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# EFFECTS OF DISTANCE TO THE SEA AND GEOMORPHOLOGICAL CHARACTERISTICS ON THE QUANTITY AND DISTRIBUTION OF MICROPLASTICS IN BEACH SEDIMENTS OF GRANADA (SPAIN)

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## ABSTRACT

Microplastics became an unprecedented challenge and mapping their contamination all over the world is needed in order to establish baseline levels and identify the polymers in order to enhance adequate legislation and policy. The main objective of this study is to demonstrate the existence of microplastic pollution on three beaches on the coast of Granada (Spain), namely La Herradura, Motril Beach and La Rábita, characterizing the particles and the relationships in their distribution. This may contribute supporting the studies carried out at a national level in accordance with the Directive on Marine Strategy (2008/56/EC). The results showed a greater median concentration of particles/kg of dry sediment in La Herradura (45.0 ± 24.7) than in Motril (31.5 ± 21.5) and La Rábita (22.0 ± 23.2). These data revealed a higher contamination by microplastics in an enclosed bay-type beach (La Herradura) in comparison with open delta-type beaches. The predominant morphologies were microspheres and fragments, with maximum median concentrations of 38.0 ± 23.7 and 6.0 ± 0.7 particles/kg, respectively. The distribution and size of the particles is affected by the geomorphological and sedimentary characteristics of these beaches, which are different from any other in Spain and in the Mediterranean in general. The beaches of Granada showed more microplastic contamination than Greek or Slovenian beaches, but less than other Spanish beaches. In this area of the Mediterranean, the presence of microplastics can be affected by the wind, sea currents or methodological aspects such as the pore size of the filters used. All of these factors were analysed when comparing the beaches of Granada with other Mediterranean beaches. This study shows that there is contamination by microplastics on the beaches of Granada, which have been little explored until now due to the difficult geological and granulometric characteristics, and gives support to other national studies.

**Keywords**: Microplastics; Beach pollution; Mediterranean; Marine litter; Spanish coast

## 1. INTRODUCTION

Contamination by microplastics (MPs) has become a problem due to the potential environmental damage produced and the harmful effects on organisms. These particles can be primary, manufactured by humans with a purpose, such as microbeads added to personal care products, in drilling fluids for extracting oil or gas, sandblasting for cleaning or boat paints (Duis and Coors, 2016). Microplastics can also be secondary, which result from the physical and chemical degradation of macroplastics in the environment, such as those produced by the tyres wear, washing of synthetic clothes or by physical-chemical degradation of larger microplastics (De Falco et al., 2018; Karlsson et al., 2018).

Microplastics are found in almost every marine and freshwater environment on Earth, including beach sediments, bottled water or food (Novotna et al., 2019; Shahul Hamid et al., 2018). Furthermore, microplastics are present in all types of environments, including those considered to be pristine or distant from the sources of production of these particles, such as the depths of the oceans or Arctic ice (Obbard et al., 2014; Woodall et al., 2014).

One of the coastal and marine environments most affected by plastic pollution is the Mediterranean Sea. The first study that reported the abundance of floating plastic debris in the Mediterranean Sea was conducted by Morris, 1980, who determined a concentration of about 1300 items per km$^2$ near Malta. Those small plastic fragments were found to be approximately 1.5 cm in diameter. Currently, it is estimated that the average plastic concentration in Mediterranean surface waters is 243,853 items per km$^2$ (Cózar et al., 2015) with sizes between 0.2-500 mm, being the highest concentration of particles located between 0.5-2.5 mm . Some factors that contribute to the issue of marine litter are the intense coastal and maritime tourism, the great flow of navigation, the industrial and fishing activities and also the configuration of the Mediterranean sea as a closed basin with a particular system of currents (Cincinelli et al., 2019).

Particularly, the Spanish coast is highly affected by intense tourist activity. In Spain, CEDEX (Centro de Estudios y Experimentación de Obras Públicas) has been collaborating since 2014 in the Monitoring Programme for Microparticles on beaches, according to the Framework Directive on Marine Strategy (2008/56/EC) (CEDEX, 2018). The microparticle monitoring program on beaches (Subprogram BM-6) includes the collection of samples and laboratory determinations. Within this programme, the following beaches in contact with the Mediterranean Sea were monitored: Las Azucenas (municipality of Motril, Granada), San Miguel in Cabo de Gata (Almería), La Llana in San Pedro del Pinatar (Murcia), Marenys in Tavernes de Valdigna (Valencia), Cal Francés in Viladecans (Barcelona) and La Pineda, in Vila Seca (Tarragona).

Under the same programme, the beach of Carchuna (Granada) was chosen in 2016 as a reference for monitoring in the Strait and Alboran Sea Demarcation. However, the granulometric conditions of the coast of Granada have degraded to such an extent that it is impossible to find sandy sediment in the area of the breaker line at present as it is mainly made up of gravel. Such conditions, which are indicative of a significant erosive action of the waves, are not suitable for microplastic particles carried by the sea to be deposited on the beach. If the sand particles themselves, of much higher density, do not remain there in a stable manner, it is less likely that microplastic particles will be retained. Kazmiruk et al., (2018) demonstrated, through sampling in different coastal areas of British Columbia, that higher concentrations of microplastics were found in fine sediments (size <250 µm). Also, Maes et al., (2017) found a negative relationship between sediment grain size and the amount of microplastics in seabed sediments through statistical analysis, which means that more particles can be found in areas with fine sediment (<500 microns).

Therefore, in the autumn of 2017, CEDEX decided to investigate whether any other beaches nearby might be eligible for inclusion in the Subprogramme. After visits to certain beaches, samples were taken and analysed at Las Azucenas beach, in a sector close to the port of Motril, and due to the good results obtained, it was incorporated into Subprogramme BM-6 to replace Carchuna beach. However, the Strait and Alboran Demarcation presents some particularly important features, such as the transition between the Mediterranean and the Atlantic, together with the special problem of pollution by plastics from intensive greenhouse agriculture in that area. Furthermore, the far western part of the Granada coast has not been studied to the best of the authors' knowledge.

Therefore, the present study aims to analyze mixed sand and gravel sediments from beaches on the coast of Granada, from the far eastern point to the far western point, considering beaches not analyzed so far by CEDEX. The main objective is to demonstrate the existence of microplastic pollution on these beaches and characterize the particles and the relationships in their distribution thus supporting the studies carried out at a national level in accordance with the Directive on Marine Strategy (2008/56/EC).

## 2. MATERIALS AND METHODS

### 2.1. Area of study

The province of Granada is located in the southeast of Andalusia, in the south of the country. Its area covers 12,635 km² and has a population of 914,678 inhabitants (2019), of which approximately 25% live in the capital. The main population centers are Granada capital and its metropolitan area (55%), followed by the Granada Coast (10.5%) with the municipalities of Motril and Almuñécar as the main population centers (INE, 2019). The main characteristic of the relief of the province of Granada is related to the difference in altitude. Throughout the province there are two major geological units: the Betic Mountain chain and the Neogene Depressions. In the mountain range of Sierra Nevada (Penibetic Range) is located the peak Mulhacén (3,479 m), the highest peak in the Iberian Peninsula. The distance between that peak and the coast line of Granada, in a straight line, is only 35 kilometers (Agencia Andaluza de la Energía, 2010).

The coastline of Granada is extremely steep. Beaches and coastal plains are unusual on this stretch of coast, where the mountains and the sea are in contact. The forms due to the active erosion of the sea front dominate to the deposit and sedimentation formations. At present, these coasts are experiencing an intense transformation of the landscape due to the competition between land uses: tourist development, the expansion of greenhouse and crop agriculture, and the road and transport infrastructure.

La Herradura is a town that belongs to the municipality of Almuñécar, located in the western end of the region of Granada coast, four kilometers from the border with the province of Malaga. In 2018 it had a population of 3,995 inhabitants. La Herradura is located on the shores of the Mediterranean, in a bay that has a single beach, divided into three parts and surrounded by cliffs. Motril is located near the estuary of the Guadalfeo River, dominating the plain of this river, an area of numerous tropical crops. It has a population of 58 020 inhabitants (INE, 2019) and its geographical position makes it an important industrial and commercial centre in which the manufacture of paper and the commercial and fishing port of Motril stand out. Finally, La Rábita is a town belonging to the municipality of Albuñol, located on the eastern side of the coast of Granada, four kilometers from the border with the province of Almeria. In 2018 it had a population of 1,985 inhabitants. It is located in a semi-desert area, on the plain at the estuary of the Albuñol wadi, surrounded by mountains and cliffs.

The climate along the entire coast of Granada is subtropical, with 320 sunny days per year. It is protected by the Lújar mountain range to the north, which slows down the cold winds of Sierra Nevada, and the warm Alboran sea to the south, which acts as a thermal regulator. The average annual temperature is between 17 and 18 ˚C, with warm summers (around 25-30 ˚C) and mild winters (around 12-15 ˚C). As for rainfall, it is usually scarce throughout the year, with December and January being the rainiest months with rainfall of around 40 mm.

Sand samples were taken from three beaches located in the described municipalities: La Herradura (municipality of Almuñécar), Motril Beach (municipality of Motril) and La Rábita (municipality of Albuñol) (Figure 1). Two beaches located at the western and eastern extremes (La Herradura and La Rábita, respectively) and one beach located in the central part of the coast of Granada (Motril Beach) were chosen, with the aim of cover all the tropical coast with representative results. Samples were taken in November 2018.

## 2.2. Sediment sampling

Sediment sampling was conducted following a modification of the method described by Besley et al., (2017). On each of the chosen beaches, four 100-metre-long lines were marked, coinciding with the following beach areas: wave breaking line, high tide line, supralittoral zone 1 and supralittoral zone 2 (close to the promenade) (Fig. 2). The separation between the lines was decided on the basis of the relief of the beaches, and was finally as follows: 2.5 metres between the first and second lines, 7 metres between the second and third lines and 10 metres between the third and fourth lines.

For the sampling, a 50x50 cm wood square was designed to avoid contamination by plastics. On each 100-metre-long line, a sampling was made every 10 metres, with a total of 11 samples per line and 44 samples per beach. Since 0.25 $m^2$ was sampled at each point, the total area sampled at each beach was 11 $m^2$. At each sampling point, 5 subsamples were taken, one at each corner of the square and one at the centre. The samples were taken with a glass beaker, deepening to the first 5 centimetres, and were sieved *in situ* with a 5 mm stainless steel sieve. At the beaches of Granada it was necessary to sieve due to differences in sediment size, in order to obtain an homogeneous sediment within the microplastic size range (< 5 mm). Finally, each sample was then stored in a sealed aluminium container and transported to the laboratory, where they were stored in a dry, dark place.

## 2.3. Microplastics extraction

First, the sediment was dried in an oven at 60ºC for 24 hours. Higher temperatures are not recommended, as the sand forms agglomerates and makes sieving difficult. All dry samples belonging to the same line were mixed in a hermetically sealed bucket. After this, two replicates of 500 g of dry sediment were taken, one from the top and another from the bottom of the bucket. This was done to prevent some polymers such as polyethylene terephthalate (PET) or polyvinyl chloride (PVC) from being neglected for their high density.

The extraction of the microplastics from the sediment was done by density difference, using a saturated solution of NaCl. For this purpose, 350 g of sodium chloride AGR ACS from Labkem were added per litre of water, using water from the public network. The solution was subsequently filtered under vacuum system, using cellulose filters FILTER-LAB 1242, with pore size of 7-9 µm, to remove possible impurities from the salt and microplastics present in the sodium chloride. The density of the obtained solution was 1.20 g/ml.

To make the extraction, the sample of 500 g of dry sediment was placed in a glass vessel and NaCl solution was added respecting a solid-liquid ratio of 1:4. The sediment was stirred manually for 5 minutes with a stainless-steel spatula and then left to rest for 24 hours. After this time, the supernatant was filtered by means of a siphoning system to avoid turbidity of the supernatant with the finer sediment. In this process, PRAT DUMAS glass fiber filters of 47 mm diameter and pore size 1.2 microns were used, and then were stored in glass Petri dishes. Although the NaCl solution was filtered using a pore size of 7-9 microns and therefore contamination could be present in the glass fiber filters, this contamination would not be detectable as subsequent analysis procedures, following the method developed by Prata et al., (2019a), only allow detection of particles larger than 50 microns. Cotton laboratory coats were used throughout the extraction process and analysis of microplastics, in order to prevent contamination from airborne textile fibers.

## 2.4. Microplastics analysis

First, the larger microplastics were removed from the filters using tweezers and analyzed in a Perkin Elmer Fourier Transform Infrared Spectrometer, model Spectrum 65, at the range of 4000-400 cm$^{-1}$ with a resolution of 2 cm$^{-1}$, to determine their polymer composition. The Carbonyl Index was also calculated for these larger microplastics, which is often used as an indicator of the presence of carbonyl groups, which may result from polymer degradation (Rodrigues et al., 2018). Peaks of the infrared spectrum of carbonyl groups are often formed when the polymer undergoes degradation, to calculate the Carbonyl Index, the absorbance of carbonyl peaks must be compared with the absorbance of a reference peak in the same sample. Therefore, the calculation of the Carbonyl Index is expressed as follows:

$$Carbonyl\ Index\ (CI) = \frac{Absorbance\ carbonyl\ peak}{Absorbance\ reference\ peak} \qquad \text{(I)}$$

The region of the spectrum where the carbonyl groups formed in the degraded polymers usually appear is between 1700-1800 cm$^{-1}$ (Fernández-Barranco, 2015; La Rubia, 2014; Rodrigues et al., 2018). Likewise, there is a zone of the spectrum that remains unaltered in most polymers even if they are exposed to the environment, which is usually found between 1450-1470 cm$^{-1}$. Within this range are the reference peaks to be considered for calculating the Carbonyl Index according to Eq (I). The relative level of photo-oxidation related to carbonyl groups can be expressed as low (CI between 0 and 0.15), medium (CI between 0.16-0.30) and high (CI > 0.31) (Rodrigues et al., 2018).

Then, filters on the filtration system were stained with 1 ml of Nile Red pigment with a concentration of 0.01 mg/ml, following the procedure described by Prata et al., (2019b). It was left to react for 5 minutes, and then the filters were washed with Milli-Q water and placed on Petri dishes again for drying. Once dried, they were examined using ultraviolet (UV) light at 254 nm which has been shown to fluoresce for the largest number of different polymers (Prata et al., 2019a). Finally, photographs were taken with a professional camera and were then analyzed using the ImageJ software, which measured the sizes of each microplastic, taking as a reference the size of the filter (which is 47 mm in diameter) to make an estimation of the size. Then, microplastics were grouped in five size categories (<100; 100-200; 200-300; 300-500; >500 µm) (adapted from Simon-Sanchez et al., 2019).

### 2.5. Statistical analysis

All concentrations of microplastics have been expressed with median values and the interquartile range (IQR), as in some cases the data did not follow a normal distribution.

When the samples were processed and the results obtained, different types of statistical analyses were carried out with the SigmaPlot 12.0 plotting and statistics software. The type of analysis carried out depends on certain factors being satisfied, and in the present study the following analyses have been performed: one-way ANOVA for samples that followed a normal distribution; Kruskal-Wallis as equivalent to ANOVA for samples that did not follow a normal distribution and equality of variances; t-student for related samples that followed a normal distribution, when only two groups needed to be compared. An α=0.05 was considered.

## 3. RESULTS AND DISCUSSION

### 3.1. Visual and FTIR identification of microplastics

Visual recognition of the sediment *in situ* along the sampling on the three beaches allowed the identification of only 14 microplastics, which were collected with metal tweezers and stored for later analysis by FTIR (for polymer identification and carbonyl index calculation). A total of 6 particles were identified in La Herradura, 5 in La Rábita and 3 in Motril. With respect to the total area sampled in each beach (11 $m^2$), the number of microplastics identified *in situ* would be equivalent to 0.56 particles/$m^2$ at La Herradura, 0.44 particles/$m^2$ at La Rábita and 0.28 particles/$m^2$ at Motril. The scarce presence of microplastics that can be visually identified is mainly due to the very thick sediment that characterizes these beaches, which makes the substrate very porous and easy to be constantly removed by the waves, so it is almost impossible for a particle to settle and remain there for some time (Bergillos et al., 2016). In addition, the density of the polymers (between 1.38 g/$cm^3$ for PVC and 0.85 g/$cm^3$ for PP) is much lower than the density of the sediments (which is between 2.5-2.9 g/$cm^3$ depending on the mineral composition of the sediment) (King and Galvin, 2002; Sharma, 2019). This means that, with a thick sediment that moves very easily when it is hit by waves, a particle of lower density such as a polymer cannot stay on a surface for long.

When the particles were analysed by FTIR a total of four different compositions were found: polyamide (PA), polypropylene (PP), polyethylene (PE) and polyethylene terephthalate (PET). Of the 14 particles found, 5 were composed of PE, 4 of PA, 4 of PP and one of PET. As the number of visually recognisable MPs on site was very low, the sampling line from which they originated was not taken into account. An example of the infrared spectrum of each of the polymers obtained can be seen in Figure 3, together with an example photograph of some of the microplastics analysed. To identify polymer type, infrared spectra of samples were compared to a library of spectra of reference materials, where the functional groups defining it could be easily identified (Chang et al., 2007; Pereira et al., 2017; Smith, 1998; Zięba-Palus, 2017).

The infrared spectra of PA, PE and PP showed peaks related to carbonyl groups in the region of 1730-1795 $cm^{-1}$, while the unaltered peaks were found between 1450-1475 $cm^{-1}$. The comparison of the absorbance of the carbonyl peaks with the absorbance of the reference peaks in the same sample reveals the photo-oxidation that these polymers may suffer when exposed to the weather. Carbonyl Index was not calculated for PET, because this polymer contained carbonyl groups in its pure condition and has a strong C=O bonds in 1730 $cm^{-1}$, which result in a high absorbance peak (Velandia-Cabra, 2017). According to the results presented in Table 1, PP was the polymer with the highest carbonyl index and, therefore, the highest degradation. However, according to the levels of degradation explained in section 2.4, PA and PE show low degradation (CI less than 0.15), while PP shows medium-low photo-oxidation (CI between 0.15-0.31).

With regards to the size of these particles, they ranged from 3 to 11 mm (Fig. 3), which would no longer be considered a microplastic, but rather a mesoplastic.

### 3.2. Analysis and interpretation of the microplastics in sand samples

The beaches chosen on the coast of Granada to be analysed for microplastic contamination have some different characteristics from each other that can influence the distribution and concentration of this contamination (Table 2). Although the three beaches have the same type of sediment because they are affected by the Betic System, two of the characteristics that can influence are the type of beach (urban or non-urban) and the topology (open beach or enclosed beach). The nature of the beach has already been analysed by other authors to see its influence on contamination by microplastics (Bayo et al., 2019). These authors found a higher concentration of MPs in urban beaches than in natural or semi-natural beaches, but without significant statistical differences.

After analysis the filters stained with Nile Red in laboratory, a total of 400 microplastics of size < 5 mm were observed, divided as follows: 189 particles in La Herradura, 128 particles in

Motril and 83 particles in La Rábita. The results showed moderate contamination on the three beaches analysed, with a greater predominance of contamination on the beach of La Herradura (Table S1), while La Rábita obtained the least contamination by microplastics. Expressed as the median values, a total of 45.0 ± 24.7, 31.5 ± 21.5 and 22.0 ± 23.2 particles/kg of dry sediment were obtained at La Herradura, Motril and La Rábita, respectively. If these results are compared with the characteristics of the beaches in Table 2, it is evident that there is more pollution on the enclosed beach than on the open beaches. However, there is no clear trend in the concentration of microplastics between urban and non-urban beaches. This may be due to the fact that Motril is not considered an urban beach but is located near touristic recreational areas.

The microplastics were collected along four different transects on each beach, divided as specified in section 2.2 of this article: L1 (wave breaker line), L2 (high tide line), L3 (supralittoral zone 1) and L4 (supralittoral zone 2), in order to see the influence of the distance to the sea on the distribution of microplastics. In La Herradura, lines 1 and 3 (wave breaker zone and supralittoral zone 1) presented the highest concentration of particles/kg. On the contrary, following this same criterion, Motril showed a higher concentration in lines 3 and 4, while La Rábita showed a higher concentration of particles in lines 1 and 4. This may be due to several factors, such as the morphology of the beach itself, the type of sea currents and winds affecting it or the proximity or distance of recurrent human activity. Figure 4 shows the distribution of microplastics per kg of dry sediment in each beach, comparing between the three selected beaches. At first glance, it should be noted that La Herradura is the one with the greatest contamination. There is not great dispersion in the data, as both the minimum and maximum values are close to the 1st (25%) and 3rd percentiles (75%). The beach where the data show the greatest disparity is La Rábita, with slightly differences between 25% and 50% of the samples. The Kruskal-Wallis test carried out to observe the differences in MPs pollution among beaches revealed no statistically significant differences (H=2.999; p=0.223). The influence of human activity and beach morphology has been statistically analysed and will be discussed later.

Comparing the three beaches, La Rábita is the one with the least contamination by microplastics in general, despite being the one closest to urban areas (Fig. 2C) and also the one with the most contamination by macroplastics (mainly plastic bottles and bags). Although macroplastics are not the subject of this study, this incidence was also observed and photographs were taken of the various rubbish found along the beach (Table S2). Moreover, on the three beaches analysed, line 2 (high tide) was the one that showed the least pollution.

The one-way ANOVA analysis carried out, which aimed to see the influence of the distance to the sea on the distribution of microplastics, obtained a F-snedecor factor of 0.566 and a p-value of 0.644, indicating that there is no clear relationship of the distance to the sea with the accumulation of microplastics (Table S3).

As for the morphologies present, fragments and small microspheres were found, the latter being much more abundant. In Table S2, the presence of these shapes can be compared among the different beaches analyzed, highlighting La Herradura as the one with the most microspheres (38.0 ± 23.7 particles/kg), while in Motril the highest number of fragments was found (6.0 ± 0.7 particles/kg). With regards to the line distribution, i.e. the influence of the proximity to the sea in the shape of MPs found, in Figure 5 it can be observed that the concentration of fragments is similar in all lines, whereas the concentration of microspheres is considerably lower in line 2 compared to the other lines. According to Browne et al., (2010), microplastics with irregular morphologies are deposited significantly faster than spherical ones. In addition, line 2 is the high-tide line, which can undergo great daily variations in terms of accumulation in marine debris in general and microplastics in particular, as has already been demonstrated by several authors (Imhof et al., 2017; Piehl et al., 2019). Since sampling on line 2 in this study was carried out just after the high tide peak, it is possible that the tide would have removed particles on that line instead of depositing them. These factors may explain the lower concentration of micro-plastics in general and of microspheres in particular on this line. The presence of the fragments is very homogeneous at any distance from the sea (Fig. 5), being a little more abundant in the area near the promenade and the urban activity. In any case, the microspheres are more abundant than the fragments, with percentages ranging from 78% to 92%. The mentioned differences in the distribution of fragments with the distance to the sea are not statistically representative, since the results of the one-way ANOVA analysis for fragments obtained an F-snedecor value of 0.572 and a p-value of 0.649 (Table S2). Since $p > 0.05$, it can be considered that there is no effect of the distance to the sea on the fragment type morphology. The Kruskal-Wallis analysis was used to study the distribution of spheres because a normal distribution of the data was not fulfilled. The results of this analysis also revealed that there were no statistically significant differences in the microspheres with the distance to the sea ($H=3.873$; $p=0.276$).

The t-student tests were carried out with the aim of observing the influence that the factors "nature of the beach" and "topology", defined according to Table 2, have on the distribution of microplastics. In the interaction of the microplastics with the nature of the beach a critical level of $p = 0.871$ was obtained (Table S4), much higher than 0.05, so it can be accepted that the two groups compared (urban beach and non-urban beach) do not differ in a statistically significant way in the distribution of the microplastics. The data reinforce the results from statistical analysis, since in urban beaches (La Herradura and La Rábita) the median value of fragments and microspheres found was 3.0 and 30.0 MPs/kg, respectively, while in the non-urban beach (Motril Beach) these concentrations were 6.0 and 27.0 MPs/kg.

The t-student test for the beach topology obtained a critical level of $p = 0.064$, also higher than 0.05, so it can be considered that the two groups compared (open deltaic system beach and bay beach) do not differ significantly from a statistical viewpoint in the distribution of microplastics (Table S5). The data obtained were median values of 4.0 and 5.0 fragments/kg for open beaches (Motril and La Rábita) and the enclosed beach (La Herradura), respectively. A higher median value of microspheres was obtained in the enclosed beach (38.0 microspheres/kg) than in the open beaches (23.5 microspheres/kg), but these are not significantly different from a statistical point of view.

With regards to the size of the microplastics analyzed, in general it can be concluded that more than 50% of the particles found in each line are smaller than 100 microns (Figure 6). There are other studies that also report similar sizes for the majority of microplastics found in beach sediments (Claessens et al., 2011; Hengstmann et al., 2018). Beyond that size, the most abundant were the MPs between 100 and 300 microns, especially in the wave breaker line. Similar sizes were also abundant on the line near the seafront. The size distribution between the different lines is homogeneous and follows a decreasing exponential trend (Fig. 6). Due to this generalized trend between sampling lines, the Kruskal-Wallis analysis carried out revealed that there were no significant differences in the size distribution between lines 1-4 (H=7.805; p=0.05) (Table S5). In general, the microplastics analyzed in this study have smaller sizes than those reported for beach sediments by other authors (Antunes et al., 2018; Dowarah and Devipriya, 2019; Sagawa et al., 2018; Simon-Sánchez et al., 2019). This may be due to increased friction and abrasion of the microplastics on the beaches of Granada caused by the large size of the sediment. Among the three beaches, La Rábita presented larger microplastic sizes than the other two, with a 30% of the total amount of particles in the size range of 100-200 microns. The Kruskal-Wallis analysis carried out also revealed no significant differences in the size distribution between beaches (H=4.498; p=0.106).

### 3.3. Comparison with other Mediterranean beaches and environmental implications

The concentrations of microplastics on beaches vary greatly in space, even within a single beach, and in time, throughout the year. Therefore, in order to have an accurate estimation of the microplastic contamination produced on a beach, it would be necessary to carry out samples during several consecutive years and in different seasons, for example spring and autumn. Likewise, it is difficult to identify the origin of microplastics on beaches, especially when it comes to particles as small as those obtained in this study. However, despite these challenges, there are some factors that may be critical in the distribution and origin of this pollution.

Both La Rábita and Motril are open beaches affected by marine delta systems (Table 2), which receive wind currents of between 3-6 m/s on average on days of regular weather, as were the sampling days, with maximum speed values of between 10-20 m/s (Puertos del Estado, n.d.). La Herradura is a beach enclosed by two cliffs, shaped like a bay, and also with finer sediment than Motril and La Rábita, although all are classified as sand and gravel. After consulting the historical data from (Puertos del Estado, n.d.), it was found that the wind conditions on La Herradura beach during that month were very similar to those of Motril and La Rábita. Another factor is the influence of the waves and currents. The wave information consulted shows that in La Herradura the average wave height in October 2018 was 1.08 m, while in Motril and La Rábita it was 0.8 m. This information, together with the geomorphology of the beach and its smaller sediment size, could justify the presence of more microplastics in La Herradura, which can be carried away by stronger waves and remain seated for longer, also conditioned by the enclosed beach morphology it has.

Another factor that may affect the concentration of pollution on these Mediterranean beaches is the configuration of marine currents typical of this sea. The existing climate in the Mediterranean means that this is a concentration basin that suffers from high evaporation and a chronic water deficit, which is only compensated by the entry of water from the Atlantic through the Strait of Gibraltar, a point that is about 200 km from the sampling area of this study. This main current that enters the Mediterranean is attached to the African coast due to the Coriolis force, and moves between the African coast and the coast of Almeria and Granada in the form of circular currents (Balbín et al., 2014). This maritime behaviour may also affect the dispersion of pollution, but further study focusing on this aspect would be necessary.

The quantities of microplastics obtained in this study are higher than those obtained by other authors for Mediterranean beaches (Table 3), as is the case of two beaches studied on Salamina Island (Greece), in which a total of 98 particles were obtained (Tziourrou et al., 2019), mostly composed of PE. On these beaches there was a greater presence of pellets, which was associated with nearby industrial activity. This could also apply to the beach of Motril, despite being classified as "non-urban", it is close to the port of Motril. This port is home to more than 20 companies dedicated to the trade of diesel and gasoline, as well as cement and fertilizers.

The microplastics obtained are also greater than the results obtained in most Slovenian beaches analyzed by Korez et al., (2019), who reported an average concentration of 7.2 ± 1.9 MPs/kg with a maximum of 44.6 ± 6.0 MPs/kg in the month of March, compared to an average of 10.9 ± 6.0 MPs/kg with a maximum of 82.1 ± 21.0 MPs/kg in the month of August. In this study, most of the MPs obtained were smaller than 2 mm, which coincides with the beaches of Granada. Following the comparison in Mediterranean beaches, also the results obtained in this study were showed higher values than Greek beaches studied by Piperagkas et al., (2019), who reported average concentrations between 5 and 20 MPs/kg in almost all beaches except one.

The results are much lower than those found by Simon-Sánchez et al., (2019) in beaches of the Ebro delta, which were 422 ± 119 MPs/kg of dry sediment, or those reported by Yabanlı et al., (2019) who found up to 1,500 particles/kg of dry sediment in beaches of the Datça peninsula, affected by the intersection of the Mediterranean Sea and the Aegean Sea. In the study by Simon-Sánchez et al., (2019), which includes Spanish beaches, authors found more fragments in the river waters than in the sediments themselves, a sign once again of the spatial variability present in microplastics. The amounts obtained in the present study are also much lower than those found by Alomar et al., (2016) on beaches on the islands of Mallorca and Cabrera (Balearic Islands, Spain). In that study, the MPs found varied from 100.78 ± 55.49 MPs/kg of dry sediment to 897.35 ± 103.31 MPs/kg, in areas far away from densely populated areas. Laglbauer et al., (2014) reported average values of 133 MPs/kg in the wave breaker area on Slovenian beaches, which is much higher than the data obtained for Line 1 in this study.

Recently, Bayo et al., (2019) sampled and studied the presence of microplastics on beaches and sediments in the Mar Menor, obtaining concentrations ranging from 8 to more than 160 MPs/kg, with an average concentration of 53 particles/kg, slightly higher than the results obtained in this study. Another interesting aspect to consider is the pore size of the filters used in the different studies (Table 3). The studies that found high amounts of microplastics were those that used filters with very small pore size (0.7 microns). The study by Laglbauer et al, (2014) obtained a considerable amount of MPs using a very large pore size filter, but the authors stated that more than 70% of the MPs found were larger than 1 mm. This may be due to the pore size of the filter (250 µm). Something similar occurs in the study by Alomar et al., (2016), which despite not reporting the pore size used, stated that all the MPs found were larger than 0.5 mm. In the present study a very small pore size was used (1.2 microns) and more than 50% of the microplastics found were smaller than 100 microns. However, relatively low quantities were found as in other Spanish beaches (Alomar et al., 2016; Bayo et al., 2019; Simon-Sánchez et al., 2019). This indicates that it is important to use a filter with the smallest possible pore size, but also indicates that the beaches of Granada are not as polluted as other Spanish beaches.

Following the comparison with other Spanish beaches, CEDEX (Centro de Estudios y Experimentación de Obras Públicas) has been collaborating since 2014 in the Monitoring Programme for Microparticles on beaches, within Subprogramme BM-6, belonging to the Monitoring Programme organised in accordance with the Framework Directive on Marine Strategy (2008/56/EC) (CEDEX, 2018). Within this programme, the following beaches analyzed are in contact with the Mediterranean Sea: Las Azucenas (municipality of Motril, Granada), San Miguel in Cabo de Gata (Almería), La Llana in San Pedro del Pinatar (Murcia), Marenys in Tavernes de Valdigna (Valencia), Cal Francés in Viladecans (Barcelona) and La Pineda, in Vila Seca (Tarragona). In the 2018 autumn sampling campaign, the same year in which the sampling of the present study was carried out, concentrations of MPs higher than those obtained in this study were obtained for the beaches of Cal Francés (47 particles/kg) and La Pineda (199 particles/kg).

On the contrary, the amount of MPs found at Las Azucenas beach in Motril was much lower (10 particles/kg) than that reported in this study for Motril beach (32 particles/kg), even for the same year of sampling and the same municipality. In terms of morphology, fragments and pellets were the most detected.

However, as the CEDEX report itself informed, the coast of Granada is the most peculiar of the Spanish Mediterranean due to its geomorphological characteristics (Bergillos et al., 2016). Its granulometric conditions are not the most suitable for this type of sampling and study, as the beaches have been deteriorating to such an extent that it is currently impossible to find a sandy sediment in the area of the breaker line and it is mainly made up of gravel. Such conditions, which are indicative of a significant erosive action of the waves, are not suitable for the microplastic particles that could be transported by the sea to be deposited on the beach. If the sand particles, of much higher density, do not remain there in a stable manner, the less likely microplastic particles will be retained. This would explain why in the present study hardly any large microplastics were found on the three beaches (only the 14 described in section 3.1) and only particles smaller than 2 mm, whose occurrence and distribution may be much more random. It would also explain the differences in the number of particles found by the CEDEX team compared to those in this study, even if it is the same year and the same season as the sampling.

In addition, this area has another peculiarity that makes it interesting to study in depth the pollution by macro, meso and micro plastics, such as the massive use of agricultural plastics for greenhouses and phytosanitary containers, which generates large amounts of waste that are not always managed correctly and that can undoubtedly lead to greater pollution of beaches (Junta de Andalucía. Consejería de Medio Ambiente, 2019). The common plastic types used in agriculture are low-density polyethylene, high-density polyethylene, polypropylene, and high-strength polystyrene. This information is consistent with the particles found *in situ* in this study, with regards to polyethylene and polypropylene. Therefore, given the absence of any notable industrial activity in the areas studied (except the port of Motril) and the presence of intense agricultural activity, it is possible that most of the granular fragments and particles found in this study come from the degradation of macroplastics used in agriculture.

Due to the controversy generated by all these factors and the difficulties already exposed to consider the existing contamination in the Granada coast and the abundance of microplastics, the authors consider it essential to continue carrying out studies of contamination by plastics and micro-plastics in this area, deepening in all these geomorphological aspects and of human activity.

## 4. CONCLUSIONS

On the three beaches analysed (La Herradura, Motril Beach and La Rábita), moderate contamination by microplastics was found, with La Herradura being the most contaminated (189 particles in total, a median concentration of 45.0 ± 24.7 particles/kg d.w.)) and La Rábita the less contaminated (83 particles in total, a median concentration of 22.0 ± 23.2 particles/kg d.w.). These data revealed a higher contamination by microplastics in an enclosed bay-type beach (La Herradura) in comparison with open delta-type beaches, although no statistically significant differences between beaches were found. On the three beaches, the high tide line was the least contaminated, while on the other three transects the distribution was homogeneous. This may be related to do the sampling after the high tide peak, as the tide may remove the particles present.

The most common morphologies were fragments and granules of sizes <100 microns (approximately 55-75% of particles), 100-200 microns (15-25%) and 200-300 microns (<20%). The predominance of size less than 100 microns could be related to increased friction and abrasion of the microplastics caused by the large size of the sediment. Compared with other Mediterranean beaches, Granada's beaches showed a higher contamination by MPs than those found in Greek or Slovenian beaches, for example. On the contrary, they presented lower pollution than many other coastal sites in Spain, such as the Catalan coast, the Mar Menor coast or the coast of the Balearic Islands.

Despite the differences between beaches shown in the data, there is no clear and significant relationship from a statistical point of view between the accumulation of microplastics and the geomorphological characteristics of the beaches, such as the distance to the sea, the type of the beach (enclosed bay or open delta) and the urban or natural character of the beaches. However, the distribution of microplastics can vary spatially and temporally. Therefore, it would be important in future studies to monitor these beaches more continuously and thoroughly over time and to provide more information regarding the type of plastic contamination on them.

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## FIGURES

Figure 1. Geographical location and coordinates of the three beaches chosen for sampling on the coast of the province of Granada (Spain).

Figure 2. Area sampled on the three selected beaches of the Costa de Granada: Line 1 (wave breaker), Line 2 (high tide), Line 3 (supralittoral zone 1) and Line 4 (supralittoral zone 2, near the promenade).

Figure 3. Example of some microplastics found in situ at the sampling site on the three analyzed beaches, and their corresponding infrared spectra. A) PA; B) PE; C) PET; D) PP.

Figure 4. Concentration of microplastics in the three beaches analyzed. The X symbol indicates the average value (n = 4). The tails show both the maximum and minimum concentrations obtained, and the box represents the upper (75%) and lower (25%) quartiles.

Figure 5. Concentration of fragments and microspheres found in the different transects analysed, expressed as median values of particles/kg and percentage of each shape per line (L1=wave breaker; L2=high tide; L3=supralittoral zone 1; L4=supralittoral zone 2, near the promenade).

Figure 6. Comparison of the frequency of different size classes between the different beach lines analysed (L1=wave breaker; L2=high tide; L3=supralittoral zone 1; L4=supralittoral zone 2, near the promenade).

**TABLES**

Table 1. Calculation of Carbonyl Index for the polymers identified in the visually recognisable microplastics of the beaches of Granada.

| | PA | PE | PP |
|---|---|---|---|
| Carbonyl peak wavelength (cm$^{-1}$) | 1,777 | 1,794 | 1,731 |
| Reference peak wavelength (cm$^{-1}$) | 1,450 | 1,471 | 1,455 |
| Absorbance of carbonyl peak | 0.0084 | 0.0096 | 0.0104 |
| Absorbance of reference peak | 0.0807 | 0.085 | 0.0638 |
| **C.I*** | **0.10** | **0.11** | **0.16** |

Table 2. Main geomorphological characteristics of the beaches analysed on the coast of Granada (adapted from Ministerio de Medio Ambiente) and microplastic concentrations (medians ± IQR) found in the current study.

| Beach | Beach nature | Type of sediment | Topology | Minimum width (m) | Maximum width (m) | Concentration (particles/kg) |
|---|---|---|---|---|---|---|
| La Herradura | Urban | Sand and gravel | Enclosed (bay type) | 16 | 62 | 45.0 ± 24.7 |
| Motril | Not urban | Sand and gravel | Open (estuary type) | 12 | 116 | 31.5 ± 21.5 |
| La Rábita | Urban | Sand and gravel | Open (estuary type) | 20 | 65 | 22.0 ± 23.2 |

Table 3. Concentrations of microplastics found in Mediterranean beach sediments.

| Location | Concentration (MPs/kg d.w) | Predominant shapes | Extraction method | Filter pore size (µm) | Reference |
|---|---|---|---|---|---|
| Coast of Granada (Spain) | 20-48 | Fragments and spheres | Density separation | 1.2 | Present study |
| Psili Ammos beach (Salamina Island, Greece) | 62 | Pellets and fragments | Density separation | n.d. | Tziourrou et al., 2019 |
| Kanakia (Salamina Island, Greece) | 36 | Pellets and fragments | Density separation | n.d. | Tziourrou et al., 2019 |
| Slovenian beaches | 7-82 | Fibres and films | Density separation | 100 | Korez et al., 2019 |
| Beaches in Northern Crete (Greece) | 5-25 | Fibres and fragments | Fluidization and flotation | 42 | Piperagkas et al., 2019 |
| Sandy beaches in Ebro River Delta (Spain) | 422 | Fibres and fragments | Density separation | 0.7 | Simón-Sánchez et al., 2019 |
| Coastline of Datça Peninsula (Slovenia) | 592-2,072 | Fragments | Density separation | 0.7 | Yavanli et al., 2019 |
| Mallorca and Cabrera (Balearic Islands) | 100-897 | Filaments and fragments | Density separation | n.d. | Alomar et al., 2016 |
| Coastal area of the Mar Menor lagoon (Spain) | 8-166 | Fragments and films | Density separation | 0.45 | Bayo et al., 2019 |
| Slovenian beaches | 133-155 | Fibres | Density separation | 250 | Laglbauer et al., 2014 |

CREDIT AUTHOR STATEMENT

**V. Godoy**: Methodology, Formal analysis, Investigation, Writing-original draft; **J.C. Prata**: Methodology, Software, Data curation; **G. Blázquez**: Investigation, Resources, Supervision; **A.I. Almendros**: Formal analysis, Investigation; **A.C. Duarte**: Supervision, Funding Acquisition; **Teresa Rocha-Santos**: Supervision, Writing-Review and Editing; Project Administration; **M. Calero**: Project Administration, Funding Acquisition, Writing-Review and Editing; **M.A. Martín-Lara**: Conceptualization, Investigation, Writing-Review and Editing.

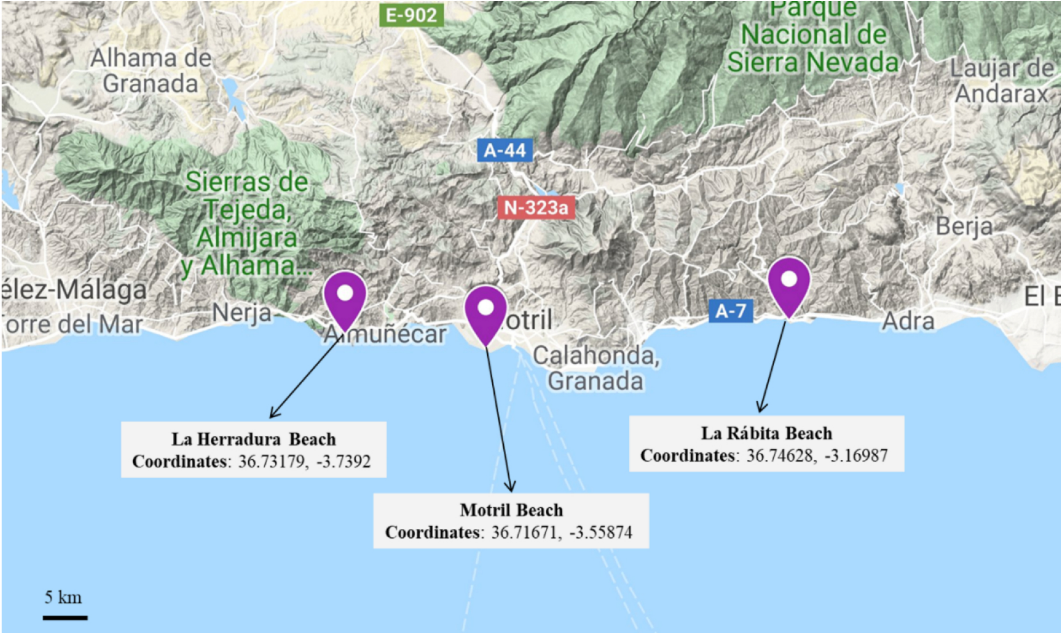
**Declaration of interests**

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

## HIGHLIGHTS

1. The beaches of Granada had been poorly studied because of their complex geology.
2. The highest contamination by microplastics was found in an enclosed bay-type beach.
3. The morphology and size more common were microspheres with a size <100 microns.
4. On the three beaches analyzed, the high tide line was the least contaminated.
5. The beaches of Granada presented lower pollution than other coastal sites in Spain.



**La Herradura Beach**
**Coordinates**: 36.73179, -3.7392

**Motril Beach**
**Coordinates**: 36.71671, -3.55874

**La Rábita Beach**
**Coordinates**: 36.74628, -3.16987

Figure 1

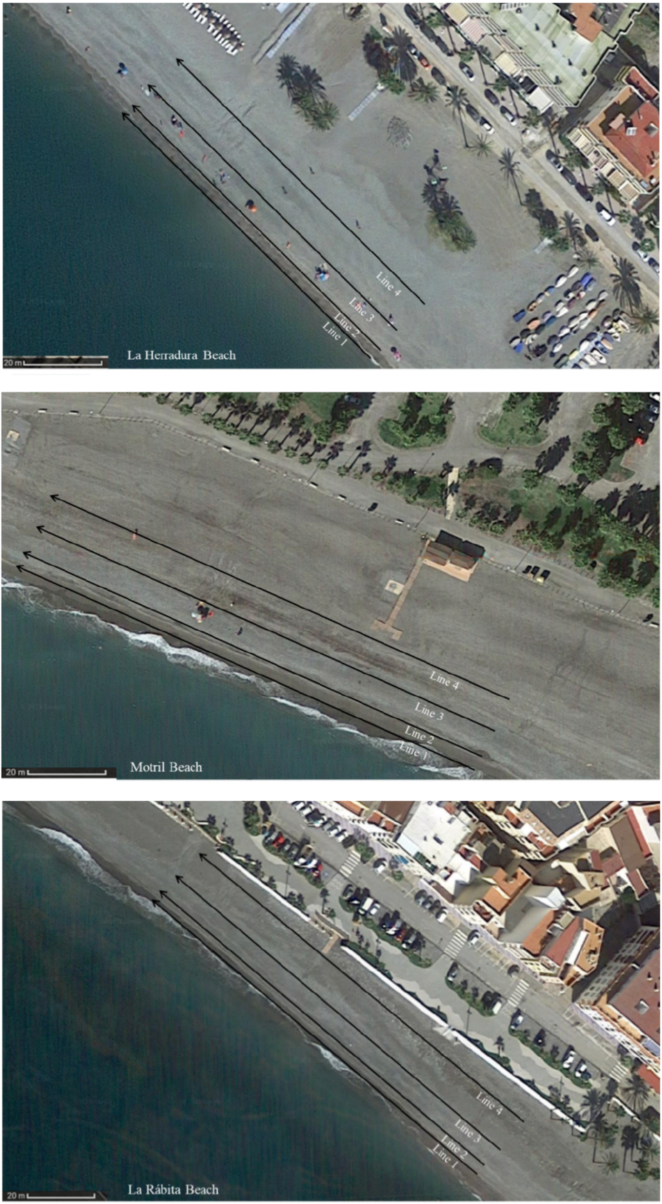

Line 4
Line 3
Line 2
Line 1
20 m
La Herradura Beach
Line 4
Line 3
Line 2
Line 1
20 m
Motril Beach
Line 4
Line 3
Line 2
Line 1
20 m
La Rábita Beach

Figure 2

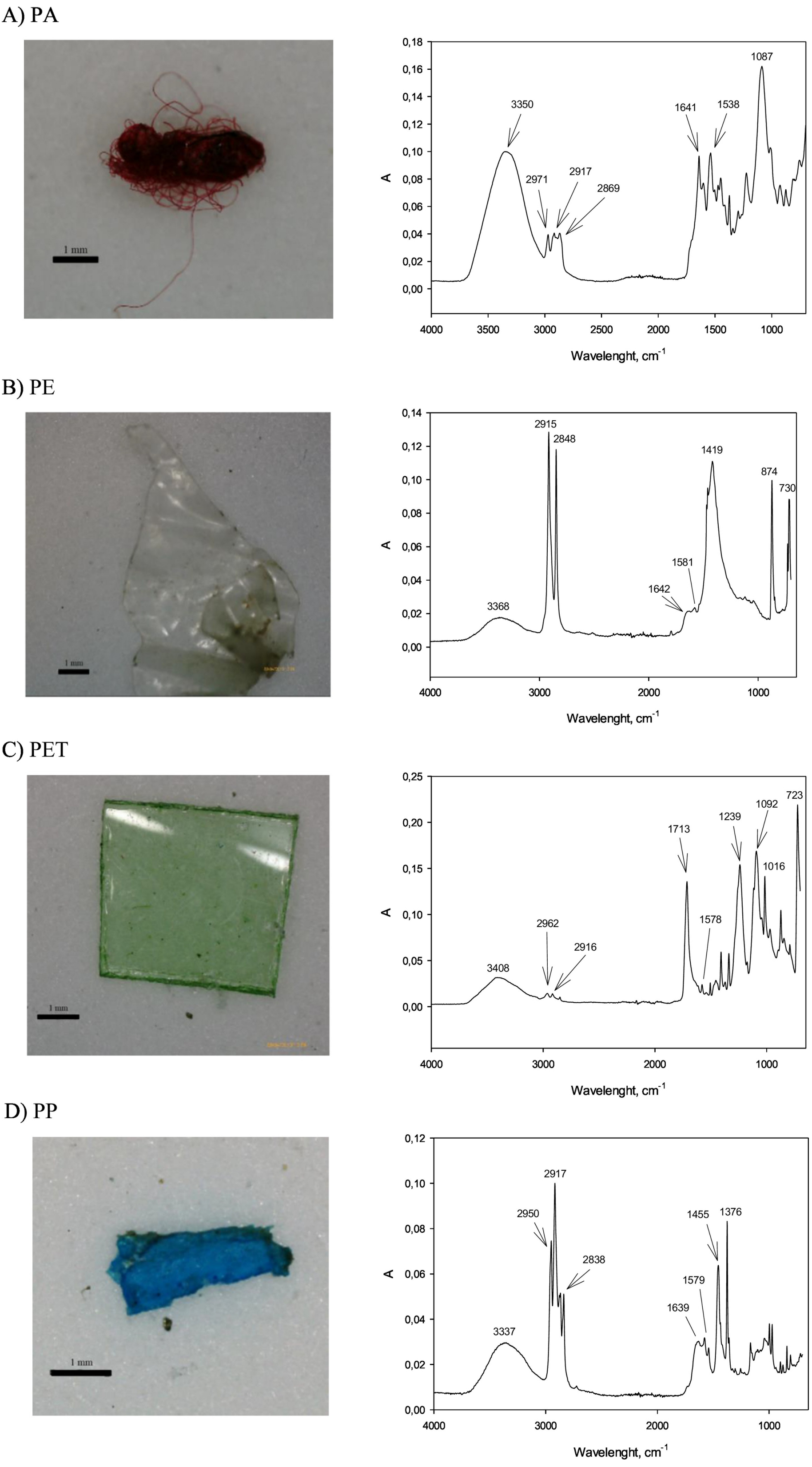

A) PA
1 mm
3350
2971
2917
2869
1641
1538
1087
A
Wavelenght, cm-1
B) PE
1 mm
3368
2915
2848
1642
1581
1419
874
730
A
Wavelenght, cm-1
C) PET
1 mm
3408
2962
2916
1713
1578
1239
1092
1016
723
A
Wavelenght, cm-1
D) PP
1 mm
3337
2950
2917
2838
1639
1579
1455
1376
A
Wavelenght, cm-1

Figure 3

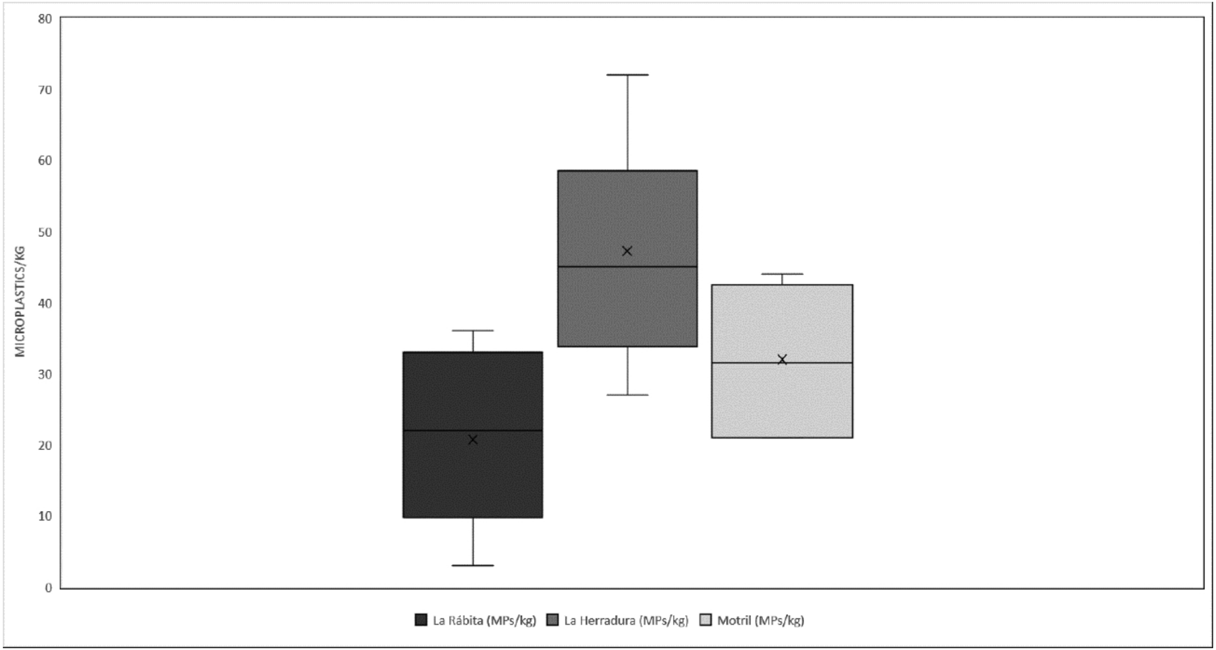
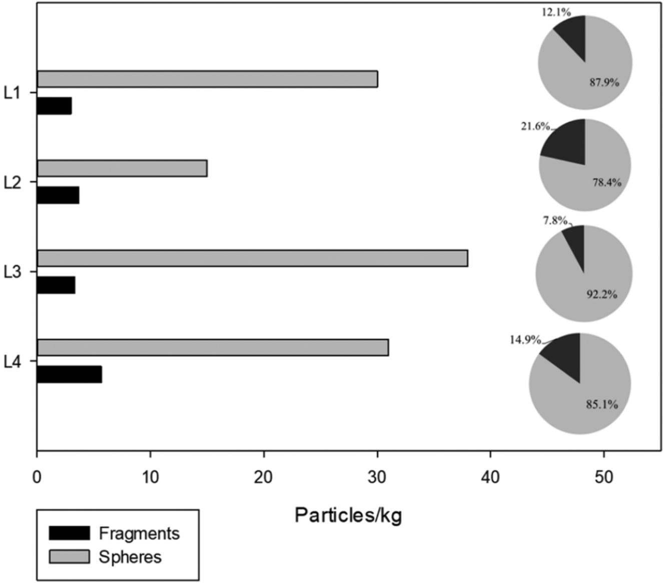

80
70
60
50
40
30
20
10
0
MICROPLASTICS/KG
La Rábita (MPs/kg)
La Herradura (MPs/kg)
Motril (MPs/kg)

Figure 4



L1
L2
L3
L4
12.1%
87.9%
21.6%
78.4%
7.8%
92.2%
14.9%
85.1%
0
10
20
30
40
50
Particles/kg
Fragments
Spheres

Figure 5

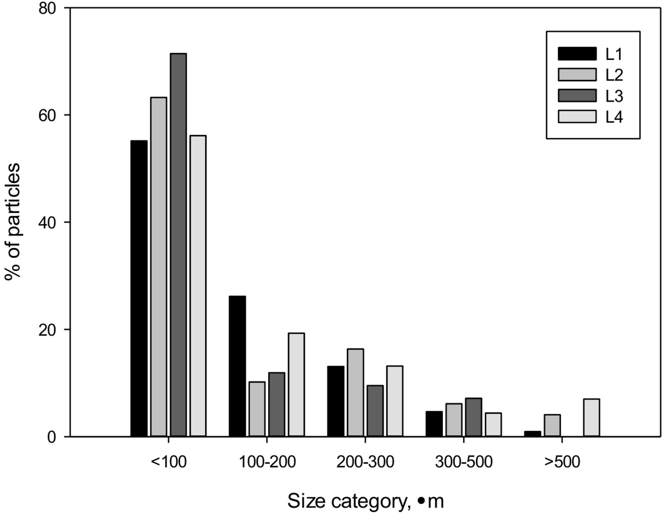

80
60
40
20
0
% of particles
L1
L2
L3
L4
<100
100-200
200-300
300-500
>500
Size category, •m

Figure 6