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EDITED BY

André Ricardo Araújo Lima,
Center for Marine and Environmental
Sciences (MARE), Portugal

REVIEWED BY

Levent Bat,
Sinop University, Türkiye
Fernanda Avelar Santos,
Federal University of Paraná, Brazil

*CORRESPONDENCE

Sonja M. Ehlers
✉ sonja.ehlers@awi.de

†PRESENT ADDRESS

Sonja M. Ehlers,
Shelf Sea System Ecology, Alfred Wegener
Institute Helmholtz Centre for Polar and
Marine Research, Helgoland, Germany

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Plastitar records in marine coastal environments worldwide from 1973 to 2023

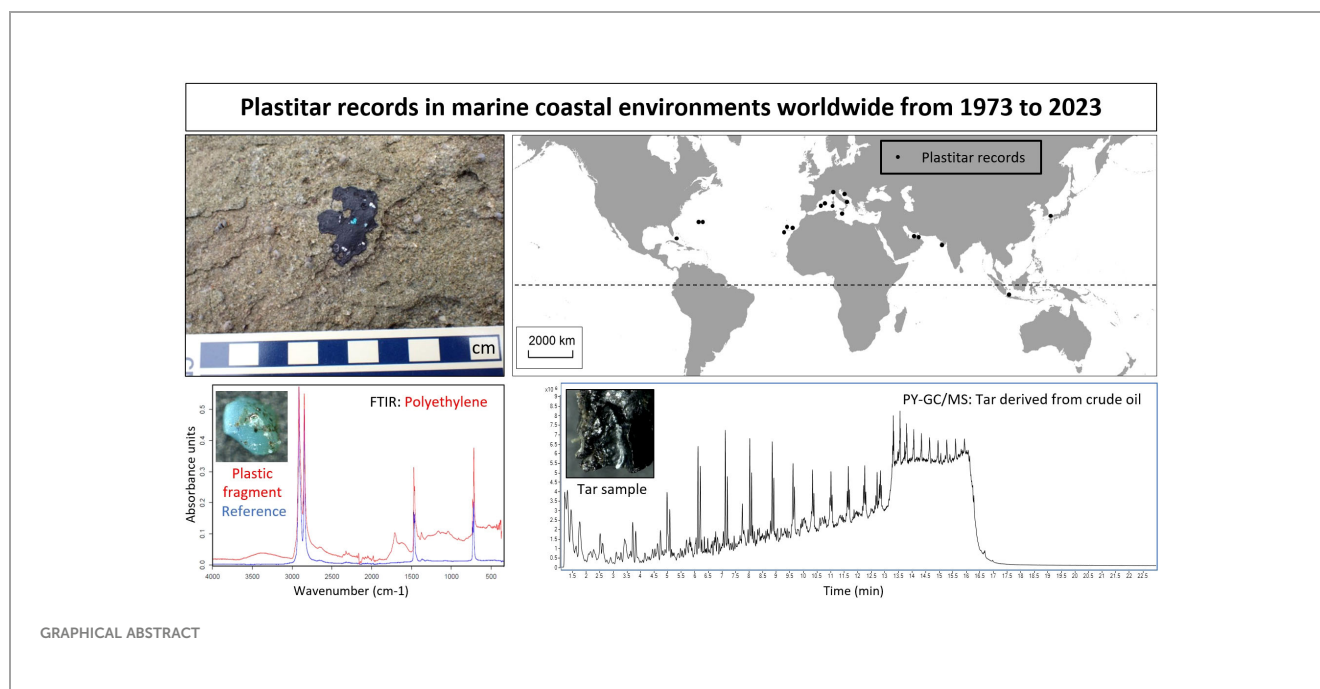
Julius A. Ellrich¹, Sonja M. Ehlers^{2*†} and Shunji Furukuma³

¹Shelf Sea System Ecology, Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research, Helgoland, Germany, ²Department of Animal Ecology, Federal Institute of Hydrology, Koblenz, Germany, ³Independent Researcher, Ube, Yamaguchi, Japan

Plastics embedded in tar residues encrusting rocky coastlines were discovered on the Canary Islands (NE Atlantic Ocean) and termed “plastitar” in 2022. As plastic and tar pollution is widespread in marine pelagic and benthic habitats, it was predicted that this novel plastic form could occur on other coasts as well. To test this prediction, we reviewed the literature on plastic embedded in tar and examined rocky coastlines in our regions for plastitar. We found eight studies which reported plastic embedded in tar under several descriptions and terms (including “tar-bonded beach-conglomerate tarcrete”, “plasto-tar crust”, “plasto-tarball” and “plastitar”) in India (Arabian Sea), Bermuda (Sargasso Sea), the Bahamas (NW Atlantic Ocean), the United Arab Emirates (Arabian Gulf and Gulf of Oman), Malta (central Mediterranean Sea), Croatia (Adriatic Sea), Italy (Adriatic and Ligurian Sea), Cabrera island, Menorca island and Sardinia island (western Mediterranean Sea) and Indonesia (Java Sea) from 1973 to 2023. We also detected plastitar during our field surveys in Japan (Sea of Japan) and Mallorca island (western Mediterranean Sea) in 2022 and 2023, respectively. Using Fourier-transform infrared spectroscopy (FTIR), we verified the contained plastics as polyethylene (PE). Pyrolysis-gas chromatography-mass spectrometry (PY-GC/MS) indicated that the tar residues derived from crude oil. Furthermore, plotting all these plastitar records in a global map revealed that almost all plastitar findings were made along the major oil transportation routes across the northern hemisphere suggesting that plastitar resulted from tanker-released crude oils. Overall, our study shows, for the first time, that plastitar has been a widespread and long overlooked plastic form in coastal systems worldwide.

KEYWORDS

plastic, tar, tarcrete, plasto-tar crust, plasto-tarball, crude oil, plastic forms



1 Introduction

Recently, plastic pellets and microplastics (plastic particles < 5 mm) embedded in tar residues encrusting rocky coastlines were detected on the Canary Islands, Atlantic Ocean (Domínguez-Hernández et al., 2022). By combining regional field surveys and regular monitorings with state-of-the-art laboratory analyses, including Fourier-transform infrared spectroscopy (FTIR) and gas chromatography-flame ionization detection (GC-FID), these authors verified the main components of their findings as plastic and tar. Based on these components, Domínguez-Hernández et al. (2022) termed their findings “plastitar”. Since all plastitar findings on the Canary Islands were made along onshore wind-exposed coasts facing nearby oil transportation routes, they proposed that plastitar may result from plastics interacting with tanker-released crude oil residues, that are commonly known as “tarballs” (Peters and Siuda, 2014; Warnock et al., 2015), being washed ashore. Domínguez-Hernández et al. (2022) also discussed their findings in relation to other recently discovered plastic forms (geochemically or -physically altered plastic; Ellrich et al., 2023a) including, for example, “pyroplastics” (incompletely combusted, melted and weathered plastics with a rock-like appearance; Turner et al., 2019) and “plasticrusts” (plastics encrusting intertidal rocks; Gestoso et al., 2019) that derive from plastics being abraded on rocky coasts by water motion (Ehlers et al., 2021; Ellrich et al., 2023b). Finally, based on their regional findings on the Canary Islands and the worldwide co-occurrence of marine plastic and tar pollution, Domínguez-Hernández et al. (2022) predicted that plastitar must occur on other coasts as well and that it is, therefore, important to identify such coasts to study the occurrence, formation and accumulation of this new plastic form.

To examine this prediction, we reviewed the literature for combinations of plastic and tar as these two pollutants are well documented in marine pelagic and benthic habitats since the 1970s (Tables S1–S3). We also surveyed coasts in our regions for plastitar and analyzed our findings using FTIR and pyrolysis-gas chromatography-mass spectrometry (PY-GC/MS). Including the findings by Domínguez-Hernández et al. (2022), our review detected nine studies reporting plastic embedded in tar (under various descriptions and terms) on coastlines worldwide from 1973 to 2023. Our field surveys found plastitar in the Sea of Japan and the western Mediterranean Sea in 2022 and 2023, respectively. Furthermore, including all these plastitar records in a global map indicated that plastic embedded in tar occurred almost exclusively along the major oil transportation lines across the northern hemisphere from the Bahamas (NW Atlantic) to Kanda (Sea of Japan) over half a century from 1973 to 2023. Altogether, these findings indicate that plastitar is not a new but long-overlooked and widespread plastic form in coastal environments worldwide.

2 Materials and methods

2.1 Literature review

For our review, we first searched the literature using combinations of the keywords ‘plastitar’, ‘plastic’, ‘oil’, ‘tar’, ‘pellet’, and ‘spill’ in Google Scholar, Web of Science, DuckDuckGo and Google and examined the detected studies for these keywords. We focused on studies written in English and Japanese. When a study reported combinations of plastic and tar, we differentiated between plastitar records (e.g., plastic embedded in tar), plastic and tar co-occurrence

records (e.g., plastic associated with tar) and tarred plastic records (e.g., plastic smeared with tar). Since these searches also detected the terms “tar-bonded beach-conglomerate tarcrete” (Gregory, 1983), “plasto-tar crust” and “plasto-tarball” for plastic embedded in tar (Wilber, 1987), we added these terms as keywords to our second literature search. For maximum data transparency, we included all original terms and descriptions used for plastitar (Table S1), plastic and tar co-occurrence (Table S2) and tarred plastic (Table S3) from the literature. We also checked the references section of each study for earlier articles reporting on combinations of plastic and tar. Finally, we examined all newer articles which had cited the respective study using the ‘cited’ function in Google Scholar and Web of Science. We performed this literature review from March to September 2023.

2.2 Field surveys and laboratory analyses

To examine whether plastitar occurs in our regions, we surveyed wave-exposed rocky intertidal habitats in Kanda, Yamaguchi Prefecture, Japan, Sea of Japan (34.333792, 130.890457) on 24 July 2022 and Cala Rajada, Mallorca, Balearic Islands, Spain, western Mediterranean Sea (39.707647, 3.460500) on 18 February 2023 for plastic embedded in tar. Plastic is common off Kanda (Enamul-Kabir et al., 2020) and Cala Rajada (Compa et al., 2020) and tar has repeatedly been reported in both regions (Zsolnay, 1987; Ruiz-Orejón et al., 2016; JMA, 2023). During our surveys we carefully examined the entire intertidal zone in Kanda (1550 m²; Figure S1) and Cala Rajada (1275 m²; Figure S2) for plastitar, collected our findings using a metal spatula and stored them in sealed and padded individual plastic bags for transportation to the lab in Koblenz, Germany (Ellrich and Ehlers, 2022).

At the lab, we separated the plastic and tar components using metal tweezers. Next, we used FTIR to identify the polymer types of the plastic components (Ellrich and Ehlers, 2022) and PY-GC/MS to verify the tar components (Mun and Ku, 2010). For our FTIR analyses, we used a Vertex 70 (Bruker, Ettlingen, Germany; equipped with a diamond crystal) in attenuated total reflectance (ATR) mode in a wavenumber range between 4000 and 370 cm⁻¹ with eight co-added scans and a spectral resolution of 4 cm⁻¹ (Ellrich and Ehlers, 2022). We then compared the measured spectra with the Bruker spectral library using Opus 8.5 software (Bruker, Ettlingen, Germany). For our PY-GC/MS analyses, we used a Multi-Shot Pyrolyzer EGA/PY-3030D (Frontier Laboratories, Saikon, Japan) and an Auto-Shot Sampler AS-1020E (Frontier Laboratories, Saikon, Japan). The pyrolyzer was attached to an Agilent 7890B gas chromatograph (Santa Clara, CA, USA) equipped with an Ultra ALLOY® UA-5(MS/HT) metal capillary separation column (Frontier Laboratories, Saikon, Japan). We pyrolyzed the tar at 600°C using a split ratio of 1:50. An Agilent MSD 5977B (Santa Clara, CA, USA) in scan mode (40–800 Da) was used for detection (Pojar et al., 2021).

3 Results and discussion

Including the findings by Domínguez-Hernández et al. (2022), our study showed that plastics embedded in tar residues were

reported (under various descriptions and terms) by nine studies in several coastal locations worldwide from 1973 to 2023 (Table S1). The earliest record was made in Gosabara, India where plastic beads embedded in beached tarball deposits were detected in 1973 (Dwivedi et al., 1974). In the 1980s, Gregory (1983) reported plastic pellets attached to tarballs and plastic incorporated in tar encrusting rocks (termed: tar-bonded beach-conglomerate tarcrete) on Bermuda, Saragossa Sea. Wilber (1987) recorded plastics embedded in pelagic and beached tarballs (termed: plasto-tarballs) and plastics incorporated in tar encrusting rocks (termed: plasto-tar crusts) off/on Bermuda and the Bahamas, NW Atlantic Ocean. In the 1990s, plastic spherules agglomerated with tarballs were found along the Arabian Gulf coast and the Gulf of Oman coast (Khordagui and Abu-Hilal, 1994). In the 2010s, plastic pellets embedded in tar deposits encrusting rocks were reported on Malta island, central Mediterranean Sea (Turner and Holmes, 2011) and the term plasto-tarball (for aggregations of plastic pellets, plastic fragments and tar) was used in a review (Eriksen et al., 2017). In 2020, beached plasto-tarballs (aggregations of plastic debris, pellets and fragments with tarballs) were collected on Žirje island, Croatian Adriatic Sea coast (Fajković et al., 2020). Very recently, a single non-floating tar matrix containing a plastic cap, sand, leaves, coral and shell fragments was reported as plastitar on Panjang island, Indonesian Java Sea coast (Utami et al., 2023) and plastitar encrusting coastal rocks was detected in Italy (Adriatic and Ligurian Sea), Cabrera island, Menorca island and Sardinia island (western Mediterranean Sea; Saliu et al., 2023; Table S1).

Additionally, we provide first records of plasto-tar crusts on intertidal rocks in Kanda, Sea of Japan (Figure 1A) and Cala Rajada, Mallorca, western Mediterranean Sea (Figure 2A). Using FTIR, we verified that the embedded plastics (Figures 1B, 2B) consisted of polyethylene (PE, Figures 1C, 2C) which is a common polymer type in waters off Kanda (Enamul-Kabir et al., 2020) and Cala Rajada (Compa et al., 2020). We also present the first FTIR spectra of the tar components (Figures 1D, 2D) of these plasto-tar crusts (Figures 1E, 2E). Furthermore, using PY-GC/MS, we confirmed that the tar components in Kanda and Cala Rajada derived from crude oil (Figures 1F, 2F; Table S1). Due to flash pyrolysis at 600°C short-chain alkenes were formed during the analysis and, together with the corresponding alkanes, can be seen as doublets in the pyrograms (Figures 1F, 2F). Crude oil mainly consists of hydrocarbons. Alkanes and alkenes are different hydrocarbon subgroups. Therefore, these results confirm that the analyzed tar components derived from crude oil.

From a methodological perspective, one could certainly argue that the plastic and tar analyses performed by Domínguez-Hernández et al. (2022) are more thorough than those performed by the previous studies which in most cases (except Fajković et al., 2020) lacked chemical plastic and tar verification (Table S1). Still, one has to take into account that most of the earlier studies (except Fajković et al., 2020) focused on reporting either pollution by tar (Dwivedi et al., 1974) or plastic pellets (Gregory, 1983; Wilber, 1987; Khordagui and Abu-Hilal, 1994; Turner and Holmes, 2011), which are two visually very easily recognizable materials (e.g., Gregory, 1977; Gregory, 1978; Shiber, 1979; Morris, 1980; Shiber, 1982; Gregory, 1983; Wilber, 1987; Shiber and Barrales-Rienda,

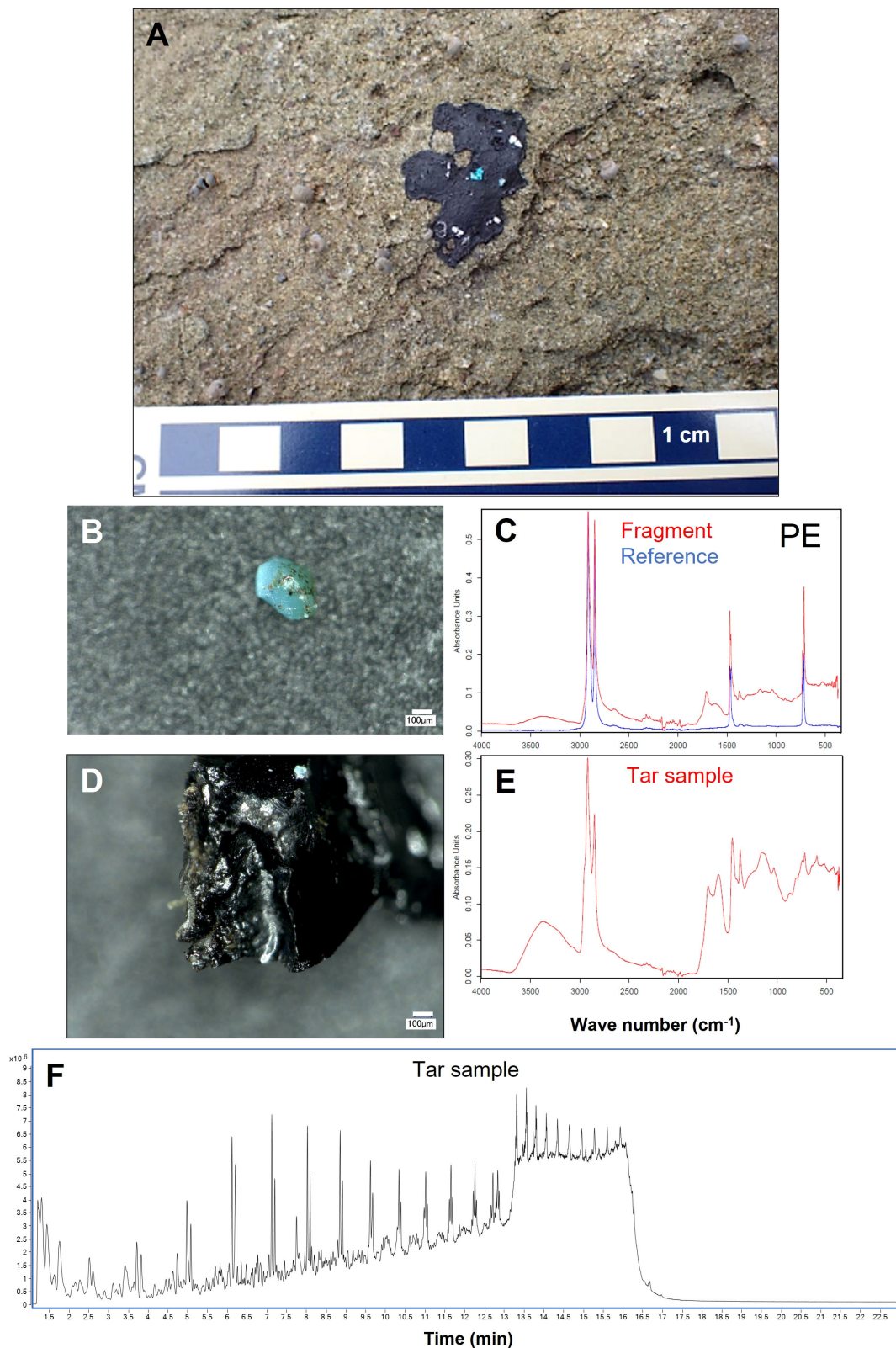


FIGURE 1
(A) Top view on a plasto-tar crust firmly encrusting the natural rocky substrate at Kanda, Houhoku-cho (Shimonoseki city, Yamaguchi Prefecture, Honshu, Japan) in May 2022. **(B)** A blue plastic fragment extracted from the plasto-tar crust. **(C)** The blue plastic fragment consisted of polyethylene (PE). The measured FTIR spectrum is depicted in red and the reference spectrum from the Bruker database is depicted in blue. **(D)** A black tar sample extracted from the plasto-tar crust. **(E)** The FTIR spectrum of the black tar sample depicted in red. **(F)** The PY-GC/MS pyrogram of the black tar sample. Further details on plasto-tar crusts in Kanda are provided in [Figure S1](#).

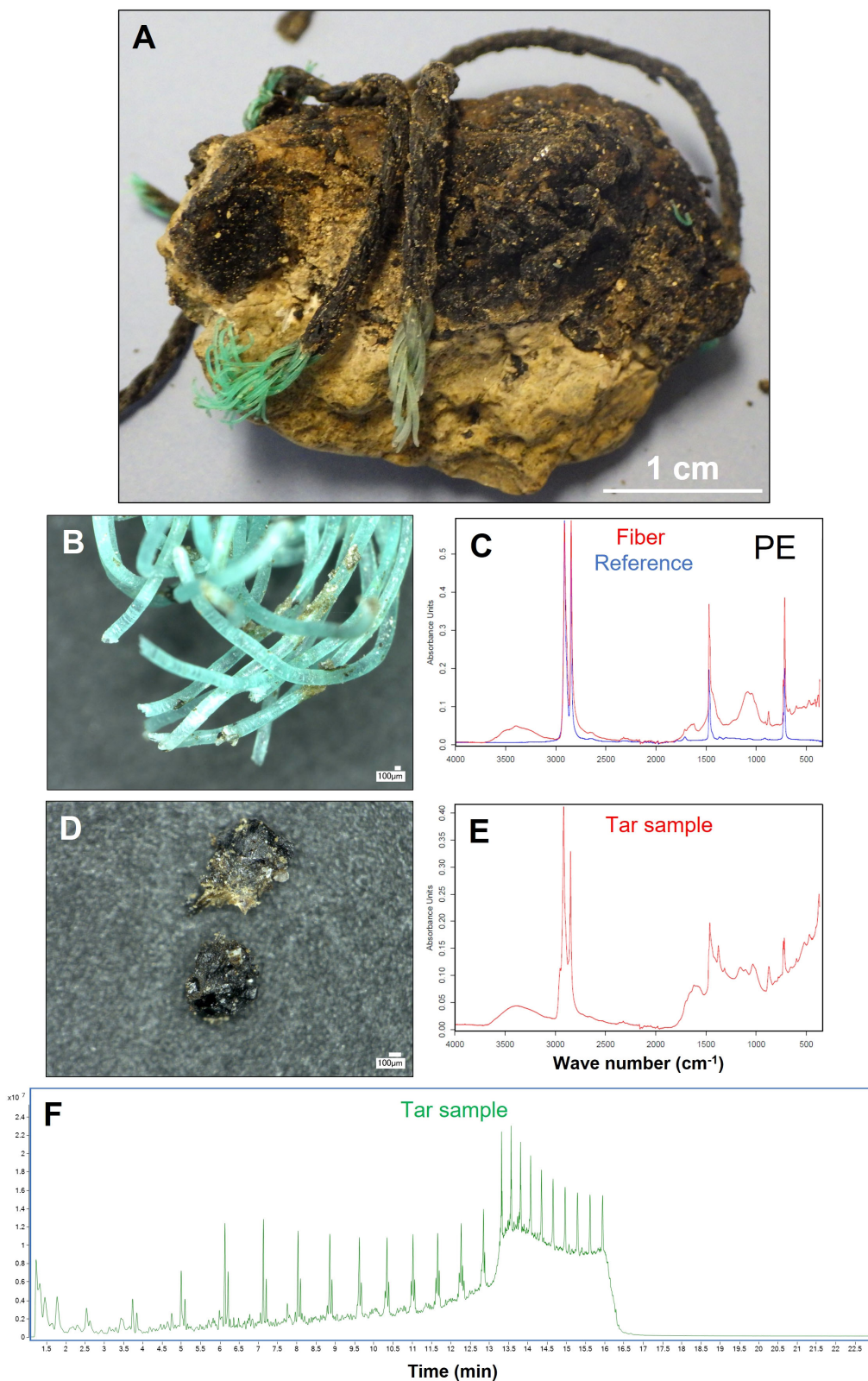


FIGURE 2
(A) Top view on a plasto-tar crust collected in Cala Rajada, Mallorca (Balearic Islands, Mediterranean Sea) in February 2022. This plasto-tar crust was removed from the rocky substrate and photographed at the lab. It contained black tar, green plastic rope, a brown pebble and sand grains. **(B)** Green plastic fibers extracted from the plasto-tar crust. **(C)** The green plastic fibers consisted of PE. The measured FTIR spectrum is depicted in red and the reference spectrum from the Bruker database is depicted in blue. **(D)** A black tar sample extracted from the plasto-tar crust. **(E)** The FTIR spectrum of the black tar sample depicted in red **(F)** The PY-GC/MS pyrogram of the black tar sample. Further information on plasto-tar crusts in Cala Rajada is provided in [Figure S2](#).

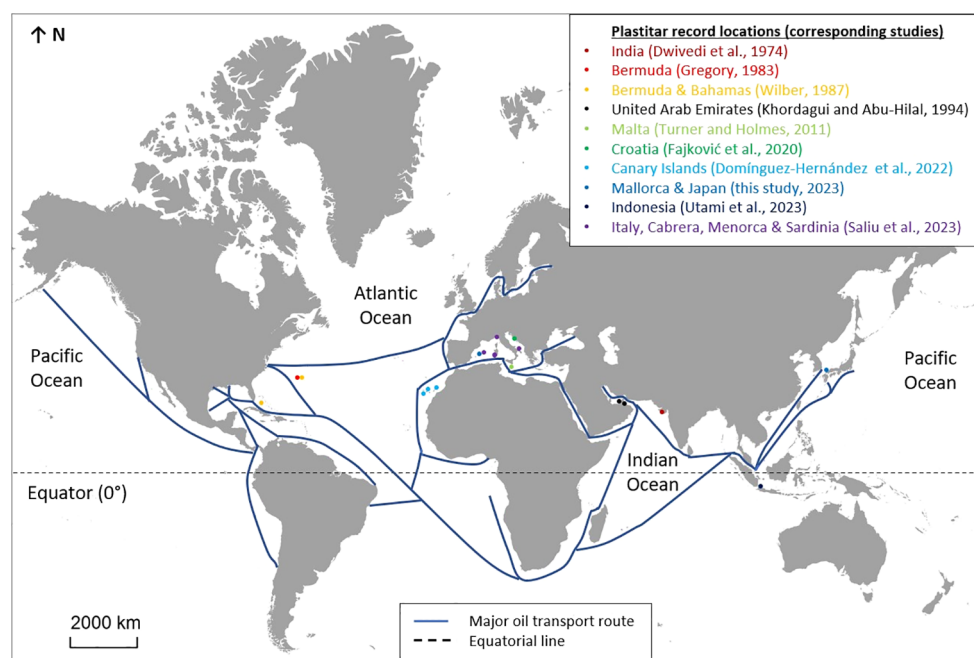


FIGURE 3

Plastitar records in marine coastal environments worldwide from 1973 to 2023. The color coded dots represent the locations of the plastitar records and the corresponding studies. The major oil transportation routes were adopted from the literature (Gupta et al., 1993; EIA, 2017; Rodrigue, 2020).

1991; Turner and Holmes, 2011; Wang and Roberts, 2013; Peters and Siuda, 2014; Warnock et al., 2015; Stachowitsch, 2019), and in most cases reported plastic embedded in tar only as an additional finding (except Fajković et al., 2020). In addition, we note that numerous field observations worldwide documented plastic-tar co-occurrence (Wellman, 1973; Dwivedi and Parulekar, 1974; Dwivedi et al., 1974; Dwivedi et al., 1975; Unnithan et al., 1981; Shiber, 1989; Abu-Hilal and Khordagui, 1993; Dufault and Whitehead, 1994; Herrera et al., 2017; Hernández-Sánchez et al., 2021; Table S2) and tarred plastics indicating plastic-tar interactions in coastal and marine environments (Shiber, 1979; Shiber, 1982; Gregory, 1983; Wilber, 1987; Shiber, 1989; van Franeker, 1989; Shiber and Barrales-Rienda, 1991; Dufault and Whitehead, 1994; Abu-Hilal and Al-Najjar, 2009; Table S3). Considered together, these records of plastitar, plastic-tar co-occurrences and plastic-tar interactions suggest that plastitar is a widespread phenomenon since the 1970s.

Interestingly, plotting all plastitar records (Table S1) in a global map showed that most plastitar records (except Fajković et al., 2020 and Utami et al., 2023) were made along the major oil transportation routes (Figure 3). This observation supports the notion that plastitar derives from tanker-released oil (Dominguez-Hernández et al., 2022) and suggests that plastitar may, to some extent, contain ship-released plastic (Vauk and Schrey, 1987; Unger and Harisson, 2016). Oil is also transported across the Adriatic Sea (Liubartseva et al., 2023) and extracted in the Java Sea (Setiawan et al., 2017) suggesting that the nearby plastitar records in Croatia (Fajković et al., 2020) and Indonesia (Utami et al., 2023) could derive from these oil sources. To further examine

potential links between plastitar and tanker-released oil, it would be interesting to compare beached plasto-tarball samples and benthic plasto-tar crust samples with pelagic tanker-released oil and tarball samples. Furthermore, our map shows that almost all plastitar records (except Utami et al., 2023) were made in the northern hemisphere (Figure 3). Yet, pelagic tarballs have repeatedly been reported off the South African coast (Eagle et al., 1979; Shannon and Chapman, 1983a; Shannon and Chapman, 1983b; Shannon et al., 1983) along which oil is transported (Figure 3). Thus, surveying this coast for plasto-tarballs and plasto-tar crusts could be useful to examine whether plastitar occurs in the northern and southern hemispheres with similar frequencies. Plastic and tar have repeatedly been reported washed up on the western coastline of India (Nair et al., 1972; Dwivedi and Parulekar, 1974; Dwivedi et al., 1974; Dwivedi et al., 1975; Quasim, 1975; Dhargalkar et al., 1977; Unnithan et al., 1981; Kaladharan et al., 2004; Rekadwad and Khobragade, 2015; Suneel et al., 2016; Shinde et al., 2017; Sivadas et al., 2021; Mishra et al., 2023; TAM Database, 2023) along which large oil amounts are transported (Gupta et al., 1993; EIA, 2017; Rodrigue, 2020). Here, intense tar strandings are associated with the monsoon season during which strong onshore winds prevail (Nair et al., 1972; Dhargalkar et al., 1977; Unnithan et al., 1981; Kaladharan et al., 2004; Shinde et al., 2017). Together with the aforementioned plastitar observations along wind-exposed coasts of the Canary Islands (Dominguez-Hernández et al., 2022), these findings suggest that the occurrence, formation and accumulation of beached plasto-tarballs and plasto-tar crusts is related to regional tar and plastic pollution levels at sea and the prevailing wind

directions. Similarly, several recent plastitar records were made along the SW coast of Sardinia island (Saliu et al., 2023) which is a coastline exposed to strong offshore surface water circulation (Millot and Taupier-Letage, 2005), on- and alongshore currents (Pinardi et al., 2015) supporting the notion that plastic and tar may accumulate in such circulation centers before being transported coastward (Wilber, 1987). Therefore, exploring regional relationships between prevailing winds, currents and plastitar could help to examine whether onshore winds and currents are environmental drivers of coastal plastitar occurrence, formation and accumulation.

Overall, our study found that plastitar descriptions, terms and occurrence records were scattered across the marine pollution literature and that information on the plastitar generation process is still missing. Therefore, we hope that this study will help developing plastitar studies to understand the occurrence, formation and accumulation of this widespread and long-overlooked plastic form in coastal systems worldwide.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material. Further inquiries can be directed to the corresponding author.

Author contributions

JE: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. SE: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Validation, Visualization, Writing – review & editing. SF: Investigation, Methodology, Writing – review & editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmars.2023.1297150/full#supplementary-material>

SUPPLEMENTARY FIGURE 1

Plastitar findings in Kanda, Yamaguchi Prefecture, Honshu, Japan. (A) Northwest view on the surveyed wave-sheltered rocky coast facing the Sea of Japan in April 2022. Tsunoshima island can be seen in the background. (B–D) Top view on examples of plastitar findings on the rocky substrate facing the Sea of Japan in July 2022. In total, five such plastitar findings were made in Kanda. All plastitar findings were firmly attached to the substrate, relatively thin (millimeters thick), hard and rough. Tar color ranged from black to gray. Plastics, such as pink, green and white fibers as well as pink fragments (indicated by red arrows) were firmly embedded in the tar matrix and appeared weathered. The tar also contained sand grains. (E) Map showing the location of the surveyed rocky coast in Kanda. (F) Enlargement indicating the surveyed rocky coast. The map and the enlargement show that the surveyed rocky coast is sheltered from incoming waves by the nearby Tsunoshima and Futago islands and the adjacent Nabeshima peninsula.

SUPPLEMENTARY FIGURE 2

Plastitar findings in Cala Rajada, Mallorca, Balearic Islands, Spain in February 2023. (A) Southeast view on the surveyed wave-exposed rocky coast facing the open Mediterranean Sea directly. (B–D) Top view on examples of plastitar findings detected among the rocks. In total, we made approximately 30 plastitar findings in Cala Rajada. All plastitar findings were firmly attached to the rocky substrate. The tar components usually occurred in rock depressions and crevices in wave-sheltered spots that were protected from incoming waves by adjacent larger rocks suggesting that such spots facilitate tar deposition on wave-swept shores. Tar color ranged from black to gray. Gray tar surfaces were somewhat harder and rougher than black tar surfaces suggesting that these differences in color, elasticity and texture may reflect tar drying and weathering from relatively new (black) to older (grey) tar by sunlight and temperature. Tar thickness ranged from millimeters (tar on rock surfaces) to centimeters (tar in rock depressions and crevices). Plastics (including green fibers, blue, red and gray fragments indicated by red arrows), small rocks, wood pieces, sand grains, mollusk and barnacle shells (not shown) were frequently embedded in the tar. During plastitar collection, we noted that black (but not gray) tar samples had a smooth and kneadable core. Partially tar-embedded plastics appeared more weathered than fully tar-embedded plastics suggesting that the tar matrix slows down plastic weathering. (E) Map showing the location of the surveyed rocky coast in Cala Rajada. (F) Enlargement of the surveyed rocky coast.

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