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Distribution and abundance of microplastics in coastal sediments depends on grain size and distance from sources

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ABSTRACT

Microplastic deposition in marine sediments is a geographically widespread problem. This study examines microplastics in intertidal and subtidal sediments at 87 locations in habitats designated as Special Areas of Conservation (SACs) and Special Protection Areas (SPAs) on the coastline of Ireland. Established methodological approaches including, organic matter digestion, density separation, particle extraction and polymer identification were applied. Microplastic abundance was closely related with distance from known sources and concentrations were greater in intertidal as opposed to subtidal sediments. Colourless, polyethylene fibres and polypropylene fragments were the most abundant MP recorded and finer grained sediments were shown to entrap more MPs than coarser sediments. The results demonstrate that an understanding of potential sources of pollution, sediment type and hydrodynamic conditions are very important in terms of MP abundance and distribution in marine sediments and also in terms of effective waste management strategies and policy aimed at reducing the global plastics problem.

1. Introduction

Microplastics (MPs) are described as any synthetic, polymeric matrix (fibres either in their individual or composite forms) or solid particle of regular (e.g., beads, pellets) or irregular shape (e.g., fragments) which are insoluble in water, with a size range from 1 micron (μm) (Arthur et al., 2009) to 5 millimeters (mm) (Gigault et al., 2018), of either primary or secondary origin (Frias et al., 2018). The inputs of microplastics to the marine environment, is the result of mismanagement of anthropogenic sources (e.g. public littering, sewage treatment effluent and combined sewer overflows, recreational boats, cruise ships, commercial and fishing vessels, etc.) with the larger pieces breaking into smaller fragments due to ultraviolet (UV) light and mechanical abrasion (secondary MPs) prior to deposition in marine sediments (Auta et al., 2017). The deposition of such debris is also dependent on variables such as source, density, currents, wind, sediment, biofouling, or faunal excretion (Veerasingam et al., 2016; Ling et al., 2017; Gonçalves et al., 2019; Hitchcock and Mitrovic, 2019; Wieczorek et al., 2019) which have negative impacts on ecosystems, human safety, and the socio-economic health of coastal communities (Sheavly and Register, 2007). The presence of microplastics in marine sediments in response to anthropogenic contamination is globally recognised (Browne et al., 2011; Hitchcock and Mitrovic, 2019; Yao et al., 2019).

Sediments in enclosed waters and low-energy environments, e.g. shallow estuarine areas and bays that receive significant land-based inputs retain more MPs than higher-energy, deeper and further from source environments (Wang et al., 2020; Uddin et al., 2021). Relatively few studies have examined MP abundance in sediments in relation to grain size and conclusions regarding variations with grain size are frequently conflicting. Microplastics deposition has been positively correlated with increased total organic carbon (TOC) content in sediments (Maes et al., 2017) and increases in TOC has also being reported with decreasing grain size (Bergamaschi et al., 1997) which indicates that finer grains trap more particles (Green and Johnson, 2020). In addition, Strand and Tairova (2016) found a strong correlation between MPs and the %TOC of fine (<63 μm) sediments. Enders et al. (2019) examined the relationship between sediment granulometry and MPs and reported a strong correlation between MPs and the finer fraction (<63 μm) of sediment, a high percentage (88.59%) of the total microplastic debris detected by Blaškovic et al. (2017) were extracted from the <63 μm to 1 mm fraction, as opposed to 11.24% of total plastic items recovered from the 1–2 mm fraction and just 1.17% from the larger 4–2

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mm grain size. However many studies failed to establish a relationship between grain size and microplastic concentrations (Martins and Sobral, 2011; Romeo et al., 2015; Alomar et al., 2016; Fastelli et al., 2016; Blašković et al., 2017; Peng et al., 2017; Renzi et al., 2018). Adding to this, many studies found that proximity to a known source (e.g. wastewater treatment plant, ports, human settlements) did not increase the concentration of micro-debris but their distribution can be influenced by factors, such as volcanic eruptions, currents, sediments and aeolian processes (Martins and Sobral, 2011; Oliveira et al., 2015; Alomar et al., 2016; Fastelli et al., 2016; Wang et al., 2020). Much uncertainty remains regarding the relationship between grain size, MP abundance and distance to sources which warrants further investigation (Alomar et al., 2016; Blašković et al., 2017; Wang et al., 2020). Given the numerous potential sources, means of dispersal and deposition, several preliminary studies and reviews have reported the presence of microplastics in coastal sediments to be a ubiquitous global problem (Van

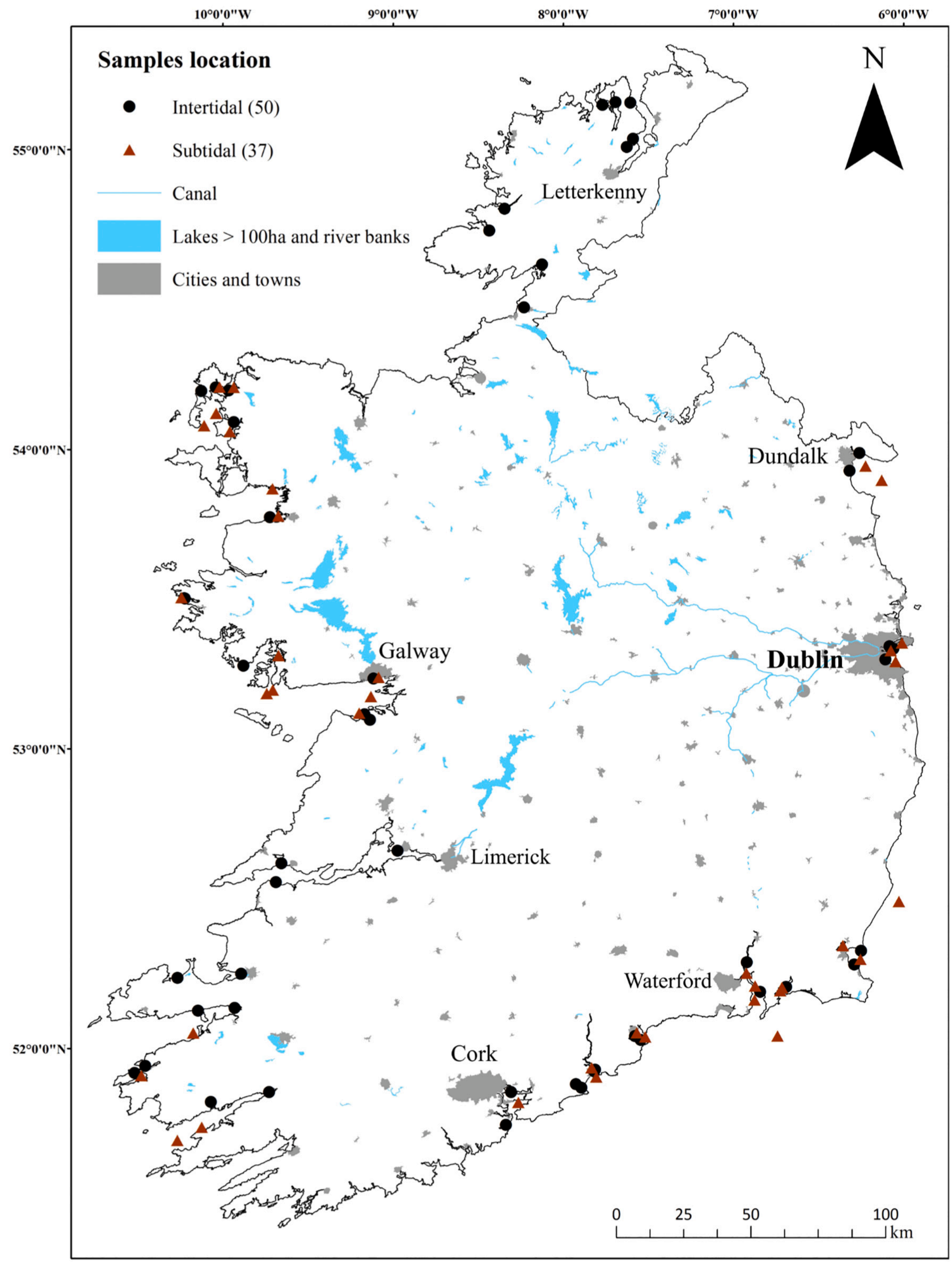

Fig. 1. Map presenting sediment sampling locations collected from the intertidal and subtidal zones, along the coastline of Ireland.

Cauwenberghe et al., 2015; Graca et al., 2017; Bissen and Chawchai, 2020; Tata et al., 2020; Yao et al., 2019).

Ireland is confronting the microplastic problem with a variety of studies from organisms (Lusher et al., 2016; Wieczorek et al., 2018; Doyle et al., 2019) coastal waters (Frias et al., 2020), continental sediments (Martin et al., 2017), freshwater (Cedro and Cleary, 2015; Mahon et al., 2014; Mateos-Cárdenas et al., 2021), sewage (Mahon et al., 2017; Ó Briain et al., 2020) and atmospheric levels (Roblin et al., 2020), but these investigations are usually focused over small spatial scales. The ability to provide a broader geographical overview of the problem can be advantageous as it can provide a benchmark for the levels of marine sediment contamination and guide future policies and legislation aimed at reducing the plastic burden in coastal environments (Maes et al., 2017; Green and Johnson, 2020). The objective of this study was to determine the distribution and abundance of microplastics in different marine sediment types from the coastline of Ireland (approximately 4.000 km). It was hypothesized that the type, distribution, and abundance of microplastics in a wide variety of different sediment types from the coastal environment is a function of increasing plastic production, increasing coastal population growth, and proximity to urban wastewater outflows and local hydrodynamics.

2. Materials and methods

2.1. Sediment sampling and classification

Surface sediment samples were collected from 87 intertidal and subtidal locations (Fig. 1) in habitats designated as Special Areas of Conservation (SACs) and Special Protection Areas (SPAs) on the coastline of Ireland (Fig. SM1) between May and November 2016. Collection of intertidal sediment samples was undertaken by coring (metal core) to a depth of 5 cm and subtidal samples were subsampled from the centre of a grab (day grab), also to a depth of 5 cm. The retrieved samples and subsamples were carefully transferred to 1 L glass bottles, with a sealing layer of aluminium foil between the sample and the cap to prevent plastic shedding contamination and 70% ethanol was added to each sample. The core and grab were thoroughly cleaned with deionized water to avoid cross contamination between samples. Sediment classification was determined by separation of dry sediments by grain size using a set of sieves ranging from 63 μm to 2 mm (Table 1) and a classification scale from Wentworth (1922).

2.2. Environmental controls

In order to avoid background contamination during the analysis, the following preventive measures were implemented during the extraction procedure: ethanol evaporation was performed inside a clean room (class 1000: ISO 6) and extractions were performed inside a laminar flow hood (AirClean600 R: ISO 5); all materials and vessels were covered with aluminium foil after each experimental step and washing. To assist in the identification of potential contamination sources, laboratory cotton coats dyed pink were always used, from extraction to analyses and identification of polymers (apart from the Clean Room facilities where a non-shed, splash-proof laboratory coat was worn). Filter blanks were run in parallel to assess and correct for contamination during both sample extraction and subsequent processing. Particles detected on filter (Whatman glass fibre filters, 47 mm Ø, 1 μm mesh) blanks were analysed for colour, size and chemical composition and compared to particles from environmental samples to avoid erroneous results.

Table 1
Classification scale as suggested by Wentworth (1922) and categorize by grain size.

Sediment category	Grain diameter
Very coarse/maërl	>1 mm
Coarse sand	1 mm–500 μm
Medium sand	500–250 μm
Fine sand	250–63 μm
Silt/mud	<63 μm

2.3. Microplastic extraction

Sediment samples were placed in a water bath at 70 (±2) °C under a fume hood (class 1000: ISO 6 clean room) until the ethanol evaporated and the sediment was transferred to aluminium trays and dried at 70 °C. Each sample was weighted, transferred to a 1 L beaker with 100 mL of hydrogen peroxide (H_2O_2; Merck Scientific, UK) and mixed with a glass rod (1 min) for the removal of organic matter (OM digestion) and left to settle overnight. This process was repeated as many times as necessary until removal of the organic matter. Subsequently, the sediments were washed through a fine mesh sieve (32 μm) and dried at 70 °C.

Dry samples were weighed and zinc chloride ($ZnCl_2$; Fisher Scientific, UK) was added (to three times the sample volume) with a density of 1.5 g cm^{-3}; stirred for 1 min and left to sit for ~2 h prior to filtration, this step was repeated two additional times. A summary diagram of the extraction procedures may be seen in Fig. SM2.

Particles retained on each filter paper were examined and counted under a stereoscope with a magnification of 0.8–4× (M165C, Leica Microsystems, Germany). Microplastics were characterized by their shape, surface texture and colour according to the criteria established by Free et al. (2014) and McCormick et al. (2014) and categorized into five classes, namely "pellets", "fragments", "fibres", "foams" and "films". Pellets included both virgin plastic pellets and microbeads.

2.4. Polymer identification

Polymer identification was conducted by separating the microplastics using stereomicroscopy, and by recording their colour (transparent, white, red, orange, blue, black, grey, brown, green, pink, tan, yellow and multicoloured) and shape (fibre, pellet, fragment, film, granules, styrofoam) before polymer identification with Raman Spectroscopy (Horiba LabRAM II, Horiba Jobin-Yvon, France). The Raman Spectrometer was equipped with a 600 groove mm^{-1} diffraction grating, a confocal optical system, a Peltier-cooled CCD detector, and an Olympus BX41 microscope (Ó Briain et al., 2020; Loughlin et al., 2021) and spectra were obtained at a range of 100–3500 cm^{-1} using a 532 nm laser for polymer identification. Spectra were compared to a spectral reference library (KnowItAll, Bio-Rad), an in-house extension of the library with additional spectra from environmental plastics collected from the intertidal zone and known virgin polymer types (purchased from CARAT GmbH, Bocholt, Germany). In addition, SLoPP and SLoPP-E libraries (Munno et al., 2020) were employed, and the 'fingerprint' region of each spectra was used to identify the polymer type.

2.5. Cleaning procedures and contamination

All laboratory material was cleaned with laboratory grade detergent (DECON 90) and rinsed with ultrapure water (18.2 Ω) [Milli-Q Element System™, Merck Millipore, USA] before and after each sample extraction. Density separation liquid ($ZnCl_2$) was filtered and reused after their density (1.5 g cm^{-3}) was checked.

Alongside the drying and filtration procedures blanks were performed and checked at the end of each day. Filters were placed in the fume hood and in the glove box to capture any airborne MPs, water blanks when performing the H_2O_2 digestion and $ZnCl_2$ when performing the MP extraction.

2.6. Mapping and determination of distances to potential MPs sources

Spatial analysis techniques were applied in a geographical information systems environment to investigate whether a relationship between potential sources and observed concentrations of microplastics could be established. The "proximity" toolset was applied for analysing distances between the sampling points and potential sources. Potential sources were identified as rivers/waterways, urban settlements and wastewater treatment facilities. From the "proximity" toolset, the 'buffer' tool was utilized to create buffer zones surrounding each of the sampling points with an extent of ≤40 km (this extent was selected arbitrarily). The "select by location" tool was then applied to create new datasets of the potential sources within the 40 km buffer zones. The "near" tool was then used to calculate distances between the input features (sampling points) and the closest features (sources within 40 km). Distances to sources were rounded to nearest 100 m and further categorized by distance bands for analysis purposes. Maps were built using ArcGIS® (10.2 and Pro 2.7) software by Esri®. ArcGIS® and ArcMap™ are the intellectual property of Esri® and are used herein under license. Copyright© 1995–2013 Esri.

2.7. Microplastic determination and standardization

A method validation was performed on 19 samples (8 intertidal and 11 subtidal) representing different sedimentary types (mud, fine, medium, coarse sand/maërl), with five extractions instead of the normal three, with 80% of particles recovered in the first three separations, validating the three-density separation approach.

Notwithstanding the employment of the contamination prevention measures, control samples (total 134) contained fibres (average of 1.5 ± 2.4), the majority of which were colourless (61%) and <1 mm in length, these fibres also had a clear consistent shape and unlike fibres from the environment these were not weathered making them easy to separate from any other fibres present. Under Raman spectroscopy blank fibres were checked and spectral ID for cellulose/cotton was matched. These were subtracted from the values for each sediment sample to account for contamination. Microplastic counts were standardised per kg of dry sediment sample.

2.8. Statistical analysis

All statistical analyses were performed using the R software and PERMANOVA^{+} add-on PRIMER 6 (Plymouth ROutines in Multivariate Ecological Research) software. All tests were performed at a significance level with a p-value less than 0.05, and when necessary were based on 1999 permutations.

2.8.1. Microplastic abundance and distribution

Each sample was classified according to three different factors: relative tidal position (2 levels: subtidal, intertidal), sediment grain size (4 levels: mud, fine sand, medium sand, coarse sand/maërl; SM3) and distance from potential sources of microplastics (2 levels: near, far). In general, the concentration of microplastic for the different combinations of factors did not accomplish normality and homoscedasticity according to Shapiro-Wilks and Levene's tests. In light of this, non-parametric tests were used instead of traditional parametric approaches. Furthermore, as all the different scenarios were not always present, the data were analysed in a sequential manner. Initially differences between tidal positions were assessed for each available combination of sediment type and distance from source. Subsequently, the effect of distances from sources in relation to sediment type was analysed and finally, differences between sediment types in relation to distance from sources of microplastics.

2.8.1.1. Subtidal vs. intertidal. To assess differences between subtidal and intertidal locations, near (i.e. <2 km) and distant (i.e. 2 km) to sources of microplastic in different sediment types (i.e. mud, fine sand, medium sand and coarse sand/maërl), non-parametric U-Mann Whitney tests were performed for six out of eight different possible combinations of scenarios.

2.8.1.2. Distance from sources. In order to assess the influence of microlitter sources (urban settlements and waste water treatment plants) on the concentration of microplastics in different sediment types, U-Mann Whitney tests were applied. Two different categories were considered for these analyses, near (i.e. <2 km) and distant (i.e. ≥2 km) to the microplastic source. While overall, four different types of sediments were found (i.e. mud, fine sand, medium sand and coarse sand/maërl), coarse sand/maërl sediments were not present close to potential sources of microplastics, and the effect of distance from source was only tested in three of the four different sediment types in this scenario.

2.8.1.3. Sediment types. To assess differences in the concentration of microplastics between the four different sediment types (i.e. mud, fine sand, medium sand, coarse sand/maërl) present within the two defined distance classes, Kruskal-Wallis tests were performed. When significant differences were identified by the Kruskal-Wallis, a Nemenyi-Damico-Wolfe-Dunn post-hoc test was applied to assess the difference between sediment types. In the case of sediment close to sources only three different types of sediment were found as mentioned above.

2.9. Microplastic composition and distribution

To assess the role of the sedimentary environment and the influence of litter sources on the composition and abundance of microplastics a two-way permutational analysis of variance (PERMANOVA) was developed. This analysis was based on a matrix of Euclidean distances between samples, according to the number and colours of microfibres and microfragments.

3. Results

3.1. Descriptive statistics of microplastics in Ireland

From the 87 sediment samples extracted (corresponding to 87 different locations), an average of 393 (±105.6) and 352 (±91) grams of intertidal (n = 50) and subtidal (n = 37) sediments were processed which produced a total of 728 filters. A sum of 1390 micro-particles was retrieved, generating an average of 14 (±21.3) fibres, 2 (±3.8) fragments and 0.01 (±0.1) pellets per sample. Micro particles were not detected in 8 samples, two from the intertidal (Fig. 2a) and six from the subtidal (Fig. 2b) zone. For the homogeneity of the results, the numbers reported from here on out, unless otherwise specified, will be per kg (kg^{-1}) of dry weight (d.w.) sediment.

Intertidal sampling points (n = 50) presented an average of 53 (±100.1) fibres (Fig. 2c) and 7 (±11.7) fragments (Fig. 2e) kg^{-1} d.w. sediment. Intertidal debris distribution was uneven along the Irish coastline with the Northwest of the country presenting a higher concentration with up to 544 fibres and 77 fragments per kg d.w. and the coastline to the Southwest and Southeast of the country presenting less than 200 fibres and 22 fragments per kg d.w. (Fig. 2c, e). Subtidal samples contained a lower concentration of debris when compared with intertidal sediments (Fig. 2b), however they were not void of litter, out of the 37 subtidal sediment cores analysed, an average of 42 (±53.7) microparticles were retrieved, distributed as 33 (±39.8) fibres (Fig. 2d) and 9 (±19.6) fragments (Fig. 2f) kg^{-1} d.w. sediment. Microfibre concentration was, also, lower in the subtidal sediments with 24% of the samples containing zero fibres and the highest number, 153 fibres per kg, on the west coast and the lowest, 2 fibres per kg, on the southwest of the country (Fig. 2d). Microfragments concentration was even lower with 51% of the samples containing no fragments (Fig. 2f).

Colour distribution for microplastics recovered in this study exposes a bias towards colourless particles (56%), followed by blue (19%), black (13%) and red (6%) colouring with white, pink, green and orange

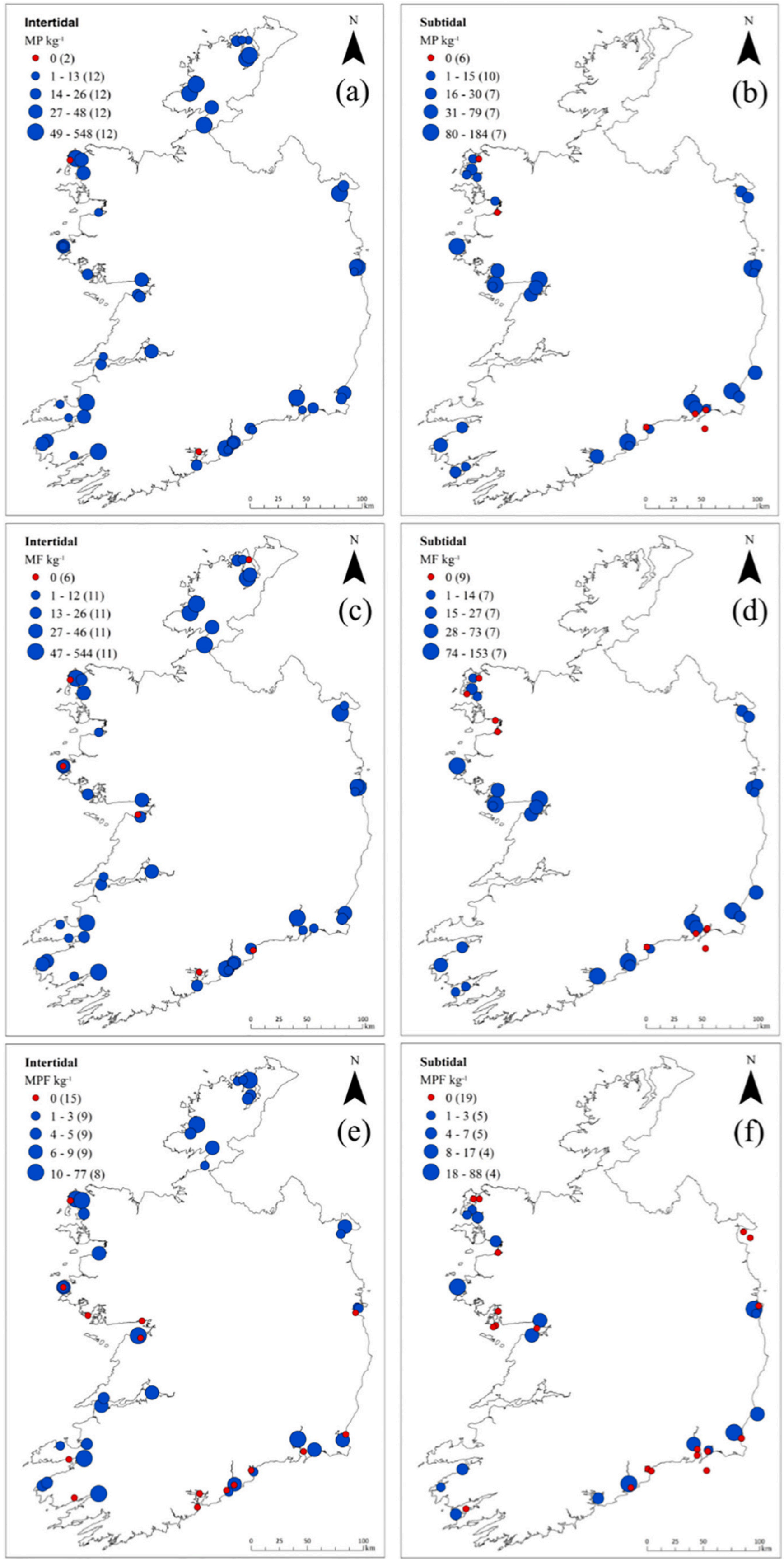

Fig. 2. Spatial distribution of microparticles (MP; a, b), microfibres (MF; c, d) and microfragments (MPF; e, f) per kg^{-1} in intertidal (a, c, e) and subtidal (b, d, f) sediments; red indicates locations where no particles where detected. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

representing the remaining colours. When separating fibres from fragments, a similar distribution was observed with colourless (62%), black (15%), blue (14%) and red (7%) fibres recorded. However, for the fragment particles the colour blue (55%) was the highest reported, followed by colourless (16%), white (10%) and green (9%). The only plastic pellet present in this study displayed an off-white colour. A similar colour distribution can be observed when intertidal and subtidal samples are analysed separately.

Out of the 1390 particles extracted from 87 cores, 689 particles (from 45 cores - 27 intertidal and 18 subtidal) were randomly selected for polymer identification using Raman spectroscopy (Fig. SM4). Most of the fibres (23%) from the intertidal samples were identified as polyethylene terephthalate (PET), follow by polyethylene (PE [19%]) and polypropylene (PP [16%]) similarly subtidal fibres comprised the same polymers although at different concentrations with PET accounting for 38% of the total fibres identified followed by PE (28%) and PP (22%). Fibres comprised of other polymers were also present in intertidal and subtidal samples including polyurethane [PU], nylon and cotton while some only appear in the intertidal (polyethylene/propylene copolymer [PE-co-PP], polystyrene [PS], rubber, polyacrylonitrile [PAN], polyacrylamide [PAM] and acrylonitrile butadiene styrene [ABS]) and others only in the subtidal (polyamide [PA]) and all at concentrations below 4%. In relation to the identification of the fragment particles, in intertidal and subtidal sediments, PP was the most abundant polymer with 64% and 83% respectively, follow by PE (29%), PS (4%) and cotton (4%) in the intertidal, and PE (8%) and PU (8%) in the subtidal samples. When relating polymer identification with colour, it is noticeable that colourless PET and PP fibres are the most abundant, alongside the blue PP fragments found around the entire coastline of Ireland and in both intertidal and subtidal samples (Fig. SM5).

3.2. Factors determining distribution of microplastics in Ireland

3.2.1. Subtidal vs. intertidal

No differences in total microplastic, microfibre and microfragment abundances were found in any of the 6 available scenarios resulting from the combination of the different sediment types and distance from source (U-Mann Whitney tests; p-values > 0.05), indicating little or no effect of the relative tidal position in the distribution of microlitter at regional scales. Due to the non-significant effect of this factor in the distribution of microplastics, all samples were pooled for this factor in subsequent statistical analyses.

3.2.2. Distance from sources

The results of the U-Mann Whitney to assess the effect of the distance from a possible source of marine microlitter revealed significant differences in the case of sediments < 63 μm (mud); total microplastic (W = 50; p-value = 0.008), microfibre (W = 49; p-value = 0.011) and microfragment (W = 46; p-value = 0.028) abundances (Fig. 3a). In all cases, higher microlitter abundances were found with proximity to sources for the total microplastics, microfibres and microfragments. No differences in microlitter abundances were found for fine and medium sands (U-Mann Whitney tests; p-values > 0.05), although it followed the expected pattern with higher medium values in sites close to potential sources of microplastics (Fig. 3b and c). Due to the absence of coarse sands in the proximity of the sources (<2 km), it was not possible to assess the effects of this factor for this kind of sediment (Fig. 3d).

3.2.3. Sediment types

The Kruskal-Wallis tests indicated significant differences in the abundance of total microplastics (H (2) = 10.11, p-value = 0.006), microfibres (H (2) = 9.35, p-value = 0.009) and microfragments (H (2) = 8.68, p-value = 0.013) between sediments close to possible sources of microlitter. Microplastics were higher in muddy sediments than in the other types of sediments, however, significant differences were only found between mud and fine sand in the case of total microplastics and

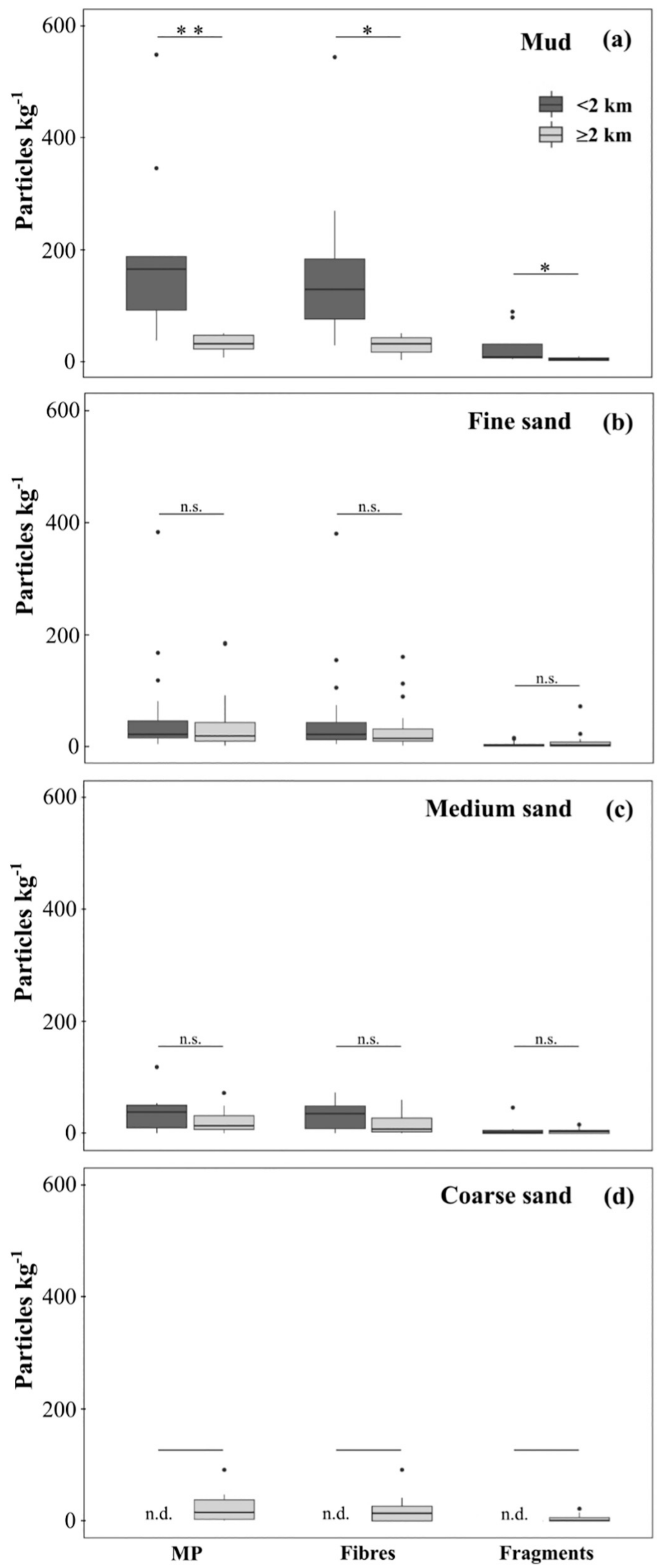

Fig. 3. Boxplot comparing, total of microplastics (MP), fibres and fragments regarding distance to a source of microlitter in the four types of sediment analysed. **p-value = 0.01–0.001; *p-value = 0.01–0.05.

microfragments and significant differences were also observed between mud and medium sand (Fig. 4a). No significant differences were found in any sediment type greater than 2 km from a possible source for any variable related to microplastic abundance (Kruskal-Wallis tests > 0.05) (Fig. 4b).

4. Discussion

The present study introduces microplastic distribution along Irish marine intertidal and subtidal sediments and within Special Areas of Conservation and Special Protection Areas. The concentration of marine micro-litter debris varied between sampling locations with a concentration as low as 0 for both inter- and subtidal sediments and as high as 548 and 184 particles kg^{-1} in one intertidal and one subtidal location, respectively. This variation between intertidal and subtidal concentrations was previously observed by Claessens et al. (2011) and Laglbauer et al. (2014); the concentration of micro particles in marine sediments vary depending on sediment particle size, wind, wave exposure, freshwater inputs, and human behaviour, among other factors. A high range of variation can be seen across Europe with a subtidal sample in Italy containing as high as 2175 particles kg^{-1} (Vianello et al., 2013) and samples from Greece with 0 particles detected (Kaberi et al., 2013). The differences in concentrations observed between studies is not only due to environmental conditions or human influence, but also due to sampling location (e.g., dunes, intertidal, subtidal), direction (e.g., perpendicular vs. horizontal to shoreline) sampling techniques and extraction methodologies applied for the recovery of the particles. The different methodologies applied for the sampling (core vs. quadrat; use of sieve vs. non-use) and extraction (e.g., chemical [NaCl vs. $ZnCl_2$] and density [1.2 vs. 1.5 g cm^{-3}]) of particles from the sediment can lead to a bias of the type, shape and polymer found in each study and their concentrations (see Table 2). Considering these discrepancies, it is difficult to compare microplastic contamination from previous studies, however the

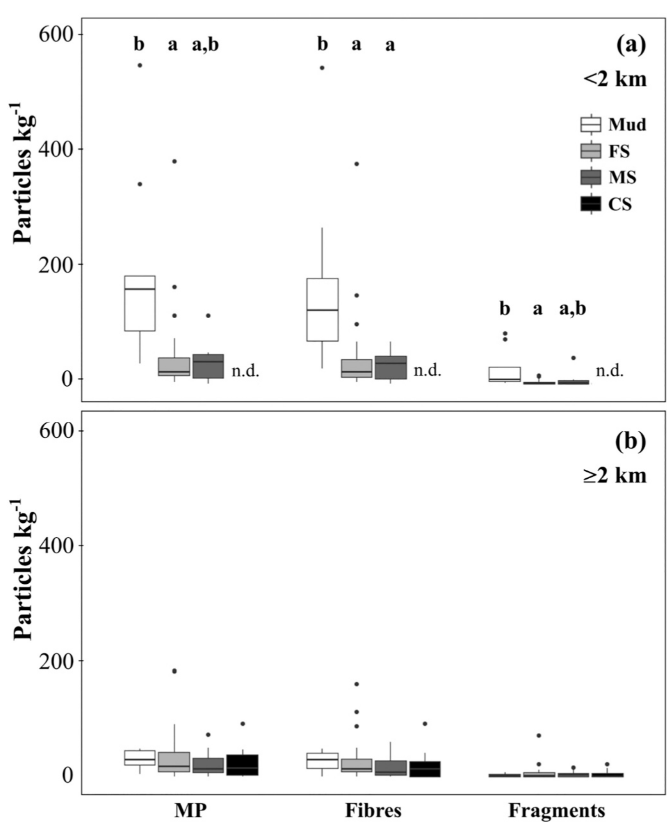

Fig. 4. Boxplot comparing, total of microplastics (MP), fibres and fragments regarding sediment type in the two distances analysed.

Table 2
Abundance, location and particle size of microplastics in European sediments adapted from Van Cauwenberghe et al., 2015.

Country	Location	Particle size	Abundance particles	Unit	Reference
UK	Beach[a]	1.6 μm–5 mm	8	kg[b]	Thompson et al., 2004[b]; Claessens et al., 2011; Laglbauer et al., 2014
	Estuary[a]		31	kg[b]	
	Subtidal[a]		86	kg[b]	
UK	Beach	1.6 μm–1 mm	<1–8	50 mL	Browne et al., 2010
Belgium	Harbour	38 μm–1 mm	166.7	kg	Claessens et al., 2011
	Continental Shelf		97.2	kg	
	Intertidal		92	kg	
	Subtidal		52.8	kg	
Portugal	Beach	1.2 μm–5 mm	1–137	m^2	Martins and Sobral, 2011
Germany	Beach	1.2 μm–5 mm	671	kg	Liebezeit and Dubaish, 2012
Italy	Lake Garda (North)	9 μm–5 mm	1108	m^2	Imhof et al., 2013
	Lake Garda (South)		108	m^2	
Greece	Beach	1 mm–4 mm	0–977	m^2	Kaberi et al., 2013
		2 mm–4 mm	10–575	m^2	
Belgium	Beach	35 μm–1 mm	7.2–20.4	kg	Van Cauwenberghe et al., 2013[a]
Italy	Subtidal	32 μm–1 mm	672–2175	kg	Vianello et al., 2013
Germany	Beach	<1 mm	1.3–2.3	kg	Dekiff et al., 2014
Slovenia	Shoreline	0.25–5 mm	177.8	kg	Laglbauer et al., 2014
	Infralittoral		170.4	kg	
Portugal	Subtidal	1 μm–500 μm	0.055–0.26	g	Frias et al., 2016
Germany	Beach (N2)	63 μm–5 mm	106.39	kg	Hengstmann et al., 2018[c]
	Beach (W)		76.27	kg	
	Beach (E)		94.41	kg	
	Beach (N1)		63.11	kg	
France	Intertidal	50 μm–1 mm	67	kg	Phuong et al., 2018[c]
Spain	Rías Baixas and Minõ river shelf	0.5–1 mm	70.2	kg	Carretero et al., 2021[c]
Ireland	Intertidal	63 μm–5 mm	**0–553**	**kg**	**This study**
	Subtidal		**0–172**	**kg**	

[a] Only fibre concentrations were reported.
[b] Original unit: fibres per 50 mL.
[c] Mean values reported.

list provided in Table 2 can be used as a reference guide. A more detail review of the methods used for the sampling and detection of microplastics in sediments is provided by Prata et al. (2019) and Birch et al. (2020).

In this study, only one spherical microplastic granule was found, with all the remaining particles being of secondary origin, fibres and fragments, but no film or foam was recorded, as previously reported for the Irish continental shelf (Martin et al., 2017). Fibres were the most abundant form of debris, comprising 86% of the total particles extracted, a finding similar to other studies (Frias et al., 2016; Martin et al., 2017). However, fragments were observed to be the most abundant shape in sediments from China (Xu et al., 2020) and Portugal (Rodrigues et al., 2020). Colour also varied when compared with studies elsewhere, with clear, followed by blue, white and black being the most abundant. Comparable results were reported by Martin et al. (2017) for Irish coastal shelf sediments. However other studies recorded a different colour variation, for example Rodrigues et al. (2020) recorded 11 distinct colours, in sediments from Portugal with the most common being clear and white. Hosseini et al. (2020) reported black (21.5%), transparent (20.8%) and green (17.5%) as the most abundant from sediments in Iran, while Mehdinia et al. (2020) only recorded colour particles (black-grey colour [51%] followed by blue/green [25%] and yellow/orange [11%]) with an absence of clear particles in sediments from Iran. It is also important to establish that the particles separated from the sample are in fact synthetic polymers using advanced techniques, as the margin for error from visual identification alone is very high (Song et al., 2015). Therefore, the use of techniques like Raman spectroscopy is essential. This study found that the most common polymer was polypropylene (34%) followed by PET (26%) and PE (26%), comprising of a total of 76% of the samples analysed, similar to results presented by Martin et al. (2017). These results appear consistent with other findings for example, in a recent review, Xu et al. (2020) reported that most studies found PP (27.2%) and PE (25.7%) to be the most common polymers in marine sediments. This is agreement with results reported by Rodrigues et al. (2020), Hosseini et al. (2020) (PE [38%] and PET [29%]) and Mehdinia et al. (2020) (PS and PE).

The variation in methodological approaches between studies, from sampling and extraction techniques to the materials found, indicates that micro-litter and in particular microplastics are not uniform in shape, size, colour, or polymer type. Standardised techniques for microplastic research can be beneficial in harmonizing the discrepancies among studies (Birch et al., 2020; de Ruijter et al., 2020). The abundance of colourless fibres in this report, may be due to the particular attention and experience that the researchers have in microplastic extraction (Ó Briain et al., 2020). Although the colour of the particles can change in the environment due to degradation from bleaching and erosion, it has been demonstrated that colour diversity indicates a wide range of microplastic sources (Martin et al., 2017). The appearance of clear PET fibres may indicate grey-water sources, as polyethylene is common in clothing, while polypropylene clear fibres are likely from commercial and/or recreational fishing materials (Govender et al., 2020; Mehdinia et al., 2020).

Sampling location is a known important factor that affects MP concentration in terms of proximity to wastewater treatment plants, storm runoff and untreated sewage outlets which are well established source of MP contamination (Birch et al., 2020; Ó Briain et al., 2020; Uddin et al., 2021). Local factors such as total organic carbon (TOC) content, hydrodynamic conditions or benthic fauna (Strand et al., 2013; Coppock et al., 2017 ; Maes et al., 2017; Ghayebzadeh et al., 2020; Sun et al., 2021) can influence the deposition of such debris. The current study observed a relationship between grain size, MP abundance and distance from known sources. A higher concentration of microplastics in finer sediments (<63 μm) within a 2 km distance from a known source, was observed with MP concentration decreasing with an increase in sediment grain size and/or distance from a possible source of microparticles. This emphasises the need to further investigate and monitor the microplastic content of sediments with a grain size <63 μm, accounting for the distance from known sources, as the availability and impact of such small particles benthic communities can be very high.

In the current study, an association was established between finer sediment particles and a greater burden of micro debris; in particular, this was noted in locations rich in organic matter, indicating the key role depositional material may play in the dispersal and accumulation of microplastics as observed by other authors (Browne et al., 2010; Kaberi et al., 2013; Strand et al., 2013; Martins and Sobral, 2011; Harris, 2020; Uddin et al., 2021; Wilson et al., 2021). A recent review by Harris (2020) stated that the dispersal of MPs is similar to that of natural sediments with coarse-grained and dense particles deposited close to sources, while less dense, finer particles remain in suspension and settle in low energy environments (Gob et al., 2010; Uddin et al., 2021). This finding was also supported by Wilson et al. (2021) and in addition, they highlighted the importance of depositional environments in determining microplastic abundance in coastal environments. Other studies, however, have cited no observed relationship between microplastic abundance and sediment type. Out of the 952 micro-debris particles ($n = 30$) found in the Tamar Estuary (NE Atlantic, UK) (65% were microplastics), a relationship between microplastic density and sediment type could not be established, however the deposition of denser polymer materials, with higher concentrations recorded downwind (e.g., polyvinylchloride) demonstrated a propensity for wind directional influences (Browne et al., 2010).

The choice of where to sample beach sediment for microplastic studies is a critical decision which has a strong impact on the results observed. Heo et al. (2013) analysed sediments from the upper to the lower shore of a South Korean beach and their results indicated that, unlike macroplastics, which accumulated at the high tide line, microplastics (2–10 mm) were most abundant in the upper intertidal zone, closer to the sand dunes. This indicates that the mechanisms influencing macroplastic distribution on beaches, including wind and currents, affect microplastic distribution in a unique way. As a result, choosing the appropriate site or zone for microplastic assessment on beaches may not be as straight forward as previously thought. In the current study, samples were collected from both the intertidal and subtidal zones. The abundance of microplastics in the intertidal zone is partly influenced by wave action and oscillations, whereas the subtidal zone is a much more stable environment and could be considered a sink for microplastics. In the current study the compositional structure of the sediment greatly influenced the capture of particles e.g., mudflats would have a greater propensity to capture objects as opposed to substrates comprised of maërl.

The results presented in this study follow a previously observed pattern for the deposition, shape, colour distribution and polymer type of microplastics in sediments from the offshore Irish continental shelf (Martin et al., 2017), although the present study provides a broader assessment of microplastic abundance by representing 87 locations inshore along the coastline of Ireland. A total of 1390 micro debris units were recovered, with a median (IQR) density of 21 (11 to 48) items kg^{-1}. All samples containing micro-litter (8 locations with 0 particles extracted), contained secondary microplastics (i.e., fibres and fragments) which were recorded at a median concentration of 25 (12 to 48) particles kg^{-1} in the intertidal, and 19 (4 to 50) particles kg^{-1} in the subtidal zone. A higher density of micro-litter particles was found in the mud/silt and silt associated sediment samples. In addition, the 8 samples which contained no micro-litter particles were categorized as sand samples; indicating that sediment type is a significant factor affecting micro debris deposition. It is expected that areas with high industrialization and urbanization also have higher concentrations of microplastics, although other factors such as exposure, wind, waves, fishing fleet activity, and riverine inputs should be considered when investigating microplastic deposition in coastal sediments.

Standardised debris monitoring protocols are required to establish comparable baselines and to monitor the pollution of coastlines worldwide. Information relating to the dispersal and deposition pattern of polymers is crucial for management purposes as strategies for microplastic prevention should differ according to source (Arthur et al., 2009). This study provides an insight into the state of microplastic debris in Irish coastal sediments in relation to sources and granulometric variation, and the baseline assessment described herein can be used to guide future research and policies relating to marine litter and in particular micro-litter in sedimentary environments.

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CRediT authorship contribution statement

Ana Mendes and Nessa Golden processed, and analysed the sediment samples for microplastic content, and wrote the core text. Ricardo Bermejo contributed by conducting the statistic analyses. Liam Morrison was responsible (in conjunction with the other authors) for the initial planning, experimental design, development of the concepts and overall supervision including the analytical process, data interpretation and editorial review on the overall manuscript.

Declaration of competing interest

The authors declare they have no conflicts of interest.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2021.112802.

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