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Microplastic Pollution in the Coast of Tarragona, Spain: A Western Mediterranean Study

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Abstract The Institut Rambla Prim, in collaboration with the Institut-Escola del Treball of Barcelona, conducted a study on marine microplastics present in the seawater along the coast of Tarragona (Balearic Sea, Western Mediterranean), specifically in the towns of l'Ampolla and Altafulla, as well as in the sand of Altafulla beach. The study involved collecting water samples using the passive filtering prototype SB-Buoy, analyzing them in the laboratory, and manually sieving the beach sand, as a citizen science project conducted by students and teachers from professional degrees. The concentrations observed varied considerably depending on the sampling locations and periods. Significant preliminary results should be highlighted: tiny microplastics dominate the samples from seawater (Ø < 3 mm), and plastic pellets in the sand accounted for 52% of the anthropogenic waste by weight in the sampling area.

Keywords Marine microplastics; SB-Buoy; Plastic pellets; Waste; Coastal ecosystems; Citizen science

Currently, microplastics (MP) are found in every place and every terrestrial ecosystem, from the abyssal depths of the oceans to the snows of the mountains (GESAMP, 2015). Simultaneously, considerable concentrations of MP have been observed in the tissues of living organisms (Van et al., 2015). The presence of MP in the marine environment presents an increasingly alarming problem due to the continuous production of plastics and their rapid transformation into waste, which often ends up in the seabed where they degrade, fragment, and disperse (GESAMP, 2015). MPs are defined as plastic particles with a size smaller than 5 mm (Viršek et al., 2016), and can be of primary or secondary origin. Primary origin are directly manufactured, such as the small particles used in the cosmetic industry for certain products and/or creams, or in the petrochemical industry, like pellets that serve as raw material for manufacturing other specific plastic products through pellet extrusion (Endo et al., 2005). On the other hand, secondary origin MPs are formed through the degradation of plastics and macroplastics in the environment exposed to light, heat, oxygen, water, or living organisms. In other words, they have undergone physical, chemical, biological degradation, or a combination of some of these factors at once, if not all (Viršek et al., 2016). The small size of MP allows them to enter the food web through ingestion by organisms in marine ecosystems. The ingested particles can cause physical damage or block the digestive system of animals, in addition to possibly carrying persistent organic pollutants (POPs) (Masura et al., 2015). Moreover, it has been widely described in the literature that both plastics and associated or adhered contaminants can act as endocrine disruptors in organs and tissues of living organisms (Manikkam et al., 2013; Pinson et al., 2017; Nelms et al., 2018). These pseudo hormones have already begun to cause serious alterations in people and children around the world, by triggering erroneous or uncontrolled immune responses (Pinson et al., 2017; Yamashita et al., 2018). Furthermore, it is imperative to underscore the profound implications that arise for human enterprise stemming from these pressing environmental challenges, as illuminated by the work of Rees (2023) and the comprehensive research efforts by Richardson et al. (2023).

The Institut Rambla Prim, in collaboration with the Institut-Escola del Treball of Barcelona, conducted a comprehensive study focused on the presence of MP within the marine surface environments of l'Ampolla and Altafulla beaches, as well as within the sand of Altafulla Beach, situated in the Tarragona region of the Western Mediterranean (Figure 1). This study engaged students and educators from Higher Vocational Training Course

specializing in Environmental Education and Control Technician, alongside those pursuing the Higher Vocational Training Course in Laboratory Technician for Analysis and Quality Control. Together, they actively participated in a prospective research endeavor, investigating the critical environmental concern of MP pollution within the marine ecosystem, thereby establishing a citizen science initiative. The study was primarily rooted in the collection of water and sand samples, followed by their meticulous analysis and classification within the confines of a specialized chemical and environmental laboratory. Additionally, the research enjoyed collaborative support from other esteemed public entities, including the Water Research Institute of Catalonia (ICRA) and the municipal authorities of l'Ampolla and Altafulla, who made significant contributions to the study.

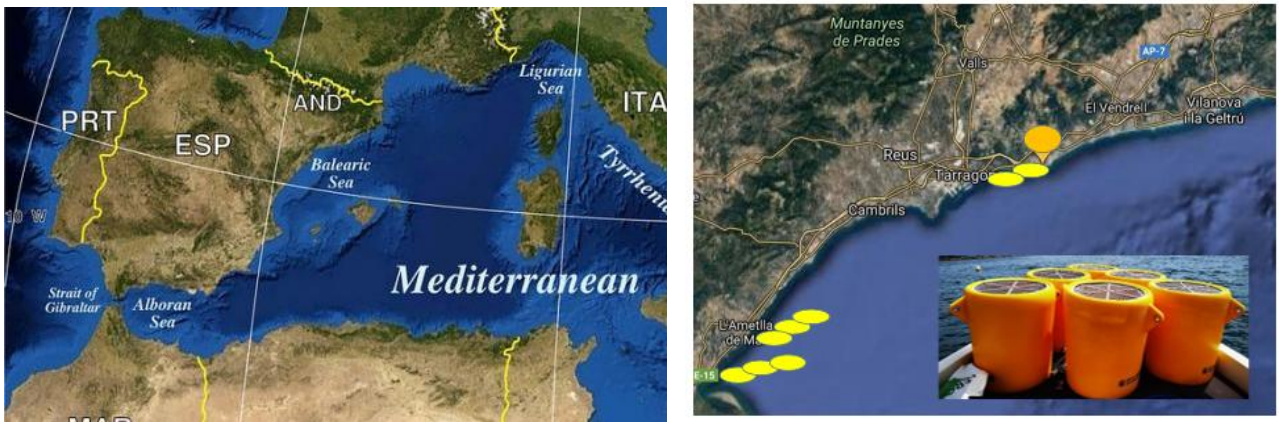

Figure 1 Location of the Buoys (yellow) and sand sampling points (orange) on the Coast of Tarragona
Note: This study was conducted in the Western Mediterranean at Balearic Sea. A: Depicts the area of study. B: Highlights the specific locations; Yellow dots represent buoys in l'Ampolla and Altafulla towns. An orange dot designates sand sampling points in Altafulla sand beach. It also showcases prototypes of sampling buoys (SB-buoy)

1 Results

1.1 Preliminary results

Within the framework of this citizen science study, an exhaustive collection of samples was conducted, and some of them have been analyzed to obtain significant data and develop representative graphs for initial prospective research. Due to the large number of samples, locations, and results, only a few clear examples and general conclusions derived from them will be presented here. It is essential to note that this work is an ongoing study, and further results are continually being elaborated and analyzed to delve into these or other hypotheses and complementary conclusions. The following are some of the most elucidating results.

1.2 Identification of sea microplastics

The distribution of MP by weight for different size ranges in sample 5 and 23 (Figure 2), obtained from the seawater of Altafulla Beach during July and August of 2021. In sample 5, from July, the analysis reveals the predominance of MP with a diameter less than 3 mm. The smallest observed diameter is 0.5-1.6 mm, representing 15.5% of the total, which is the lowest percentage recorded. In contrast, the size range from 1.6 to 2 mm shows the highest percentage, accounting for 24.4% of all identified MP. The next size range from 2 to 3 mm comprises 19.8%. Finally, the larger size ranges; 3 to 6 mm Ø, and greater than 6 mm Ø, accumulate 24.2% and 16.1%, respectively. In sample 23, obtained from Altafulla Beach during August 2021, once again, the analysis reveals that most MP have a diameter less than 3 mm. The smallest observed diameter is 0.5~1.6 mm, representing the highest percentage at 30.9% of the total. The size range from 1.6 to 2 mm Ø shows the next highest percentage, at 17.9% of all identified MP. The two subsequent size ranges; 2 to 3 mm and 3 to 6 mm Ø, present similar quantities as the previous one, at 16.7% and 17.5% respectively. Finally, the size greater than 6 mm Ø and MP fibers appearing in this sample represented 13.3% and 3.8% respectively.

These data highlight the prevalence of small-sized MP in the two analyzed samples. MP with a diameter less than 3 mm account for the majority identified particles, 59.7% in Sample 5 and 65.5% in Sample 23. It should be noted

that with a more accurate classification of MP, limiting the maximum size to 5 mm Ø, the percentages of particles smaller than 3 mm would increase significantly (Ø < 3 mm). Due to the characteristics of the laboratory sieves, these data are not entirely accurate, and the maximum particle sizes are not limited to 5 mm Ø.

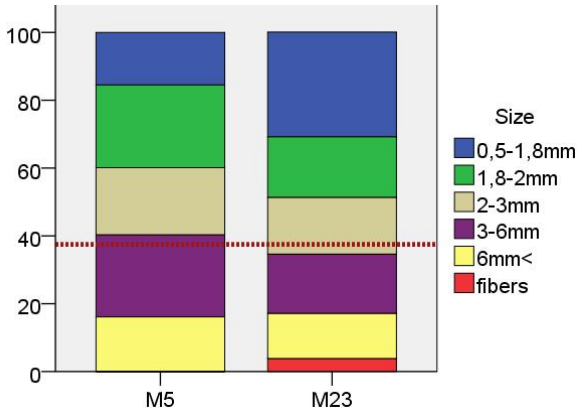

Figure 2 Weight percentage histogram for sample fractions

Note: Histograms displaying weight percentages for two distinct samples, Sample 5 (on the left) and Sample 23 (on the right). To delineate the size distribution of MP within these samples, we have overlaid red dashed lines, serving as an approximate demarcation between smaller and larger MP particles (Ø < 3 mm)

1.3 Identification of waste in sand

The composition of the beach sand based on different samples (n=14) obtained at the following coordinates and date: 41.134 607, 1.38 4632; 01/11/2021 (Figure 3 A). The analysis reveals that 75% of the total samples consist of inorganic matter such as sand and stones. The 14% corresponds to organic matter, including algae, trees wood and leaves, among others. Finally, the remaining 11% represents the identified anthropogenic waste (ẋ; n=14). The composition of this 11% of waste is detailed, mainly comprising anthropogenic plastics (Figure 3 B). Notably, the majority of this 11% of human-made waste consists of 52% plastic pellets, 33% macroplastics with a size greater than 5 mm Ø, and 15% MP with a size smaller than 5 mm Ø (ẋ; n=14). This information highlights the significant presence of anthropogenic residues in the beach sand, with a considerable proportion of plastics. The plastic pellets, in particular, constitute the majority of these residues in the study area.

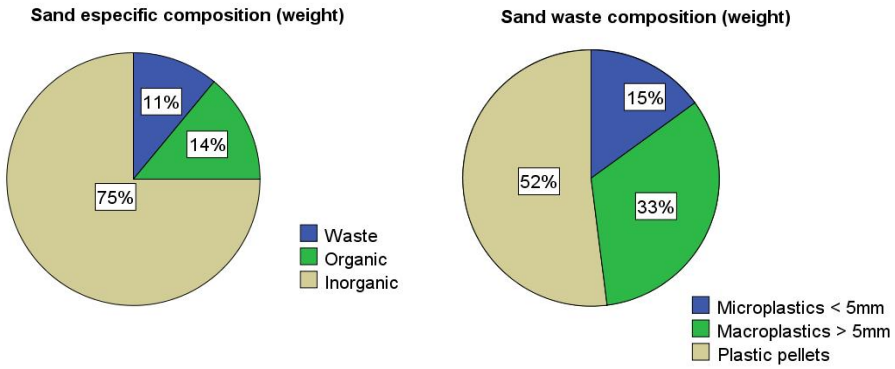

Figure 3 Percentage of waste on Altafulla Beach

Note: (A: n=14) and typology of waste (B: n=14)

2 Discussion

This study has conducted prospective research in citizen science to understand the composition of anthropogenic waste on beach sand and the presence of MP in coastal marine waters in the province of Tarragona, Spain. The results obtained have provided significant insights into this environmental issue in the area. In the context of identifying MP in water, the distribution was analyzed in two samples collected at Altafulla Beach in July and August 2021 (Figures 2). Each figure represents a distinct size range distribution. In both cases, it has been

observed that the majority of MP have a diameter smaller than 3 mm. These results align with previous studies and reinforce the prevalence of small-sized MP in marine ecosystems (Nelms et al., 2018). Moreover, a mere 3.8% of Sample 23 comprises plastic fibers. This finding leads us to infer a terrestrial origin for the suspended particles in marine water, as opposed to an atmospheric source, given that the predominant description of atmospheric MP aligns with a fibrous nature (Dris et al., 2015). Finally, the study outcomes have the potential to elucidate the existence of a distinct trituration mechanism, as described, within both marine and terrestrial/limnological environments. This underscores a more significant contribution to the generation of minute MP from sources within these ecosystems rather than from atmospheric origins.

MP, in general, exhibit high bio-availability to marine organisms, either through direct ingestion or indirectly through trophic transfer from contaminated preys (Covernton et al., 2021). The latter pathway has been observed in low trophic level organisms under laboratory conditions, but there is less empirical evidence in high trophic level taxa. In situ studies face challenges in dealing with contamination and distinguishing between MP ingested directly or indirectly (Nelms et al., 2018). Studies suggest that trophic transfer would represent an indirect but potentially significant pathway of MP ingestion for any species with a feeding ecology involving the consumption of whole prey, including humans. Other studies point to empirical evidence that MP particles can be transferred between trophic levels, from fish to higher-level marine predators. For example, a study revealed a higher presence of plastic in the stomachs of harbour seals (*Phoca vitulina*) compared to other parts of the gastrointestinal tract or faeces. This suggests that the stomach can act as a selective trap for non-nutritive items such as MP and other anthropogenic debris (Rebolledo et al., 2013). In situ investigations outlined MP ingestion in commercially relevant demersal fish species sourced from the Spanish Atlantic and Mediterranean coasts-specifically, the lesser spotted dogfish (*Scyliorhinus canicula*), the European hake (*Merluccius merluccius*), and the red mullet (*Mullus barbatus*). The study revealed that 17.5% of the sampled fish exhibited MP ingestion. On average, 1.56 ± 0.5 MP items were found per fish, ranging in size from 0.38 to 3.1 mm—the same size range that predominantly characterizes the present study (Bellas et al., 2016). Recent studies have unequivocally substantiated the occurrence of MP ingestion in the Spanish Mediterranean Sea waters, as exemplified by findings from the Mar Menor coastal lagoon in Southeast Spain. A study focused on the gilthead seabream (*Sparus aurata L.*), a commercially significant species. Notably, 40.32% of the total isolated microparticles were identified as MP (<5 mm) using FTIR spectroscopy. The average MP concentration was recorded at 20.11 ± 2.94 MP kg−1 per fish, with concentrations of 3912.06 ± 791.24 and 1562.17 ± 402.04 MP by kg in the stomach and intestine, respectively. MP sizes ranged from 91 µm to 5 mm, with an average of 0.83 ± 0.04 mm, reinforcing the findings of the present study regarding the small size range. Nine polymer types were identified, with high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene polypropylene (PEP), and polyvinyl (PV) emerging as the most abundant polymers. The study underscores heightened levels of MP ingestion in the Mar Menor, surpassing averages reported in most Mediterranean Sea studies, owing to its semi-enclosed ecosystem. In this case, the presence of fiber forms in the area is primarily attributed to local fishery activities by the authors (Bayo et al., 2021).

In relation to human health, the discovery that MP can transfer from fish to higher-level predators carries important implications (Rist et al., 2018). For instance, significant concentrations of MP have been observed in whole shellfish, which are subsequently ingested by humans (Murray and Cowie, 2011; Rochman et al., 2015). A clear example of detected material in the composition of MP is ethylene-propylene. This polymer represents the main ingested material in high-level marine animals and even in humans. It is important to note that significant implications regarding the action of endocrine disruptors or pseudo-hormones remain to be evidenced in future research (Rist et al., 2018).

Regarding the identification of waste in the sand, it is relevant to point out that a large portion of this waste consists of 52% plastic pellets, followed by 33% macroplastics with a size greater than 5 mm Ø, and 15% MP with a size smaller than 5 mm Ø. These results emphasize the significant presence of anthropogenic residues, especially plastics, in Altafulla Beach sand, with a notable abundance of plastic pellets. Furthermore, it should be

noted that the majority of macroplastics present in these samples (33%) will eventually become bioavailable MP in the ecosystem due to the actions of physical, chemical, and biological agents in the natural environment.

In the context of sand waste, pellets refer to plastic particles characterized by a cylindrical shape and a diameter ranging from approximately 1 to 2 millimeters. The majority are white, with a few being transparent, and a small minority exhibit various basic and specific colours due to the extrusion of diverse products (e.g., blue, red, green, yellow, etc.). These plastic particles serve as raw materials utilized worldwide and are mass-produced in petrochemical industries. Consequently, they pose a significant problem, as they can unintentionally be released during manufacturing and transportation processes, ultimately finding their way into rivers, seas, and oceans (Ogata et al., 2009). The production and transportation cycle of pellets entail losses and critical points at different stages. This cycle is controversial due to multiple factors, initiating at industrial and petrochemical plants where unintentional pellet losses occur, and they subsequently infiltrate sewage systems and eventually reach rivers. Furthermore, during terrestrial and maritime transport, substantial losses and spillages have been detected, eventually dispersing throughout beaches worldwide (Ogata et al., 2009; Ohgaki et al., 2021). Additionally, a compelling phenomenon occurs as consequence—the absorption of chemical contaminants existing in the environment, which are evidently discernible in the collected pellet samples (Endo et al., 2005). With certainty, these granules contain traces of PCBs and pesticides, among other substances (Teuten et al., 2009). PCB's, in particular, constitute one of the most critical contaminants, known to act as endocrine disruptors or pseudo-hormones upon contact with tissues and organs of living organisms (Pinson et al., 2017). Unfortunately, pellets and their associated substances are found on beaches worldwide, primarily in regions experiencing severe weather conditions, along with other types of organic and anthropogenic debris, as described in this study. Concerning the Tarragona coast, it has been an excessive example of pellet concentration, awaiting the results of analyses of contaminants adhered to these pellets (86.4 grams of pellet samples sent to Fuchu, Tokyo, Japan; International Pellet Watch).

In summary, this study reaffirms the issue of MP and the presence of anthropogenic residues in coastal beaches and marine waters of the Tarragona coastline. The high concentration of small MP and the prevalence of pellets underscore the urgency of implementing measures to address this environmental concern. The study provides a fundamental scientific basis for further research and the implementation of management and mitigation strategies aimed at preserving the marine ecosystems in the Tarragona coastal region and safeguarding the environmental health of the investigated areas.

3 Materials and Methods

The SB-buoys prototipe were sampled monthly from June 2021 to September 2021 with the assistance of auxiliary boats (Figure 1). The contents of the buoys were deposited in glass jars, noting the collection date, buoy number, municipality where the buoy is located, and sample number (Viršek et al., 2016). Samples were stored in a dark space with freshwater during the months of August and September. Subsequently, the cleaning and selection of the samples were carried out during the month of October of the same year. Samples were filtered through a mesh, and using laboratory forceps and spatulas, the MP were separated from persistent organic and inorganic matter. The MP obtained during the selection and purification process were placed in glass jars with 70% alcohol. These samples were labelled with information from the primary sample and then sent to the Institut-Escola del Treball of Barcelona for a comprehensive analysis of composition, size, and shape through sieving, classification, and weighing (Viršek et al., 2016). Finally, a visual identification with a microscope and/or binocular loupe was performed to regroup the plastic microfibers (Figure 4). The classification of water MP was done up to a maximum size of 6 mm and above, derived from the mesh size of the laboratory sieves used. Other samples were manually collected using hand dip nets to complete the series of samples from the summer of 2021 at Altafulla Beach. This last series of added samples followed the same preservation, labelling, and analysis protocol as previously described, to be eventually sieved, classified, and weighed as well (Viršek et al., 2016).

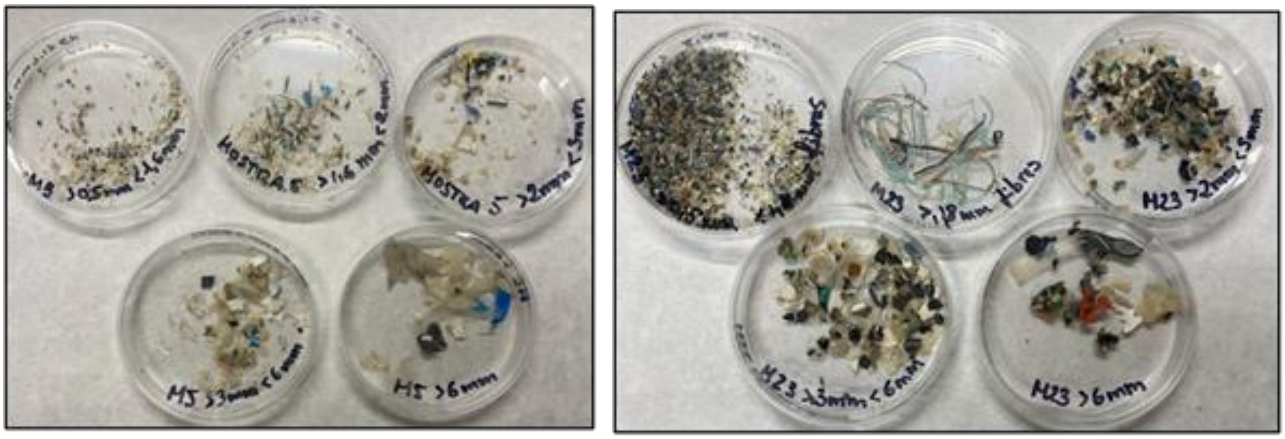

Figure 4 Classification of microplastics (MPs) by size and shape

Note: A: Depicts sample 5 collected at the beginning of summer 2021 along the Altafulla coast. B: Represents sample 23 collected at the end of summer 2021, also along the Altafulla coast

3.1 The sand waste collection protocol

For the sand analysis, samples were taken at Altafulla Beach from an area with abundant waste presence (Altafulla, Tarragona. Coordinates: 41.134 607, 1.384 632. Date: 01/11/2021). Subsequently, in the 21-22 academic year, second-year students of Environmental Education and Control at Institut Rambla Prim of Barcelona worked on the classification of plastics included in sand samples. The classification was divided into three main groups: macroplastics (Ø > 5 mm), MPs (Ø < 5 mm), and plastic pellets (Ø ≈ 1~2 mm). The plastic pellets were recognized as predominantly white spheres, some transparent, and occasionally of various colors (Figure 5 C). The samples of macroplastics and MPs were stored, and the plastic pellet samples were divided into 37 bags, each containing 100 pellets (2~3 g per bag; total 86.4 grams) to be sent to the Laboratory of Organic Geochemistry at the University of Agriculture and Technology in Fuchu, Tokyo, Japan (International Pellet Watch). There, the state of chemical contamination adhered to the pellets will be determined and data used for future research (Ogata et al., 2009; Ohgaki et al., 2021) (Figure 5 C).

Figure 5 A comprehensive classification of plastic waste derived from sand samples

Note: A: Highlights larger plastic debris found in the sand samples (Macroplastics). B: Focuses on the classification of minute plastic particles microplastics (MPs), facilitating the understanding of smaller plastic elements present in the sand. C: Displays the segregation of plastic pellets, contributing to the understanding of various plastic pellets types within the sand samples

4 Conclusions

This preliminary study investigates the composition of anthropogenic residues in beach sand and MP in marine waters along the Tarragona coast, yielding significant results that underscore the widespread presence of these

contaminants in marine ecosystems. Through the collection of samples and subsequent analyses, valuable insights have been gained into the magnitude and characteristics of this issue. This emphasizes the urgent need to address the escalating problem of MP, particularly those of smaller dimensions. To effectively tackle this issue, it is imperative to comprehend their sources, distribution, and potential environmental and human health effects, laying the foundation for the development of appropriate management strategies and mitigation efforts. As mentioned earlier, the significance of certain fish species as biomonitors within the Spanish Marine Pollution Monitoring Programme (SMP) cannot be overstated. Given the established evidence of microplastic ingestion in these species, they emerge as promising candidates for monitoring spatial and temporal trends in ingested litter. This is especially pertinent considering the complexities of current protocols, which use passive filtering SB-buoys, and their relevance to human health. The implications of present study contribute essential baseline data for implementing the Marine Strategy Framework Directive descriptor 10 in Spain. This underscores the imperative need for comprehensive assessments and effective management strategies to address the pervasive issue of MP contamination in commercial fish populations (Bellas et al., 2016).

On the other hand, the escalating environmental issues associated with conventional plastics have spurred a growing interest in bioplastics as promising alternatives. While acknowledging that bioplastics are not without their environmental concerns, including greenhouse gas emissions and potential land use change, current research underscores that the environmental impact of bioplastics is generally less severe compared to traditional counterparts (Atiwesh et al., 2021; Shah et al., 2021). As new-generation bioplastics continue to be developed, it becomes paramount for policymakers to rely on comprehensive studies that consider life cycle assessments. This strategic approach will facilitate well-informed decisions on the integration of bioplastics, offering a potential solution to mitigate the adverse effects of marine MP. Furthermore, considering the triturating effect on marine MPs mentioned earlier, bioplastics could emerge as a crucial mechanism for waste degradation and ecosystem integration. This further emphasizes their potential role in sustainable waste management practices.

Regarding the identification of beach sand residues, a substantial proportion of anthropogenic plastic residues has been detected, with pellets being the most prevalent. Notably, pellets have accounted for the largest proportion of these residues, surpassing the combined presence of macroplastics and MPs. This worrisome presence of pellets on beaches signifies a persistent and long-lasting impact. Despite persisting for several decades, it is only in recent years that increased attention has been directed towards this environmental concern by various entities and associations, both in Spain and on a global scale. Controlling the pervasive presence of plastic pellets in marine environments necessitates the implementation of more stringent controls within the petrochemical industry. By enforcing stricter regulations and oversight on production processes, including the transportation and handling of raw materials and finished products, we can minimize the unintentional release of plastic pellets into marine ecosystems (International Pellet Watch, 2023). Furthermore, it is imperative to establish comprehensive studies and monitoring protocols to systematically track the distribution, concentration, and ecological impact of plastic pellets. This proactive approach ensures a continuous and adaptive monitoring system, allowing for timely interventions and adjustments in industrial practices to mitigate environmental harm. The collaboration between regulatory bodies, industries, and scientific communities is crucial to developing and implementing effective strategies that address the issue of plastic pellet pollution while fostering a sustainable and responsible petrochemical industry (International Pellet Watch, 2023). In the present study, samples of sand pellets are dispatched to the International Pellet Watch, Laboratory Organic Geochemistry located in Fuchu, Japan. This initiative aims to facilitate the monitoring of chemical aggregation and distribution within the studied area. Subsequent studies stemming from these current results will ascertain the requirements for implementing new and refined protocols in pellet monitoring within the area. This evaluation will be pivotal in enhancing the precision and scope of future monitoring endeavors.

In conclusion, this study has highlighted the problem of MP and the presence of anthropogenic residues in the analyzed beaches and coastal marine waters, on Western Mediterranean. The obtained results emphasize the need for immediate action to address this urgent environmental matter. Implementing management and mitigation

strategies is imperative to reduce the presence of MP and residues in marine ecosystems, thereby preserving the health of seas and oceans and marine biodiversity. These actions must be guided by principles of sustainability, promoting responsible plastic production and usage practices, fostering appropriate waste management, and increasing awareness and education on reducing plastic usage and combating plastic pollution. This study provides a fundamental scientific basis for future research and initiatives aimed at conserving and protecting our coastal ecosystems along the Tarragona coast.

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