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Research Paper

Beneath the water column: Uncovering microplastic pollution in the sublittoral coastal sediments of the Canary Islands, Spain

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HIGHLIGHTS

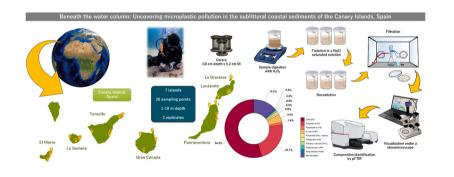
G R A P H I C A L A B S T R A C T

- First prevalence study of microplastics in coastal sediments of the Canary Islands.
- Colorless/translucent cellulosic and polyester fibers were the types of particle most found.
- A correlation of microplastic concentration/wave height was observed for most locations.
- Similar morphotype and composition patterns have been observed in other samples of region.

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ABSTRACT

Marine ecosystems pollution by microplastics (MPs) is a global problem of special concern. The present study examines the prevalence and distribution of MPs and cellulosic particles in sublittoral coastal sediments of the Canary Islands archipelago (Spain). At twenty-six different locations alongside seven islands, three samples were taken parallel to the shoreline between 1 and 10 m depth (n = 78). Sediment samples were primarily digested

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Plastic µFTIR Macaronesia North Atlantic with a H_2O_2 solution followed by four flotations in a saturated NaCl solution. The mean concentration obtained was 3.9 ± 1.6 items/g of dry weight. A similar distribution pattern was observed across all islands concerning particles morphology, color, size and composition: mainly colorless/translucent and blue fibers (60.0%). Additionally, fragments were also found, and to a much lesser extent microbeads, films and tangled messes. Micro-Fourier Transform Infrared spectroscopy analysis of 12.5% of the fibers, showed that they were mainly cellulosic (54.5%) -either natural or semisynthetic- followed by polyester (22.7%) and acrylic (4.5%). The potential correlation between particle distribution in nearshore sediments and wave intensity was also explored. This work provides the first comprehensive report on the current MPs content of the seabed of the region.

1. Introduction

Plastic pollution is one of the most important environmental problems that humanity must face in the present and near future, caused mainly by an extremely high dependency on plastic materials, as well as the mismanagement of plastic waste. It is well known that plastic residues, in particular those in the form of micro and nanoplastics (MNPs), have reached all environmental compartments [1-3] and, in extension, those organisms that live in them [4,5], including humans [6-15].

Research on MNPs is constantly evolving and focusing on different relevant aspects to better understand their impact on the environment and human health. One of those research lines is currently focused on pollution and source assessment, in which studies seek to identify the main sources of MNPs in different ecosystems, as well as their quantity, distribution and composition, in order to elucidate the magnitude of the problem and its origin [16-18]. Another important research line is also related with MNPs transport and dispersion in the environment, whether through ocean currents, winds, rivers, or other mechanisms, which involves tracking dispersal routes and understanding the factors that influence their movement [19-21].

In the case of the marine environment, according to different estimations, approximately up to 23 million tons of plastic are being annually released [22,23]. Plastic pollution makes its way into the oceans via various pathways, including direct input from coastal communities, river and stream runoff carrying plastic debris, and the discharge of wastewater, often containing plastic materials, among other sources. Once in the ocean, meso (5–25 mm) and macroplastics (> 25 mm) fragment over time due to thermal, atmospheric, photo or mechanical degradation processes into microplastics (MPs, size between 1 μ m and 5 mm in their largest dimension) and nanoplastics (size < 1 µm) [24], undergoing long-range transport through ocean currents, though they can also be directly released with that size (i.e. MPs present in cosmetic microbeads or plastic pellets). Particles denser than seawater sink, traveling significant distances through the water column before eventually reaching the ocean floor [25,26]. However, low density MNPs may also increase their density by biofouling processes, and they can also be transported in the water column as a result of their behavior as passive drifters being dragged by the subduction processes of the water mass itself; such movement is associated with the physical processes that condition the transport and ocean dynamics [27,28]. Once in the sediments, which are widely recognized as sinks for MPs [29], they can mobilize, or return to the water column, becoming accessible in the benthic suspensions and therefore to sediment-dwelling organisms, among others [30-32].

Nowadays, it is well known that textile fibers are among the anthropogenic particles mostly distributed in the marine environment, especially in the water column and sediments [33,34]. This includes 100% synthetic fibers like polyester (PES), polyamide (PA), and acrylic (ACR), primarily from laundering [35]. Additionally, it involves cellulosic fibers, with natural ones like cotton and linen, as well as regenerated cellulose fibers like rayon or viscose, often referred to as semisynthetic fibers [36,37]. According to the 2021 Market Report of Textile Exchange [38], in 2021 the global fiber production reached 113 million tons, among which 54% corresponded to PES fibers, followed by cotton (22%) and PA (5%), the second most used synthetic fiber. Viscose

compiles for a 5.1% of the total, while other synthetic fibers like PP, ACR or elastane, represent 5.2% of total production; therefore, it is not surprising that these fibers are the ones mostly found in the marine environment.

Results of a previous work developed in sublittoral seabed sediments of the island of La Palma (Canary Islands, Spain) revealed the presence of relatively high concentration of transparent, blue, black and red fibers most of which had a cellulosic nature (either natural or manufactured cellulose), but also PES, PA or ACR fibers, among others, were found, suggesting as the possible main source the different wastewater discharge points located around the islands [39]. Similar fiber types and distribution patterns were observed in fish and sea urchins from the island of Tenerife [40,41] and recycled wastewater in the island of Fuerteventura [42]. Agricultural soils irrigated with this water also showed the presence of these fibers. These findings highlight the importance of ongoing monitoring studies in the region to understand connections between different environmental compartments, rather than focusing solely on specific sample types or areas.

In an attempt to globally understand MPs pollution in a relatively isolated Atlantic archipelago, we have studied the MPs concentration, morphotype and composition as well as the organic matter content and characteristics of seabed sediments collected from 26 locations in the Canary Islands archipelago. Though it is well known that natural or manufactured cellulose are not considered plastics, they have also been determined in this study, since they are frequently dyed and also released with wastewaters, constituting an additional source of anthropogenic contamination. The potential relationship between particles distribution in sediments near the shore and the intensity of waves was also studied as well as a possible relationship between particles concentration and sediment characteristics. These results, together with those from our previous work developed in the island of La Palma (4 samplings) [39], provides the first general and complete distribution map of anthropogenic particles morphology, size and composition in the whole archipelago, allowing a better understanding of the degree of plastic pollution of the region and, in extension, in other insular territories. This work represents the second study of these characteristics developed in the region and one of the very few studies of this type carried out in Spain (see Table S1 of the Supplementary Material).

2. Materials and methods

2.1. Study area and field work

In this study, we examined 26 locations across seven Canary Islands: Tenerife, La Gomera, El Hierro, Gran Canaria, Fuerteventura, Lanzarote, and La Graciosa. Each island was sampled in 4 locations, except Tenerife, which had 6, Lanzarote, which had 3, and La Graciosa, the smallest, with one location. Fig. 1 and Table S2 of the Supplementary Material show the sampling locations and sampling points characteristics, respectively.

Sublittoral coastal sediment samples were collected using the same sampling method as Villanova-Solano et al. [39]. Briefly, samples were collected in triplicate at each location by scuba divers from July 2020 to May 2021 using stainless-steel cores of 10 cm length and 5 cm diameter at a depth between 1.2 and 10.2 m on a uniform sandy bottom (a Cressi

Leonardo dive computer was used for depth measurements). A total of 78 samples were taken parallel to the shoreline and separated 10 m from each other.

2.2. Sediment samples characterization

Once at the laboratory, the full content of each core was deposited into a glass beaker, mixed and homogenized and covered with aluminum foil. Afterwards, a subsample of ~ 10 g from each core was weighed in a 250 mL glass beaker for MPs determination using an analytical balance (Mettler Toledo XS205 Dual Range with a maximum weighting capacity of 220 g and 0.1 mg of resolution). In order to determine the water content of the sediments, another subsample of 10 g of wet sediment were accurately weighed in ceramic capsules and placed into an oven (Conterm, LED digital, J.P. Selecta®) at 105 °C for 24 h (3 replicates per sample). After that time, the capsules were allowed to cool at room temperature in a desiccator and weighed again until constant weight. The water content was calculated by weight difference. Once the water content was determined, the same sample was used to determine the organic carbon content following the loss on ignition (LOI) method [43,44]. All samples were calcined at 450 °C for 8 h in a muffle furnace (Nabertherm GmbH, Bahnhofstr, Germany). After calcination, samples were allowed to cool at room temperature in a desiccator and again re-weighted to obtain the loss of organic matter after combustion. The particle size distribution was evaluated by sieving 100 g of the previously dried sediment during 5 min on an analytical sieve shaker (Retsch AS 200 digit cA) working at 2.0 mm amplitude, through a standard series of ten sieves with the following mesh sizes: 8, 4, 2, 1, 0.5, 0.25, 0.125, 0.063 and 0.032 mm. The textural group and

sample statistics (median, mean, sorting) were calculated using the software GRADISTAT Version 9.1 following Blott and Pye [45].

2.3. Sediment samples treatment for microplastics extraction

Sediment samples were treated following the methodology applied by Villanova-Solano et al. [39]. Briefly, the full content of each core was mixed (since no significative differences were observed between sediments collected every 2.5 cm) and 10 g of each were accurately weighted in an analytical balance and digested during 2 h with 40 mL of 33% (w/v) of H₂O₂ in order to remove the organic matter (constant stirring at 300 rpm 60 °C). Afterwards, 100 mL of a NaCl saturated solution (approximate density of 1.2 g·cm⁻³) were added and stirring for 1 min; the solution was left for an hour and filtrated under vacuum through a 50 µm stainless-steel filter previously washed with Milli-Q water obtained from a Milli-Q Gradient A10 system from Millipore (Burlington, MA, USA) and introduced in a muffle furnace (Nabertherm GmbH, Bahnhofstr, Germany) for 4 h at 450 °C. The flotation procedure was repeated 4 times. Afterwards, the filters were immediately introduced in Petri dishes and visualized under a trinocular light stereomicroscope with magnifications \times 0.65 – \times 5.5 (Euromex Nexius Zoom EVO, Arnhem, The Netherlands) and with an image analysis system (Levenhuk M1400 PLUS-14Mpx digital camera with the Levenhuk Lite software version x64, 4.10.17659.20200906) to identify and classify particles according to their size, color, and shape. The lower size limit of the particles studied was \sim 90 μm and the viewing time per filter was between 1 and 2 h, depending on the sample. To visually establish if a particle is made of plastic, the criteria of Hidalgo-Ruz et al. was met [46, 47], even though, a subset of samples was confirmed by microFourier

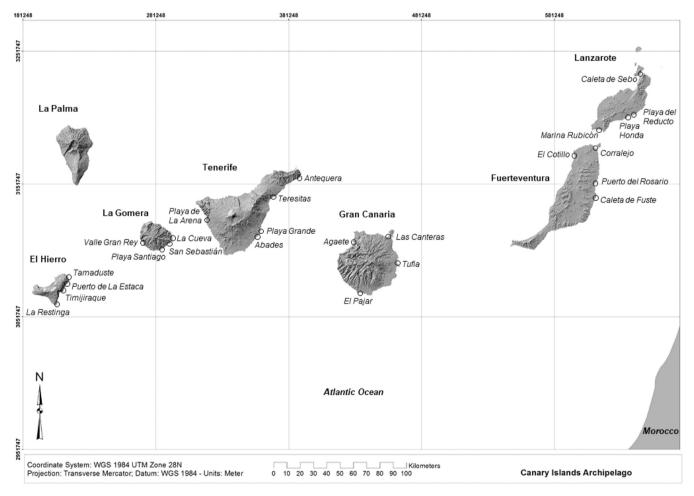


Fig. 1. Geographical location of the Canary Islands archipelago and the sampling locations (marked with cycles).

Transform Infrared Spectroscopy (µFTIR), as it will be later indicated.

2.4. Precautions to avoid sample contamination

One of the fundamental problems of MPs determination at this level is precisely that contamination may occur during sample handling, so it was necessary to take certain precautions to minimize environmental contamination. All sample treatment was carried out in glove boxes located in a "clean room". All material used was plastic-free. Before their use, nonvolumetric glassware was washed with Milli-Q water obtained from a Milli-Q Gradient A10 system, covered with aluminum foil and heated up to 550 °C for 4 h in a muffle furnace, while volumetric glassware was cleaned using a NoChromix® solution from Godax Laboratories (Cabin John, MD) in sulfuric acid (95% w/w, VWR International) for 24 h. Fifty µm stainless-steel filters were also heated up to 450 °C for 4 h in a muffle furnace to remove any contamination. Milli-Q water was also used to prepare the NaCl saturated solution. Both 33% (w/v) H₂O₂ and NaCl saturated solutions were filtered through a polyvinylidene fluoride (PVDF) 0.22 µm filter. A procedural blank (full sample pre-treatment without sediments) was conducted for every set of three samples in parallel to account for potential contamination during the extraction procedure [48]. Table S3 in the Supplementary Material compiles a subset of some of the results of the procedural blanks as an example. In case contamination was found, MPs with the same color and shape as those found in the corresponding control batch were subtracted for each sample, though if a high contamination took place, the complete batch of samples was discharged and the analysis repeated with another portion of sediment. Also, during treatment, the samples were covered with aluminum foil previously cleaned with filtrated Milli-Q water to avoid laboratory contamination during the whole process. Finally, white laboratory coats were also replaced by orange coats to easily detect fibers transfer, since orange fibers were not previously found in the studied samples. In addition, an air purifier (Mi Air Purifier 2 H, Model:AC-M9-AA) equipped with a high efficiency particulate air (HEPA) filter was also used.

2.5. MicroFTIR analysis

A randomly distributed subsample of microparticles (n = 220, n = 20) \sim 12.5% of the total found) that included fibers of each filter, was analyzed by µFTIR using a Perkin-Elmer Spotlight[™] 200 Spectrum Two instrument with a mercury cadmium telluride detector. Microparticles were placed on KBr, which was used as a slide, and their spectra were recorded in micro-transmission mode using the following parameters: spot 50 µm, 32 scans, and spectral range 550-4000 cm⁻¹. All spectra were compared with the Omnic 9.1.26 database (ThermoFisher Scientific Inc., Massachusetts, USA) and with spectra from our own database. Microparticles were considered plastics when the match confidence was at least 70%. Natural (i.e. cotton and linen) and semi-synthetic fibers (i. e. rayon/viscose/cellophane, lyocell/Tencel) as well as cotton and linen with non-natural colors that consist of cellulose, were classified as cellulosic since their spectra are practically identical and, therefore, they are difficult to differentiate, especially in the case of the microparticles found in the environment due to weathering processes. Polyethylene terephthalate (PET) was classified as "polyester" (PES) since it is a thermoplastic polymer resin of the PES.

2.6. Statistical analysis

Statistical methods were implemented using R statistical software and its extension RStudio [49]. The significance level for all tests was set at p < 0.05. The normality of the data was analyzed using the Shapiro-Wilk test, and the homogeneity of variance with Bartlett's test. Since the distribution of the data was not normal, the Kruskal-Wallis non-parametric U-test and post hoc paired *t*-test with Bonferroni correction were applied to determine significant differences in particles concentration (items/g DW of marine sediment) between islands. Spearman's correlation coefficients were calculated to determine significant relationships between MPs concentration and sediment characteristics (i.e., organic matter and particle size parameters).

2.7. Physical oceanographic environment

While various physical processes can transport floating particles from the open ocean to the coast [50], the accumulation of plastic debris near the coast is likely to be predominantly influenced by the effects of the wave field. Specifically, the Stokes drift, a significant physical forcing associated with waves, plays a crucial role [50]. In essence, the Stokes drift results from the asymmetric motion of particles as they are advected by waves: particles experience a vertical orbital oscillation with a forward speed at the sea surface greater than the backward speed at depth, leading to a net forward displacement [51].

The significant height of open ocean waves in the vicinity of the Canary Islands is diagnosed using satellite-based sensors, given the limited availability of in situ measurements. Utilizing sensors installed in satellites, significant wave height data is obtained along the satellite path with a spatial resolution of 7 km. This valuable dataset is acquired from the Copernicus Marine Service, specifically from the product 'Global Ocean L3 significant wave height from NRT satellite measurements' [52], which amalgamates observations from three missions to enhance spatial coverage and temporal resolution.

The fundamental hypothesis driving this study is that oceanic waves can play a significant role in the long-term transportation and accumulation of plastic debris in coastal areas. Fig. 2 depicts the locations where wave observations were recorded from 2020 to 2021, while the red dots indicate the positions of plastic sampling stations. Additionally, circles are drawn to delineate the area influenced by the waves around each specific beach.

To characterize the wave conditions at each sampling point, the average significant wave height is computed by taking the mean of the significant wave height values within the respective area of influence.

3. Results and discussion

3.1. Particle abundance and morphotype

After sample treatment, a total of 2943 particles were identified under a trinocular light stereomicroscope among the 78 sublittoral coastal sediments samples collected in this work. Particles were classified according to their size, color and shape, in the last case as fibers, fragments, microbeads, tangled messes or films.

Table 1 provides a summary of the total items count and average concentrations (items/g DW) for each island, with the average abundance across all islands being 3.9 ± 1.6 items/g DW. In the case of the island of La Palma [39] the average concentration was 2.7 ± 0.8 items/g DW) which aligns with the findings of our study. Fig. S1 in the Supplementary Material offers a visual representation of locations with the highest concentrations. Additionally, Table S4 in the Supplementary Material compiles the quantification and characterization results for particles on each island. Notably, the islands of La Graciosa and El Hierro exhibit the highest particle concentrations among all the islands, primarily due to the abundance of fragments near fishing harbors in Caleta de Sebo (La Graciosa) and La Restinga beaches (El Hierro). These harbors are believed to be the primary sources of plastic pollution in these locations, a phenomenon not observed in other sampling sites.

As can be seen in Table 1, the most common morphotypes found were fibers (60.0%) followed by fragments (39.2%). Microbeads, tangled messes and films had the lowest presence: 0.2%, 0.3% and 0.3%, respectively. Fig. 3 shows images obtained under the stereomicroscope of some of the particles identified, including fragments (Fig. 3a), a tangled mess (Fig. 3b), fibers (Figs. 3d and 3f), a microbead (Fig. 3c) and a film (Fig. 3e).

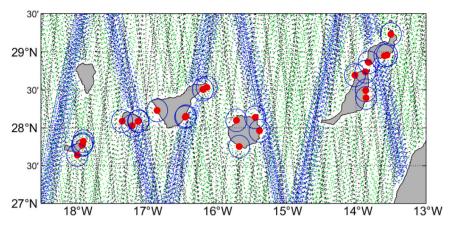


Fig. 2. Locations where significant wave height observations are available, indicated by blue, green, and black dots, representing the respective satellite missions. Additionally, red dots represent the locations where sediment samples were taken. Circles are used to denote the area of influence for each beach, within which significant wave height observations are spatially and temporally averaged.

Table 1

Total items, shape and percentage of the particles found, average concentrations and standard deviation of the microparticles found in the seabed sediments of the Canary Islands (Spain).

Island	Total items	Shape of the particles found	Average concentration (items/g DW)
La Graciosa	838	Fibers (14.2%, $n = 119$) Fragments (85.4%, n = 716) Films (0.2%, $n = 2$) Tangled mess (0.1%, n = 1)	16.5 ± 5.6
Lanzarote	39	Fibers (82.1%, $n = 32$) Fragments (17.9%, $n = 7$)	0.3 ± 0.2
Fuerteventura	201	Fibers (91.1%, $n = 183$) Fragments (8.9%, $n = 18$)	1.1 ± 0.3
Gran Canaria	133	Fibers (100.0%, n = 133)	0.7 ± 0.3
Tenerife	377	Fibers (91.0%, $n = 343$) Fragments (8.7%, $n = 33$) Film (0.3%, $n = 1$)	1.4 ± 0.8
La Gomera	200	Fibers (97.5%, n = 195) Fragments (1.5%, n = 3) Films (0.5%, n = 1) Microbeads (0.5%, n = 1)	1.1 ± 0.4
El Hierro	1155	Fibers (66.0%, $n = 762$) Fragments (32.7%, $n = 377$) Tangled mess (0.6%, $n = 7$) Microbeads (0.4%, $n = 5$) Films (0.3%, $n = 4$)	6.2 ± 3.3
Total	2943	Fibers (60.0%, $n = 4$) Fibers (60.0%, $n = 1767$) Fragments (39.2%, n = 1154) Tangled mess (0.3%, n = 8) Microbeads (0.2%, $n = 6$) Films (0.3%, $n = 8$)	3.9 ± 1.6

* The total items correspond to the particles found across all locations on each island, with triplicate samples (3 cores per location). Fibers longer than 5 mm have been excluded from the counting.

Concerning the color distribution, a similar pattern can be observed within the different locations and islands suggesting a common origin for these contaminants including the case of the island of La Palma previously published in Villanova-Solano et al. [39]. Fig. 4 displays the distribution color of the fibers per island (upper figure) as well as that of the fragments (lower figure). As it can be seen, most fibers were colorless/translucent (68.8% of the total), followed by blue (18.3%) and black (6.2%). Red, pink, yellow, green, grey, brown and purple fibers were also found, although in a lesser amount (see Fig. S2 of the Supplementary Material). However, with respect to the fragments, the color pattern changes; a high percentage of them (62.2% of the total) were blue, followed by green (21.9%) and red (10.9%). Yellow, white, pink, orange, black and purple fragments were found in a lesser extent (see Fig. S3 of the Supplementary Material).

Regarding the size of the fibers, Fig. 5 displays the histogram of the size and color distribution per island, while Fig. S4 of the Supplementary Material exhibits the overall size and color distribution. As can be seen from the figures, the size pattern is similar between islands, being significantly most abundant those with sizes between 500 and 1000 μm (25.0% of the total, n = 600 fibers). The average fibers length was 1573 \pm 1028 μm . It is worth noting that 62 of all the fibers found had a size larger than 5 mm with the longest fiber being 14,685 μm (these 62 items were not considered for counting purposes).

For comparison purposes, Fig. 6 exhibit the box and whiskers plots of the concentration of (items/g DW) of the sampling points of the seven islands while Figs. S5-S16 of the Supplementary Material show the box and whiskers plots distributed by shape and location at each island. As it can be seen in the Fig. 6, a significantly higher concentration of particles (p < 0.05) was found on the island of La Graciosa compared to the rest of the islands. It should be noted that on the island of La Graciosa only one location, Caleta de Sebo, was sampled. However, the amount of particles was extremely high 16.5 \pm 5.6 items per g of DW sediment. It is also worth noting that the concentrations at the island of El Hierro, although not significantly different, were also high, with the locality of La Restinga standing out (corresponding to the outliers, for more information see Fig. S16 of the Supplementary Material). This could suggest a possible difference in the accumulation or origin of the anthropogenic particles at each island. Furthermore, it coincides with the fact that the two sampled locations Caleta de Sebo and La Restinga are harbors so MP contamination could be of local origin, as previously commented.

3.2. Microplastics composition analysis

A total of 220 fibers (~12.5% of the total fibers) were analyzed by μ FTIR. Fig. 7 displays the fiber composition, revealing that 54.5% (120 particles) were cellulosics, of which 69 were colorless/translucent fibers and 51 were colored fibers. As indicated in the experimental section, natural fibers (e.g. cotton or linen) and semi-synthetic fibers (e.g. rayon or viscose) were classified as cellulosic because their spectra are practically identical especially when it comes to the MPs found in the environment due to weathering processes. Weathering causes a decrease in the mechanical properties of cellulose, by weakening it [53], being one of the most noticeable effects of the weathering processes the

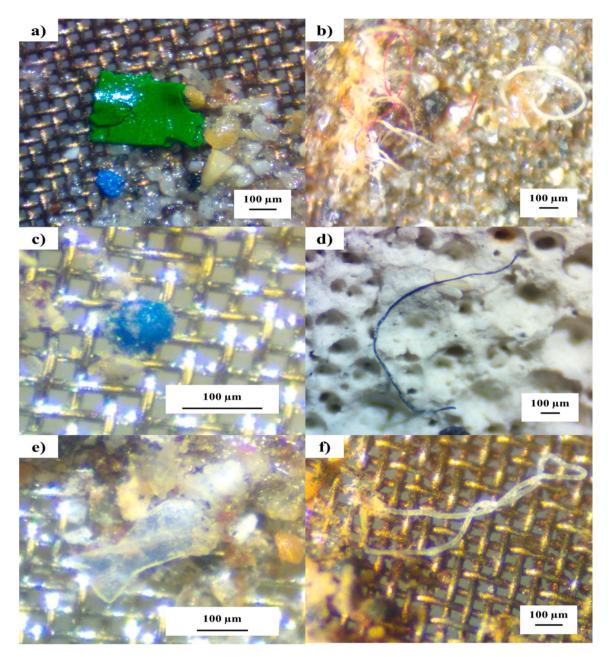


Fig. 3. Stereomicroscope photographs of the particles found in sublittoral coastal sediments of the Canary Islands: a) green and blue fragments from La Graciosa, b) red and transparent tangled messes from El Hierro, c) a blue microbead from La Gomera, d) a blue fiber from Fuerteventura, e) a transparent film from La Gomera and f) a transparent fiber from El Hierro.

formation of carbonyls groups in the cellulose chain [54]. Nevertheless, in another work such as the study conducted by Mušič et al. [55] it was confirmed that the FTIR spectra exhibited no shifts in peak positions; instead, they revealed differences in the behavior of the aged samples [55].

Concerning the rest of the fibers, 22.7% (50 particles out of 220) were identified as PES fibers. The remaining fibers varying from 4.5% to 0.5% included acrylic, PE, PA (including Nylon), PS, PVC, PP and PUR with 13.2% (twenty-nine particles) remaining unidentified. Consequently, 32.3% of the analyzed particles consisted of entirely synthetic fibers, mirroring a similar pattern among the islands. These results also align with our previous research carried out on the sediments of the island of La Palma [39].

Since 65.0% of the world fiber production are synthetic fibers (28.0% are plant fibers like cotton, 1.6% are animal fibers like wool and 6.4% are manmade cellulosic fibers), it is not surprising that they appear

in the environment, in particular, in the marine environment.

3.3. Comparison with previous studies

Regarding the composition of the microfibers, the morphotype and color distribution, they all followed a similar pattern to other previously reported studies from the region, including our previous work in sediments of the island of La Palma [39], as well as biota (sea urchins and cultivated fish) and recycled wastewater and soils irrigated with such water. In the work of Sevillano-González et al. [41] authors studied the presence of anthropogenic particles in individuals of *Diadema africanum* sea urchins collected at two sampling points in Tenerife, being fibers the prevalent morphotype (97.5%) followed by fragments (1.9%) and films (0.6%), which were all mainly blue (43.3% and 47.0% in the two sampling points) and colorless/white (32.5% and 39.5%). Concerning the composition, µRaman analysis showed that they were mainly

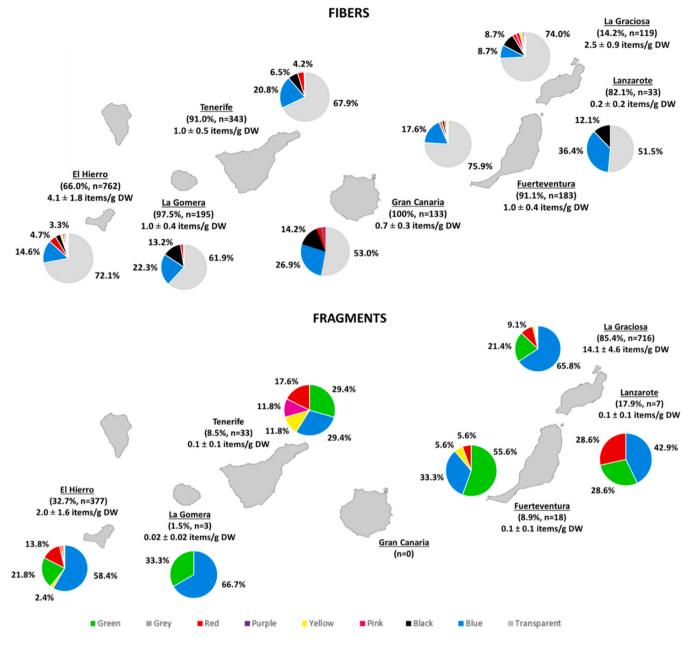


Fig. 4. Color distribution of the fibers (upper map) and fragments (lower map) and average concentrations in each island of each morphotype of the particles found in sublittoral coastal sediments of the Canary Islands.

cellulosic (46.0%), followed by PP (24.3%) and PET, a PES (24.3%). This study, together with the data reported in the present work, suggest that a transfer of such particles could be taking place from the seabed sediments to sea urchins, as a result of their grazing activity. A similar pattern was also observed by Sánchez-Almeida et al. [40] in the gastrointestinal tracts of individuals of cultivated European sea bass (Dicentrarchus labrax, n = 45) and gilt-head sea bream (Sparus aurata, n = 41) from Tenerife. Most of the items found were colorless (47.7% for Dicentrarchus labrax and 60.9% for Sparus aurata) and blue (35.3% vs. 24.8%) microfibers, with an average length of 1957 \pm 1699 μm and $1988 \pm 1853\,\mu\text{m},$ respectively. μFTIR spectroscopy also showed the prevalence of cellulosic fibers together with PES, PAN, and poly (ether-urethane). Both fish species were cultivated near the sea surface, which suggests the existence of the same distribution pattern in the sea water column. Though it was not possible to analyze the sea water in this work, in a previous work from Pérez-Reverón et al. [42], the occurrence of anthropogenic particles in four recycled wastewaters

(RWWs) used for soil irrigation in the island of Fuerteventura was studied. Results showed the prevalence of cellulosic and PES fibers (between 84.4% and 100%) of blue and colorless colors (up to 55.6% and 33.3%, respectively), with an average length of 787 \pm 812 μm in the water samples. Since the Canary Islands region is surrounded by a high number of wastewater discharge points, wastewater could represent a main source of this type of particles in the marine habitat. Besides, studies carried out by our research group in which MPs have also been determined in several wastewaters (data not published yet) also agree with this issue.

Table S1 of the Supplementary Material compiles some of the most recent works in which MPs have been determined in seabed sediments. In most of the studies fibers have been the prevalent morphotype [56-66], though in some cases fragments have prevailed. This is the case of deep-sea sediments (1655–3062 m depth) in Australia [67] or in South China (1–38 m) [68] probably associated with the fact that the sampling points are close to areas with a high human activity, as pointed

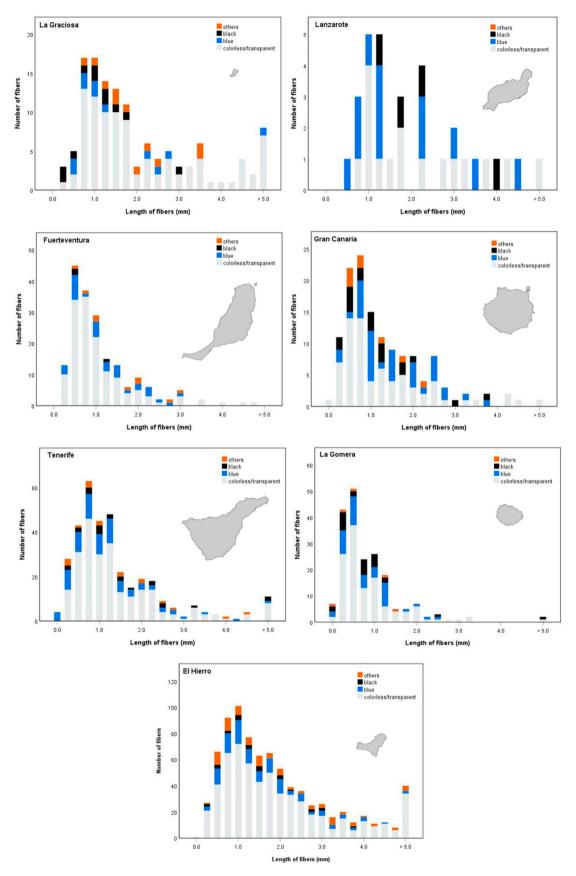


Fig. 5. Histograms of the size and colors distribution of fibers found in sublittoral coastal sediments per island (n = 1829). Particles longer than 5 mm have also been shown for comparison purposes only.

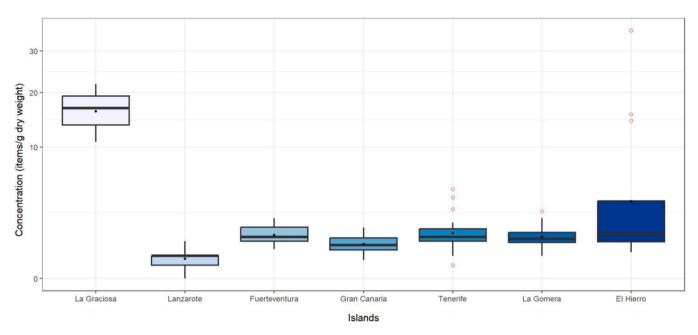


Fig. 6. Box and whisker plots of the concentration (items/g DW) of the anthropogenic particles found in the sublittoral coastal sediments of seven Canary Islands.

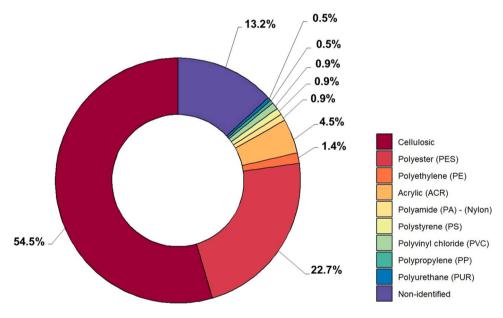


Fig. 7. Distribution of the composition of the fibers found in the sublittoral coastal sediments of the Canary Islands (n = 220). "Cellulosic" includes natural and semisynthetic cellulose (see Experimental Section for more details).

by the authors. Concerning the colors, colorless/transparent and/or blue have prevailed in many cases [57,59,63,64,68,69]. If the composition is considered, cellulosic fibers have only been studied on few occasions [56,61], cases in which they accounted for the highest percentage, while 100% synthetic polymers like PES have also prevailed in several works [59,60,69]. Regarding works previously carried out in Spain, those carried out in the Mediterranean coast, [57,60,61] and in NW Spain [70] exhibit similarities with our work concerning the morphotype and color, but not concerning the composition since PE, PP and PS (low density polymers) have been mostly found, except in the work of [60], in which PES prevailed followed by PA.

Finally, it should also be mentioned that the concentrations found in our work are higher than the ones reported in most of the works compiled in Table S1, probably as a result of the four flotations carried out which could improve the recovery percentage of the particles. Despite several differences and similarities have been found, it should be taken into consideration the existing differences concerning sampling strategies, sample treatment as well as MPs identification techniques.

3.4. Relationship between particles concentration and sediment characteristics

Various physicochemical characteristics of sediment samples from the different selected locations, grouped by islands, are presented in Table S5 of the Supplementary Material. The average organic matter content in the sediments was 17.3 g kg⁻¹, ranging from a minimum of 3.4 g kg⁻¹ at Timijiraque Beach (El Hierro) to a maximum of 60.6 g kg⁻¹ at Marina Rubicón (Lanzarote). No significant correlation was observed between organic matter content and MPs concentration (r = -0.023; p = 0.912) indicating that these two components may be linked to different sources and/or subject to different transport and sedimentation mechanisms. Surprisingly, the area with the highest organic matter content, Marina Rubicón in Lanzarote, had the lowest average concentration of particles (0.2 \pm 0.2 items/g DW). This lack of correlation in marine sediments has been noted by various authors in different regions of the planet [71-75]. Regarding grain size distribution, the sediments ranged from sandy-to-sandy gravel textures, and particle sorting varied from poorly sorted to moderately well sorted. Similarly to what occurred with organic matter, none of the granulometric statistical parameters showed a significant correlation with MPs contents (for example for the median values r = 0.125; p = 0.542). This suggests that the size and distribution of the particles comprising the sediments do not appear to be critical factors influencing the retention of these contaminants. Similar results were also found in our previous study carried out in La Palma [39]. In this regard, Harris [76] hypothesizes that in the case of particles with very low density, such as fibers, there should not be a relationship since both the sediment and MPs are transported to the same location by different processes.

3.5. Physical mechanism behind particles distribution

The distribution of MPs in the marine environment is influenced by various forcing mechanisms operating at different spatio-temporal scales [50]. For our study, focusing on MPs located near the shore and within sediments, the primary process likely responsible is wave action in the coastal zone. Waves play a significant role in transporting MPs from the open ocean to coastal areas, where they can either be driven towards the swash zone or become buried within the sediments due to wave impacts.

In this section, we investigate the potential relationship between MP distribution in sediments near the shore and the intensity of waves. Specifically, we analyze the significant height of waves, a parameter indicative of wave intensity. Our samples were collected from diverse locations such as beaches and within harbors. Since the distribution of MPs and their abundance within harbors might be influenced by long-term accumulation effects rather than wave action, these observations are excluded from our comparison.

Additionally, we excluded a few locations (Antequera in Tenerife, La Cueva and Puerto Santiago in La Gomera, Puerto la Estaca and La Restinga in El Hierro, Agaete in Gran Canaria, Corralejo, Puerto del Rosario and Caleta de Fuste in Fuerteventura, and Caleta de Sebo in La Graciosa) where the wave significant height was lower than 1.2 m, as the accumulation of plastics in these areas could be influenced by a different dominant physical forcing that still needs further investigation. With the resulting database, we can now explore the potential relationship between wave effects and particles accumulation within sediments in the coastal area (Fig. 8). The analysis reveals a discernible trend, wherein higher concentrations of particles are observed in sediments at locations with elevated significant wave heights. When fitting this data to a linear regression model, it yields a correlation coefficient of 0.59 (p < 0.05). This suggests that waves likely play a crucial role in driving MPs towards the near shore zone and facilitating their efficient burial within the sediments.

4. Conclusions

The analysis of sublittoral seabed sediments across the Canary Islands archipelago, situated in the Atlantic Ocean, has unveiled a consistent morphological and compositional pattern among anthropogenic particles across all the islands, despite the geographical distances separating them. Notably, colorless/transparent, and blue fibers composed of cellulose and PES were predominant. However, in two fishing harbors on the islands, a notable concentration of fragments of various colors was also observed. In the first instance, drawing from prior research within the islands, it is probable that these fibers originate from the numerous wastewater discharge points encircling the islands.

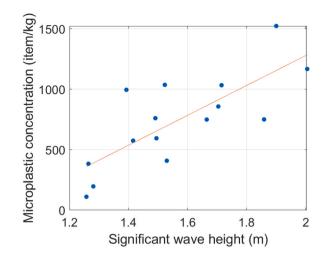


Fig. 8. Relationship between the significant wave height and the accumulation of MPs within the sediments at several beaches in the Canary Islands.

In the second scenario, relating to the fragments, their likely origin is local, especially in proximity to the harbors. Furthermore, locations experiencing elevated significant wave heights exhibited higher concentrations of particles in the sediments. This study confirms the presence of MPs and cellulosic microparticles in marine sediments on oceanic islands, providing the initial assessment of the sublittoral seabed status in the region.

When comparing these findings with previous studies in the same region, which involved the analysis of living organisms such as sea urchins and cultivated fish, it suggests a dynamic exchange of these particles between sediments and seawater, as well as their incorporation into the biota. Notably, a consistent morphological and compositional profile has been observed in these organisms, reinforcing the interconnected nature of this relationship.

Environmental implication

Microplastics constitute nowadays an important group of emerging and ubiquitous contaminants since they are present in all environmental compartments and cause important environmental problems also to biota. In the particular case of humans, there are still important issues that are not yet fully known. This work provides a better overview of the current state of sublittoral coastal sediments from oceanic islands concerning plastic pollution, establishing also a correlation between the distribution of anthropogenic particles in sublittoral coastal sediments and wave intensity, providing valuable insights into the relationship between these factors.

CRediT authorship contribution statement

Cristina Villanova-Solano: Conceptualization, Methodology, Validation, Formal Analysis, Investigation, Data curation, Writing Original-Draft, Writing -Review & Editing, Visualization. Francisco J. Díaz-Peña: Conceptualization, Methodology, Formal Analysis, Resources, Data curation, Writing Original-Draft, Writing -Review & Editing. Cintia Hernández-Sánchez: Conceptualization, Methodology, Validation, Formal Analysis, Writing Original-Draft, Writing -Review & Editing. Javier González-Sálamo: Conceptualization, Methodology, Validation, Formal Analysis, Writing Original-Draft, Writing -Review & Editing. Carlos Edo: Methodology, Formal Analysis, Investigation, Data curation, Writing Original-Draft, Writing -Review & Editing. Daura Vega-Moreno: Methodology, Software, Formal Analysis, Investigation, Data curation, Writing Original-Draft, Writing -Review & Editing, Visualization, Project administration, Funding acquisition. Sonia Fernández-Martín: Investigation, Writing Original-Draft, Writing -Review & Editing. Eugenio Fraile-Nuez: Methodology, Software, Formal Analysis, Investigation, Data Curation, Writing Original-Draft, Writing -Review & Editing, Visualization. Francisco Machín: Methodology, Software, Formal Analysis, Investigation, Data Curation, Writing Original-Draft, Writing -Review & Editing, Visualization. Javier Hernández-Borges: Conceptualization, Methodology, Validation, Formal Analysis, Resources, Data Curation, Writing Original-Draft, Writing -Review & Editing, Visualization, Supervision, Project administration, Funding acquisition.

Conflict of interest

Authors declare no conflict of interest.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.133128.

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