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Short Communication

Plastitar: A new threat for coastal environments

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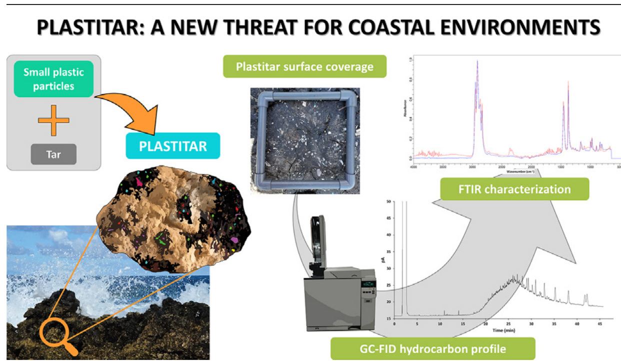
HIGHLIGHTS

- A new plastic formation was found in the Canary Islands, Spain.
- A combination of tar and microplastics covers rocky shores.
- A new term, plastitar, is proposed.
- Plastitar characteristics suggest that it does not meet any prior classification.
- Effects on the coastal environments should be further investigated.

GRAPHICAL ABSTRACT



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ABSTRACT

Oil residues have been frequently found on the coasts all over the world as a result of different accidental releases. Their partial evaporation and solidification onto the coastal rocks can produce the formation of a new solid structure forming an agglomerate with other materials, mainly microplastics (though wood, glass, sand and rocks were also found), yielding to a new plastic formation, name herein for the first time as "plastitar". These new formations have been found in several of the islands of the Canary Islands archipelago (Spain). Their study has shown that these new formations can be permanently attached to the rock, occupying even a 56% of the sampled area with an heterogeneous distribution. It was also observed that the studied plastitar was composed mainly of tar and polyethylene (90.6% of the studied particles) and polypropylene (9.4% of the studied particles) microplastics, primarily fragments (82.5%), pellets (15.7%) and lines (1.8%). The ever more frequent presence of plastics and, in particular, microplastics in coastal environments can lead to the common occurrence of these new plastic formations (probably present in other parts of the world), which long-term effects on the coasts should be further investigated.

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1. Introduction

Nowadays, it is evident that plastic pollution constitutes one of the main global environmental problems, which has been originated by the exponential growth of plastic production since the 1950s (reaching 367 million of tons in 2020 (Plastics Europe, 2021)), the high dependency of our society on plastics, the massive irrational use of many plastic items (i.e. single use plastics), as well as an inappropriate and insufficient waste management, among others relevant issues.

In this context of continuous anthropogenic pressure onto the environment at different levels, plastics can be found in all the environmental compartments (water, soil and air) (Gola et al., 2021; Hernández-Sánchez et al., 2021b; Vighi et al., 2021), also under the form of new plastic formations resulting from the interaction of plastic debris with other wastes, anthropogenic processes, and environmental elements (De-la-Torre et al., 2021). Very recently, such new plastic formations, which have been classified as *plastiglomerates*, *plasticrusts*, *pyroplastics* or *anthropoquinas*, have been reviewed and discussed by De-la-Torre et al. (De-la-Torre et al., 2021). These materials have been named and classified in that way since their characteristics clearly suggest that they do not meet any prior classification and could potentially have a different impact than that of common plastics.

Plastiglomerates were first described in 2014 by Corcoran et al. as the combination of melted plastic, sediments, basaltic lava fragments, and organic debris (Corcoran et al., 2014). They were first found in Kamilo beach (Hawai) and later observed in other regions like Indonesia, United States of America (USA), Portugal, Canada (Corcoran et al., 2018; Corcoran and Jazvac, 2020; Ellrich and Ehlers, 2022) or Japan (Furukuma, 2021). The main source of this type of marine litter is the uncontrolled burning of waste (Corcoran et al., 2014). Plastiglomerates include a wide variety of plastic wastes in their structure, such as pellets, food packaging, bottle tops, fragments of unknown origin and remnants of fishermen's ropes. This waste assembly may form a homogeneous melt that contain partially melted parts, even intact plastic parts embedded in a molten matrix. X-ray fluorescence analysis of the plastiglomerates has shown that they are mainly composed of plastic (up to now only polypropylene (PP) has been identified (Ellrich and Ehlers, 2022)) and that they also contain a low percentage of inorganic matter such as sand (Turner et al., 2019).

On the other hand, the term *anthropoquinas* has been described by Fernandino et al. as sedimentary rocks formed from the cementation of a variety of materials available in a natural environment (Fernandino et al., 2020). These materials can be of natural and anthropogenic origin, such as wood, plastic, burnt waste, glass, sand, sediments, and organic materials. These formations have been found in Río Grande do Sul (Brasil) as well as in the eastern Cantabrian coast (Spain) (Irabien et al., 2015). The formation process of these type of rocks is yet to be determined, but it is known that the genesis is not mediated by human activity; instead, factors such as the formation time, distribution, or oceanographic conditions, among others, play an important role (Fernandino et al., 2020). Clearly the presence of this type of pollution has a significant visual impact and could represent a risk to coastal ecosystems. Furthermore, anthropoquinas are also an example of how various types of anthropogenic waste enter the geological cycle (De-la-Torre et al., 2021).

Plasticrusts consist of plastic encrusting wave-exposed rocky intertidal habitats which are presumably generated by waves smashing plastic debris against intertidal rocks. They are a novel form of plastic debris first reported on the island of Madeira (Portugal) in 2016 (Gestoso et al., 2019), and later observed on the island of Giglio, Tyrrhenian Sea (Italy) (Ehlers and Ellrich, 2020). There have not been direct observations of plasticrusts formation, though Ehlers et al. (Ehlers et al., 2021) suggested that high rock surface temperatures in summer contribute to this process. Due to the recency of these findings, the available data on the formation process are scarce, but abundance of plasticrusts is known to be increasing, as well as the fact that they are mainly formed from maritime ropes (Ehlers et al., 2021). In fact, Gestoso et al. (Gestoso et al., 2019) have confirmed its increased coverage over time since 2016 in recent surveys conducted in January 2019. Reported plasticrusts consisted principally of polyethylene (PE), PP and polyethylene terephthalate (PET), being the first two of them the most produced types of plastics worldwide and also the ones mostly found in the coastal marine environment as a result of their low density (Gola et al., 2021; Plastics Europe, 2021).

Concerning *pyroplastics*, they are melted plastic with a rock appearance that were first observed in Whitsand Bay (Cornwall, UK) and on beaches from Scotland (Orkneys), Ireland (County Kerry), northwest Spain and Canada (Vancouver) (Turner et al., 2019). Later, these plastic formations were also reported on the island of Giglio, Tyrrhenian Sea (Italy) (Ehlers and Ellrich, 2020), in the Ariho River Estuary (Japan) (Furukuma, 2021) and in Madeira (Portugal) (Ellrich and Ehlers, 2022). They are formed when plastic waste is burnt and cooled down, which creates greyish agglomerates with encapsulated plastic material that may leach toxic substances. Pyroplastics have been described as amorphous matrixes with a neutral colour (black-charcoal-grey, off white or brown) and a smooth surface due to weathering processes. They float in water as a result of their lower density, which is their distinctive property that allows differentiating them from conventional rocks without further analyses (Turner et al., 2019). De-la-Torre et al. (2021) have suggested that such buoyancy, together with their durability during transport, could turn them into vectors of alien invasive species, toxic chemicals and pathogens. Up to now, pyroplastics of PE and PP (Turner et al., 2019) or PET (Ehlers and Ellrich, 2020) have been reported. Open campfires and plastic waste burning in beaches are the main sources of pyroplastics. Very recently, their formation as well as that of plastiglomerates has also been reported in pebble beaches (composed of small rocks and pebbles) which contribute to their shaping and degradation -pebble casts prints are formed- (Ellrich and Ehlers, 2022).

As it is well-known, petroleum residues have been frequently found on the coasts all over the world as a result of different accidental releases, such as operational discharges from tankers, tanker spills, pipeline releases, the extraction of petroleum, or sunken vessels. According to MARPOL 73/78 Annex I legislation (International Maritime Organization, 2021), which became effective in 1983, oil discharges from ships can only be developed under certain controlled conditions. Such regulation has positively influenced these practices, by highly reducing them as documented by different studies (Carpenter, 2019). The Canary Islands archipelago (Spain) was also designated as a highly sensitive marine area due to its vulnerability to damage from international shipping, being the main problem the intense traffic of large oil tankers whose cargo destination is the Persian Gulf (Marine Environment Protection Committee, 2005). Despite such important international achievements, oil spills still occur, and those contaminants reach the coasts around the globe.

Once oil is present in the ocean, part of it evaporates and weathers, reaching the shore with a high viscosity. The final product, which has a black or dark brown colour, is named as *tar* or *pitch*, and it often comes ashore in the form of balls, a fact that has been documented in a good number of occasions (Shirneshan et al., 2016; Suneel et al., 2013). In a rocky coast, tar may attach to rocks and dry, sometimes covering large surface areas, also retaining other materials such as plastics.

In this work, we report for the first time, a new plastic formation composed of plastic (in particular, microplastics) and tar, which have been found in different locations on the coast of the Canary Islands (Spain). Evidence suggests that they do not meet any previous classification (e.g. plastiglomerates, pyroplastics, plasticrusts or anthropoquinas), being a unique heterogenous mixture that should be studied in more depth, taking into account the potential negative impacts that they may have on the environment.

2. Experimental

2.1. Study area and field work

Plastitar formations were observed in different locations of the Canary archipelago: Playa Grande beach (Tenerife), the coast of Malpaís de Güímar (Tenerife), Arenas Blancas beach (El Hierro) and the coast of Famara

(Lanzarote), which have currently been classified as hotspots due to the large amount of plastic waste that reaches these beaches. Fig. 1 shows study sites, while Table 1 compiles the location, observation periods, orientation and main wind incidence. In general, each location was visited once in each indicated month.

2.2. Plastitar surface coverage study

In order to study plastitar composition and coverage surface, a sampling area located at the eastern end of Playa Grande was selected since it is a beach which receives high amount of plastic wastes all over the year. Quadrants of 20 × 20 cm were placed on five different places of the rocks of Playa Grande beach in January 2022, maintaining a distance between sampling points of 2 to 3 m covering a total sampling area of around 20 m^2. Each point was photographed and georeferenced. A piece of tar was collected using stainless-steel tweezers and introduced in 10 mL vials previously calcinated in a Carbolite CWF 11/13 muffle (Sheffield, United Kingdom) at 550 °C for 4 h. The number, shape and colour of the microplastic items found (sizes between 1 and 5 mm in their largest dimension) were also visually determined in situ. Any other material apart from plastic having a size bigger than 5 mm was also reported. Considering their shapes, microplastics were classified as foams, films, pellets, fibres, or lines and fragments (Crawford and Quinn, 2017). For Fourier-transform infrared (FTIR) spectroscopy analysis, an average of 25 microplastics of different shapes (127 in total) were collected from each quadrant with stainless-steel tweezers, with particular emphasis on the collection of fragments and pellets, the most common microplastic found. Each particle was introduced in previously calcinated 20 mL vials. ImageJ® Image Processing and Analysis in Java software was used for estimating tar affected area, as well as to count and measure the collected microplastics (Ferreira and Rasband, 2012).

2.3. Contamination control

Though the studied plastics had a size bigger than 1 mm, to prevent possible contamination of the samples, the use of plastic materials in sample processing was avoided, stainless-steel tweezers were used for sample handling while glass vials and Petri dishes were also used for sample storage.

2.4. Infrared spectroscopy analysis

A FTIR spectrometer Cary 630 equipped with a single reflection diamond Attenuated Total Reflectance (ATR) module (Agilent Technologies, California, USA) was used to determine the composition of the plastic particles embedded in the plastitar, with a ZnSe beamsplitter and a 1.3 mm diameter thermoelectrically cooled deuterium triglycine sulphate (dTGS) detector. FTIR spectra were collected with 32 scans per spectrum (Happ-Genzel apodization function was applied) at a resolution of 8 cm^{-1} in the range 4000 and 650 cm^{-1}. Agilent MicroLab PC FTIR software was used to acquire and identify spectra. The minimum matching for positive identification was set at quality values $\geq$0.70 over 1.00, which corresponds to a 70% of positive identification. Such criterium was set following the indications of the Guidance of Marine Litter in European Seas of the European Commission (Galgani et al., 2013).

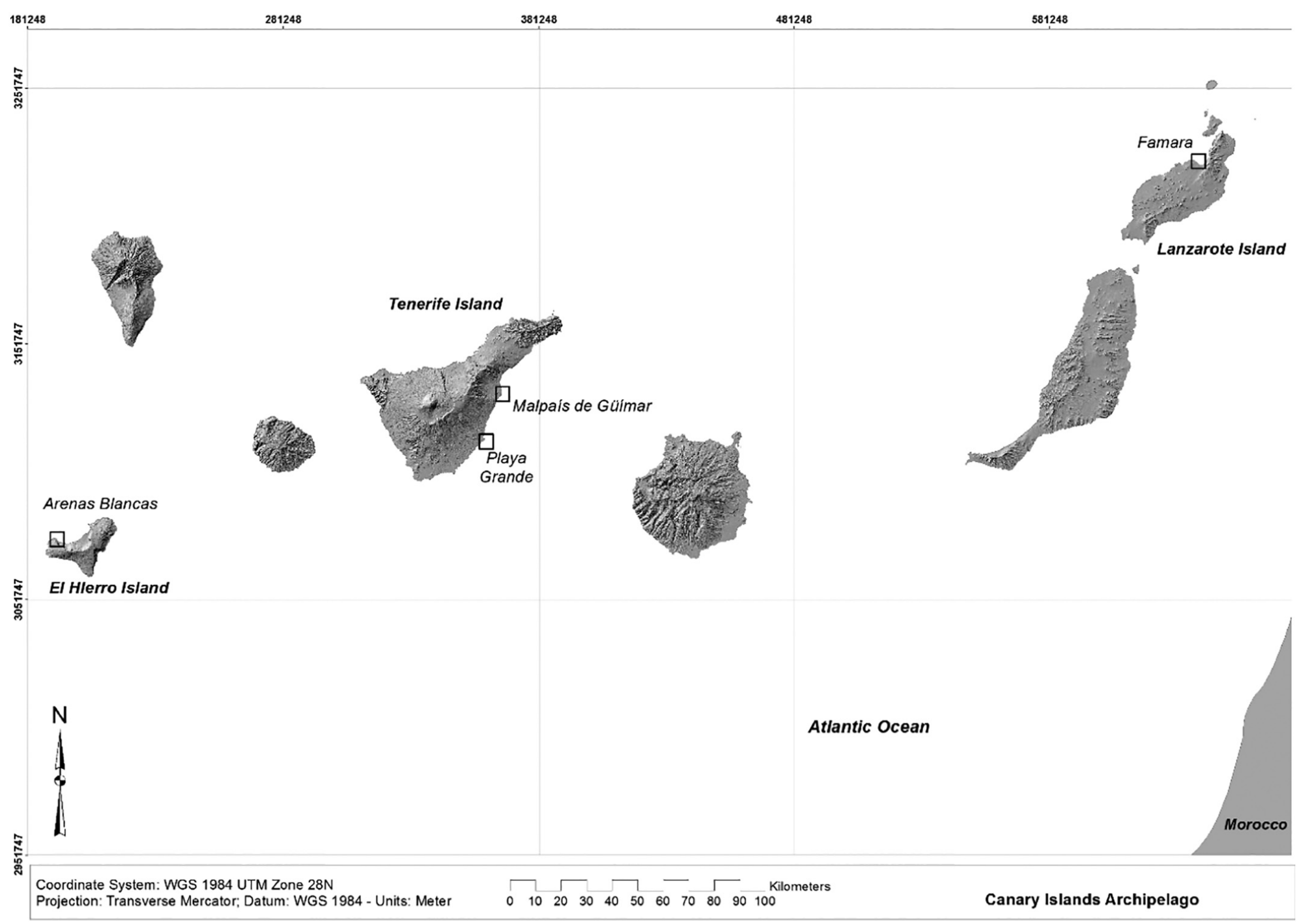

Fig. 1. Location of different areas of the Canary Islands where plastitar has been observed: Arenas Blancas beach (El Hierro), the coast of the Malpaís de Güímar (Tenerife), Playa Grande beach (Tenerife) and the coast of Famara town (Lanzarote).

Table 1
Location, observation periods, orientation and main wind incidence where plastitar was found.

Beach name	Playa Grande beach	Coast of the Malpaís de Güímar	Arenas Blancas beach	Coast of Famara
Municipality and island	Arico (Tenerife)	Güímar (Tenerife)	La Frontera (El Hierro)	Teguise (Lanzarote)
Observation period (once a month)	September 2019 January 2020 June 2020 September 2020 January 2021 April 2021 June 2021 September 2021 January 2022	January 2021 April 2021 June 2021 September 2021 January 2022	June 2020 September 2020 January 2021 April 2021 June 2021 September 2021 January 2022	June 2020 April 2021 June 2021 September 2021 January 2022
Coordinates	28° 09′ 10.03″ N 16° 25′ 53.80″ W	28° 18′ 28″ N 16° 21′ 43″ W	27° 46′ 00.66″ N 18° 07′ 18.38″ W	29° 07′ 12.83″ N 13° 34′ 00.81″ W
Orientation	Northeast	Northeast	East	North
Main wind incidence	North/Northeast	Northeast	East/Northeast	North/Northeast

Prior to FTIR analysis, microplastics were washed with cyclohexane (purity 99.7%), purchased from VWR International Eurolab (Barcelona, Spain), in an ultrasonic bath (Ultrasonic Cleaner USC-600 T working at 45 kHz and 120 W and purchased from VWR International Eurolab) for 15 min in order to remove tar that covered the microplastics and that could preclude the correct identification of the particles. In some cases, it was necessary to scratch the polymer surface or cut it in two parts to obtain a spectrum with enough quality.

2.5. Gas chromatography-flame ionization detection

A 7820A gas chromatographic (GC) system acquired from Agilent Technologies equipped with an autosampler, an HP-5 capillary column ((5%-phenyl)-methylpolysiloxane, 30 m × 0.32 mm × 0.25 μm) and a flame ionization detector (FID) was used. The temperature program used started at 90 °C, which was maintained for 2 min, and then it was increased up to 280 °C at a rate of 8 °C/min, which was kept for 20 min, reaching a total run time of 45.75 min. Nitrogen was used as carrier gas at a flow of 1.5 mL/min and also as make-up gas at 25 mL/min. In addition, hydrogen at a flow of 30 mL/min and air at 400 mL/min were used for stable flame formation. The injector and detector temperatures were set at 280 and 300 °C, respectively. The injection volume was 1 μL, which was injected in splitless mode, and, after 0.75 min, the split was opened with a purge flow of 40 mL/min. The injection program included three needle washes with cyclohexane and one with the sample pre-injection, and five washes with the same solvent post-injection. GC ChemStation software from Agilent Technologies was used to control the GC-FID system.

The gas GC-FID profile of tar was obtained by dissolving 100 mg of tar in 25 mL of cyclohexane, keeping it for a few minutes in an ultrasonic bath until total dissolution. After filtration through a 0.2 μm polyvinylidene fluoride filter from Whatman™ (GE Healthcare, United States), 1 μL of the extract was injected in the GC-FID system.

3. Results and discussion

Within the Interreg-Mac project IMPLAMAC ("Evaluation of the impact of microplastics and emerging contaminants in the coasts of the Macaronesian region") (IMPLAMAC Project, 2019), our group has been studying since October 2019 the arrival of microplastic pollution to different beaches of the Canary Islands, in particular, in the province of Santa Cruz de Tenerife. Our recent studies have shown the existence of two important hotspots of microplastic arrival in the province: one located at Arenas Blancas beach (El Hierro) (Hernández-Sánchez et al., 2021a) and another one at Playa Grande beach (Tenerife) (Álvarez-Hernández et al., 2019; González-Hernández et al., 2020). Both locations, together with Famara beach on the island of Lanzarote (Herrera et al., 2018), are the three main hotspots of the arrival of microplastic debris in the Canary archipelago. These areas are exposed to dominant north/northeast winds that bring high amounts of plastic debris to the coast, in particular microplastics (Álvarez-Hernández et al., 2019; Baztan et al., 2014; González-Hernández et al., 2020; Hernández-Sánchez et al., 2021a).

In those studies, FTIR measurements have revealed that, as likely occurs in the marine environment, most microplastic particles (between 69 and 85%) are composed of PP and PE (Álvarez-Hernández et al., 2019; Baztan et al., 2014; González-Hernández et al., 2020; Hernández-Sánchez et al., 2021a), the two polymers most commonly produced in the world (Plastics Europe, 2021), which, due to their low density, float on sea water. Most reported microplastics have been fragments and pellets, the later found at percentages of 44.3% and 49.9% on the beaches of Famara (Herrera et al., 2018) and Arenas Blancas (Hernández-Sánchez et al., 2021a), respectively. Notably, tar has also been found together with microplastics as tiny pieces of the same size (1–5 mm in their largest dimension) (Hernández-Sánchez et al., 2021a; Herrera et al., 2018).

During the previously mentioned sampling campaigns on the beaches of Arenas Blancas and Playa Grande, it was noted the arrival of large amounts of tar which has attached to the rocks surface. In the case of Famara beach, such issue, which has been widely known by local people, has been mainly observed in the rocky coast of Famara village, at one side of the main beach. Besides, during the examination of the coast of Tenerife within the same project, similar formations were also found along the coast of Malpaís de Güímar. In all cases, it was evident that tar had attached to the rocks over time and that it had agglomerated with other materials that the sea brought to the shore, immobilizing them.

Fig. 2 shows images of such formations found at the rocky parts of Playa Grande beach (Tenerife), while Figs. S1–S4 of the Supplementary Material shows analogous images taken on the coast of Arenas Blancas beach (El Hierro), Famara village (Lanzarote) and Malpaís de Güímar (Tenerife). In the particular case of the pictures taken at Playa Grande, it can be clearly observed that a high number of microplastics (mesoplastics, though to a lesser extent, can also be distinguished) have been embedded in the tar matrix, which occupies a very large surface, as well as small pieces of wood or coarse sand. Since the characteristics and properties of this new formation really differ from those previously reported as plastiglomerates, pyroplastics, plasticrusts or anthropoquinas, we proposed to name it as "plastitar", attending to its two major components.

To characterize this new material in more depth, five quadrants of 20 × 20 cm were randomly placed at five different points on the rocks of Playa Grande beach (see Fig. S5 of the Supplementary Material). Table 2 shows the results of visualization of each quadrant. Concerning the covering surface of the tar, it was estimated that plastitar occupies a 56% of the sampled area, showing a very heterogeneous distribution.

Fig. 3 shows the distribution of shapes and colours of the microplastics found at the sampling points (sizes between 1 and 5 mm). They can be mostly classified as fragments (82.5%, n = 373), pellets (15.7%, n = 71), lines (1.8%, n = 8) and films (0.2%, n = 1) of white/transparent colours. Apart from microplastics, small pieces of highly degraded ropes of 4–10 cm (n = 5) were also observed, as well as other meso and macroplastics of different shapes (n = 54).

Fig. 2. Images of plastitar in Playa Grande beach (Tenerife). Photographs taken in December 2021.

Concerning the composition of the microplastics collected from the plastitar from the five quadrants (85 fragments, 34 pellets and 8 lines), it is important to highlight that different washing solvents were tested (Milli-Q water, methanol and cyclohexane) in order to remove the rests of tar from their surface and improved the quality of the spectra obtained. The best results were obtained with cyclohexane, so it was used as cleaning solvent. FTIR analyses revealed that 90.6% (n = 115) of all microplastics were PE while 9.4% (n = 12) were PP, only these two types of low-density polymers were distinguished. Regarding fragments and pellets, which are the most abundant morphological types, Fig. 4 shows the composition for both microplastic forms, as well as the FTIR spectra of a PP pellet and a PE fragment, respectively, compared with those of the reference materials. As can be seen, the distribution of both polymers is identical for both types of morphologies. Collected data with regards to microplastics composition, shape and colour highly agree with those reported in previous microplastic studies developed in the same coastal area (Álvarez-Hernández et al., 2019; González-Hernández et al., 2020; Hernández-Sánchez et al., 2021a; Herrera et al., 2018).

It should also be taken into consideration that concerning the number, type and composition of microplastics indicated above, these are the only ones found on the surface of plastitar. Depending on its thickness, which can be in some cases of several centimetres, other size debris may be found, being, in general, a quite complex and variable in composition formation.

Regarding tar, the GC-FID profile of the tar samples collected at each quadrant was determined by dissolving a small amount of tar in cyclohexane and injecting it into a GC-FID system as indicated in the Experimental Section (Blanco et al., 1992; Cislak, 1943). Fig. S6 of the Supplementary Material shows the GC-FID chromatogram of one of the samples. In that figure, the typical profile of aliphatic hydrocarbons (which elute at equidistant retention times) could be distinguished as a result of their high concentration (Cortes et al., 2010; Wang et al., 1964). Similar chromatograms were obtained for the five tar samples that were collected. The individual components of the tar as well as the differences or similarities between them, were not determined since this issue was out of the scope of this work.

Figs. S1–S4 of the Supplementary Material correspond to plastitar at Arenas Blancas beach (El Hierro), Famara village (Lanzarote) and the coast of the Malpaís de Güímar (Tenerife). Appearance is quite similar showing a variable composition between the agglomerated materials in terms of size, shapes and colours. Pieces of wood or coarse sand grains as well as small rocks appear in most of the samples. Moreover, in a good number of occasions, small pieces of ropes (1–7 cm length) were found

Table 2
Percentage of plastitar area per quadrant, number of macro and mesoplastic items and types and number of microplastics found.

	Quadrant 1	Quadrant 2	Quadrant 3	Quadrant 4	Quadrant 5	Average (sd)
Plastitar area (%)	60	64	24	91	41	56.0 (25.0)
Number of pellets	20	21	3	11	16	14.2 (7.4)
Number of fragments	49	104	63	65	92	74.6 (22.6)
Number of lines	8	0	0	0	0	1.6 (3.6)
Total number of microplastics	77	125	66	76	108	90.4 (24.9)
Total number of macro/mesoplastics	6	22	3	14	17	12.4 (7.8)

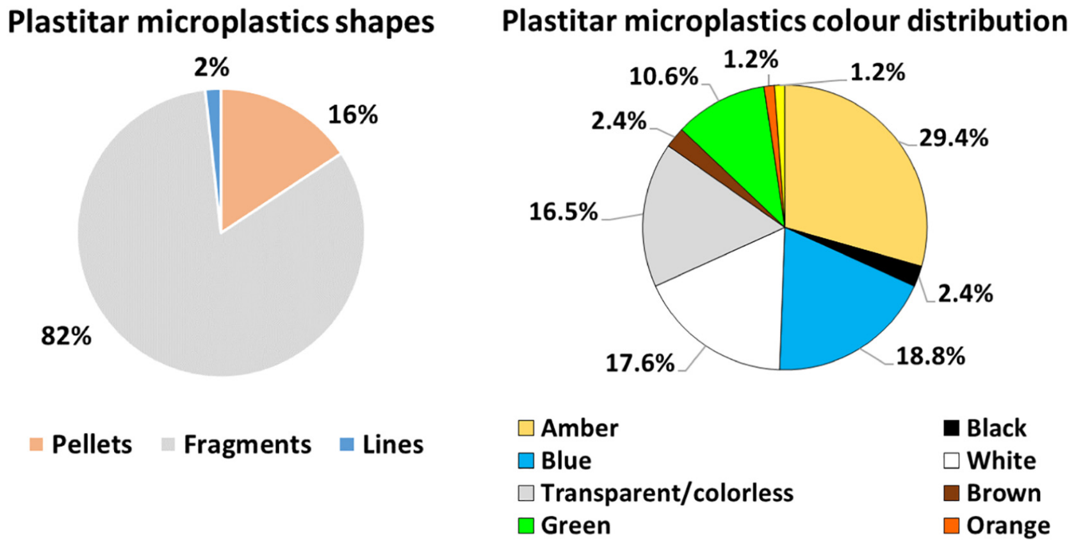

Fig. 3. Distribution of colours and shapes of the microplastics extracted from tar.

completely embedded in the tar. At Malpaís de Güímar and Famara, drier and harder agglomerates than the other two beaches were found, probably as a result of the aging of such debris. In fact, highly degraded plastics could be observed in both cases. In Arenas Blancas (see Fig. S1 of the Supplementary Material), some rocks were completely covered by a thick layer (>3 cm) of tar mixed with an extremely high amount of microplastics of different shapes and colours. Probably, the arrival of both materials took place simultaneously or at least in a very short period of time, yielding to a "carpet" aspect, which is distinctively different from the plastitar found in the other three sites. Judging by all the in-situ observations, a wide variability of shapes and plastic content can be found, being a highly heterogenous material, but the general combination of solidified tar and plastic remains in all of them.

The four study sites have in common that they are exposed to the all year along dominant north/northeast winds (see the rose winds of Figs. S7–S10 of the Supplementary Material), which is a distinctive feature of coastal microplastic contamination hotspots of the Canary Islands archipelago (Álvarez-Hernández et al., 2019; González-Hernández et al., 2020; Hernández-Sánchez et al., 2021a). Therefore, a significant amount of debris (tar included) arrives to these places all

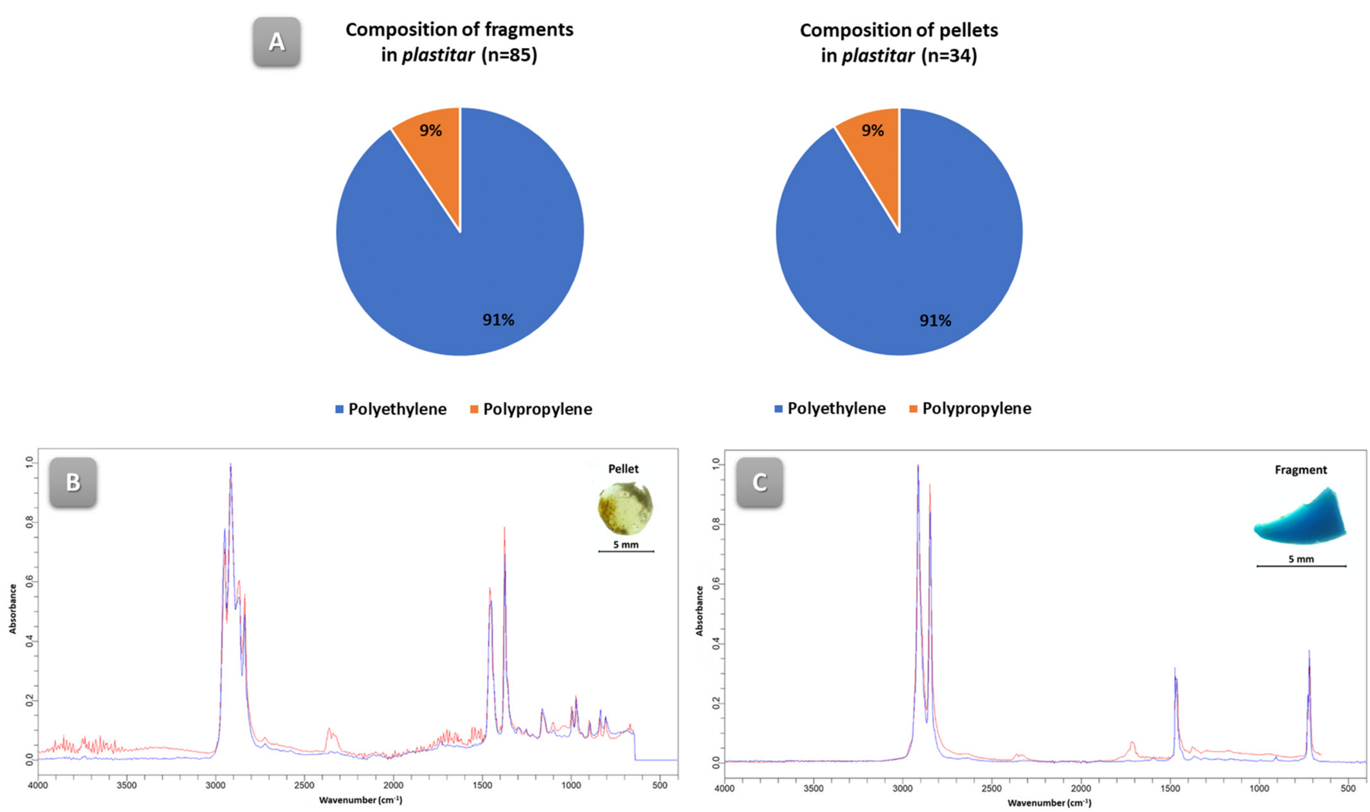

Fig. 4. A) Distribution of the composition of fragments and pellets found in plastitar, B) ATR-FTIR spectra of a pellet identified as polypropylene (red) and ATR-FTIR spectra of the spectral library (blue), and C) ATR-FTIR spectra of a fragment identified as polyethylene (red) and ATR-FTIR spectra of the spectral library (blue).

Table 3
Comparison between previously reported plastic formations and plastitar.

Name	Description	Composition of the plastic material	Places reported	Reference
Plastiglomerate	Anthropogenic multi-composite matrix consisting of melted plastic, beach sediment or sand, basaltic lava debris, bottle tops, and pieces of organic material. Its formation mainly derives from the burning of plastic material during, for instance, campfires or illegal waste burning.	PP (FTIR analysis)	Hawaii, USA	(Corcoran et al., 2014)
			Indonesia USA Portugal Canada	(Corcoran et al., 2018; Corcoran and Jazvac, 2020; Ellrich and Ehlers, 2022)
			Japan	(Furukuma, 2021)
Anthropoquina	Sedimentary rock containing objects of anthropogenic source, including plastic, wood, waste burning, glass, sand and organic materials.	–	Spain	(Irabien et al., 2015)
			Brazil	(Fernandino et al., 2020)
Plasticrust	Plastic pieces encrusted in the texture of intertidal rocks that may persist over time. It might be caused by the coastal wave-induced crash of larger plastic items against rock outcrops. The high summer rock surface temperatures contribute to such process.	PE, PP and PET (FTIR analysis)	Madeira, Portugal	(Gestoso et al., 2019)
			Giglio, Italy	(Ehlers and Ellrich, 2020)
Pyroplastic	An amorphous matrix that appears to be formed by the burning or melting of plastic and that is usually characterized by a single, neutral colour (black-charcoal-grey, off-white or brown), with occasional hues of green, blue, pink or yellow. It has cracks and fractures, pits and cavities. Open campfires and plastic waste burning in beaches are their main sources.	PE and PP (FTIR analysis)	England Scotland Ireland Spain Canada	(Turner et al., 2019)
		PET (FTIR analysis)	Giglio, Italy	(Ehlers and Ellrich, 2020)
		–	Japan	(Furukuma, 2021)
		–	Madeira, Portugal	(Ellrich and Ehlers, 2022)
Plastitar	An agglomerate of tar and mainly microplastics of 1–5 mm size (wood pieces, glass, small rocks and sand grains were also detected) which are attached to the rock surface immobilizing both materials.	PE and PP (FTIR analysis)	Canary Islands, Spain	This work

over the year causing the combination of both materials and their solidification onto the rocks.

Table 3 compiles the characteristics of previously reported plastic formations (i.e. plastiglomerates, pyroplastics, anthropoquinas and plasticrusts) and those of plastitar. As can be inferred from that table, as well as by a visual comparison of the photographs shown in this work with those of the studies compiled in Table 3, tar was not present in any of them or at least it has not been previously reported. Besides, only plasticrusts were the ones retained on the coastal rocks but as a result of the impact of plastics on them, not caused by the previous presence of a material that sticks to the rock (tar). The only feature shared for all those agglomerates is that plastic items of similar composition (i.e. PP or PE) are present. Since PP and PE are the most commonly microplastics found in the marine environment, a coincidence between the types of plastics is expected. All these issues clearly suggest that a new marine-coastal plastic formation has been found.

A relevant aspect about the presence of tar on coastal environments is the fact that it contains hydrocarbons that can be photo-oxidized and impact negatively the marine ecosystem altering the ecological equilibria (Langangen et al., 2017; Zhou et al., 2019). As an example, polycyclic aromatic hydrocarbons (PAHs) present in tar, which are persistent organic pollutants that can bioaccumulate, have moderate to high acute toxicity to aquatic organisms. They can also act as endocrine disruptors and be carcinogenic, among others (Honda and Suzuki, 2020). Its combination with plastic materials clearly supposes a double threat to the marine ecosystem with unknown environmental consequences, since plastics can be ingested by marine organisms causing intestinal blockage, internal injuries, oxidative stress and damage, inflammatory responses, among other important issues (Prokić et al., 2019). Therefore, further research is necessary to fully understand the potential effects of this particular plastic formation, which is probably present in many parts of the globe.

4. Conclusions

A new plastic formation named for the first time as “plastitar” has been reported in several places of the coast of the Canary Islands (Spain), in particular, in three microplastics hotspots of this region. This new formation corresponds to agglomerates of tar and mainly microplastics, in particular, amber, blue and white fragments, pellets and lines of sizes between 1 and 5 mm, which are attached and immobilized to the surface of marine coastal rocks. The combination of two widely known marine contaminants (plastic and tar) has a negative visual impact in the line rocky coastal environment and could represent an unassessed threat for its conservation. This new aggregated formation is probably present in other coastal parts of the world receiving tar spills over the years. Therefore, it could be of high interest to identify new affected areas by this material and monitor its formation process and accumulation rate as well as their potential effects on the marine coastal environment.

CRediT authorship contribution statement

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Declaration of competing interest

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.

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Appendix A. Supplementary data

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References

Álvarez-Hernández, C., Cairós, C., López-Darias, J., Mazzetti, E., Hernández-Sánchez, C., González-Sálamo, J., Hernández-Borges, J., 2019. Microplastic debris in beaches of Tenerife (Canary Islands, Spain). Mar. Pollut. Bull. 146, 26–32. https://doi.org/10.1016/j.marpolbul.2019.05.064.

Baztan, J., Carrasco, A., Chouinard, O., Cleaud, M., Gabaldon, J.E., Huck, T., Jaffrès, L., Jorgensen, B., Miguelez, A., Paillard, C., Vanderlinden, J.P., 2014. Protected areas in the Atlantic facing the hazards of micro-plastic pollution: First diagnosis of three islands in the canary current. Mar. Pollut. Bull. 80, 302–311. https://doi.org/10.1016/J.MARPOLBUL.2013.12.052.

Blanco, C.G., Canga, J.S., Domínguez, A., Iglesias, M.J., Guillén, M.D., 1992. Flame ionization detection relative response factors of some polycyclic aromatic compounds: Determination of the main components of the coal tar pitch volatile fraction. J. Chromatogr. A 607, 295–302. https://doi.org/10.1016/0021-9673(92)87088-P.

Carpenter, A., 2019. Oil pollution in the North Sea: The impact of governance measures on oil pollution over several decades. Hydrobiologia 845, 109–127. https://doi.org/10.1007/s10750-018-3559-2.

Cislak, F.E., 1943. Coal tar solvents. Ind. Eng. Chem. 35, 1020–1022. https://doi.org/10.1021/ie50406a002.

Corcoran, P.L., Jazvac, K., 2020. The consequence that is plastiglomerate. Nat. Rev. Earth Environ. 1, 6–7. https://doi.org/10.1038/s43017-019-0010-9.

Corcoran, P.L., Jazvac, K., Ballent, A., 2018. Plastics and the Anthropocene. In: Dellasala, D.A., Goldstein, M.I.B.T.-E. of the A. (Eds.), Elsevier, Oxford, pp. 163–170 https://doi.org/10.1016/B978-0-12-809665-9.10000-X.

Corcoran, P.L., Moore, C.J., Jazvac, K., 2014. An anthropogenic marker horizon in the future rock record. GSA Today 24, 4–8. https://doi.org/10.1130/GSAT-G198A.1.

Cortes, J.E., Rincon, J.M., Jaramillo, J.M., Philp, R.P., Allen, J., 2010. Biomarkers and compound-specific stable carbon isotope of n-alkanes in crude oils from Eastern Llanos Basin, Colombia. J. S. Am. Earth Sci. 29, 198–213. https://doi.org/10.1016/j.jsames.2009.03.010.

Crawford, C.B., Quinn, B., 2017. Microplastics, standardisation and spatial distribution. Microplastic Pollut. 101–130. https://doi.org/10.1016/B978-0-12-809406-8.00005-0.

De-la-Torre, G.E., Dioses-Salinas, D.C., Pizarro-Ortega, C.I., Santillán, L., 2021. New plastic formations in the anthropocene. Sci. Total Environ. 754, 142216. https://doi.org/10.1016/j.scitotenv.2020.142216.

Ehlers, S.M., Ellrich, J.A., 2020. First record of ‘plasticrusts’ and ‘pyroplastic’ from the Mediterranean Sea. Mar. Pollut. Bull. 151, 110845. https://doi.org/10.1016/j.marpolbul.2019.110845.

Ehlers, S.M., Ellrich, J.A., Gestoso, I., 2021. Plasticrusts derive from maritime ropes scouring across raspy rocks. Mar. Pollut. Bull. 172, 112841. https://doi.org/10.1016/j.marpolbul.2021.112841.

Ellrich, J.A., Ehlers, S.M., 2022. Field observations in pebble beach habitats link plastiglomerate to pyroplastic via pebble clasts. Mar. Pollut. Bull. 174, 113187. https://doi.org/10.1016/j.marpolbul.2021.113187.

Fernandino, G., Elliff, C.I., Francischini, H., Dentzien-Dias, P., 2020. Anthropoquinas: First description of plastics and other man-made materials in recently formed coastal sedimentary rocks in the southern hemisphere. Mar. Pollut. Bull. 154, 111044. https://doi.org/10.1016/j.marpolbul.2020.111044.

Ferreira, T., Rasband, W., 2012. ImageJ User Guide IJ 1.46r.

Furukuma, S., 2021. A study of ‘new plastic formations’ found in the Seto Inland Sea, Japan. Int. J. Sci. Res. Publ. 11, 185–188. https://doi.org/10.29322/IJSRP.11.06.2021.P11427.

Galgani, F., Hanke, G., Werner, S., Oosterbaan, L., Nilsson, P., Fleet, D., Kinsey, S., Thompson, R.C., van Franeker, J., Vlachogianni, T., Scoullos, M., Veiga, J.M., Palatinus, A., Matiddi, M., Maes, T., Korpinen, S., Budziak, A., Leslie, H., Gago, J., Liebezeit, G., 2013. Guidance on Monitoring of Marine Litter in European Seas. https://doi.org/10.2788/99475.

Gestoso, I., Cacabelos, E., Ramalhosa, P., Canning-Clode, J., 2019. Plasticrusts: A new potential threat in the Anthropocene’s rocky shores. Sci. Total Environ. 687, 413–415. https://doi.org/10.1016/j.scitotenv.2019.06.123.

Gola, D., Kumar Tyagi, P., Arya, A., Chauhan, N., Agarwal, M., Singh, S.K., Gola, S., 2021. The impact of microplastics on marine environment: A review. Environ. Nanotechnol. Monit. Manag. 16, 100552. https://doi.org/10.1016/j.enmm.2021.100552.

González-Hernández, M., Hernández-Sánchez, C., González-Sálamo, J., López-Darias, J., Hernández-Borges, J., 2020. Monitoring of meso and microplastic debris in Playa Grande beach (Tenerife, Canary Islands, Spain) during a moon cycle. Mar. Pollut. Bull. 150, 110757. https://doi.org/10.1016/j.marpolbul.2019.110757.

Hernández-Sánchez, C., González-Sálamo, J., Díaz-Peña, F.J., Fraile-Nuez, E., Hernández-Borges, J., 2021a. Arenas Blancas (El Hierro island), a new hotspot of plastic debris in the Canary Islands (Spain). Mar. Pollut. Bull. 169, 112548. https://doi.org/10.1016/j.marpolbul.2021.112548.

Hernández-Sánchez, C., González-Sálamo, J., Ortega-Zamora, C., Jiménez-Skrzypek, G., Hernández-Borges, J., 2021b. Microplastics: An emerging and challenging research field. Curr. Anal. Chem. https://doi.org/10.2174/1573411016999201029194655.

Herrera, A., Asensio, M., Martínez, I., Santana, A., Packard, T., Gómez, M., 2018. Microplastic and tar pollution on three Canary Islands beaches: An annual study. Mar. Pollut. Bull. 129, 494–502. https://doi.org/10.1016/J.MARPOLBUL.2017.10.020.

Honda, M., Suzuki, N., 2020. Toxicities of polycyclic aromatic hydrocarbons for aquatic animals. Int. J. Environ. Res. Public Health 17, 1363. https://doi.org/10.3390/ijerph17041363.

IMPLAMAC Project, 2019. IMPLAMAC – Evaluación del impacto de microplásticos y contaminantes emergentes en las costas de la Macaronesia. https://implamac.com/ (accessed 1.21.22).

International Maritime Organization, 2021. MARPOL annex I - prevention of pollution by oil. https://www.imo.org/en/OurWork/Environment/Pages/OilPollution-Default.aspx (accessed 1.24.22).

Irabien, M.J., García-Artola, A., Cearreta, A., Leorri, E., 2015. Chemostratigraphic and lithostratigraphic signatures of the anthropocene in estuarine areas from the eastern cantabrian coast (N. Spain). Quat. Int. 364, 196–205. https://doi.org/10.1016/j.quaint.2014.08.056.

Langangen, Ø., Olsen, E., Stige, L.C., Ohlberger, J., Yaragina, N.A., Vikebø, F.B., Bogstad, B., Stenseth, N.C., Hjermann, D.Ø., 2017. The effects of oil spills on marine fish: Implications of spatial variation in natural mortality. Mar. Pollut. Bull. 119, 102–109. https://doi.org/10.1016/j.marpolbul.2017.03.037.

Marine Environment Protection Committee, 2005. Description of the Canary Islands Particularly Sensitive Sea Area.

Plastics Europe, 2021. Plastics-the facts. https://plasticseurope.org/knowledge-hub/plastics-the-facts-2021/.

Prokić, M.D., Radovanović, T.B., Gavrić, J.P., Faggio, C., 2019. Ecotoxicological effects of microplastics: Examination of biomarkers, current state and future perspectives. TrAC Trends Anal. Chem. 111, 37–46. https://doi.org/10.1016/j.trac.2018.12.001.

Shirneshan, G., Bakhtiari, A.R., Memariani, M., 2016. Identification of sources of tar balls deposited along the Southwest Caspian coast, Iran using fingerprinting techniques. Sci. Total Environ. 568, 979–989. https://doi.org/10.1016/j.scitotenv.2016.04.203.

Suneel, V., Vethamony, P., Zakaria, M.P., Naik, B.G., Prasad, K.V.S.R., 2013. Identification of sources of tar balls deposited along the Goa coast, India, using fingerprinting techniques. Mar. Pollut. Bull. 70, 81–89. https://doi.org/10.1016/j.marpolbul.2013.02.015.

Turner, A., Wallerstein, C., Arnold, R., Webb, D., 2019. Marine pollution from pyroplastics. Sci. Total Environ. 694, 133610. https://doi.org/10.1016/j.scitotenv.2019.133610.

Vighi, M., Bayo, J., Fernández-Piñas, F., Gago, J., Gómez, M., Hernández-Borges, J., Herrera, A., Landaburu, J., Muniategui-Lorenzo, S., Muñoz, A.R., Rico, A., Romera-Castillo, C., Viñas, L., Rosal, R., 2021. Micro and nanoplastics in the environment: Research priorities for the near future. Springer, Cham, pp. 163–218.

Wang, Z., Fingas, M., Yang, C., Christensen, J.H., 1964. In: Morrison, R.D., Murphy, B.L.B.T.-E.F. (Eds.), 16 - Crude oil and refined product fingerprinting: Principles. Academic Press, Burlington, pp. 339–407.

Zhou, Z., Li, X., Chen, L., Li, B., Wang, C., Guo, J., Shi, P., Yang, L., Liu, B., Song, B., 2019. Effects of diesel oil spill on macrobenthic assemblages at the intertidal zone: A mesocosm experiment in situ. Mar. Environ. Res. 152, 104823. https://doi.org/10.1016/j.marenvres.2019.104823.