higher MP abundances than three other sites within the same zone in Denmark (Figure 1). This location, in Klaipéda, is at the outlet of the freshwater Curonian Lagoon, into which several rivers flow creating a unidirectional flow (Christian et al. 2008). The lagoon has high concentrations of agricultural and industrial pollution (Christian et al. 2008). Previous research on MP contamination in lagoons showed varied results. For example, a study in Italy found high levels of MP contamination, which was attributed to significant freshwater inputs and the low-energy environment (Vianello et al., 2013). In contrast, three studies conducted in and around the Vistula Lagoon bordering Poland and Russia found low concentrations of MPs, ranging from 1-39 MPs kg^{-1} d.w. (Table 3). Although Klaipedá is located close to this area, it has an average abundance roughly 30 times greater.

Table 3. An overview of studies examining MP contamination in marine sediment in Europe. The location, sampling location, size definition of microplastics, along with abundance in microplastics per kg of dry weight are noted. Abundances in italics have been converted[a]. Zones are as follows: I Mediterranean, II Atlantic, and III Baltic. Table S2 gives further climatic and demographic details of these regions.

Reference	Zone	Country	Sampling location	Size definition	Abundance (#/kg d.w.)
Alomar et al. (2016)	I	Spain	Subtidal	< 5 mm	100.78-897.35
Baztan et al. (2014)	II	Canary Islands (Spain)	Beach	< 5 mm	109, 90 and 30[b]
Blašković et al. (2017)	I	Croatia	Subtidal	≤ 5 mm	32.3-377.8
Claessens et al. (2011)	II	Belgium	Harbour	< 1 mm	166,7
			Subtidal		97,2
			Beach		92,8
Dekiff et al. (2014)	II	Germany	Beach	< 1 mm	23-213 fibers
					4-25 coloured fibers
					0-4 particles
Esiukova (2017)	III	Russia	Beach	< 5 mm	1.3-36.3
Faure et al. (2015)	-	Switzerland	Beach	< 5 mm	*0.3-90*
Fischer et al. (2016)	-	Italy	Beach	< 5 mm	112 and 234
Frère et al. (2017)	II	France	Subtidal	< 5 mm	1
Graca et al. (2017)	III	Poland	Subtidal	≤ 5 mm	15
			Beach		39
Kaberi et al. (2013)	I	Greece	Beach	< 4 mm	*1.5-15.7 (1-2 mm)*
					0.3-15.0 (2-4 mm)
Laglbauer et al. (2014)	I	Slovenia	Shoreline	≤ 5 mm	177,8
			Infralittoral		170,4
Leslie et al. (2017)	II	The Netherlands	Subtidal	< 5 mm	100-3600
Liebezeit and Dubaish (2012)	II	Germany	Beach	< 5 mm	461 fibers
					210 granules
Martins and Sobral (2011)	II	Portugal	Beach	< 5 mm	*0.7-11*
Norén (2007)	II	Sweden	Subtidal	N/D	*16-2590*
Popa et al. (2014)	-	Romania	Beach	N/D	*1000-5500*
Stolte et al. (2015)	III	Germany	Beach	< 2 mm	2-11 fibers
					0-7 particles
Strand and Tairova (2016)	II	Denmark	Subtidal	≤ 5 mm	192-675
Thompson (2004)	II	United Kingdom	Beach	< 5 mm	*8*
			Estuarine		*31*
			Subtidal		*86*
Vianello et al. (2013)	I	Italy	Subtidal	< 1 mm	672-2175
Zobkov and Esiukova (2017)	III	Russia	Subtidal	< 5 mm	34

[a] To increase the comparability of these studies, the units were converted to MPs kg^{-1} of dry weight (d.w.) where possible. An average sediment density of 1600 kg m^{-3} was used as per Claessens et al. (2011) and Ballent et al. (2016) to convert units of volume or area to kg. The latter could only be done if the sampling depth was reported. An average dry/wet ratio of 1.25 was used (Van Cauwenberghe et al. 2015). If the weight of the MPs was reported rather than a count, the unit was not converted.

[b] Reported in g/L

In the Mediterranean zone, we found that western coasts are less prone to MP accumulations, although this result was not statistically significant. This is in agreement with a recent study, which modelled the effects of circulation on plastic accumulation in the Mediterranean, finding that the accumulation on coastlines in the western basin was considerably lower (Mansui et al. 2015). The accumulation in the eastern basin could indicate that currents and water circulation play an important role in the distribution of MP abundance in the Mediterranean. Other studies conducted in the Balearic Islands, Croatia, and Slovenia found MP concentrations on the same scale as the results reported here (Table 3). In this study, we found high abundances in Greece, which contrasts with the lower abundances found in a previous study (Kaberi et al. 2013). However, in Kaberi et al. (2013), MPs smaller than 1 mm were not counted, which in our study accounted for the majority of MPs (Figure 2). The high concentration found in the Lagoon of Venice is likely caused by the urban estuarine environment, as discussed above. The highest MP abundance was found in the small coastal village Lido di Dante, Italy, situated between the mouths of two rivers. This contrasts with results from San Mauro nearby, which was among the least polluted sites. This highlights the importance of small-scale factors such as river mouths (Rech et al. 2014), waste water treatment plants, and densely populated zones adjoining rivers (Mani et al. 2016). Several of the reviewed studies have attributed high MP concentrations to river discharge (Claessens et al. 2011; Faure et al. 2015), although this may not be the case in all circumstances (Clunies-Ross et al. 2016).

The high population density along the Mediterranean coast (Gazeau et al. 2004; Table S2) did not result in significant higher levels of microplastics. Population density has been shown to be positively correlated with MPs abundance, suggesting that the spatial distribution of MPs is influenced primarily by source proximity (Browne et al. 2011). However, Nel and

Froneman (2015) did not find this correlation and identified water circulation as a dominating mechanism.

The Atlantic zone showed the lowest average MP abundance. Relatively low concentrations were found off the continental mainland. The levels we detected in Belgium and Germany were comparable to previous studies (Table 3). Interestingly, Iceland's southernmost village, Vik, is located in a rural setting, yet MP concentrations were significantly higher than other locations. The comparatively low anthropogenic activity in this area could indicate that the MPs originated from the North Atlantic Current. Recent studies have shown accumulation of plastics in the North Atlantic branch of the thermohaline circulation (Cózar et al. 2017).

4.2 Microplastics characterization

Overall, MPs identified in this study were predominantly blue/black or red fibres. Several studies similarly found that blue/black and red are the most common fibres (Nel and Froneman 2015; Alomar et al. 2016; Strand and Tairova 2016; Frère et al. 2017). The high proportion of fibrous MPs reported in our study was comparable to other studies (Thompson 2004; Claessens et al. 2011; Dekiff et al. 2014; Alomar et al. 2016; Graca et al. 2017; Zobkov and Esiukova 2017). Some studies find that over 90% of MPs are fibrous, which is similar to the scale found here (Laglbauer et al. 2014; Strand and Tairova 2016; Blašković et al. 2017). Microfibres generally derive from the machine washing of synthetic fabrics (Browne et al. 2011; Hernandez et al. 2017). Up to 700 000 fibres can be released per standard wash load (Napper and Thompson 2016). They are introduced to the aquatic environment via wastewater (Murphy et al. 2016). With wastewater believed to be a likely origin of many of these fibres, the finding of these fibres on marine beaches highlights the potential for widespread distribution of MPs once within the environment. Fibres can also enter the marine environment through the fragmentation of fishing ropes and nets (Thompson 2004), which is may account for 18% of marine debris, and is commonly made of PE, PP, and nylons (Andrady 2011). Only one particle was a potential primary MP based on the spherical shape; low quantities of primary MPs were also commonly reported in other studies (Laglbauer et al. 2014; Graca et al. 2017; Zobkov and Esiukova 2017).

In the current study we used Raman spectrometry as a secondary method of MP characterization. This resulted in a 4.5% success rate in matching a MP to a specific polymer and a 4.5% success rate in matching to dyes. This detection rate was comparable to other studies. For example, Horton et al. (2017) had a polymer identification rate of 8.3%, while Frère et al. 2017 had a success rate of 13%. Other studies examining MP pollution in beach sediment have found higher confirmation rates (e.g. Ballent et al. 2016; Clunies-Ross et al. 2016). There are many factors that likely contributed to the low success rate. A common problem in Raman spectroscopy is fluorescence, when strong light intensities are emitted, obscuring relevant peaks (Bart 2006). This is usually the result of biological material from the environment on the MP surface, but it may also be the result of plasticisers and additives (Purcell and Bello 1990; Löder and Gerdts 2015). In this study, fluorescence was an important cause of poor quality spectra. Additionally, additives such as dyes and pigments can mask the spectrum so that it does not match directly to a polymer type in the reference library (Lenz et al. 2015).

For the fraction of fibres that we could identify with the Raman spectrometry we distinguished three types of polymers: polyethylene (PE), polypropylene (PP), and polyester (PEST). Studies in Portugal, Germany, Italy, Greece, Switzerland, and France all found PE and PP the most common polymer types (Martins and Sobral 2011; Kaberi et al. 2013; Vianello et al. 2013; Faure et al. 2015; Frère et al. 2017). In addition, several visually identified MPs were matched to dyes, which was also comparable to other studies (Horton et al. 2017; Imhof et al.

2016). Given that the response signals of polymers are easily masked by dyes and that in the environment they usually occur as a composite, it is reasonable to assume that particles identified as dyes will usually be polymers (Horton et al. 2017). Some studies have used dyes as an indicator of plastic. In this study, several suspected MPs were matched to dyes that have been found in other MP studies, such as phthalocyanine dyes which are commonly used as plastic additives. These particles were thus inferred to be MPs, except for Drimaren navy blue, an azo dye which is commonly used to dye both plastic and non-plastic fibres (Lenz et al. 2015). The Indigio dye is commonly used to dye cellulosic fibres used in fabric for blue jeans (Wiesheu et al. 2016). The dye may therefore not relate to MPs but to cotton. This indicates that although many dyes can be related to polymers, there is some uncertainty surrounding others.

4.3 Citizen Science

The incorporation of citizen science in MP research is often challenging because of difficulties with collecting, sorting, and distinguishing plastics from other marine debris and materials (Zettler et al. 2017). Here we demonstrated that by providing simple instructions that only pertain to the collection of samples, these problems can be successfully avoided. Nevertheless, citizen science does result in limited accompanying field observations, information on which may have helped explain some of the high MP abundances found in the current study. Important factors which could result in higher MP loads include space available for deposition (Baztan et al. 2014), human activity (Ng and Obbard 2006; Yu et al. 2016), and weather evens such as storms or heavy winds (Graca et al. 2017). We therefore suggest future studies and participating citizen scientists to make note of such factors.

5. Conclusions

This study found that MPs, particularly secondary, blue fibres, are ubiquitous within European beach sediments. The abundance of MPs differs geographically, with locations in the Eastern Mediterranean and on Iceland showing particularly high concentrations. By using citizen science we were able to examine the large-scale distribution of MP contamination in European beach sediment, thereby taking an important step in providing a coherent overview of the nature and extent of MP contamination in Europe beach sediments.

Competing interests

The authors declare that they have no competing interests.

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Supplementary Information:

Table S1 GPS Coordinates of each sampling location.

Table S2. Characteristics of three European coastal zones. Adapted from Gazeau et al. (2004).

Table S3. Abundance of microplastics in beach sediment used in Figure 1 based on available literature.

Figure S1 Pictures of a spherical particle (a), a yellow particle (b), a red fibre (c), blue fibres (d, e), a multi-coloured fibre (f) and a purple fibre (g).

Table S1 GPS Coordinates of each sampling location.

			Given		Estimated	
Beach ID	Region	Country	Latitude	Longitude	Latitude	Longitude
005_16		Bosnia			42.92	17.62
001_16	Kalundburg	Denmark			55.69	11.09
002_16	Fyns Hoved	Denmark			55.61	10.59
003_16	Bjerge Nord	Denmark			55.59	11.15
009_15	Klaipéda	Lithuania			55.70	21.14
002_15	Normandy	France	49.38	-0.89		
022_15	Pilion	Greece			39.44	23.05
017_15	Vik	Iceland	63.26	-19.00		
008_16	Tel Aviv	Israel			32.11	34.86
024_15	Sicily	Italy	36.76	15.10		
032_15	Lido di Dante	Italy	44.38	12.32		
012_15	Tromsø	Norway	69.78	18.54		
016_15	Smøla	Norway	63.29	8.14		
026_15	Drøbak	Norway	59.64	10.64		
007_15	Porto	Portugal	41.18	-8.69		
020_15	Madeira	Portugal	33.05	-16.34		
021_15	Barcelona	Spain	41.40	2.21		
029_15	Denia	Spain			38.84	0.11
018_15	Rottumeroog	The Netherlands	53.54	6.61		
027_15	Dikili	Turkey			39.07	26.89
006_16	Cassis	France	43.21	5.54 *		
002_17	Normandy	France	50.00	1.39 *		
001_17	San Mauro	Italy	44.17	12.44		

* These values were converted from degrees to meters using an online converter (https://www.fcc.gov/media/radio/dms-decimal) and subsequently checked with google maps.

Table S2. Characteristics of three European coastal zones. Adapted from Gazeau et al. (2004).

	Coast		
Characteristic	Baltic	Mediterranean	Atlantic
Median wind speed (m/s)	7,4	6,5	8,3
Median precipitation (mm/yr)	720	679	1022
Median of monthly averaged surface water temperature (°C)	7,7	19,5	10,6
Median wave height (m)	2.5-3.5	2.5-3.5	3.5-4.5
Coastal population density (inhabitants per km^2)	13,1	133	19,4

Table S3. Abundance of microplastics in beach sediment used in Figure 1 based on available literature.

Reference	Location	Abundance	
Claessens et al. (2011)	Locations estimated from Fig. 1. KB and GZ visualised together.	Average value for all beaches was used for both locations (KB/GZ and KZ)	92.8
Dekiff et al. (2014)	Coordinates of middle location obtained from paper	Sum of fibres, coloured fibres and particles. Average of particles was taken across 3 sampling locations	131.8
Esiukova (2017)	A general location (near Kaliningrad) was estimated from Fig. 1.	Average was calculated from Table 2 abundances.	9.2
Graca et al. (2017)	Middle location of 3 beach sampling locations was estimated from Fig. 1	Taken from paper	39.0
Kaberi et al. (2013)	Due to its small size, coordinates of centre of island were estimated	Average for all locations in both size categories was taken and these two averages were added	15.8
Laglbauer et al. (2014)	Coordinates from Izola were estimated, in the middle of the sampling area (Slovenian coast)	Average of coast and infralittoral reported overall averages was taken	174.1
Liebezeit and Dubaish (2012)	Average coordinates for both islands were estimated from Fig. 1	Reported average fibre and particle abundance was added; one number for both islands	671
Martins and Sobral (2011)	Coordinates were obtained from paper. Fonte and Cova were grouped together using Cresmina's coordinates.	Abundances were taken from Fig. 3. Average was taken for Cresmina, Fonte and Cova.	0.7; 2.6;7.5
Stolte et al. (2015)	Beege's coordinates were estimated as reference location for all sampling locations in theMecklenburg-Vorpommern province. Paper's coodinates for Dangast were used for remaining locations.	Averages were calculated from Appendix 1.A values	7.0; 6.4
Thompson (2004)	General coordinates of Plymouth were estimated	Taken from paper	8.0

Comments	Only sampling locations on beaches were taken into account (Table 1). If coordinates were not provided by the paper, the sampling locations were estimated from provided figures and city names. This was deemed sufficient due to the large scale of the map.

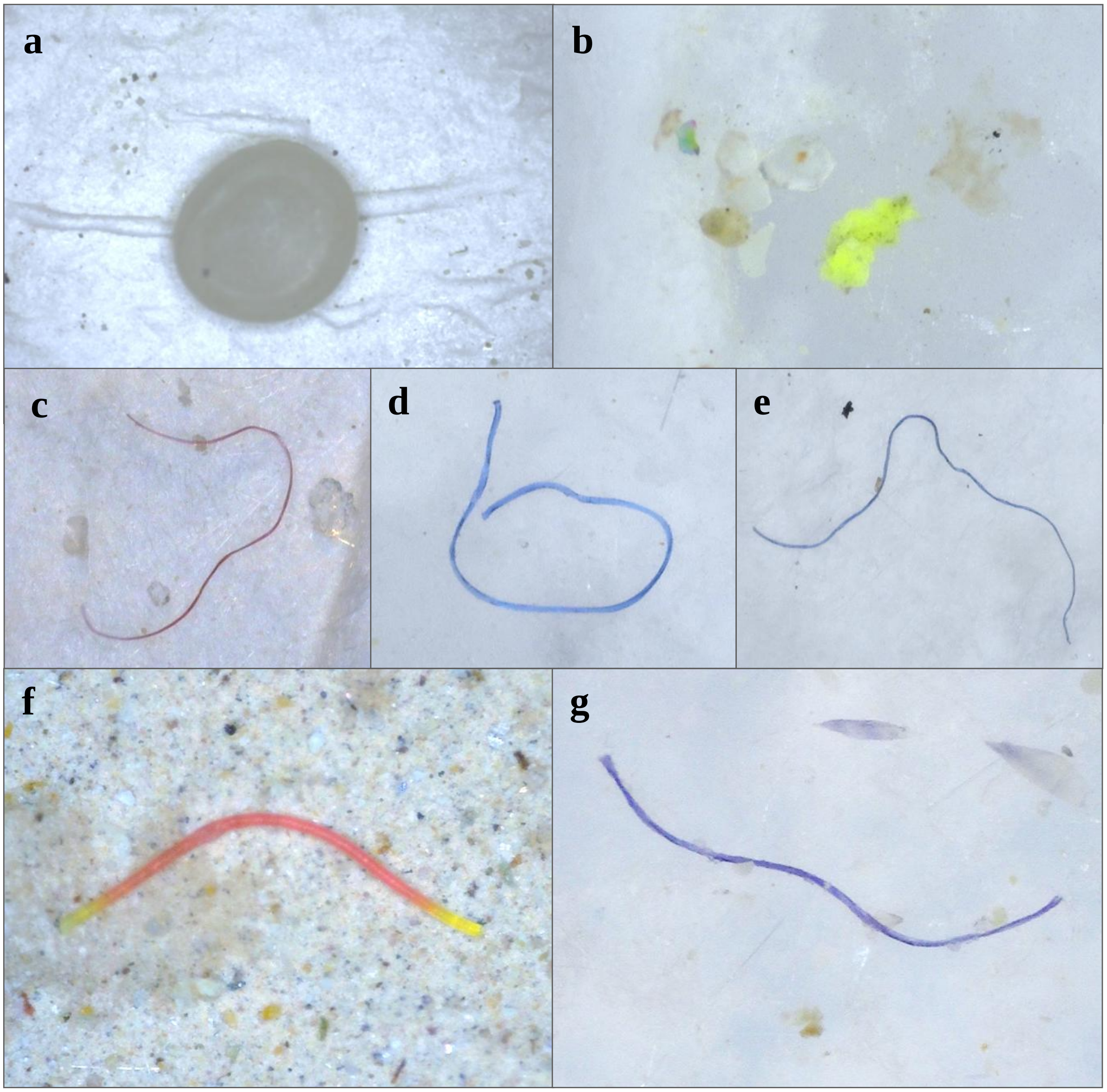

Figure S1 Pictures of a spherical particle (a), a yellow particle (b), a red fibre (c), blue fibres (d, e), a multi-coloured fibre (f) and a purple fibre (g).