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4 **A LARGE-SCALE INVESTIGATION OF MICROPLASTIC CONTAMINATION:**  
5 **ABUNDANCE AND CHARACTERISTICS OF MICROPLASTICS IN EUROPEAN**  
6 **BEACH SEDIMENT**  
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2 **Abstract**

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

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

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15 **1. Introduction**

16 Since the first commercial manufacture of plastics in the 1940s, plastic production and  
17 consumption have increased rapidly (Cole et al. 2011), with approximately 322 million tonnes  
18 (Mt) of plastic produced in 2015 (PlasticsEurope 2016). Approximately 5 to 13 Mt of plastic  
19 waste entered the ocean in 2010 (Jambeck et al. 2015), where it will persist and accumulate  
20 (Barnes et al. 2009). One subgroup of plastic that has raised particular concern are microplastics  
21 (MPs), commonly defined as pieces of plastic smaller than 5 mm (Thompson 2004; Arthur et al.  
22 2009; Cole et al. 2011). MPs are now ubiquitous in the marine environment (Eriksen et al. 2014):  
23 their presence has been recorded near densely-populated areas, remote regions, and in different  
24 types of marine environments, such as beaches (e.g. Besley et al. 2017), estuaries (e.g. Leslie et  
25 al. 2013), surface water (e.g. Lusher et al. 2015) and deep sea sediment (e.g. Van Cauwenberghe  
26 et al. 2015).

27 A distinction is commonly made between primary and secondary MPs. Primary MPs are  
28 manufactured to be of microscopic size and are often purposefully added to products (Derraik  
29 2002; Napper et al. 2015) or can be used as raw material in industry. These MPs likely enter the  
30 environment via wastewater treatment plants and industrial drainage systems (Derraik 2002;  
31 Napper et al. 2015). Secondary MPs are the result of the gradual weathering or abrasion of larger  
32 plastics, mainly through prolonged exposure to solar UV radiation resulting in photo-  
33 degradation, or mechanical abrasion (Barnes et al. 2009; Andrady 2011; GESAMP 2015).  
34 Weathering is particularly evident on beaches, where temperatures and oxygen concentrations  
35 are higher than in water (Andrady 2011; GESAMP 2015).

36 As fragmentation and weathering decreases the size of plastics, their potential to be  
37 ingested by marine biota increases (Browne et al. 2008). The bioavailability of MPs in the

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38 marine environment has been demonstrated in different studies. MPs have been found in mussels  
39 (Santana et al. 2016), demersal and pelagic fish species (Bellas et al. 2016; Rummel et al. 2016),  
40 worms and seabirds (Cole et al. 2013). The direct effects of MP ingestion include reduced  
41 feeding, blocking of the intestinal tract leading to starvation and impaired bodily functioning,  
42 and translocation to the circulatory system (Browne et al. 2008; Cole et al. 2013; Wright et al.  
43 2013). Furthermore, a limited number of studies have demonstrated the trophic transfer of  
44 MPs have raised concerns about MPs and their possible negative impact on the health of marine  
45 food webs and humans (Farrell and Nelson 2013; Setälä et al. 2014; Van Cauwenberghe and  
46 Janssen 2014; Rochman et al. 2015).

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

59 Besley et al. (2017) investigated the major sources of variation in sampling and extraction  
60 procedures. The main source of variation resulted from the extraction procedure, and not the

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

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83 **2. Methodology**

84 **2.1 Sampling, extraction and identification procedure**

85 *Sample collection* – Five samples per beach were collected between June 2015 and January  
86 2017. Beach sediment was collected from 23 different locations across 13 different countries  
87 (Table S1). Samples from Israel and Turkey were also included, because they adjoin the  
88 Mediterranean Sea, which is a specific area of interest due to possible trapping of MPs.  
89 Participation in sample collection for this study was volunteer-based, with recruiting  
90 predominantly via social media. Within Leiden University, participants were also recruited via  
91 personal emails. The participants were provided with 6 re-sealable plastic bags and a link to the  
92 sampling instructions. The only other materials needed to obtain the samples were a metal spoon  
93 and a smartphone to take a picture of the sampling location, and note the GPS coordinates. For  
94 details on the sample collection protocol see: [www.lucmicroplastic.wordpress.com](http://www.lucmicroplastic.wordpress.com). Participants  
95 were first asked to look for the high tide line, described as the line of deposition, take a picture  
96 and note the GPS coordinates if possible. Five replicate samples were obtained from a 40 m  
97 stretch of beach at the high tide line. Every 10 m, approximated by 10 large steps, a zip-lock bag  
98 was filled with roughly 100 g of sand of the top 5 cm of the beach using the metal spoon.

99 *Extraction* – All samples were sent by mail or transported in person back to Leiden University  
100 for extraction. A standardised, density separation method of extraction was used to extract the  
101 MPs from the sediment (Besley et al. 2017). A total of 100 g of the sediment was weighed, put  
102 into a glass dish and dried for 48 hours at 60 °C. The dried sediment was sieved through a 5-mm  
103 sieve. Next, a 250 mL flask was filled with 50 g of dry sediment and 200 mL of a fully-saturated,  
104 filtered salt solution (358.9 g of NaCl in 1 L of demineralized water; water density of 9,043  
105 kg/m<sup>3</sup> at 20 °C). Finally, it was sealed with Parafilm. If <50 g of sand was provided by the

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106 participants all of the available sediment was used, and the final abundance was adjusted  
107 accordingly. The mixture was then stirred at 900 RPM for 2 minutes, after which it was left to  
108 settle. After a minimum of 8 hours, approximately 75-100 mL of the supernatant was poured off  
109 the surface and filtered through a vacuum pump covered with 47 mm Millipore, 0.45 µm filter  
110 paper (Fisher scientific, the Netherlands). The filter paper was transferred to a covered petri dish  
111 to avoid contamination and left to dry at room temperature. This extraction process was repeated  
112 three times for each sample to increase the recovery rate (Besley et al. 2017).

113 *Visual identification* -- The filter papers were examined under a stereo-microscope (Motic  
114 Classmag 41, Motic, Germany); at up to 40x magnification and MPs counted. This process  
115 allowed for quantification of MPs in the range of 0.3 – 5 mm (NOAA 2015). This was done  
116 systematically by dividing the filter paper up into four quadrants with the top clearly marked. The  
117 approximate location on the filter paper, the colour and shape (fibre, film or particle) were noted  
118 for all MPs. Colours were then grouped in the categories ‘blue/black’ and ‘red’, as these were the  
119 most abundant, with all other colours grouped within the category ‘other’. The visual  
120 identification was partially guided by a set of rules reported by Hidalgo-Ruz et al. (2012). They  
121 mention three important characteristics of MPs: i) there should be no cells or organic structures  
122 visible, ii) fibres should be equally thick throughout their entire length, and iii) they should  
123 exhibit clear and homogenous colour throughout. However, there are exceptions to these rules.  
124 For example, biofouling and bleaching can change the colour and apparent thickness of a fibre  
125 (Marine & Environmental Research Institute 2015). Therefore, the identification was  
126 additionally guided by a visual comparison to pictures of MPs from other publications (Leslie et  
127 al. 2013), and the observed colour (perceived as bright or unusual, as depicted in Dekiff et al.  
128 2014).



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

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

143 **2.2 Polymer identification**

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

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4 151 200 - 1800  $\text{cm}^{-1}$ . For each item analysed, laser intensity was adjusted using an inbuilt filter, as  
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7 152 dark-coloured items can be damaged by the laser.  
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10 153 The spectra were analysed using the Bio-Rad KnowItAll® Informatics System – Raman  
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12 154 ID Expert (2015) software (Bio-Rad Laboratories, California, USA). The software matches the  
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15 155 sample spectra to several potential spectra from a database of known compounds, and it ranks  
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17 156 and rates these matches (for a more detailed description see Horton et al. 2017). Given a  
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20 157 selection of possible matches, the most suitable match was selected based on peak position. The  
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22 158 version of the software used provided limited spectrum editing capabilities, therefore most  
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24 159 spectra were manipulated with the spectrum acquisition software LabSpec 6.0 (Horiba, Japan)  
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27 160 before they were analysed with the BioRad KnowItAll® matching software. These  
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30 161 manipulations consisted of baseline corrections and truncating the spectrum to eliminate noise  
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32 162 that may interfere with the interpretation.  
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### 35 163 **2.3 Data analysis**

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37 164 *Classification of zones and subzones* -- To examine large-scale trends, data was aggregated into  
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40 165 zones, similar to Hidalgo-Ruz and Thiel (2013). In the study by Hidalgo-Ruz and Thiel (2013)  
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42 166 zones were classified according to climate and water regime. Similarly, we classified our  
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45 167 samples into 3 zones: Zone I covers all beaches bordering the Mediterranean; Zone II covers the  
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47 168 beaches adjacent to the Atlantic Ocean and North Sea; and, Zone III those adjacent to the Baltic  
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50 169 Sea (see Table S2 for the coastal attributes of these zones). These zones have different  
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52 170 characteristics. For example, the Atlantic coast has the highest average wind speed, waves and  
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55 171 annual precipitation, while the surface water temperature is highest along the Mediterranean  
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57 172 coast, which is also most densely populated (Gazeau et al. 2004; Table S2). Furthermore, the  
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59 173 Mediterranean Sea has been shown to contain particularly high concentrations of plastic due to  
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174 its semi-enclosed structure and large plastic input (Cózar et al. 2015). The Baltic Sea is similarly  
175 semi-enclosed. The Mediterranean Sea is commonly divided into an eastern and western basin  
176 that are divided near the Tunisian and Sicilian coast (International Hydrographic Organization  
177 1953). The hydrological characteristics of these basins can lead to different behaviours of plastic  
178 in the marine environment. In our study we also make a distinction between the eastern and  
179 western Mediterranean coasts. The Atlantic zone was similarly divided into the North Sea and  
180 Atlantic, the former of which is boreal whereas the Atlantic is warm-temperate (Dauvin 2008).  
181 The main European ports are situated in the southern North Sea and maritime traffic in the  
182 northern English Channel is the busiest in the world (Dauvin 2008). As a result, MP abundance  
183 will therefore be examined within 3 zones and 5 subzones.

184           Some locations are situated in transition regions between zones (one) and subzones (two).

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

190 *Statistical analysis* – MP concentrations for sampling locations were reported as mean  $\pm$  SEM of  
191 the 5 replicates expressed in MPs per kg of dry weight sediment. We conducted an analysis of  
192 variance (ANOVA) (using R version 0.98) on the 23 sampling locations (with 5 replicate  
193 samples per location) with significance set at  $\alpha < 0.05$ . A nested ANOVA with the same  
194 significance level was performed on the aforementioned zones and subzones. The data was  
195 checked for normality and homogeneity of variance using Shapiro-Wilk’s W-test and Levene’s  
196 test respectively. Although ANOVAs are robust for the violation of these assumptions, if they

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197 are violated, results need to be interpreted with caution when p-values are close to  $\alpha$ , which was  
198 noted in the results section where applicable. If significant differences were observed, a Tukey's  
199 post-hoc test was conducted.

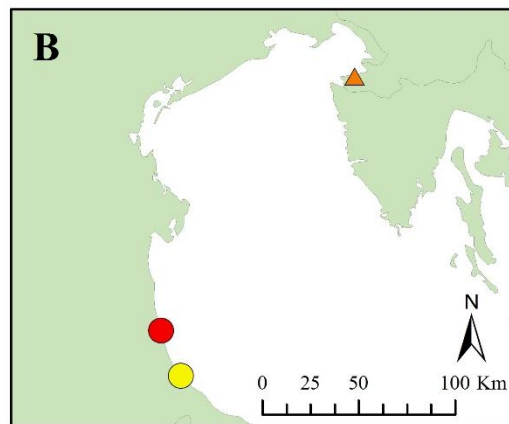
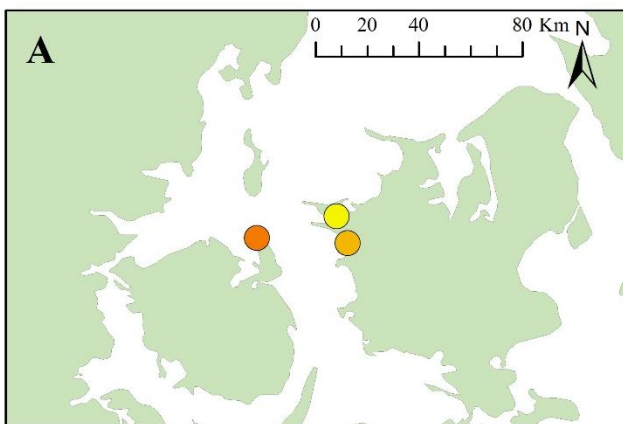
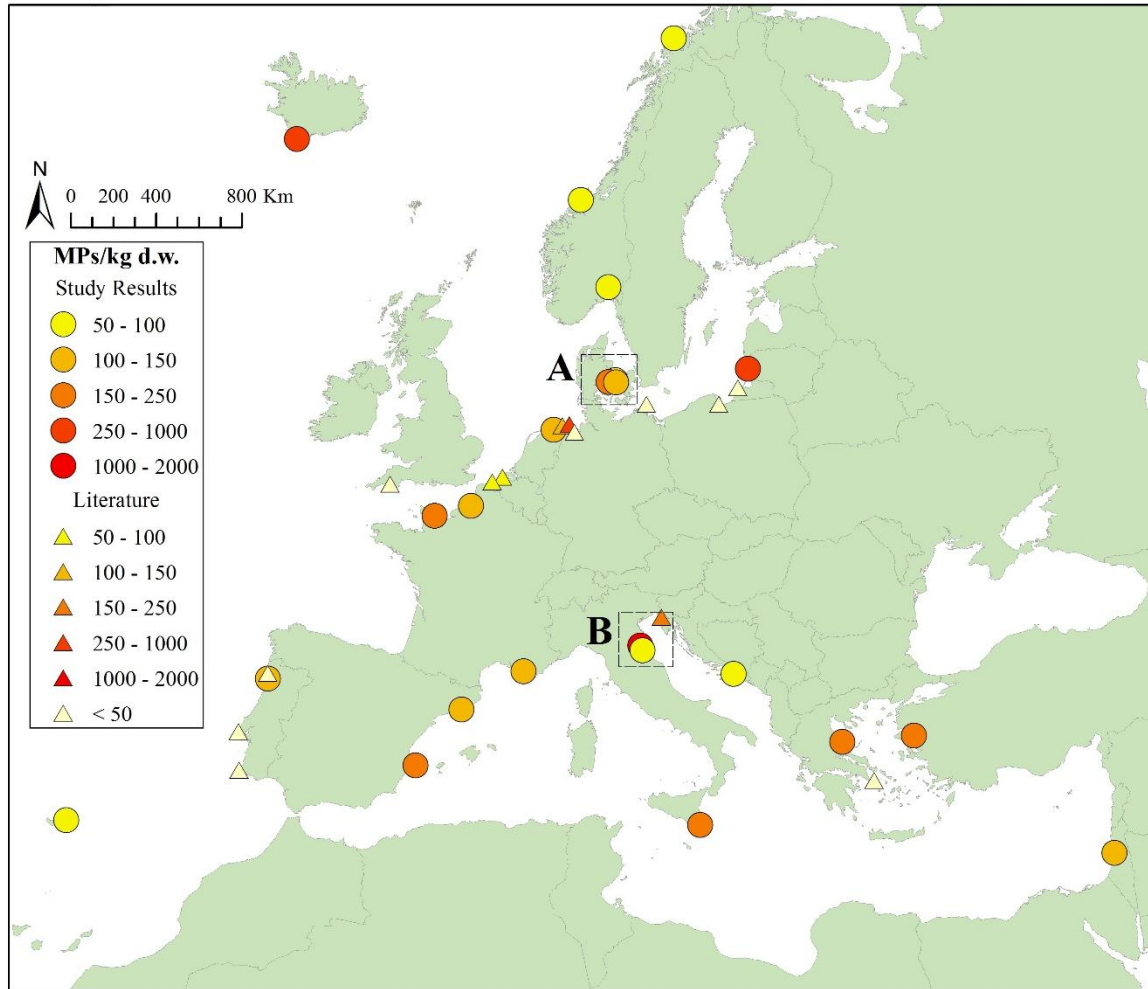
### 3. Results

#### 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  $72 \pm 24$  MPs  $\text{kg}^{-1}$  d.w. in Tromsø, Norway, to  $1512 \pm 187$  MPs  $\text{kg}^{-1}$  d.w. in Lido di Dante, Italy. The majority of locations had abundances below 248 MPs  $\text{kg}^{-1}$  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  $\text{kg}^{-1}$  d.w., respectively (see Table 2 for more details). The Atlantic zone was the least polluted with a mean of 190 MPs  $\text{kg}^{-1}$  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  $\text{kg}^{-1}$  d.w., respectively (Table 2). The levels of microplastics in the western subzone were relatively low and homogeneously 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  $\text{kg}^{-1}$  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.

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**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 ( $\pm$  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.

Location	Group		Abundance (MPs/kg d.w.)		Length (mm)		Colour (%) <sup>b</sup>		
	Zone	Subzone <sup>a</sup>					Blue/black	Red	Other
Sicily, IT	I	W	160 $\pm$ 31	c	1,32 $\pm$ 0,30	a	70,0	20,0	10,0
Denia, ES	I	W	156 $\pm$ 29	c	1,96 $\pm$ 0,71	a	79,5	12,8	7,7
Barcelona, ES	I	W	148 $\pm$ 23	c	1,13 $\pm$ 0,36	a	81,1	8,1	10,8
Cassis, FR	I	W	124 $\pm$ 36	c	1,28 $\pm$ 0,32	a	87,1	9,7	3,2
Lido di Dante, IT	I	E	1512 $\pm$ 187	a	1,38 $\pm$ 0,37	a	72,0 *	11,2 *	16,8 *
Dikili, TR	I	E	248 $\pm$ 47	c	1,01 $\pm$ 0,17	a	62,9	14,5	22,6
Pilion, GR	I	E	232 $\pm$ 93	c	0,93 $\pm$ 0,48	a	77,6	10,3	12,1
Tel Aviv, IL	I	E	168 $\pm$ 16	c	0,94 $\pm$ 0,31	a	81,0	9,5	9,5
San Mauro, IT	I	E	84 $\pm$ 12	c	1,42 $\pm$ 0,58	a	90,5	9,5	0
Bosnia	I	E	76 $\pm$ 13	c	1,54 $\pm$ 0,33	a	73,7	26,3	0
Vik, IS	II	A	792 $\pm$ 128	b	1,80 $\pm$ 0,33	a	84,8	8,1	7,1
Porto, PT	II	A	140 $\pm$ 26	c	1,34 $\pm$ 0,32	a	74,3	8,6	17,1
Smøla, NO	II	A	92 $\pm$ 21	c	0,96 $\pm$ 0,24	a	78,3	8,7	13,0
Madeira, PT	II	A	92 $\pm$ 15	c	1,98 $\pm$ 0,73	a	91,3	4,3	4,3
Tromsø, NO	II	A	72 $\pm$ 24	c	1,60 $\pm$ 0,48	a	77,8	16,7	5,6
Normandy, FR	II	NS	156 $\pm$ 29	c	0,91 $\pm$ 0,28	a	92,3	5,1	2,6
Normandy, FR	II	NS	143 $\pm$ 13	c	1,36 $\pm$ 0,42	a	78,8	12,1	9,1
Rottumeroog, NL	II	NS	124 $\pm$ 27	c	1,28 $\pm$ 0,54	a	80,6	16,1	3,2
Drøbak, NO	II	NS	100 $\pm$ 21	c	1,50 $\pm$ 0,36	a	80,0	12,0	8,0
Klaipėda, LT	III	B	700 $\pm$ 296	b	1,42 $\pm$ 0,29	a	75,0 *	14,4 *	10,6 *
Fyns Hoved, DK	III	B	164 $\pm$ 21	c	1,26 $\pm$ 0,44	a	82,9	9,8	7,3
Bjergje Nord, DK	III	B	128 $\pm$ 31	c	1,34 $\pm$ 0,44	a	84,4	12,5	3,1
Kalundburg, DK	III	B	88 $\pm$ 33	c	1,55 $\pm$ 0,45	a	81,8	13,6	4,5

<sup>a</sup> E = Mediterranean-East, W = Mediterranean-West, A = Atlantic Ocean, NS = North Sea and B = Baltic Sea.

<sup>b</sup> and \* indicates a subsample was taken due to high MP abundance.

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238 **Table 2.** A summary of the mean abundance ( $\pm$  SEM), mean length ( $\pm$  SEM), and colour per  
239 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 $\pm$ 62	1.29 $\pm$ 0.13	77.5	13.2	9.3
West	147 $\pm$ 14	1.43 $\pm$ 0.22	79.4	12.7	7.9
East	387 $\pm$ 100	1.20 $\pm$ 0.16	76.3	13.6	10.2
II: Atlantic	190 $\pm$ 35	1.41 $\pm$ 0.14	82.0	10.2	7.8
North Sea	131 $\pm$ 12	1.26 $\pm$ 0.20	82.9	11.3	5.7
Atlantic	238 $\pm$ 62	1.54 $\pm$ 0.20	81.3	9.3	9.4
III: Baltic	270 $\pm$ 90	1.39 $\pm$ 0.20	81.0	12.6	6.4

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

### 247 **3.2 Microplastics characterization**

248 *Physical characteristics* – The majority of MPs identified in this study were fibrous (98.7 %).  
249 Other types of MPs found were films (5 items, 0.35 %) and particles (13 items, 0.91 %). Only  
250 one particle was identified as a potential primary MP because of its spherical shape (Figure S1a).  
251 Other particles were more angular and irregularly shaped (Figure S1b), suggesting they resulted  
252 from breakdown of larger plastics. As a proportion of MPs, blue/black MPs were 77.5-82.9%,  
253 red MPs was 9.3-13.6% (Table 1). Other colours that were identified were green, orange, purple,  
254 grey, white, and multi-coloured (photographic examples fibres identified were shown in Figure  
255 S1c-g). The average length of the MPs ranged from 0.91 mm in Normandy to 1.97 mm in  
256 Madeira (Table 1). These results were not statistically significant (ANOVA,  $F_{22,207} = 0.51$ ,  $p =$   
257  $0.967$ ). Among different zones, the average length ranged from 1.26-1.54 mm (Table 2). Zones  
258 and subzones showed no statistically significant differences (nested ANOVA,  $F_{\text{sub}, 2,20} = 0.22$ ,  $p =$   
259  $0.719$ ,  $F_{\text{zone}, 4,18} = 0.52$ ,  $p = 0.801$ ). The majority of the MPs measured (54.8%) were < 1 mm in  
260 size. The distribution of MPs within size categories was shown in Figure 2, and follows an  
261 exponentially decreasing number of MPs with increasing size.

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

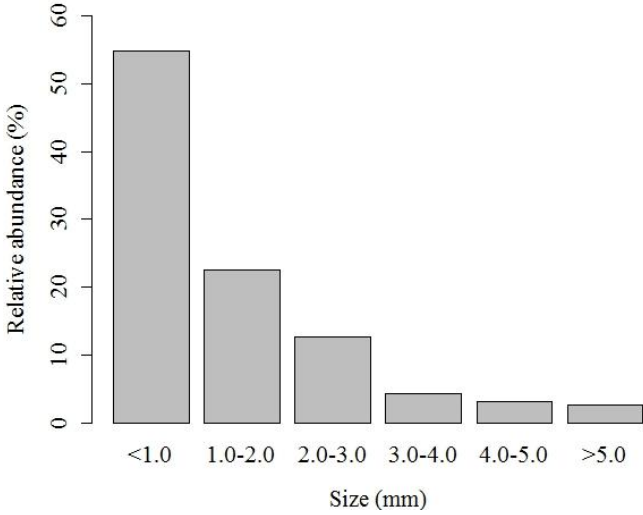
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264 Of the remaining 129 visually confirmed MPs, only 10 (4.5%) were matched to a specific  
265 polymer type. The three types of polymer that were identified are polyester (7 items),  
266 polypropylene (2 items) and polyethylene (1 item). Additionally,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

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**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).

## 279 **4. Discussion**

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

### 284 **4.1 Microplastics abundance and spatial distribution**

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

291 Within the Baltic Sea, one sampling location in Lithuania showed much higher MP  
292 abundances than three other sites within the same zone in Denmark (Figure 1). This location, in  
293 Klaipėda, is at the outlet of the freshwater Curonian Lagoon, into which several rivers flow  
294 creating a unidirectional flow (Christian et al. 2008). The lagoon has high concentrations of  
295 agricultural and industrial pollution (Christian et al. 2008). Previous research on MP  
296 contamination in lagoons showed varied results. For example, a study in Italy found high levels  
297 of MP contamination, which was attributed to significant freshwater inputs and the low-energy  
298 environment (Vianello et al., 2013). In contrast, three studies conducted in and around the  
299 Vistula Lagoon bordering Poland and Russia found low concentrations of MPs, ranging from 1-  
300 39 MPs kg<sup>-1</sup> d.w. (Table 3). Although Klaipėda is located close to this area, it has an average  
301 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<sup>a</sup>. 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 <sup>b</sup>
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	<i>0.3-90</i>
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	<i>1.5-15.7 (1-2 mm)</i>
					<i>0.3-15.0 (2-4 mm)</i>
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	<i>0.7-11</i>
Norén (2007)	II	Sweden	Subtidal	N/D	<i>16-2590</i>
Popa et al. (2014)	-	Romania	Beach	N/D	<i>1000-5500</i>
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		<i>31</i>
			Subtidal		86
Vianello et al. (2013)	I	Italy	Subtidal	< 1 mm	672-2175
Zobkov and Esiukova (2017)	III	Russia	Subtidal	< 5 mm	34

<sup>a</sup> To increase the comparability of these studies, the units were converted to MPs kg<sup>-1</sup> of dry weight (d.w.) where possible. An average sediment density of 1600 kg m<sup>-3</sup> 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.

<sup>b</sup> Reported in g/L

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

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

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

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

346 **4.2 Microplastics characterization**

347 Overall, MPs identified in this study were predominantly blue/black or red fibres. Several studies  
348 similarly found that blue/black and red are the most common fibres (Nel and Froneman 2015;  
349 Alomar et al. 2016; Strand and Tairova 2016; Frère et al. 2017). The high proportion of fibrous  
350 MPs reported in our study was comparable to other studies (Thompson 2004; Claessens et al.  
351 2011; Dekiff et al. 2014; Alomar et al. 2016; Graca et al. 2017; Zobkov and Esiukova 2017).  
352 Some studies find that over 90% of MPs are fibrous, which is similar to the scale found here  
353 (Laglbauer et al. 2014; Strand and Tairova 2016; Blašković et al. 2017). Microfibres generally  
354 derive from the machine washing of synthetic fabrics (Browne et al. 2011; Hernandez et al.  
355 2017). Up to 700 000 fibres can be released per standard wash load (Napper and Thompson  
356 2016). They are introduced to the aquatic environment via wastewater (Murphy et al. 2016).  
357 With wastewater believed to be a likely origin of many of these fibres, the finding of these fibres  
358 on marine beaches highlights the potential for widespread distribution of MPs once within the  
359 environment. Fibres can also enter the marine environment through the fragmentation of fishing

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ropes and nets (Thompson 2004), which 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.



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383 2016). Given that the response signals of polymers are easily masked by dyes and that in the  
384 environment they usually occur as a composite, it is reasonable to assume that particles identified  
385 as dyes will usually be polymers (Horton et al. 2017). Some studies have used dyes as an  
386 indicator of plastic. In this study, several suspected MPs were matched to dyes that have been  
387 found in other MP studies, such as phthalocyanine dyes which are commonly used as plastic  
388 additives. These particles were thus inferred to be MPs, except for Drimaren navy blue, an azo  
389 dye which is commonly used to dye both plastic and non-plastic fibres (Lenz et al. 2015). The  
390 Indigio dye is commonly used to dye cellulosic fibres used in fabric for blue jeans (Wiesheu et  
391 al. 2016). The dye may therefore not relate to MPs but to cotton. This indicates that although  
392 many dyes can be related to polymers, there is some uncertainty surrounding others.

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### 394 **4.3 Citizen Science**

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

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## 5. Conclusions

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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.

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### Competing interests

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The authors declare that they have no competing interests.

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624 **Supplementary Information:**

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626 Table S1 GPS Coordinates of each sampling location.

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

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

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

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633 **Table S1** GPS Coordinates of each sampling location.

Beach ID	Region	Country	Given		Estimated	
			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	Klaipėda	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.

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

Characteristic	Coast		
	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 <sup>2</sup> )	13,1	133	19,4

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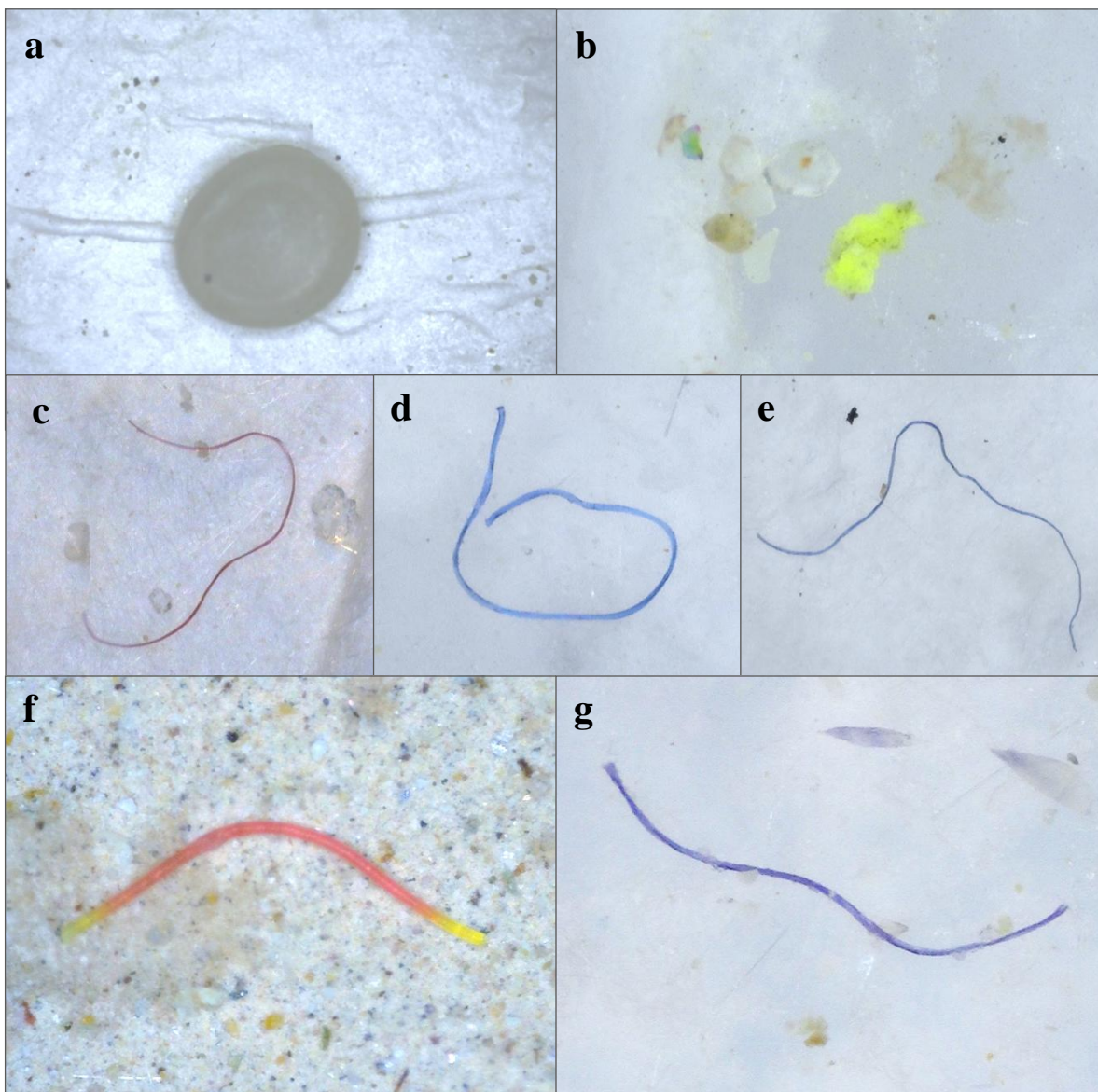
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639 **Table S3.** Abundance of microplastics in beach sediment used in Figure 1 based on available  
 640 literature.

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 the Mecklenburg-Vorpommern province. Paper's coordinates 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

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 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.

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**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).