Monitoring plastic pellet pollution in coastal environments through handheld Raman spectroscopy: Data from the Mediterranean coasts (Southern Italy)

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ABSTRACT

This paper examines the distribution and chemical properties of beached plastic pellets along the Ionian and Tyrrhenian coasts of Southern Italy. Three locations have been sampled: Agnone Bagni (SR) and Paradiso (ME) on the Ionian coast of Sicily, Baia del Tono in Milazzo (ME) on the Sicilian Tyrrhenian coast, and Pizzo Calabro (VV) in Calabria on the Tyrrhenian coast. Variations in shape, size, compactness, color, and other physical features, correlated with residence times and transport, has been highlighted. Raman spectroscopy, used in a portable configuration, enabled rapid identification of polymer types, demonstrating its utility for on-site plastic pollutant monitoring. Polyethylene and polypropylene were the predominant polymers. Principal component analysis of the spectra determined the optimal chemometric classification of pellets by composition, avoiding interference or distortion. In conclusion, the study provided preliminary insights into pellet abundance, composition, weathering extent, and distribution across these shorelines, underscoring the importance of regular beach monitoring.

1. Introduction

Plastic pellets, commonly known as nurdles, are small lentil-sized pieces of pre-production plastic that are the raw materials used in the manufacture of various plastic products (Sewwandi et al., 2023; Jiang et al., 2021; Giugliano et al., 2022). Due to their small size (typically from 3 to 5 mm) and shape, nurdles can be easily mistaken for small stones or other debris. They are generally rounded and often perfectly symmetrical, unlike natural objects, and can come in various colors, though clear or white are the most common. These small plastic particles serve as raw materials for manufacturing a wide array of plastic products. After a period of stagnation in 2020 due to the Covid-19 pandemic, global plastic production increased to 390.7 million tons in 2021, with 90.2 % being fossil-based. In the same year, European plastic production amounted to 57.2 million tons (Plastics Europe, 2022). Accidental spillage during transport, handling, and manufacturing processes has significantly increased plastic pellet pollution in the world's oceans, including the Mediterranean Sea (GESAMP, 2015; Fendall and Sewell, 2009; Sharma et al., 2021). This pollution poses grave threats to marine ecosystems and organisms, causing physical damage, chemical contamination, and ingestion by marine species. Studies have demonstrated that plastic pellets can absorb and concentrate toxic substances, thereby acting as vectors for these contaminants when ingested by marine life (Teuten et al., 2009; Mato et al., 2001; Brennecke et al., 2016; Pelamatti et al., 2022). Ingestion can also lead to blockages in the digestive tract, potentially resulting in starvation and death for marine organisms (Wright et al., 2013; Carson, 2013; Izar et al., 2022a, 2022b; Okoye et al., 2022).

The presence of plastic pellet pollution has been documented in marine environments worldwide, even in remote areas such as the Arctic

(Bergmann et al., 2017; Bergmann et al., 2022) and Antarctica (Waller et al., 2017; Lozoya et al., 2022).

The Mediterranean Sea is particularly susceptible to plastic pollution, given its semi-enclosed nature, high coastal population density, and substantial maritime traffic (UNEP/MAP, 2015; Boucher and Bilard, 2020). Suaria et al. (2016) identified the Mediterranean as one of the global regions with the highest concentrations of microplastics, notably including plastic pellets. Marine currents and coastal geomorphology lead to the accumulation of these pellets in specific "hotspot" areas (Sanchez et al., 2014; Sharma et al., 2021; Dunphy, 2020). The detection of plastic pellets in the Mediterranean's deep-sea sediments (Alomar et al., 2016) and their ingestion by various marine organisms (Deudero and Alomar, 2015) underscore the extent and severity of this issue. Suaria et al. (2016) reported an average of 1.07 pellets per square meter of beach across all coastal areas in the Mediterranean Sea studied, highlighting the ubiquity of this pollution. Addressing plastic pellet pollution necessitates immediate action from both governments and industries, including enhanced waste management, promotion of sustainable consumption, and stricter regulations on plastic production and transport (Jambeck et al., 2015; Galloway, 2015). Moreover, is needed a properly regulation for preproduction pellets. The prices for virgin plastics have decreased reducing the use of recycled plastic, as the cost is currently not competitive (Syberg et al., 2021). The EU Marine Strategy Framework Directive (MSFD) specifically addresses the issue of plastic pellet pollution in coastal environments, underlining the need for EU member states to assess and manage the impact of these pollutants on marine ecosystems. Through the MSFD, the EU aims to ensure that measures are implemented to reduce the presence and impact of plastic pellets, thus preserving biodiversity and the health of marine habitats (EU Marine Strategy Framework Directive website).

While plastic pollution is a widely studied global phenomenon, this research provides new insights by conducting a robust chemical characterization and spatial analysis of plastic pellets across four beach sites in the understudied Ionian and Tyrrhenian coastal locations of Southern Italy: three Sicilian sites - Agnone Bagni (SR), Baia del Tono (Milazzo, ME), Paradiso (ME) and a Calabrian site near Pizzo Calabro (VV).

This research employs, for the first time, a portable spectrometer for a rapid identification of pre-production plastic pollutants in coastal settings. Raman spectroscopy, recognized as a state-of-art method for qualitative polymer characterization, plays a crucial role in both plastic manufacturing and environmental surveillance (Dąbrowska, 2021; Lv et al., 2020; Sagitova et al., 2016; Andreassen, 1999; Guo et al., 2019; Nava et al., 2021; Stephens et al., 2004). However, benchtop Raman instruments restrict analyses to the laboratory setting. The recent development of handheld Raman technology now enables in-situ polymer characterization. The handheld design provides a lightweight, portable tool that can be easily transported and utilized in the field for on-site plastic monitoring. This enables immediate, in-situ chemical characterization of plastic debris without requiring sample transportation or laboratory benchtop equipment. The ability to rapidly detect and identify polymer types across sampling sites makes handheld Raman spectroscopy an invaluable advance for on-site pollution surveillance, mitigation, and source tracking.

With its simplicity of operation, lack of sample preparation, and rapid non-destructive measurement, handheld Raman is ideal for field rapid identification of plastic pellet chemical composition. The technique's portability and accessibility allow expanded sampling and continuous monitoring of plastic pollution not previously feasible. This work demonstrates the power of handheld Raman as a breakthrough technology for real-time and in-situ analysis of plastic pollutants in challenging environments.

Employing a methodical approach that integrates Raman spectroscopy with spatial sampling and statistical analysis, this work sheds light on the microscale patterns and chemical properties of pellet pollution.

Therefore, the method employed in this study exhibits high reproducibility, ensuring reliable and consistent results across different environmental samples and conditions. This aspect underscores the technique's potential for widespread application in ongoing environmental monitoring efforts.

Looking ahead, the method holds promise for scaling from macroscopic to microscopic levels of plastic analysis, providing a comprehensive view of plastic pollution from larger debris to minute particulates.

As such, the findings contribute crucial new evidence elucidating pellet abundance, polymer variability, accumulation zones, and degradation profiles along the shorelines of Southern Italy. This sets a precedent for expanded sampling and advanced spectroscopic characterization that can be replicated, also using a portable instrumentation, across broader geographic areas.

2. Materials and methods

2.1. Distribution of sampled sites

Nurdles collections were conducted at four chosen sites: Paradiso (Messina), Baia del Tono (Milazzo), Agnone Bagni (Siracusa), and Pizzo Calabro (Vibo Valentia) (Fig. 1).

The sampling strategy provides a novel perspective by collecting pellets across multiple beach sites spanning two different coastal seas. The locations were selected to allow comparison between the Ionian and Tyrrhenian coastlines as well as characterization of beaches with distinct geomorphological features. The four sampling sites represent diverse hydrodynamic conditions and coastal dynamics, enabling analysis of spatial factors driving pellet distribution. This targeted multi-site approach generates new insights into differential patterns of pellet pollution, degradation, and accumulation across the understudied shorelines of Southern Italy.

The beach of Paradiso (38°13′16.84″N, 15°33′59.26″E) is placed in the central Mediterranean on the Ionian side of the Messina Strait, an extensional basin superimposed onto the Apennine Orogen during the Plio-Pleistocene (Ghisetti, 1984; Ghisetti, 1992). The Messina Strait is one of the most impressive tide-dominated depositional system, whose sedimentary dynamics is regulated by the tidal amplification. The tidal amplitude reached in this area is peculiar and completely different from the other sampling sites. Due to a physical narrowing, opposite tidal phase inversion between the Tyrrhenian Sea and Ionian Sea produces a phenomenon of tidal amplification of a huge volume of marine water. Cyclically every 5–6 h, a phase of high tide in the Tyrrhenian Sea corresponds with a phase of low tide in the Ionian Sea, generating ca. 17 cm/km of different elevation. This volume is calculated in ca. 750.000–1.000.000 m3/s of marine water to pass from north to south of the strait and vice versa. The water mass is accelerated when it transits through the narrowest strait zone resulting of NE- and SE-flowing strong tidal currents in the Messina Strait, which move with velocity picks up to 3 m s − 1.

The difficulty in identifying a framework that summarizes the sedimentary processes and associated depositional environments for these systems stems from the great number of variables (Longhitano, 2018). The nurdles are found during sea storms within the gravelly-sandy sediments characterizing Paradiso Beach.

Baia del Tono (38°14′42″ N, 15°14′27″ E) is located in the northeastern sector of Sicily dominated by the Peloritani Mts., a mountain chain formed by metamorphic rocks unconformably covered by sedimentary layers (Scicchitano et al., 2011). Baia del Tono, located along the western coast of Capo Milazzo peninsula, consists of predominantly gravelly-sand. Here the wind from North-West (Maestrale) predominates, and secondarily the winds from North and South-East (Tramontana and Scirocco respectively) (Note illustrative della Carta Geologica d'Italia, scala 1.50000).

Agnone Bagni (37°18′49″ N, 15°06′08″ E) is situated on the Ionian coast of Sicily in the southernmost coastal segment of the Catania Plain, bordered to the South by the Neogene Quaternary Hyblean volcanites.

(a)

(b)

(c)

(d)

(e)
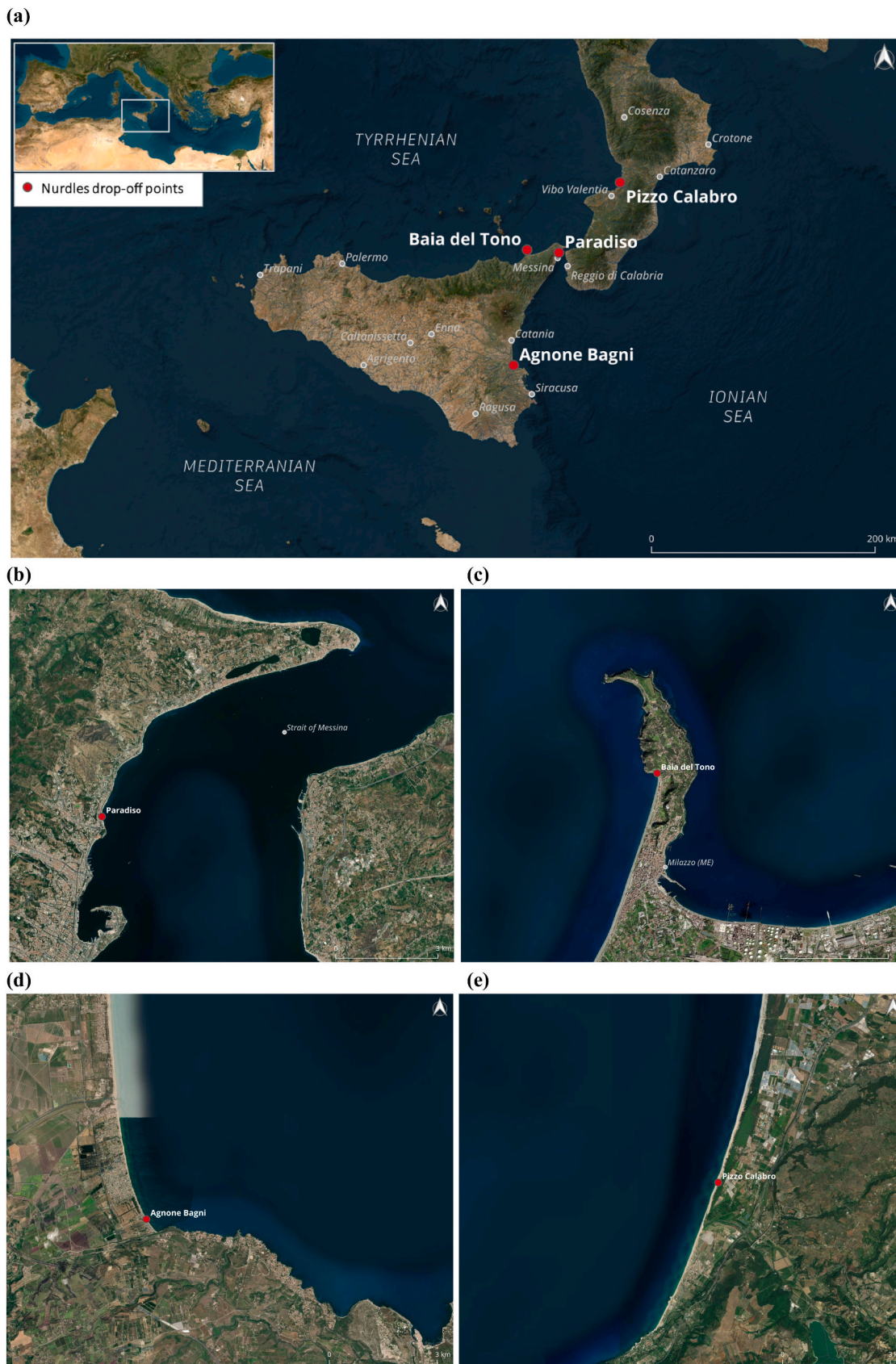
Fig. 1. – Geographic contextualization of the sampled beaches (red dots) in the Mediterranean area, across Ionian and Tyrrhenian Sea (a). Details of the coastline for each specific location: Paradiso (b), Baia del Tono (c), Agnone Bagni (d), Pizzo Calabro (e). Basemap is by Esri, World Imagery "Satellite". February 10, 2024 (Arcgis website). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The area is characterized by wide sandy beaches bordered inland by dune deposits. The sediments range from medium to fine sands (Note illustrative della Carta Geologica d'Italia, scala 1:50000, foglio 641). The area belongs to a complex hydrodynamic framework of the Ionian Sea and Messina Strait. Anti-clockwise gyres interact with the S-directed current from the Messina Strait, forming, nearshore, a quite complex circulation system (Longhitano and Colella, 2007).

Pizzo Calabro (38°47′08″ N, 16°12′11″ E) lies in the Tyrrhenian coast of central Calabria, in the portion named Santa Eufemia Littoral Province (Le Pera and Critelli, 1997). The study area develops on the edge of the Calabria-Peloritani Arc made of crystalline basement nappes partially affected by Alpine metamorphism (Ortolano et al., 2005; Pezzino et al., 2008). The beach is characterized by sand and cobbles. The study area is placed in a relatively high-energy coastal environment, where prevailing winds blow from North-West and from West-Southwest (Morrone and Ietto, 2021).

The granular analysis of samples from several beaches marks the first investigation of its scale in the region and provides a template for expanded microplastics sampling worldwide. By sampling pellets from beaches subject to varied currents, winds, and morphological factors, the findings elucidate pellet variability shaped by nearshore transport mechanisms and geomorphology.

2.2. Collection strategy

Plastic pellets were collected from the four selected beaches between January and August 2023 to analyze their spatial distribution and physic-chemical characteristics across the sites. The beach sediment was collected from the dry sand within a square area (1 m × 1 m and at 0–10 cm below the sediment surface) drawn above the high tide line after recording GPS coordinates. The sand collected was then filtered in the laboratory through a sediment sieve with a mesh size <5 m. The sieve removed sand, pebbles and any other waste that was smaller than our target (pre-production pellets). A total amount of 115 plastic pellets were collected and analyzed from the four beach sites. Specifically, 30 pellets were sampled from Baia del Tono, where a surface density of 160 pellets per m^2 was observed; 25 pellets were collected from Agnone Bagni, which had a lower surface density of 10 pellets per m^2. The highest density was found at Pizzo Calabro, where 30 pellets were gathered, corresponding to 1500 pellets per m^2 across the beach surface. Finally, 30 pellets were obtained from Paradiso beach, with a surface density of 100 pellets per m^2.

2.3. Analyzing pellets from visible to microscopy

In the laboratory, the pellets collected from each beach underwent characterization based on various attributes such as color, shape, size, surface texture, fouling, and any evident surface spots or signs of degradation. Raman spectroscopy was employed for chemical analysis to identify the type of polymer present and to conduct principal component analyses. This systematic categorization aimed to elucidate the differences and similarities in pellet properties and abundances between the four coastal environments in the Ionian and Tyrrhenian Seas.

2.3.1. Visual and physical examination

The pellets from each beach were visually inspected for macroscopic characterization and quantification of key properties. Pellets were categorized by color, with the majority being white/translucent, followed by yellow/brown. All pellets exhibited consistent discoidal or cylindrical shapes with diameters <5 mm. However, significant variation was observed in pellet surface texture and appearance. The majority displayed porous, eroded surfaces as opposed to smooth polymer surfaces. These roughened textures likely resulted from prolonged environmental weathering processes during transport in seawater and tumbling in beach sand. Many pellets had clear signs of oxidative degradation, with pitting, cracks, and substantial surface irregularities

that indicated permanence in the dynamic coastal zone over extended time periods.

A subset of pellets also exhibited visible fouling in the form of organic substance accumulation on the plastic surface. Fouling materials detected included algae, sediment, shells, and uncharacterized biological deposits that had adhered to the pellet during residence in the marine environment. Furthermore, several pellets featured unique superficial spots and stains on their surfaces. These markings may have arisen from mineral deposits or microbial biofilms forming during the biodeterioration of the plastic in seawater.

While general shape typology remained consistent, the proportion of degraded, weathered pellets with porous, fouled surfaces provided insight into the variable environmental lifetimes and exposure histories across the different beach environments. The systematic categorization and quantification of key pellet characteristics enabled informative comparisons regarding relative abundance, weathering extent, fouling, and inferred degradation patterns between the Ionian and Tyrrhenian coastal areas. This spatial analysis illuminated differential rates of pellet accumulation and weathering linked to transport mechanisms, hydrodynamics, and morphological factors unique to each beach site.

2.3.2. Macro-photography documentation

After initial classification of pellets by visual inspection and macroscopic examination, representative samples were carefully selected for detailed high-resolution photographic documentation. Samples were chosen to showcase the diversity of shapes, colors, surface features, and distinguishing characteristics observed across locations. A Canon EOS M3 digital camera equipped with a Sigma 70 mm f/2.8 macro lens was utilized to capture the pellet images. This allowed 1:1 life-size reproduction for discerning fine details. The camera was mounted on a sturdy tripod and paired with a non-reflective black matte background to minimize glare. Directional lighting from one or more angles was used to illuminate the pellets and highlight subtle surface morphology, textures, inclusions, and spots.

Sufficient exposure time settings were configured to visualize nuanced features within the plastic matrix. Multiple images per pellet were taken, adjusting the lighting angles and focus zones to fully composite all details from the exterior into the interior. The comprehensive photographic documentation enabled archiving of visual data on the macroscale properties of typical pellet specimens from each beach. The images (Fig. 2) elucidate variations in dimensions, shapes, surface degradation patterns, extent of fouling, embedded inclusions, and other discernible characteristics.

The assembled pellet image library supplements the quantitative visual examinations, creating a permanent record for in-depth comparative analysis. The photographs capture the diversity of pellet types, surface weathering, accumulation of inorganic/organic matter, and signs of prolonged coastal environmental weathering. The imaging approach provides invaluable visual evidence elucidating the pathways and lifetimes of plastic pellet pollution across Southern Italy's shorelines.

2.3.3. Raman spectroscopy

Raman spectroscopy was used to collect detailed spectra for each plastic pellet sample. The Raman spectra contain information about the intensity of scattered light at different wavelength bands, which can be used to identify the chemical composition. The measurements on the samples were carried out using the handheld Raman spectrometer BRAVO-Bruker, manufactured by DuoLaser™ (Brucker Website). The instrument is equipped with a patented SSE™ (Sequentially Shifted Excitation), an exclusive technique capable of mitigating fluorescence effects. It employs, for excitation, two different lasers centered at 785 nm and 852 nm in wavelengths, which work in tandem to minimize fluorescence effects. These lasers offer exceptional sensitivity over a wide spectral range, but it was not feasible to separate their contributions using the experimental setup. These features involve carefully

Fig. 2. Variety of plastic pellet samples collected from beaches in Southern Italy: Pizzo Calabro, Baia del Tono, Agnone Bagni, and Paradiso. The pellets display differences in shape, size, color and surface textures indicative of varying polymer types, weathering, degradation, and accumulation across the four coastal sites.

controlling temperature variations of the laser, allowing for the measurement of an extensive range of materials using handheld Raman systems, surpassing previous capabilities. In addition, the system ensures the utmost reliability in verifying materials without any ambiguity. The instrument was connected to a dedicated PC where, using Bruker OPUS software, it was possible to set the acquisition parameters (this option is not possible from the handheld) and visualize the result of each measurement. The program also allows for automatic comparison of the compounds with an internal library. The acquisition of plastic pellets' spectra was carried out using a range of 400 to 3000 cm^{-1}, and each measurement's integration time was limited to 120 s or less. The compound assignments were obtained by comparing the measured spectra with specific open databases as IRUG and Public Spectra.

3. Results

Analysis of visual characteristics revealed consistent patterns across the 115 collected pellet samples. The vast majority (82 %) were white/translucent in color, with the remaining 18 % exhibiting yellowish or brownish hues. All pellets displayed the expected discoidal or cylindrical shapes under 5 mm diameter. Surface texture proved more variable, with 73 % appearing rough and porous versus 27 % with smooth polymer surfaces. Fouling from adhered organic and inorganic matter was visible on 41 % of the pellets. Signs of degradation including cracks, pits, and substantial surface irregularities were noted on many specimens. This quantitative breakdown by visual attributes provides key baseline data on the physical properties and diversity of plastic pellet pollution across these Mediterranean coastal environments. The categorization elucidates differential weathering extents and accumulation patterns between locations.

The Raman spectroscopy analysis has yielded a comprehensive understanding of the polymer composition prevalent in the sampled coastal environments. This analysis involved baseline correction and averaging of spectra to ensure the precision and reliability of the results. Table 1 shows a summary of characteristic Raman peaks for Polyethylene (PE) and Polypropylene (PP) corresponding to different types of molecular vibrations. Fig. 3 presents a comparison of the Raman spectra acquired from pellet samples collected at the sampling sites versus commercial polymer standards. The spectra of polyethylene (PE) and polypropylene (PP) standards are displayed alongside representative spectra obtained from pellets, showing the characteristic peaks used to identify these polymers as the predominant types. Nylon 6, which was detected in a very small number of pellet samples, is not included in this figure. The strong concurrence between sample and standard peak positions enabled unambiguous assignment of polymer composition. This demonstrates the utility of Raman spectroscopy for distinguishing the main pellet polymer types despite weathering and degradation.

Table 2 represents different clusters of sites, each with specific ID numbers, identification types, and associated site names.

The samples from Pizzo Calabro were categorized into two primary groups, A and B, each exhibiting distinct Raman spectral characteristics. Group A is characterized by a series of peaks at specific wavenumbers (1063, 1130, 1169, 1295, 1366, 1415, 1441, 1458, 2723, 2852, and

Table 1

Characteristic Raman Spectral Peaks for Polyethylene (PE) and Polypropylene (PP) with Associated Vibrational Modes and Symmetry Types. The table includes the wave numbers of the peaks in cm^{-1} and classifies the type of molecular vibration and whether it is symmetric or asymmetric.

Polymer	Raman Shift (cm^{-1})	Vibration Type
PE	1060	C-C stretching
PE	1128	C-C stretching
PE	1293	CH_2 wagging
PE	1442	CH_2 bending (scissoring)
PE	2722	CH_2 stretching
PE	2848	CH_2 stretching
PE	2890	CH stretching
PP	399	CH_3 rocking
PP	811	CH_2 rocking/C—C and C–CH_3 stretching
PP	840	C-C stretching/CH_3 rocking
PP	970	C-C stretching/CH_3 rocking
PP	1150	C-C stretching/CH bending
PP	1330	CH stretching/CH_2 wagging/CH_3 bending
PP	1460	CH_2 bending/CH_3 asym. bending
PP	2722	CH_2 stretching
PP	2840	CH_2 stretching
PP	2884	CH stretching
PP	2958	CH_3 asymmetric stretching
NY6	1081	C–C stretching
NY6	1443	CNH bending
NY6	1637	C=O stretching
NY6	2904	CH_2 stretching

$2885\ cm^{-1}$), closely matching the standard Raman spectra for high-density polyethylene (PE), with a match percentage exceeding 90 %. The vibrational modes associated with these peaks provide a deep insight into the molecular structure of the polymer. The peaks at 1063 and $1130\ cm^{-1}$ are attributed to the anti-*symmetric* (*asym*(C—C)) and symmetric (sym(C—C)) stretching vibrations of C—C bonds, respectively. Furthermore, the peaks at 2852 and 2885 cm-1 correspond to the symmetric (sym(CH_2)) and anti-symmetric (*asym*(CH_2)) stretching vibrations of CH_2 groups, a characteristic moiety in PE (Sagitova et al., 2016).

Group B samples from Pizzo Calabro displayed a different set of peaks at various wavenumbers (320, 399, 453, 527, 811, 840, 901, 944, 975, 1001, 1037, 1154, 1158, 1220, 1332, 1361, 1437, 1463, 2723, 2841, 2886, 2902, and $2953\ cm^{-1}$), aligning well with polypropylene (PP) with a matching percentage above 85 % in spectral libraries. The vibrational assignments for these peaks were meticulously detailed, enhancing the understanding of the polymer's molecular structure. For instance, the peak at $399\ cm^{-1}$ is ascribed to CH_2 vibration and CH bending as plotted in Fig. 3. The peak at 811 cm-1 is linked to CH_2 rocking and stretching of C—C and C–CH_3, while the peak at $975\ cm^{-1}$ is associated with CH_3 rocking and C—C stretching. Other notable peaks at 1332 and 1463 cm-1 are attributed to CH bending and CH_2 twisting, and anti-symmetric bending of CH_3 and CH_2 bending, respectively. The peak at 2886 cm-1 is indicative of symmetric stretching of the CH_3 group in the PP polymer (Andreassen, 1999, Guo et al., 2019).

The samples from Baia del Tono were also sorted into PE (Group C) and PP (Group D), showcasing the diversity in polymer composition across different beach locations. Agnone Bagni predominantly exhibited PE spectra (Group E), with an exception in sample SR 23, which matched with PP (Group F). This variation underscores the heterogeneity of plastic pollution, even within a localized region.

Additionally, SR7 and SR8 (Group G) exhibited peaks at 933, 1059, 1081, 1130, 1172, 1200, 1279, 1308, 1383, 1443, 1470, 1637, 2723, 2880, 2904, 2929 cm-1, matching nylon 6 (Nava et al., 2021, Stephens 2004). The peak at 1637 cm-1 was related to C=O stretching. The peak at 1081 cm-1 was related to C—C stretching (Stephens et al., 2004), and the peak at $1443\ cm^{-1}$ was assigned to CNH bending. The peak at 2904 cm^{-1} was assigned to the stretching of the CH_2 group. Nylon is polymer less frequently reported in marine environments. This discovery broadens the spectrum of known pollutants and emphasizes the need for a more extensive analysis to uncover the full range of polymers present in marine debris.

The Raman analysis of plastic pellets from Paradiso (Groups H and I) further corroborates the prevalence of PE and PP in the marine environment. Most of the samples (83 %) displayed characteristic PE peaks, while a smaller fraction (13 %) aligned with PP. An additional 3 % of the samples from Paradiso presented unique spectral features, suggesting the presence of an alternate polymer, and necessitating further investigation.

Twenty-five samples exhibited Raman peaks characteristic of PE, including signals at 1063 and 1130 cm-1 corresponding to C—C stretching, and peaks at 2852 and 2885 cm-1 representing CH_2 stretching vibrations. These PE-matched samples make up 83 % of the Paradiso pellets. Four samples, accounting for 13 % of the Paradiso data, displayed Raman spectra consistent with polypropylene (PP) (Group I). Notable peaks at 811, 975, 1332, and 1463 cm-1 signify the presence of PP in these samples based on the chemical shifts of CH2, CH3, and C—C moieties.

Only one Paradiso sample (3 % of dataset) did not match either PE or PP spectral profiles. This sample showed unique peaks that may suggest an alternate polymer identity, though further analysis is required for confirmation.

In summary, the Raman spectroscopic analysis of plastic pellets from the four sampling sites show that polyethylene (PE) is the predominant polymer type, accounting for 82 % of all analyzed samples (Fig. 4). Polypropylene (PP) was the second most common polymer, representing
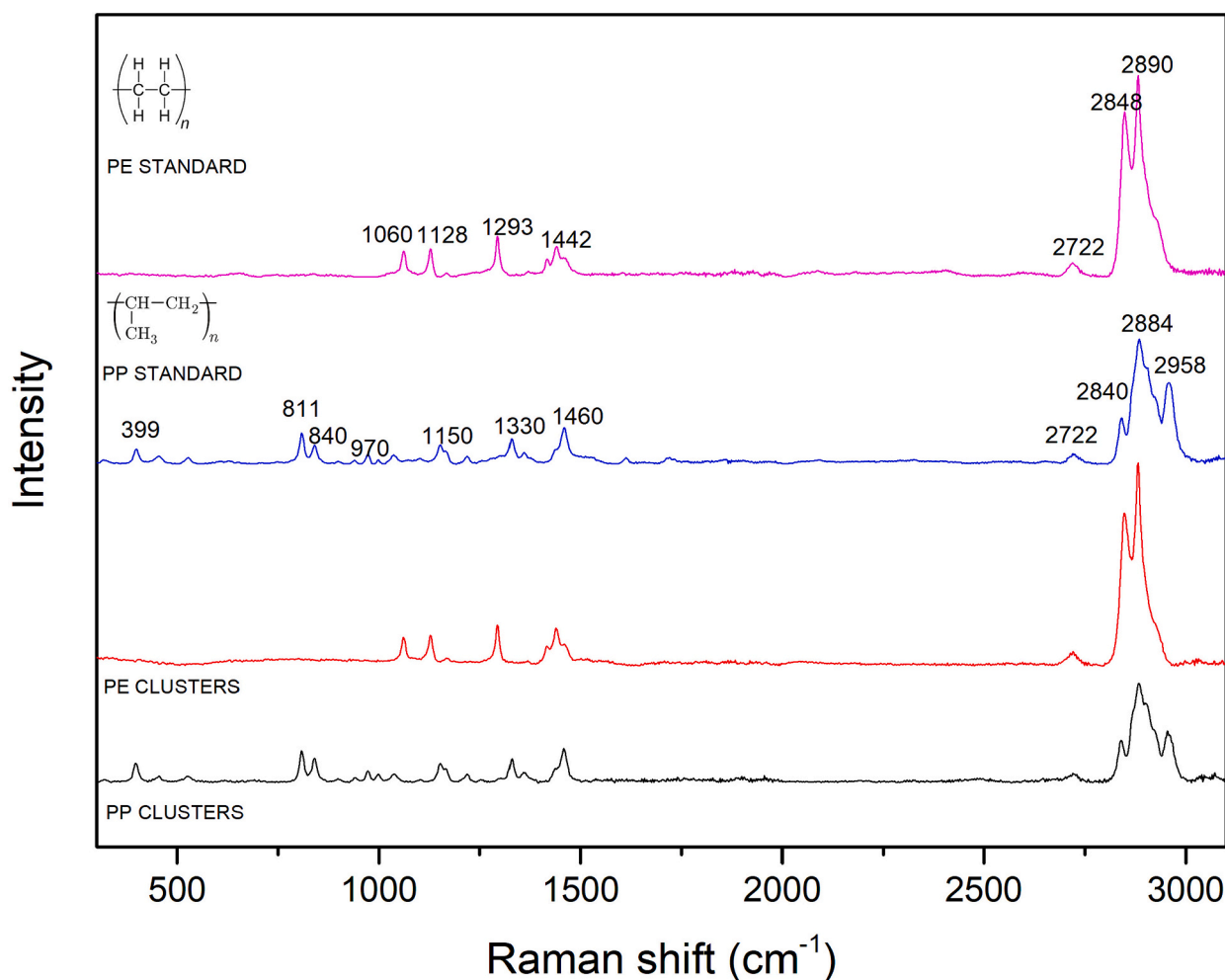
Fig. 3. Comparison of standard Raman spectra for PE (magenta) and PP (blue) materials with representative spectra of PE (red) and PP (black) clusters. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
Overview of material clusters, each with specific ID numbers, identification types, and associated site names.

Cluster	Site id	Polymer	Provenance
Group A	PIZZO_1–12,14-17,19-22,24-25,28–30	PE	
Group B	PIZZO_ 13,23,26,27	PP	Pizzo Calabro
Group C	TONO_ 1–2,5-6,8-11,13,16-22,24–26, 28,30	PE	
			Baia del Tono
Group D	TONO_ 3, 4, 12, 14, 15, 23, 27,29	PP	
Group E	SR_ 1–6, 9–22, 24–25	PE	
Group F	SR_ 23	PP	Agnone Bagni
Group G	SR_ 7–8	NY6	
Group H	PARAD_1–12, 14–21, 23–25, 27, 28, 30	PE	Paradiso
Group I	PARAD_13, 22, 26, 29	PP	

15 % of the total sample set. The remaining 3 % of samples did not match PE or PP spectral profiles, suggesting the presence of less common polymer types.

The predominance of PE at all sampling locations indicates that it is the main polymeric component of plastic materials. These pellets are increasingly used in the production of packaging, bottles, and other single-use plastic items that are widely utilized in our daily lives. Their accidental release into the environment during production and transportation phases has led to a significant increase in these "raw materials" on our coasts and those around the world.

The consistent presence of PP at lower levels is also unsurprising since PP usage is secondary to PE for many consumer applications.

The prevalence of polypropylene and polyethylene is probably related to the density of these two polymers that allows it to float on the sea' surface and then settle on the beaches during storm surges (Giugliano et al., 2022). Nurdles mix with sediments, becoming trapped, to be remobilized during subsequent tides and extreme weather events months to years after the spill. Their distribution is strongly influenced by tides, waves, and wind.

The minor fraction of other polymers highlights the utility of Raman spectroscopy to identify fewer common plastics that comprise beached marine debris. Further analysis is warranted to clarify the full range of polymers contributing to pellet pollution in these waters. Overall, this preliminary dataset provides insight into the major plastic types dispersed across southern Italian coastal environments.

4. Discussion

4.1. Statistical multivariate analysis

The interpretation and classification of such number of spectra is difficult and time consuming, but the use of statistical multivariate methods can significantly improve the process. Multivariate analysis techniques use statistical methods to analyze complex datasets with many variables. These methods allow large, multi-dimensional data to be reduced to key patterns and trends, facilitating analysis and interpretation. Specifically, we used principal component analysis (PCA) and
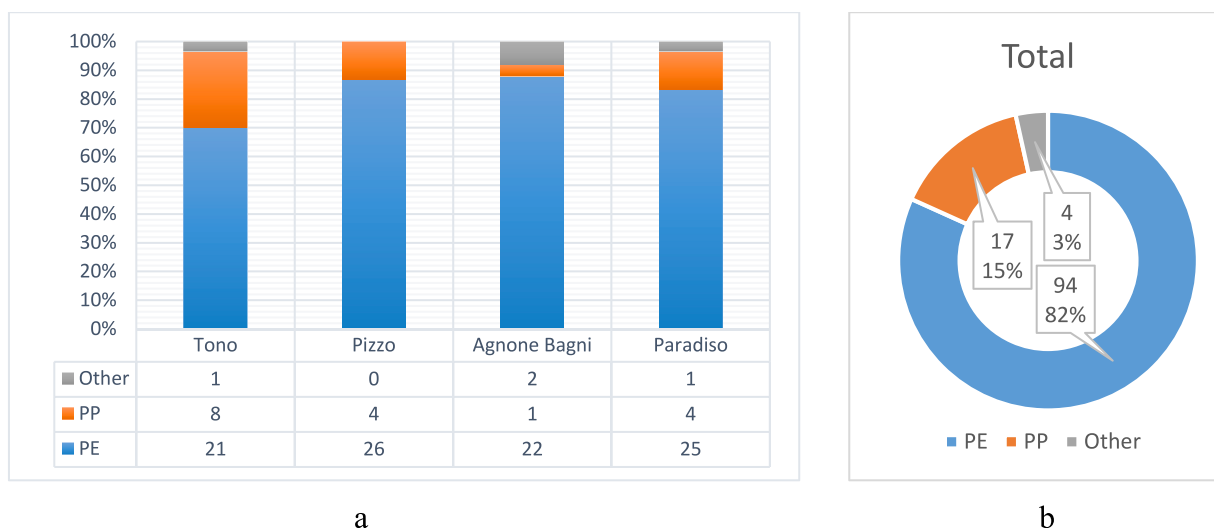
a

b

Fig. 4. Graph a: quantification of the different plastic types (PE, PP, other) across four locations: Tono, Pizzo, Agnone, and Paradiso (a). Graph b: Number of samples collected, by type of polymer: PE (94 units), PP (17 units) and others (4 units). Total number of samples collected: 115 units.

hierarchical cluster analysis (HCA).

Multivariate methods, in fact, are able to outline subtle differences among data and help to identify and to group data without the loss of important information (Armetta et al., 2023; Armetta et al., 2021; Renda et al., 2019). In this case PCA algorithm was applied to catalog spectra of the samples coming from the different sampling sites.

The results of the PCA analysis are the transformation of the original dataset, characterized by a very large number of variables, into a new one with a lower dimensionality and described in terms of principal components (PC), still retaining most of the original data information as possible. Thus, the procedure starts with the computation of a new set of variables, the principal components, after a series of successive axis rotations. To this purpose, spectra are loaded into rows of a matrix, that represents the whole dataset, each spectrum, i.e. each row, is a data point of the dataset, the variables are represented by the Raman intensities recorded at the given wavenumbers, identified by the columns. The input of the PCA analysis, in this case is then 1451×112 matrix *M*. The principal components are determined in such a way that PC1 is the one with the largest possible variance to the original data. All the other PCs are determined following the same criteria as PC1. After the procedure most of the variance is usually contained in the first few principal components, the *loadings* associated to the PCs represents the Raman intensities variations while *scores* are coefficients that consider the magnitude of the spectral changes. Thereafter, the HCA algorithm groups elements of the data set following a similarity criterium. Each spectrum can be considered as a vector, whose components are the measured intensities in the sampled spectral region. Given a number of clusters *k* HCA algorithm performs classification of the spectra in different clusters. Spectral information can be obtained by a comparison of the average spectrum of each cluster to the loadings of the most significant PCs. In Fig. 5 is reported the PC1-PC2 plot and the clustering results of the PCA analysis.

The number of *k* of the cluster is a priori unknown so the HCA procedure was performed changing the clusters number and the most reliable results was obtained with three clusters. The first two PCs account for most of the variance (87 %), the contribution of remaining PCs is very low being probably related to the experimental noise. One can observe in Fig. 5 that clusters #1 and #3 exhibit a partial overlap, encompassing spectra with scores aligned along a clearly defined direction within the PC1-PC2 plane. This suggests that for these elements, the variance is contingent upon a singular set of variables and that spectra assigned to the cluster are only slightly different. This hypothesis is validated when examining the mean spectra derived from averaging
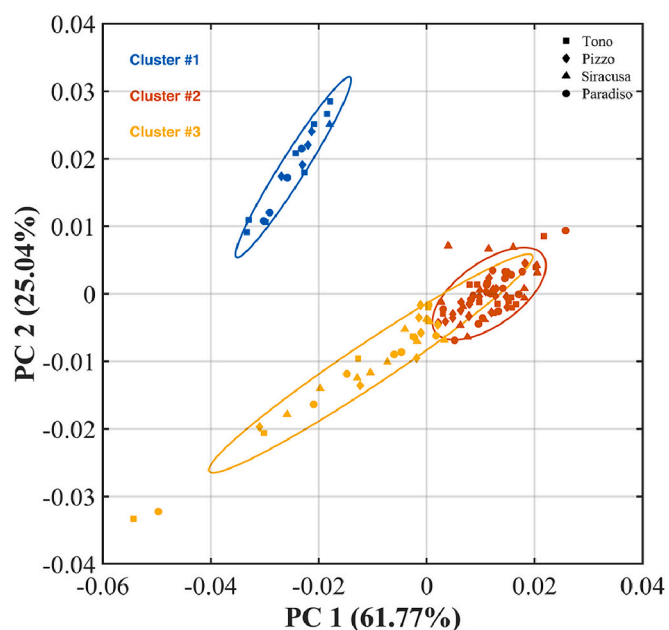

Fig. 5. PCA: PC-PC2 plot and HCA clustering results of the PCA analysis of the Raman dataset. The first two PCs accounts for about 87 % of the total variance. Markers refer to the sample collection site.

the spectra within each cluster and shown in Fig. 6, that shows the two spectral regions where Raman features occur. As it can be seen mean spectra from cluster #2 and #3 are very similar and spectra belonging to these clusters can be identified as polyethylene, with the characteristic Raman peaks due to the C—C and C—H$_2$ vibrations in the 1000–1600 cm^{-1} region. Cluster #1 encompasses spectra belonging to polypropylene as indicated by the presence of several bands to different vibrational mode of C-H$_x$ groups in the region 800–1200 cm^{-1}, and the characteristic shape of the C-H$_x$ stretching mode in the 2800–3000 cm^{-1}.

4.2. Further consideration about plastic pellet features

The visual examination of pellet characteristics including color, shape, texture and fouling revealed both commonalities and differences
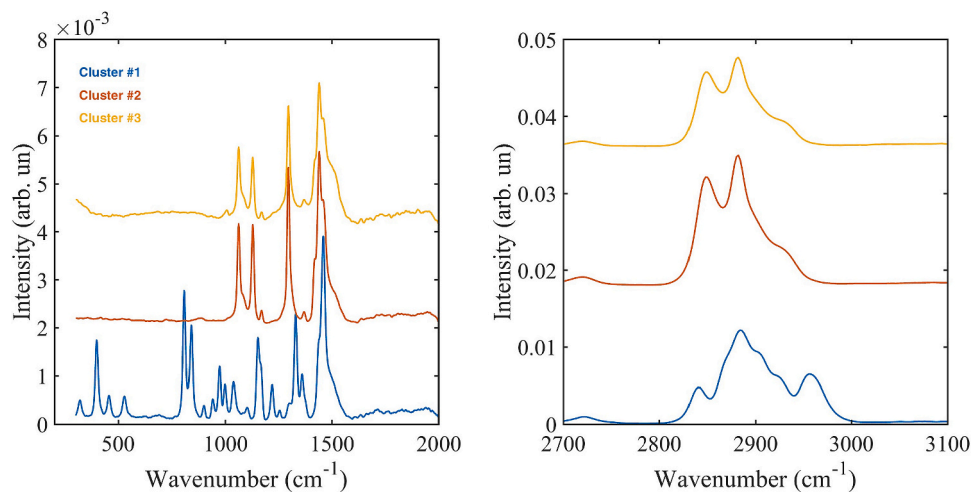
Fig. 6. Clusters averaged spectra. As discussed in the text Cluster #2 and #3 show very little differences so that their elements can be classified as similar.

across the four beach sites surveyed along the Ionian and Tyrrhenian coasts of southern Italy. The predominance of discoidal and cylindrical forms composed primarily of polyethylene (PE) and polypropylene (PP) matches widespread industrial pellet types used as pre-production plastic feedstocks. This points to the likely origin of these pellets from manufacturing processes and supply chains.

However, variations in the extent of surface degradation and fouling implied diverging residence times for the pellets in the marine environment (Abaroa-Pérez et al., 2022). As noted by Wang and Castelao (2016), rougher, fouled pellet surfaces correlate to longer durations of coastal and oceanic weathering processes. The coastal sites with higher proportions of smooth, unworn pellets likely indicate more recent inputs, whereas more degraded pellets suggest prolonged exposure and accumulation over years. Tracking surface condition provides clues to pellet age and transport history.

Unlike roughness and fouling, the inherent color of plastic pellets does not directly indicate environmental weathering or result from degradation reactions. As discussed by Giugliano et al. (2022), the distinct colors exhibited by plastic pellets are intentionally imparted during manufacturing through addition of pigments and dyes. Pellet color selection correlates to the final commercial plastic product and applications (Izar et al., 2022a, 2022b). For instance, white or translucent pellets are common for packaging materials, while black pellets would point to uses in electronic device casings. Thus, the range of pellet colors observed across the sampled beaches more likely reflects the diversity of source applications and industry usages, rather than alterations occurring during marine weathering. Tracking pellet color frequencies across locations and over time can help elucidate the contributions from different plastic industry sources and product categories.

However, as extensively documented in the literature, the coloration of plastic pellets can depend also on the absorption of organic contaminants due to environmental weathering (Teuten et al., 2009; Takada and Yamashita, 2016; Karlsson et al., 2021; Abaroa-Pérez et al., 2022; Endo et al., 2005). The aforementioned studies have demonstrated that yellowish and brown pellets tend to sorb higher levels of contaminants such as PAHs, DDT, and PCBs compared to white and translucent pellets. This suggests that darker pellet hues could indicate longer residence times in the marine environment, resulting in increased contaminant absorption. Although the wide range of pellet colors observed in this study could be derived from potential contamination, the variability of polymers limits general correlations between color and weathering. Therefore, a detailed analysis of contaminants is necessary to directly quantify the relationships between pellet color, weathering duration, and pollution levels.

The Raman spectroscopic analysis enabled detailed chemical characterization of the polymer composition of hundreds of individual pellets across the four coastal sites. The technique definitively demonstrated polyethylene (PE) and polypropylene (PP) collectively accounted for over 95 % of pellets at all locations, though in varying proportions. This aligns with multiple studies by Karapanagioti et al. (2011), Compa et al. (2019) and others showing the overwhelming prevalence of PE and PP pellets comprising plastic pollution in marine ecosystems globally. The dominance of these two polymers, which readily float due to their low density, is unsurprising given their widespread usage in ubiquitous single-use plastics and packaging. Higher density polymers tend to sink more readily when entering the oceans, reducing their abundance in surface trawls and coastal surveys.

While the overall polymer types were consistent across sites, the notably higher contribution of PP vs. PE pellets at the Agnone Bagni site merits further study to determine possible localized sources or inputs. The few samples matching nylon at this location also suggest additional area sources or accumulation factors differing from the other three beaches. As reported by Longhitano (2018), a key role in the Agnone Bagni area is certainly played by the oceanographic setting of the Simeto delta within the complex hydrodynamic framework of the Ionian Sea and Messina Strait. The complex circulation system is influenced by both the western Ionian Sea circulation and the S-directed current deriving from the Messina Strait.

As Compa et al. (2019) discussed, combining chemical characterization data with spatial distribution analysis and hydrodynamic modeling can help to identify specific pellet sources and pathways that may explain site variabilities.

4.3. Consideration about spatial distribution and accumulation patterns

The geomorphology and coastal dynamics of the sampling sites is supposed to play a key role in pellet distribution and retention patterns.

Many studies (Portman and Brennan, 2017, Prevenios et al., 2018, Merlino et al., 2020 and reference in) highlight the presence of marine litter in the Mediterranean Sea basin, probably due to its semi-closed morphology. The model transport and distribution of floating debris in the ocean of Lebreton et al., 2012 identified the Mediterranean Sea as a potentially important accumulation zone at the global scale.

Cózar et al. (2015) estimated the concentration of marine litter in the Mediterranean Sea basin, in a large range of values from 22 to 1934 g/km2, with a greatest abundance in northwestern Tyrrhenian Sea. Strong currents and winds may transport litter, especially the more buoyant, at considerable distance from the point of spillage (Oliveira et al., 2015). The combination of factors as the proximity to sources of input, the direction of the prevailing winds, the major coastal currents and the

geomorphological characteristics of the coast may contribute to different patterns of distribution and accumulation of pellets on the beaches (Moreira et al., 2016). By reviewing the beaches sampled below, we understand the geomorphologic features and hydrodynamic conditions that favor the occurrence of plastic pellets in beaches.

The Tono beach has a cross-shore width of approximately 25 m and a slope of about 20°. The sediment is composed of gravel ranging from coarse to fine. At the time of sampling, the beach exhibited two sets of storm berms, located respectively 4 and 10 m from the shoreline. Longitudinally, to the north, it is bounded by the high and rocky coast of Capo Milazzo, while to the south, it extends uninterrupted for several kilometers. Capo Milazzo promontory is a natural delimitation that, together with the currents parallel to the coastline, probably favor the distribution and accumulation of the pellets on the shoreline.

The Pizzo beach has a cross-shore width of about 60 m and a slope of approximately 15°. Here the sediment is composed of very fine gravel to very coarse sand. At the time of sampling, the beach presented one berm about 10 m from the shoreline. Longitudinally, towards the southwest, it is closed after several kilometers by a high and rocky cliff coast, while to the northwest, its extension continues uninterrupted throughout the Gulf of Lamezia Terme. This geomorphological setting of the coastal area of Pizzo could act as a pellet trap and the evolution of coastline appears to be controlled by converging currents (Morrone and Ietto, 2021).

In the Ionian sector, at the southernmost portion of the Gulf of Catania, there is the Agnone Bagni beach, that is about 40 m wide and has a slope of approximately 10°. The sediment consists of sand ranging from very coarse to medium. At the time of sampling, the beach did not exhibit any berms. Longitudinally, towards the southeast, it is enclosed by a high and rocky cliff coast, while to the northwest, it continues its extension uninterrupted for about 20 km.

As previously reported, in Agnone Bagni area, anti-clockwise gyres interact with the S-directed current from the Messina Strait, forming, nearshore, a quite complex circulation system. This littoral water movement causes the longshore drift of sand, (in part carried by Simeto river) southwards, (Longhitano and Colella, 2007). The sediments remain trapped, with floating pellets on the water, in Agnone Bagni bay, due to its peculiar shape.

The Paradiso beach has a cross-shore width of about 15 m and a slope of approximately 5–10°. The sediment consists of very coarse sand. Compared to the other beaches, it is about 500 m long, and has no natural boundaries. To reduce the longshore drift and to trap sediments, shore protection structure built perpendicular to the shoreline. In this area the plastic pellets were found only after a storm, whereas in the previous weeks no accumulation was recognized.

Characterizing accumulation zones assists monitoring and mitigation efforts (Moreira et al., 2016).

Overall, this preliminary study has chemically characterized plastic pellet pollution across distinctive coastal environments in southern Italy. Further work should expand the spatial scale of sampling to identify regional pollution hotspots and incorporate multivariate analysis and polymer degradation methods to clarify pellet sources, transport pathways, and environmental residence times.

4.4. Broader impacts and future research directions

This preliminary study has conducted an initial survey and chemical characterization of plastic pellet pollution across four distinct coastal environments in southern Italy. Further work should expand the spatial scale and scope of beach sampling to construct a more comprehensive understanding of regional pollution distribution, transport pathways, accumulation zones, and contamination gradients between the Ionian and Tyrrhenian coastal compartments.

Incorporating advanced multivariate analysis approaches and polymer degradation methods could help untangle pellet sources, environmental residence times, and coastal versus oceanic transport contributions. For instance, polymer oxidation and photodegradation profiles could reveal the timescales over which sample pellets have persisted in the marine environment.

Analyzing organic pollutants like pesticides, pharmaceuticals, and heavy metals adsorbed onto pellet surfaces could provide further insights into their influential role as contaminant transport vectors in the Mediterranean (Karlsson et al., 2021). Studies by Rios et al. (2007) and Rochman et al. (2013) demonstrated accumulation of persistent organic pollutants on plastic residues in the ocean. Additionally, Holmes et al. (2012) showed sorption of trace metals like cadmium and lead to plastic pellets immersed in seawater. As noted by Giugliano et al. (2022), the weathered, pitted texture of marine plastic debris facilitates sorption of hazardous contaminants from seawater. Characterizing the contaminant load would further elucidate potential ecological impacts of pellet pollution.

Future research should also aim to better delineate the diverse degradation processes and rates occurring across marine environments through simulated weathering experiments. Understanding pellet fragmentation and material disintegration mechanisms will inform ecosystem circulation timescales.

Additional work should examine ecological implications of pellet abundance on nesting beaches, interactions with biota, food web transfers, and toxicological effects.

Finally, a critical unanswered question remains determining the predominant origins of pellets deposited on Mediterranean coastlines. Sewwandi et al. (2023) highlighted diverse potential pathways from accidental release across all stages of plastics manufacturing and supply chains including at the production site, transportation, storage or spillages due to maritime accidents. Identifying the predominant sources of pellets will direct upstream solutions.

Overall, this first broad regional analysis has established an important framework for expanded research elucidating the nuanced issue of plastic pellet pollution facing Southern Mediterranean coastal zones. The multi-faceted methodology combining spatial surveys, chemical characterization, geomorphological context, and spectroscopic analysis provides invaluable baseline data while revealing key knowledge gaps, generating new insights into pellet abundance, composition, accumulation patterns, and degradation profiles.

This work motivates the need for continued monitoring and polymer analysis to inform mitigation strategies tailored to each unique coastal setting.

5. Conclusions

The Mediterranean Sea, particularly the southern coast of Italy, has been a focal point for understanding the distribution and chemical characteristics of marine plastic pellets. In this context, we analyzed the distribution and composition of plastic pellet pollution across four beach sites along the Ionian and Tyrrhenian coastlines. The following key conclusions can be drawn:

- Ubiquitous pellet pollution in the region and dominance of polyolefins like PE and PP (95 %), matching global trends in global marine plastic pollution.
- Variations in the extent of pellet degradation and fouling between sites implied differences in residence times and environmental exposure during transport.
- This preliminary dataset provides baseline data on the composition, relative abundance, and properties of plastic pellet pollution across these coastal environments.
- The results demonstrated how portable Raman spectrometer can be successfully used to monitor plastic pollution in situ on coastal environments.

The results highlight the need for ongoing monitoring and mitigation to address this environmental issue facing the Mediterranean region.

Further research could be conducted by expanding the number of sampled beaches across a wider area. Increased spatial sampling would provide more data to identify pellet accumulation zones shaped by coastal geomorphology and nearshore currents.

Moreover, the portability and non-destructive capabilities of handheld Raman spectroscopy demonstrate its potential for broadening plastic pollution analysis from in-field macrodebris characterization to rapid detection of microplastics. This could enable comprehensive screening across a wide size range of plastic particles, from visible pellets to microscopic fragments, providing a more complete understanding of the diverse plastic waste accumulating in marine ecosystems.

CRediT authorship contribution statement

Assunta Donato: Writing – review & editing, Writing – original draft, Validation, Resources, Investigation, Formal analysis, Data curation, Conceptualization. **Donatella Spadaro:** Writing – review & editing, Writing – original draft, Validation, Resources, Investigation, Formal analysis, Data curation, Conceptualization. **Dario Giuffrida:** Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Investigation, Formal analysis, Data curation, Conceptualization. **Giuseppe Sabatino:** Writing – review & editing, Writing – original draft, Resources, Investigation, Conceptualization. **Marcella Di Bella:** Writing – review & editing, Writing – original draft, Resources, Investigation, Conceptualization. **Sebastiano Trusso:** Writing – review & editing, Supervision, Investigation, Formal analysis, Data curation. **Rosina Celeste Ponterio:** Writing – review & editing, Supervision, Investigation, Formal analysis, Data curation.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at https://doi.org/10.1016/j.marpolbul.2024.116312. These data include the Google map of the most important areas described in this article.

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Further reading

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