

## Microplastic and tar pollution on three Canary Islands beaches: An annual study



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### ABSTRACT

Marine debris accumulation was analyzed from three exposed beaches of the Canary Islands (Lambra, Famara and Las Canteras). Large microplastics (1–5 mm), mesoplastics (5–25 mm) and tar pollution were assessed twice a month for a year. There was great spatial and temporal variability in the Canary Island coastal pollution. Seasonal patterns differed at each location, marine debris concentration depended mainly of local-scale wind and wave conditions. The most polluted beach was Lambra, a remote beach infrequently visited. The types of debris found were mainly preproduction resin pellets, plastic fragments and tar, evidencing that pollution was not of local origin, but it comes from the open sea. The levels of pollution were similar to those of highly industrialized and contaminated regions. This study corroborates that the Canary Islands are an area of accumulation of microplastics and tar rafted from the North Atlantic Ocean by the southward flowing Canary Current.

### 1. Introduction

Plastic, due its properties such as durability, impermeability and low cost production, has become essential in our daily life. Microplastics (< 5 mm) and mesoplastics (5–25 mm) includes synthetic fibres, microbeads, preproduction resin pellets and fragments derived from larger plastics. These small pieces of plastic become one of the most common and persistent pollutants of the sea and beaches around the world (Derraik, 2002; Moore, 2008; Ryan et al., 2009; Cózar et al., 2014; Eriksen et al., 2014). In the early 1970s, scientists tried to alert society about this problem (Carpenter and Smith, 1972; Carpenter et al., 1972), but their warning was largely ignored. Now, almost five decades later, the reality is worse than expected; the size of plastic particles is getting smaller, their abundance is increasing, and their distribution is becoming global (Moore, 2008; Thompson et al., 2009). In the North Pacific Central Gyre, the mass of plastic was six times higher than plankton biomass (Moore et al., 2001). Cózar et al. (2014) reported 7000 to 35,000 tons of plastic in the total ocean and Eriksen et al. (2014) estimated that 5.125 trillion particles, weighing 268,940 tons, are currently floating at sea. However, the concentration of particles < 4.75 mm is 100 orders of magnitude lower than the total estimate, based on rates of fragmentation of plastic debris that has been dumped into the sea since the 70s, thus a significant portion of

microplastics has disappeared. The question, “Where is all the plastic?” continues without answer. Here, we explore one possible answer, namely that the missing plastic has been deposited, accumulated, and buried as microplastic debris in beaches, marshes, and other coastal areas all over the world.

The southward flowing Canary Current brings plastic debris from the open North Atlantic Ocean to the coasts of the Canary Islands, mainly on the N and NE exposed beaches (Baztan et al., 2014). In the first evaluation of this phenomenon, Baztan et al. (2014), showed that the Canary Islands are highly polluted by microplastics, reaching values above 100 g per L of sand, on the most exposed areas (Fig. 1).

At Famara beach, the citizen science project, COASTAL (Communities-Based Observatories Tackling Marine Litter), is continuing its research. This effort includes the Famara Participative Observatory project that will provide long-term data on microplastic pollution in the region. In addition, it will be carrying out the important task of increasing awareness in the local population through the media social group “Agiita con el Plástico” (Baztan et al., 2015). Famara is also the beach chosen in Canary region to carry out the monitoring of microparticles on beaches (BM-6) established by the Marine Strategy Framework Directive (2008/56/CE) (CEDEX, 2016).

In order to better understand the condition that affects the microplastic, mesoplastic and other marine debris deposition in this area, we

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Fig. 1. Microplastic pollution in the Canary Islands. (a) Marine plastic debris along the high tide line in Famara beach, Lanzarote. (b) Detailed view of marine plastic debris.

aimed to determine:

- 1– The micro and mesoplastic accumulation on three beaches of the Canary Islands.
- 2– The types of debris found in the samples.
- 3– The temporal and spatial variability of marine debris accumulation.

## 2. Materials and methods

### 2.1. Study area

The study was conducted from September 2015 to September 2016, at three sandy beaches in the Canary Islands: Lambra (La Graciosa Island), Famara (Lanzarote Island) and Las Canteras (Gran Canaria Island) (Table 1, Fig. 2). The areas were selected because they are exposed to the predominant wind and swells (N-NE), have enough space to deposit plastic debris on the high tide line and are accessible to sampling (Fig. 2c, d and e).

Lambra is the most isolated of the three beaches, located on La Graciosa, a small-populated island located in the so-called “Chinijo archipelago”. These islands are at the northernmost of the Canary Islands, and therefore the first to encounter the plastics flowing with the Canary Current. Famara is located on Lanzarote Island. The nearest town is Caleta de Famara, with less than 1000 inhabitants; this beach, however, receives a large number of tourists all year around. Las Canteras is an urban beach, located in a nucleus of population of more than 350,000 inhabitants. Due to the benign climate, Canteras is daily used by many thousands of tourists throughout the year.

**Table 1**

Summary of geographical and sedimentary conditions at each beach. Data from Alonso Bilbao (1993) and Mangas et al. (2008).

	Lambra beach	Famara beach	Las Canteras beach
Location	29° 16.763'N 13° 29.736'W	29° 6.917'N 13° 33.504'W	28° 7.854'N 15° 26.775'W
Total longitude (m)	600	6000	2949
Turistic pressure	Low	Medium	High
Beach cleaning	Once a month Macrolitter	Once a month Macrolitter	Twice a day Macro and microlitter
Orientation	N-NE	N	N
Exposure	Open to NE	Open to N-NW, partially protected to NE	Open to NW, partially protected to NE
Intertidal zone (m)	20	100	60
Sediment type	Medium sands	Fine sands	Fine sands
Median sediment size (mm)	0.433	0.228	0.125

### 2.2. Field work

We have applied a slightly modified TSG-ML sampling protocol. We collected 3 replicates (instead 5 recommended) separated by, at least, 5 m, on 1 cm layer (instead 5 cm) (MSFD GES Technical Subgroup on Marine Litter, 2013). The Spanish BM-6 report (CEDEX, 2016) did not report particles under the first centimeter of sand in the beaches studied. This finding supports our decision to limit our sampling to the upper layer (1 cm). Samples were collected, every 2 weeks, in the highest tide to avoid variability due to the tidal cycle. In a square of 50 × 50 cm (0.25 m<sup>2</sup>) along the high tide line, sediments were collected from the top 1 cm of sand to exclusively collect the marine debris deposited by the last tide. At the same time, 3 L of seawater were added to each sample, mixed, and then the supernatant was filtered through a 1 mm mesh. This process was repeated three times to collect as much marine debris as possible. In Las Canteras, all sampling was done before the beach cleaning to avoid underestimation.

In the laboratory, samples were dried for 24 h at 60 ° C. For the samples containing remnants of vegetal debris (mainly composed of leaves, seeds, wood, seaweeds and seagrass), a density separation by ethanol (96%) was done to separate plastics and tar from organic material. Samples were dried again, sieved and separated in two sizes classes: large micro-debris (1–5 mm) and meso-debris (5–25 mm). After sieving each size class, the samples were weighted in a high precision balance (0.1 mg). The items in each sample were not counted, due to the large number of samples and the amount of particles present in them. In order to compare the number of items per m<sup>2</sup> with other studies, a short study was performed on three samples from each site to determine the relationship between number of items/weight in debris 1–5 mm. Ratios obtained in Lambra were 69.9 ± 16.3 items/g; in Famara, 52.7 ± 12.9 items/g; and in Las Canteras, 79.8 ± 8.1 items/g (Appendix A). We only used this data for comparison purposes because this relationship showed great variability between sites, and also between each sample studied.

### 2.3. Environmental variables

We analyzed the effect of environmental variables on monthly marine litter accumulation on each study site. The oceanographic data was provided by Puertos del Estado (2016) of the Government of Spain and included: significant wave height (m), wave direction in degrees (0 = N, 90 = E), peak wave period, primary swell wave height (m) and tidal coefficient. In addition, several meteorological variables were accounted: wind speed (km/h), maximum wind speed (km/h), wind direction in degrees (0 = N, 90 = E) and rain (L/m<sup>2</sup>), as provided by Agencia Estatal de Meteorología (AEMET, 2016) of the Government of Spain.

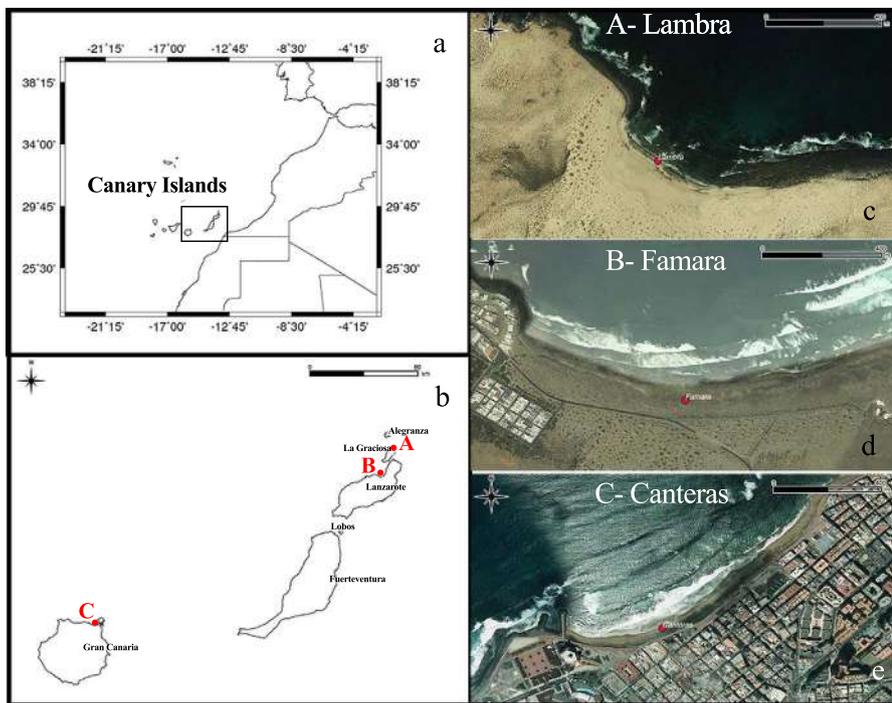


Fig. 2. Study area. (a) Location of Canary Islands. (b) Sampling sites. (c) Satellite image of Playa Lambra (location A), La Graciosa Island. (d) Satellite image of Famara beach (location B), Lanzarote Island. (e) Satellite image of Las Canteras (location C), Gran Canaria Island.

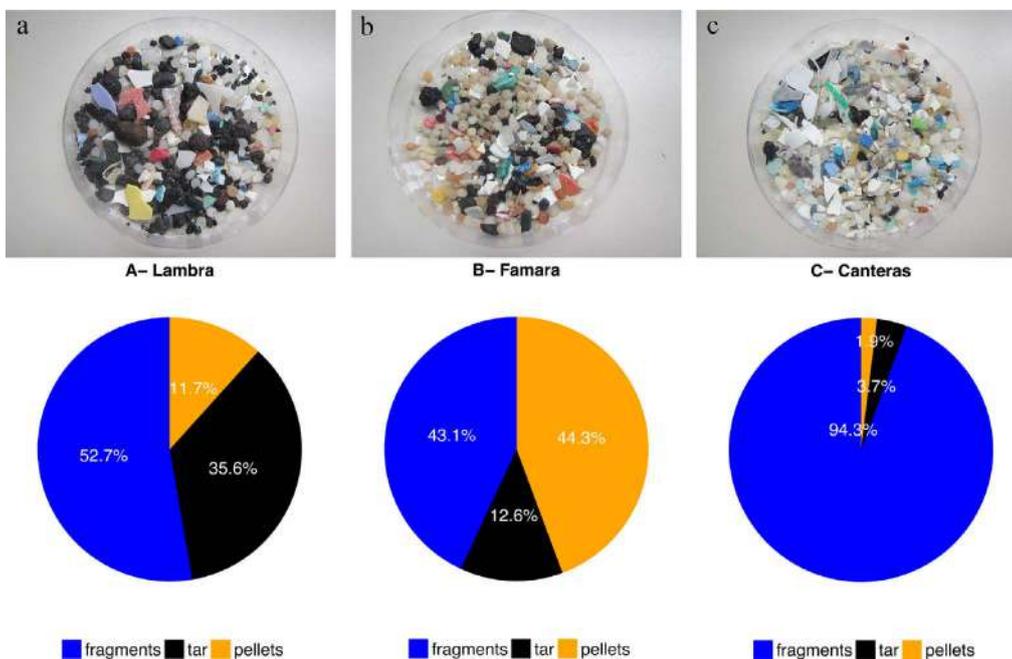


Fig. 3. Composition of marine debris. (a) Lambra beach 52.7% plastic fragments, 35.6% tar and 11.7% preproduction pellets. (b) Famara beach 44.3% preproduction pellets, 43.1% plastic fragments and 12.6% tar. (c) Las Canteras beach 94.3% fragments, 3.7% tar and 1.9% preproduction pellets.

#### 2.4. Statistical analysis

The data were analyzed using R statistical program (R Core Team, 2015). To confirm normality, meso and micro-debris concentration data were analyzed by the Shapiro Wilk test and the homoscedasticity of the residuals was assessed graphically. Meso and micro-debris concentration data were not normal and statistical differences between areas and seasons were tested using Kruskal-Wallis test and Conover posthoc test. All data are publicly available online in Mendeley Data, Herrera (2017).

### 3. Results

#### 3.1. Micro and meso-debris accumulation

Because the samples contained, not only microplastics, but also a large amount of tar, we use the terms, “micro, meso-debris and total debris” throughout the paper to include both types of contaminants.

A total of 261 samples were taken from September 2015 to September 2016 at three locations. The average concentration of large micro-debris (1–5 mm) was 23.7 g/m<sup>2</sup> in Lambra, 16.6 g/m<sup>2</sup> in Famara, and 5.4 g/m<sup>2</sup> in Las Canteras. The highest micro-debris concentration was 125 g/m<sup>2</sup>, 244.2 g/m<sup>2</sup> and 90.7 g/m<sup>2</sup> in Lambra, Famara and Las Canteras respectively. The average meso-debris accumulation (5–25 mm) was 17.9 g/m<sup>2</sup> in Lambra, 4.8 g/m<sup>2</sup> in Famara and 4.3 g/m<sup>2</sup>

in Las Canteras. Maximum values of meso-debris were 157.8 g/m<sup>2</sup>, 85.1 g/m<sup>2</sup> and 69 g/m<sup>2</sup> in Lambra, Famara and Las Canteras respectively.

### 3.2. Composition

We analyzed the composition of 10 g of 3 representative samples (largest samples) collected at each location in order to determine the composition of debris. A representative sample of 10 g contained 524 items in Lambra, 548 items in Famara and 881 items in Las Canteras. Lambra beach samples were composed of 52.7% of plastic fragments, 35.6% tar and 11.7% preproduction resin pellets. Similar values were found in Famara where the samples were composed of 44.3% pellets, 43.1% fragments and 12.6% tar. However, in Las Canteras samples were composed mainly of fragments (94.3%); tar and preproduction resin pellets comprised only 3.7% and 1.9%, respectively (Fig. 3).

### 3.3. Temporal and spatial variability

Total debris (1–25 mm) accumulation along the tide line showed significant differences between locations (Kruskal-Wallis test  $p < 0.001$ ) (Fig. 4). Lambra was the most polluted beach with a mean of 41.6 g/m<sup>2</sup> of total marine debris at the high tide line, Famara showed a mean concentration of 21.4 g/m<sup>2</sup> and Las Canteras 9.7 g/m<sup>2</sup>. The maximum values found were: 282.8 g/m<sup>2</sup> in Lambra (March 2016); 304.01 g/m<sup>2</sup> in Famara (October 2015); and 127.5 g/m<sup>2</sup> in Las Canteras (June 2016) (Fig. 5).

We found significant differences between seasons in Lambra and Famara; the greatest micro and meso-debris pollution was in winter and autumn in Lambra (Kruskal-Wallis test  $p < 0.01$ , Conover test  $p < 0.01$ ); and in autumn, winter and spring in Famara (Kruskal-Wallis test  $p < 0.01$ , Conover test  $p < 0.01$ ). In Las Canteras, there were no significant differences in debris between seasons (Kruskal-Wallis test  $p > 0.01$ ), however highest values were found in summer and spring.

The Azimuth wind and wave plots of all data show a maximum marine debris concentration related to significant wave height above 1.5 m from NW and NE (Fig. 6a) and to N-NE winds (Fig. 6b). When we analyze the temporal changes in debris concentration and local meteorological conditions, we found, in Lambra beach, the highest values related to periods of strong winds and waves in autumn and winter (Fig. 7a). In Famara, high concentrations were related to strong waves, but not related to strong winds, predominant in summer, as shown in Fig. 7b. In contrast, Las Canteras did not show a correlation between the number of plastics particles and periods of strong wave and wind (Fig. 7c).

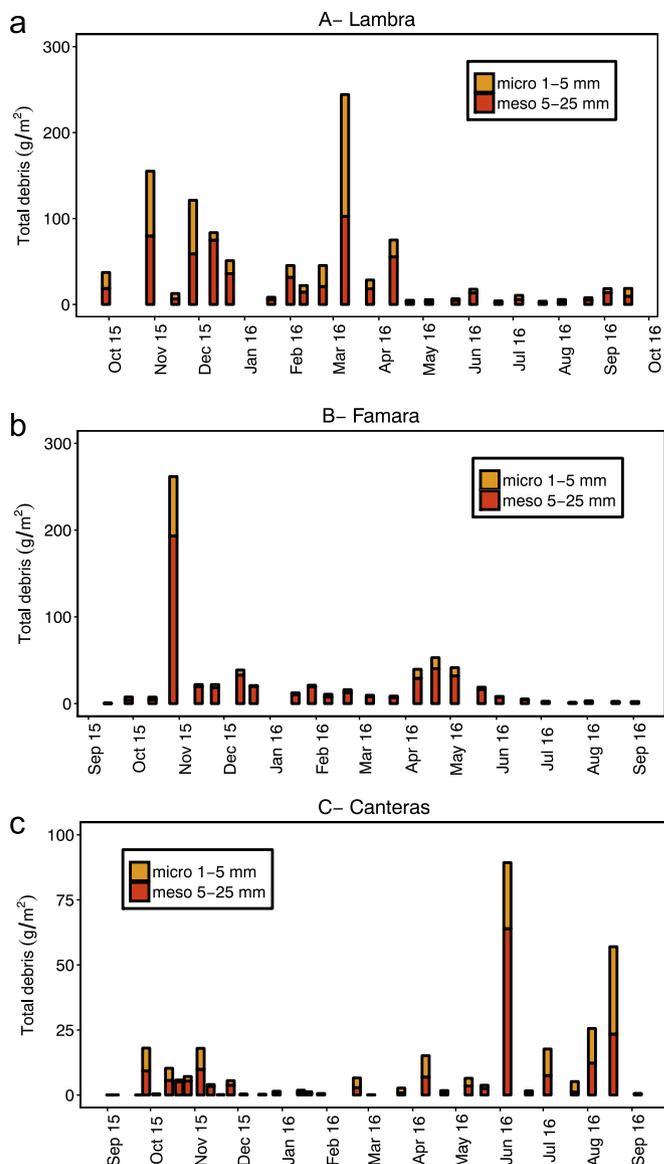


Fig. 5. Mean abundance in g/m<sup>2</sup> of micro (1–5 mm) and meso-debris (5–25 mm) collected from September 2015 to September 2016. (a) Lambra beach. (b) Famara beach. (c) Las Canteras beach.

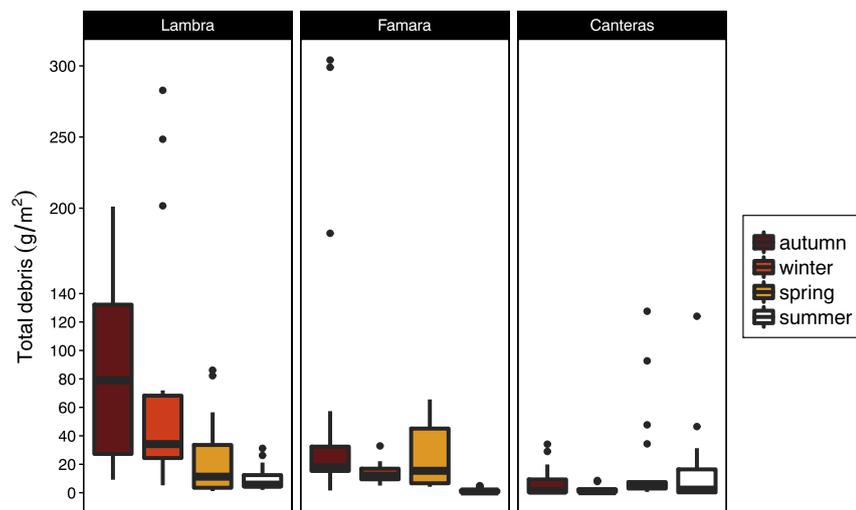


Fig. 4. Marine debris in g/m<sup>2</sup> by location and season. The central thick line of each box designates the median, the box height shows the interquartile range, and the whiskers indicate the lowest and the highest values.

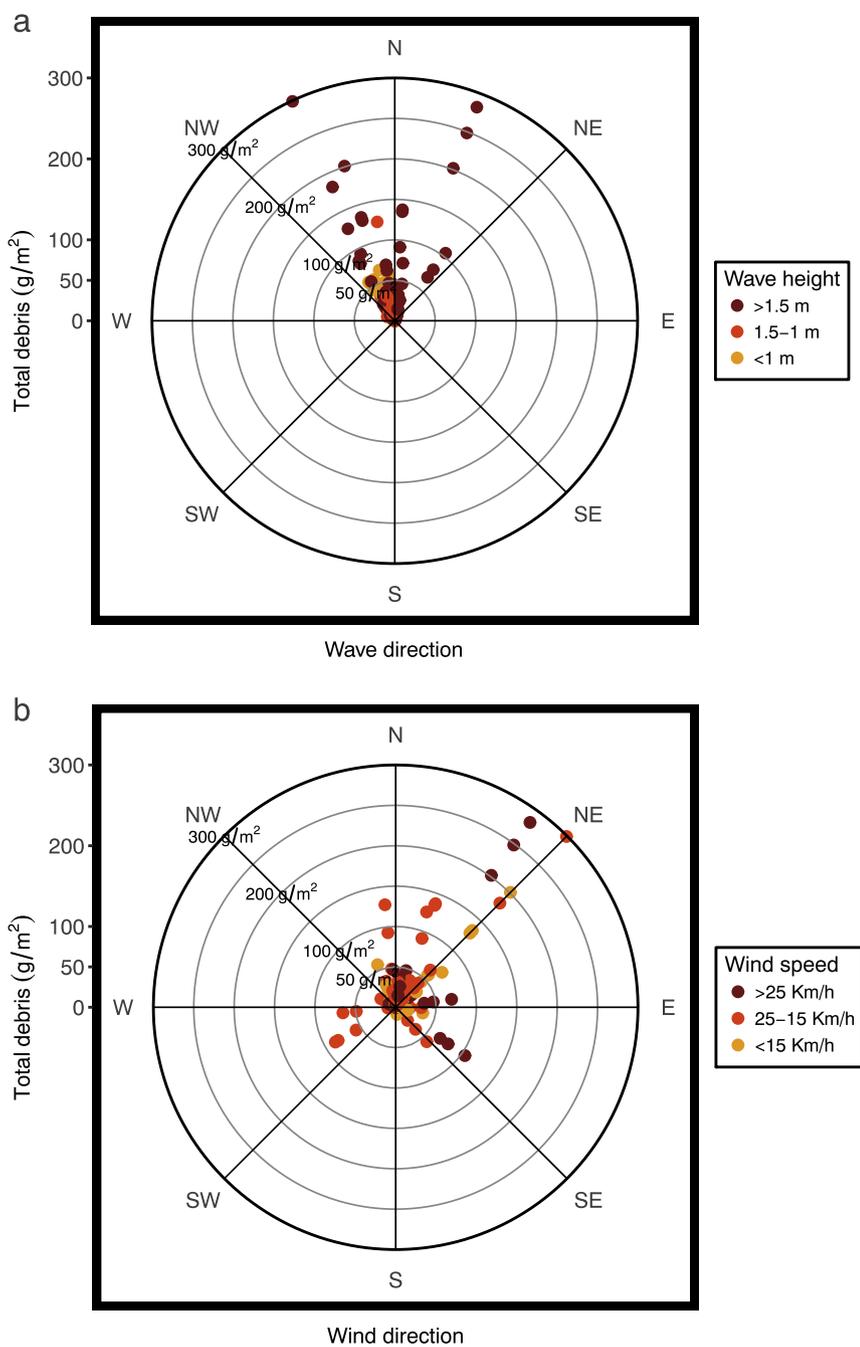


Fig. 6. Azimuth plots. (a) Wave height (m) and direction, and marine debris concentration of all samples collected. (b) Wind speed (mean in km/h) and direction, and marine debris concentration of all samples collected.

#### 4. Discussion

The plastic and tar pollution values found were very high in the three beaches studied. Lambra beach was the most affected, despite being the furthest from urban centers and the one with the smallest influx of tourists. These data and the type of marine debris found, were evidence that the pollution was not local. It came mainly from the open sea via the Canary Current. In the Lambra beach samples, 35.6% of the marine debris was tar; and in Famara, it was 12.6%. This type of waste has been reported in a Caribbean island (Debrot et al., 2013) and in a recent study from a remote island in the Maldives (Imhof et al., 2017). However, in the Canary Islands, it is surprising because the beaches of Lambra and Famara are not located near large commercial ports, as is the case of Las Canteras, in which tar pollution was not important. These tar wastes are likely to come from ships that discharge bunker oil at sea, or from old oil spills deposited on rocks and fragmented by

action of waves, producing small solid tar fragments.

It is alarming, not only because both beaches are located in protected areas (UNESCO Biosphere, Natural Park and Marine Reserve), but also because they are special protection areas for birds (ZEPA), and both microplastics and small tar spheres pose a great risk for the local bird populations. A study of Corys shearwaters (*Calonectris diomedea*) carried out in the Canary Islands showed that 83% of birds were affected, containing, on average, 8.0 plastic pieces per bird (Rodríguez et al., 2012). Plastic ingestion may cause physical damage, provoke satiation and induce starvation and general debilitation (Gregory, 2009; Ryan et al., 1988). In addition, there is a chemical hazard associated with microplastic ingestion, they concentrate persistent organic pollutants (POPs) at levels several orders of magnitude higher than those in the sea. The International Pellets Watch program analyzed polychlorinated biphenyls (PCBs), dichloro-diphenyltrichloroethane and its degradation products (DDTs), and hexachlorocyclohexanes (HCHs) in

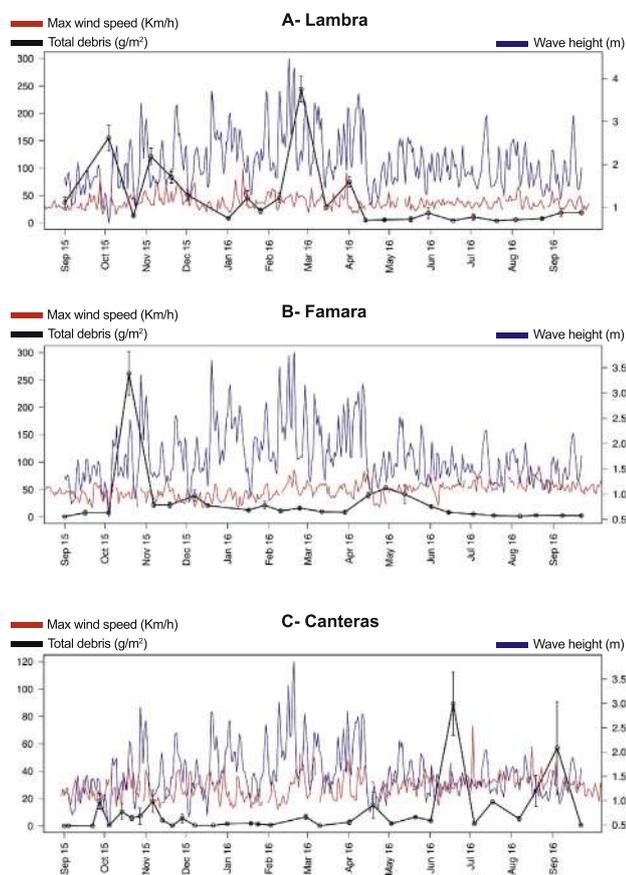


Fig. 7. Temporal variability of marine debris in  $\text{g/m}^2$  (left axis, black line), maximum wind speed in  $\text{km/h}$  (left axis, red line) and wave height in meters (right axis, blue line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

pellet samples from El Cotillo beach located in Fuerteventura, Canary Islands (Heskett et al., 2012). The median concentrations in the pellets ( $n = 5$ ) were for PCBs (sum of 13 congeners),  $9.9 \text{ ng/g-pellet}$ ; for DDTs,  $4.1 \text{ ng/g-pellet}$ ; and for HCHs,  $0.6 \text{ ng/g-pellet}$ . Baztan et al. (2017) reported higher PCBs pollution in pellets collected from Famara beach with values of  $31.15 \text{ ng/g-pellet}$  of total PCBs concentration. Once ingested, the POPs can be transferred to many organisms via predation (Hirai et al., 2011; Karapanagioti et al., 2011; Rios et al., 2007; Teuten et al., 2007, 2009).

A notable fact is the large number of resin preproduction pellets, mainly from samples collected in Famara (44.3%). These preproduction plastic pellets, also called “nurdles”, are the raw material for manufacturing plastic products. According to PlasticsEurope (personal communication) there is not plastic industry (production or transformation) in the Canary Islands. The resin pellets that wash up on the islands’ beaches are transported by the currents, coming from ships or industries in other parts of the planet. Studies since the 1970s have reported high levels of plastic waste, mainly pellets, found at sea and along coasts (Carpenter et al., 1972; Shiber, 1982, 1987). However, the amount of preproduction resin pellets on the world’s shores is increasing and these are present even in remote areas (Ogata et al., 2009; Veerasingam et al., 2016). More research efforts are needed to determine the possible source of tar and pellets, and to determine the adsorption of persistent organic pollutants (POPs) and other chemical contaminants, and to assess subsequent potential harm to marine animals in the region.

The highest pollution level in Lambra beach could have been due to the fact that it is the most exposed beach, the windiest, and the beach with the strongest waves, especially in autumn and winter when the

greatest accumulation of debris occurred. The effect of wind on marine debris deposition and accumulation has been demonstrated (Browne et al., 2010). Other authors found higher levels of debris and tar contamination in the windward beaches due to strong winds and waves (Debrot et al., 2013, 1999). Famara also has high pollution values mainly in autumn and spring, however in summer there were no high values despite it being a very windy period on this beach. Las Canteras was the beach that showed smallest amount of debris. On this beach, peaks occurred in summer when high waves and high tides caused the accumulation of marine debris. The surface current is another factor that likely affected the debris deposition. Here, this variable was not measured at each location, and data from Puertos del Estado were not available. In addition, in the present work, the oceanographic data provided by Puertos del Estado were estimated from models and refers to the open sea, not near-shore, local conditions. Spatial inconsistency in the seasonal patterns can be explained by the local wind fields and hydrodynamic conditions. These produce different patterns in the accumulation of debris coming from the open sea, even between beaches close to each other.

There is great variability in the concentration of marine debris between the different seasons of the year, and also between sampling days. For the development of more accurate models to predict the concentration of marine debris, or for the determination of the long-term trends, it is necessary to measure the current direction and velocity, the wave direction and height in situ, and to increase the sampling frequency. This requires arduous sampling work. Citizen science could help with the sample collection for long-time studies, and at the same time generate awareness and promote environmental education (Hidalgo-Ruz and Thiel, 2013; Baztan et al., 2015). In addition, improvement in quantitative methods, including meteorological and oceanographical measurements, as well as the use of standard methods and units, are necessary to facilitate comparison and evaluation of long-term, global scale, trends in marine-litter accumulation. Quantifying microplastics is currently accomplished by microscopy and by separating each particle manually, while in other fields such as medicine and oceanography measurement is accomplished by high resolution image analysis with the aid of well developed software. Research in the field of image analysis is needed to measure plastic particles automatically in order to maximize human and material resources.

The beach chosen to monitor microparticles (BM-6) in the Canary Island area was Famara beach (CEDEX, 2016). Samples were collected on the 21st November 2016. The mean was  $10.86 \text{ g/m}^2$ , lower than our average value for all data from Famara beach ( $16.6 \text{ g/m}^2$ ), and lower than our average value found on the 25th November 2015 ( $18.17 \pm 7.3 \text{ g/m}^2$ ) (Table 2). However, the maximum values obtained for the present study in Famara and Lambra beaches are slightly lower than those presented by Baztan et al. (2014).

The BM-6 report (CEDEX, 2016) and Baztan et al. (2014) did not mention tar pollution in describing their samples. Perhaps, this was because tar is not included as a category of marine litter or marine debris. However, it is an important source of marine pollution in the Canary Islands, and is likely to be important in other regions. By definition tar should be included because it is a “persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment” (Galgani et al., 2010; Scientific and Technical Advisory Panel, 2011; GESAMP, 2015; NOAA Marine Debris Program, 2017).

The comparison with studies carried out in other parts of the world is difficult due to the different objectives, size categorizations and the different methodologies and units used, as reflected in the review by Browne et al. (2015). In the present study, the number of particles was not counted, because the time invested in the processing of 261 samples would have been too large. However, the most convenient units to express the concentration in order to be comparable with other studies is  $n^\circ \text{ particles/m}^2$ . In addition, it is advisable to report the volume of sand collected, because not all studies are based on samples collected

**Table 2**

Review of microplastic abundance in sediments from different regions. \*Samples include tar and microplastics. \*\*Values estimated from mean weight of particles (Appendix A).

Area	Size (mm)		g/m <sup>2</sup>	Items/m <sup>2</sup>	References
Lambra, Canary Islands*	1–5	Mean	23.7	1656**	Present work
	1–5	Min–max	0.77–125	53.4–8737**	
Famara, Canary Islands*	1–5	Mean	16.6	874.8**	Present work
	1–5	Min–max	0–244.2	0–12,869**	
Las Canteras, Canary Islands*	1–5	Mean	5.4	430.9**	Present work
	1–5	Min–max	0–90.8	0–7245**	
Famara, Canary Islands	1–5	Mean	10.86	541.66	CEDEX (2016)
Hong Kong	0.315–5	Mean	5.6	5595	Fok and Cheung (2015)
		Min–max	0.008–249.16	16–258,408	
		Mean	0.0032		
Uruguay	> 0.3	Mean			Lozoya et al. (2016)
SE Pacific beaches, Chile	1–4.75	Min–max		< 1–805	Hidalgo-Ruz and Thiel (2013)
	1–4.75	Mean		27	
North coast Taiwan		Min–max		16–1936	Kunz et al. (2016)
South Korea	1–5	Min–max		1.6–92,217	Lee et al. (2013)
Mid-west Korea	1–5	Mean		46.7–1247	Kim et al. (2015)
Portuguese coast	1–10	Mean		28.6–392.8	Martins and Sobral (2011)
Hawaiian archipelago	1–15	Mean		1.2	McDermid and McMullen (2004)
Caribbean islands	1–5	Min–max		0.2–2500	Schmuck et al. (2017)
North Gulf of Mexico, USA	0.5–5	Mean		13.2–50.6	Wessel et al. (2016)
Southeast Brazil		Min–max		2–1300	Gomes De Carvalho and Neto (2016)
Persian Gulf, Iran	0.45–4.75	Min–max		2–1258	Naji et al. (2017)
Russian Baltic coast	0.5–5	Min–max		7–5560	Esiukova (2017)
Slovenia	0.25–5	Mean		178.8	Laglbauer et al. (2014)
Maldives Islands	1–5	Mean		22.6	Imhof et al. (2017)
	> 5	Mean		13.2	
Famara, Canary Islands	1–5	Min–max		g/L	Baztan et al. (2014)
				0–109	
China, Bohai Sea	0.1–10	Mean		Items/L	Yu et al. (2016)
				102.9–163.3	
German Baltic coast	0.1–1	Min–max		Items/kg	Stolte et al. (2015)
				1–7	
Belgium	0.038–1	Min–max		48.7–156.2	Claessens et al. (2011)
Singapore		Min–max		0–16	Ng and Obbard (2006)
Italy, Tyrrhenian Sea		Mean		151–678.7	Fastelli et al. (2016)
China	< 1–1.5	Min–max		4320–12,160	Qiu et al. (2015)

from the same depth. Furthermore, volume is more comparable than mass because sand has different densities. The BM-6 report showed that 88.7% of microplastics are in the 1–5 mm fraction size (CEDEX, 2016). From these data, and average values of mass and n° of items (10.864 g/m<sup>2</sup> or 541.66 particles/m<sup>2</sup>) we calculate an average number of particles of 1–5 mm per gram in 44 items (CEDEX, 2016). This value is in the range obtained in the present study for Famara (52.7 ± 12.9 items/g), but this estimation has a high deviation (Appendix A). We use it only for comparison purposes. The ratios obtained for Lambra (69.9 ± 16.3 items/g) and Las Canteras (79.8 ± 8.1 items/g) also showed high variability (Appendix A).

Values obtained in other regions of the world showed that accumulation of marine debris in the Canary Islands is higher than in most of the other zones, except Hong Kong (Fok and Cheung, 2015), South Korea (Lee et al., 2013) and China (Qiu et al., 2015) (Table 2). This indicates that the Canary Archipelago is a hot spot of marine litter, as previously showed by Baztan et al. (2014) and the BM-6 report (CEDEX, 2016).

## 5. Conclusions

- 1– Spatial inconsistency in the seasonal patterns of coastal pollution was found. Debris accumulation depended mainly of coastline orientation and local-wind and wave conditions.
- 2– The strong presence of resin pellets and tar pollution are evidence that contamination is not land-produced. Further research is necessary to determine their origin.
- 3– Due the large amount of tar present in the samples, and its negative impact on ecosystems and marine biota, we suggest including tar as a category of marine litter or marine debris in order to report it in monitoring programs established by the Marine Strategy Framework

Directive (MSFD 2008/56/EC).

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2017.10.020>.

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