



Environmental status of marine plastic pollution in Spain

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ABSTRACT

The excessive use of plastic in our society is causing a massive accumulation, since it is a non-biodegradable product and with still poor recycling rates. This effect can be observed in the seas, which more and more plastic waste are accumulating. The present work is a critical review, based on all currently available literature, that reports environmental status of marine plastic pollution, especially microplastic pollution, in Spain. The three Spanish water areas with the highest presence of plastics are the Alboran Sea, the Gulf of Alicante and the vicinity of Barcelona probably related to fishing and industrial activities and high population densities. With regard to microplastic contamination on beaches in Spain, annual monitoring by the Spanish government shows contamination along the entire coast of the country, with particularly high concentrations in the Canary Islands (between 800 and 8800 particles/m² in spring). Between 40 and 50% of the particles analyzed were pellets and the main factors postulated for the distribution of these particles are marine currents and the geomorphological characteristics. With regards to biota, ingestion of microplastics by fish has been intensely confirmed and, important differences were observed between the locations of the sampling, being bogues (*Boops boops*) one of the fish species more studied in Spain. Finally, the work includes a revision of European and Spanish legislation about plastics and marine pollution and some strategies to reduce this kind of contamination in Spain.

1. Introduction

Modern society needs many different types of materials to satisfy the daily requirements, and these materials are often chosen on the basis of their efficiency or synergies within a combination of materials for a system or product. One of the most widely used materials is plastic, which has become indispensable thanks to its versatility and capacity, offering customised solutions for a wide variety of needs in countless products, applications and sectors.

In 2019, the production of plastics reached approximately 370 million tons, with China being the largest producer (31% of world production). In Europe, plastics production reached almost 62 million tonnes. The most popular type of plastic is polypropylene (PP), followed by polyethylene (PE) in all its forms (low density, high density, etc.). With regard to demand by sector and by type of polymer, the packaging sector and the construction sector are the largest consumers of plastic (PlasticsEurope, 2020).

Over the past 60 years, plastic has brought economic, environmental and social benefits. However, the excessive use of disposable products has led to an exponential increase in the amount of plastic waste

resulting from land and maritime activities, resulting in considerable economic, environmental and social problems. Solving these problems and addressing the legacy of waste and pollution caused by plastics is a challenging task that requires concerted action at all levels of government and on multiple geographic scales (United Nations Environment Assembly, 2018).

Marine pollution caused by plastics has become a particularly important debate. Approximately 6.4 million tonnes of plastic are dumped into the seas and oceans each year (80% of all marine waste) in the form of “macroplastics”, pieces larger than 2.5 cm, or “microplastics”, pieces of plastic smaller than 5 mm. Microplastics (MPs) pollution has become significant due to the potential risk it means to marine fauna and humans. These particles can result from the physical and chemical degradation of larger plastics in the environment (secondary MPs) or can reach the sea directly by dumping products containing them (primary MPs) (Godoy et al., 2019). In this sense, the environmental degradation of plastics is an important factor for the formation, distribution, and accumulation of microplastics in the aquatic system. Degradation of plastics occurs primarily through abrasive forces, heating/cooling, freezing/thawing, wetting/drying, effect of

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UV light, oxidation or hydrolysis and biodegradation by bacteria, fungi or algae (Klein et al., 2018).

According to a study by the International Union for Conservation of Nature (IUCN), macroplastics from poorly managed waste make up 94% of total plastic waste in the environment. Once the plastic reaches the sea or the beaches it is gradually degraded and turned into MPs, which are mostly deposited in sediments. For primary microplastic, tyre dust is the largest source of leakage (53%), followed by textiles (33%), microbeads in cosmetics (12%), and production pellets (2%) (Boucher and Billard, 2020).

In recent years there has been a growing awareness of the importance of protecting the environment and the possible impacts of using these types of materials. Europe is addressing this problem through a series of measures intended to contribute to the circular economy and the reduction of waste. It is therefore proposed that all plastic packaging should be recyclable or reusable by 2030, that consumption of disposable plastics should be reduced and that the deliberate use of MPs should be restricted. In addition, the strategy recognises that marine litter is a growing problem (80% of it from plastics) and includes initiatives and guidelines to minimise plastic waste (European Commission, 2018a; European Commission, 2018b).

In this context the Agenda 2030 stands out, an international commitment signed in 2015 by the governments of the United Nations member countries. It consists of 17 Sustainable Development Goals (SDGs) to address the social, economic and environmental challenges of globalisation. As part of Agenda 2030, SDG 14 aims to conserve and sustainably use oceans, seas and marine resources for sustainable development. Specifically, target 14.1 states the need by 2025 to prevent and significantly reduce marine pollution of all kinds, in particular from land activities, including marine debris and nutrient pollution, making the issue of plastics pollution one of the top priorities worldwide (UNEP, 2020; MDSA, 2020).

Among these measures, it should be noted the collaboration of the Global Plastics Alliance, an agreement of 74 associations of the plastics industry worldwide in the fight against marine waste, where more than 355 projects have been or are being carried out in different parts of the world aimed at combating this problem (GBA4E, 2020). In addition, the plastics industry has promoted the Operation Clean Sweep, a voluntary programme dedicated to helping all plastic resin handling operations achieve zero plastic pellet loss (Operation Clean Sweep, 2020).

In the particular case of Spain, in 2015 was labelled Europe's most polluted country by the European Environment Agency (EEA). Now, with growing concerns over plastic and emissions pollution on the global stage, Spain becomes a country of note. For example, in 2017 Spanish researchers showed that microplastics were present in standard table salt (Iñiguez et al., 2017). In 2018 a Sperm whale washing up on the coast of Spain made international headlines (death by plastic). This event highlighted the microplastics problem of the Mediterranean Sea, which on the Spanish coastline makes up 54% of plastic pollution. In this work, a comprehensive review based on all data currently available on marine plastic pollution in Spain was provided.

This work covers and describes different aspects including: (a) sources of plastic pollution in Spain; (b) quality and quantity of (micro) plastic pollution in beaches; (c) quality and quantity of (micro)plastic pollution in wastewater treatment plants; (d) quality and quantity of (micro)plastic pollution in freshwater environments and transitional waters; (e) quality and quantity of (micro)plastic pollution in biota; (f) responses already in course in Europe and Spain to reduce plastic litter and marine litter.

2. Situation of plastic pollution in Spain

Spain is located in southwest Europe and occupies 80% of the Iberian Peninsula, which it shares with Portugal. It has a surface area of 505,957 km², including the peninsular area, the Balearic Islands, the Canary Archipelago and the Spanish cities located in North Africa: Ceuta

and Melilla.

The country has approximately 8000 km of coastline, where four marine domains can be distinguished: Mediterranean, Cantabrian, peninsular Atlantic and Macaronesian Atlantic (Canary Islands).

The Spanish regulatory system for the protection of the marine environment is set out in Law 41/2010, of 29 December, and in Royal Decree 957/2018, of 27 July, which amends Annex I to Law 41/2010. This law divided the Spanish marine environment into five marine districts (Fig. 1): North Atlantic, South Atlantic, Strait and Alboran, Levantine/Balearic and Canary Islands, for each of which a marine strategy is being developed, with an updating period of 6 years (MITECO, 2021).

According to data from the National Statistics Institute (INE, 2020), urban waste management companies collected 22.7 million tonnes of waste in 2018, 0.8% more than in the previous year. Of this, 18.3 million tonnes were mixed waste and 4.4 million tonnes were collected separately. In per capita terms, 485.9 kg of waste were collected per person and year in Spain.

With regard to plastic waste, in 2018, 2.5 million tonnes of post-consumer plastic waste were collected through official plans to be treated, 41.9% of which was recycled (13% more than in 2016), 19.3% was used for energy recovery (12.2% more than in 2016) and 38.8% was deposited in landfills (3.6% less than in 2016) (PlasticsEurope, 2020).

The new Directive (EU) 2018/852 on packaging and packaging waste sets recycling targets of 50% of plastic packaging by 2025 and 55% by 2030, where Member States had to put in place the necessary laws, regulations and administrative provisions to comply with the provisions of this Directive in 2020 (Directive (EU) 2018/852). In recent years, an increasing evolution has been observed with regard to plastic waste recycling in Spain, where in 2018 almost 42% of plastic waste was recycled, as indicated above, and 51% of plastic packaging, positioning it as the second country in Europe that recycles the most plastic packaging. Since the trend continues positive, Spain could be increasingly closer to achieving the European Commission's plastic waste targets, although changes could occur due to the current health situation (coronavirus crisis).

According to the Ministry for Ecological Transition, in 2014 a Monitoring Programme for Marine Litter was designed as part of the programmes for monitoring the environmental state of the marine environment, as a development of Law 41/2010, of 29 December, on the protection of the marine environment (MITECO, 2019). These campaigns consist of a 100-m transect sampling of the beach and a second inspection over a length of 1000 m (objects larger than 50 cm are considered), on different Spanish beaches, taking into account the marine demarcation (Fig. 1) and the possible origin of the waste, which is divided into maritime origin (mainly from navigation, fishing and aquaculture) and terrestrial origin (mainly from municipal and industrial origins) (CONAMA, 2018).

In general, in Spain, and according to the report of 2019 mentioned above, in the transect of 1000 m the highest average number of objects per sample was registered in the South Atlantic demarcation and the lowest average number was registered in the Canary Islands demarcation, with the highest quantities being registered in the winter and autumn campaigns and decreasing in spring and summer. As for the category of objects larger than 50 cm, they were mainly plastic, followed by wooden objects. The known origin of marine litter is mainly related to maritime traffic or shipping, being no less important fishing activities (North Atlantic and South Atlantic districts) and agricultural activity (Almería and Murcia - Strait and Alboran demarcation).

Regards the transect of 100 m, the highest average number of objects per sample was counted in the Strait and Alboran demarcation, and the lowest average number was recorded in the South Atlantic demarcation, with the maximum number of marine litters being counted in the autumn campaign and the minimum in the summer. In terms of the category of objects, these were mainly unidentifiable pieces of plastic between 0 and 2.5 cm and between 2.5 and 50 cm in size. Most of the



Fig. 1. Marine districts of Spain.
Source: Own elaboration.

litter found on Spanish beaches is mainly related to tourism (South Atlantic, Canary Islands, Levantine-Balearic and Strait and Alboran demarcations), not being less important the maritime traffic or navigation (North Atlantic demarcation). For 100 m transects, the report of the MSFD group (Marine Strategy Framework Directive) on the implementation of that Directive (Directive 2008/56/EC) published in 2020 (European Commission, 2020) sets the acceptable amount of marine litter at 20 objects per 100 m of beach. In the results of the 2019 report, Spanish beaches obtained an average of 327 objects per 100 m, with a minimum of 50 and a maximum of 1168 objects/100 m. The beaches with the highest presence of litter objects were those belonging to the Mediterranean. Therefore, these results show that Spanish beaches are far from complying with the acceptable level of marine litter proposed by the MSFD group but, on the other hand, according to the aforementioned report, Spain was one of the few European countries that submitted its monitoring report in 2019 with a high number of reported implementation measures, which means that progress in understanding the origin of marine litter and in implementing measures to mitigate it is positive.

Some of the results obtained by the Spanish Monitoring Programme for Marine Litter coincide with those obtained in a study carried out by a team of scientists from the University of Alicante and the Spanish Institute of Oceanography (IEO) in Murcia (García-Rivera et al., 2018), which indicates that the main component is plastic, mainly from fishing activity and maritime traffic by merchant ships, being the Mediterranean Sea one of the areas most affected by marine pollution. The researchers indicate that the three areas with the highest presence of plastics are the Alboran Sea, the Gulf of Alicante and the vicinity of Barcelona, where in the Alboran Sea the presence of marine litter is high near the coast due to the fact that the narrow continental platform that leads to an accumulation of all types of litter in the coastal areas. In addition, due to the high level of maritime traffic entering and leaving the Strait of Gibraltar in open waters, the distribution of marine litter influences this area.

With regard to MPs, estimates of the contribution of MPs to Spanish

marine waters according to the different sources considered are presented in Table 1, noting that the main sources of MPs to the marine environment in Spain are pre-production pellets and the degradation of tyres due to their use (CEDEX, 2017). These quantities are not necessarily found on Spanish beaches since, as will be seen in the next section of this article, there are several geological and meteorological factors that influence the distribution and accumulation of MPs.

Ecologists in Action, a Spanish non-governmental organization, each year carries out an annual environmental analysis of the approximately 8000 km of Spanish coastline, both peninsular and insular, covering the 10 coastal autonomous communities, plus the autonomous cities of Ceuta and Melilla. Since 2015, two flags have been assigned to each province, one for pollution and the other for environmental mismanagement, taking into account: water discharges, purification and sanitation; urban planning; industrial activities; ports and cruise ships; erosion; rubbish; biodiversity and invasive species; aquaculture; oil spills and prospecting; various.

In their last report, “Black Flags 2020” report, a total of 48 black flags (two per province) have been assigned to the different Spanish beaches (Fig. 2). The report points out that the biggest problem on the Spanish coasts is water discharges and their poor treatment.

In addition, the report takes into account the consequences that the current health situation (coronavirus crisis) is having on the Spanish coast, since the waste generated, such as masks and gloves, is

Table 1
Estimated emissions of MPs to the marine environment (CEDEX, 2017).

| Source | MPs emissions (t/year) |
|---------------------------------------|------------------------|
| Pre-production pellet | 5700 |
| Degradation of tyres due to their use | 1700–4200 |
| Paint | 425–714 |
| Washing of synthetic clothes | 35–450 |
| Artificial turf sports fields | 25–165 |
| Cosmetics | 90 |
| Detergents | 4 |



Fig. 2. Black flag.

Source: Own elaboration. [Ecologists in Action, 2020](#).

increasingly common on beaches and in the sea because it is being poorly managed. The COVID-19 pandemic has emphasised the dependence on disposable plastics. For example, if 2/3 of Spain's population throw an old plastic personal protective equipment (PPE) as two days (mainly masks), it would be equivalent to around 476 million PPE per month. If 1% of those PPE are not properly disposed, it would be equivalent to around 57 million of PPE littered per year, many of which would end up in the coasts. Spanish cities have already noticed the growing problem of coronavirus litter.

Table 2 shows a list of the Spanish beaches analyzed by Ecologists in Action, together with sources of pollution or environmental mismanagement for 2020.

In this context, what started as a health crisis has rapidly become an economic, social and environmental threat due to the fact that public health is now the top priority. The short-term negative effects of the COVID-19 pandemic are mostly related to the use and consumption of plastics and the generation of healthcare waste, where the demand for plastics is expected to increase by 40% in packaging (mainly food sector) and by 17% in other applications, including medical uses. This dramatic increase in waste is overloading the capacity of each country or municipality to manage and treat it properly, where incineration and landfilling are being prioritised, which is a major environmental problem because it contributes to the emission of greenhouse gases, as well as other potentially dangerous compounds, such as heavy metals, dioxins and furans (Patrício et al., 2021).

In the next sections, a revision of current studies on microplastic pollution in beaches, sea surfaces, wastewater treatment plants, freshwater environments and transitional waters and biota in Spain is provided.

3. Marine microplastic pollution in Spain

3.1. Microplastic pollution in beaches

There are some published articles that demonstrate the presence of MPs on beaches and various coastal areas of Spain, highlighting mainly the Canary and Balearic Islands, the Ebro River delta, the Mar Menor and the Alboran coast (Alomar et al., 2016; Álvarez-Hernández et al., 2019; Bayo et al., 2019; Godoy et al., 2020; Herrera et al., 2018; Rapp et al.,

2020; Reinold et al., 2020; Simon-Sánchez et al., 2019). These studies complement the work done annually by a government agency called CEDEX (Center for Studies and Experimentation of Public Works, Spain). Since 2013, Spain has a series of programmes managed by CEDEX in order to monitor the state of the marine environment which includes, among others, a specific sub-programme on MPs on beaches.

This sub-programme consists of taking beach sediment samples and subsequent laboratory determinations. It has been running since 2016 and currently, it carries out studies on 14 Spanish beaches (Fig. 3) distributed along the entire coast of the country (CEDEX, 2019). Of these, the beaches of Doñana, Itzurun, San Miguel and Lambra were included in the study for the first time in the 2019 campaign. All these beaches are subject to the same sampling and analysis protocol.

The sampling campaigns are carried out in spring and autumn, in order to observe differences due to seasonality. The sampling procedure used on these beaches is similar to that recommended by MSFD Group (2013) and by Maes et al. (2017), and followed by most studies of this type. Portions of sediment are taken within a square of 50 × 50 cm, which moves along the high tide line of the beaches up to a length of 100 m. In the laboratory, the treatment of the samples consists of a first sieving through 5 mm followed by a second sieving through 1 mm. Both fractions (between 1 and 5 mm and <1 mm) are separated by density with a NaCl saturated solution. The supernatant is filtered through a membrane and then stained with Nile Red pigment. The last step of this procedure, as in most studies of this type, consisted of inspection by stereo-microscopy (with blue light of a wavelength between 450 and 510 nm and an orange filter of 529 nm), counting the particles and classifying them by size, colour and shape. The classification by size ranges from those <200 µm to 4–5 mm, while the amount of micro-particles detected is expressed as a function of mass (particles/kg of dry sediment) or per unit area (particles/m² of beach).

The results obtained for each beach in the spring and autumn campaigns from 2017 to 2019 are presented in Tables 3 and 4, respectively. In general, the concentrations of MPs found are higher in autumn than in spring in all considered years. In addition, 2018 was the year in which less contamination by MPs was found. There are significant differences between spring and autumn in 8 of the 14 beaches analyzed, although not all of them show a clear trend. In spring, a tendency can be observed in Las Azucenas, Marenys, Cal Francés and La Llana. There is an increase

Table 2
List of beaches and the reason for marine pollution (Ecologists in Action, 2020).

| Autonomous communities | Province | Pollution | Environmental mismanagement |
|------------------------|------------------------|--|---|
| Andalusia | Almeria | Deretil beach, Villaricos, industrial pollution | Cala Siret, Villaricos, urban pressure |
| | Cadiz | El Carmen beach, Barbate, without wastewater treatment plant | Los Lances beach, urbanistic threats |
| | Granada | La Rábida beach, El Pozuelo, waste and toxic pollution | Beach of Peñón de Salobreña, urbanization |
| | Huelva | Huelva estuary, industrial pollution | Ría de Piedras and beaches of Lepe and Isla Cristina, purification |
| Asturias | Malaga | Estepona's coastline, purification | Nerja, impact of an urban development project |
| | Asturias | Lack of sanitation in Gijón | Bad management of the tufted cormorant |
| Canary Islands | Las Palmas | Fecal water discharges "Riu de las Dunas" hotels | Dumping and tourism on Lobos Island |
| | Santa Cruz de Tenerife | Dumping of La Nea beach | Project for the construction of the port of Fonsalía |
| Cantabria | Cantabria | Usgo beach, discharges from the Solvay company emissary | Filling of the Raos Marshes in Santander Bay |
| Catalonia | Barcelona | Inadequate sanitation system in Badalona | Airport expansion threatens La Ricarda lagoon |
| | Girona | Insufficient management in the face of storms such as the Gloria | Sos-Costa Brava: urbanism |
| Ceuta | Tarragona | Costal system of the Platja Llargà in Tarragona | Bad management in Barra del Trabucador (Ebro Delta) |
| | Ceuta | Audouin's Gull overfished and abandoned | Pelagic coastal area and north bay coastline |
| Basque Country | Biscay | Extension of the Plentzia dock | Extension of the car park in the biotope of San Juan de Gaztelugatxe |
| Galicia | Gipuzkoa | Sanitation of Pasai Donibane | Santa Klara Island: tourism and urbanism |
| | A Coruña | Lires Beach, fecal contamination | Playa de las Delicias, regenerations and threats to <i>Zostera noltii</i> |
| | Lugo | ALCOA Industrial Pollution | Mariña lucense, insufficient purification system |
| | Pontevedra | ENCE Industrial pollution | Marin port fillings |
| Balearic Islands | Balearics | Cala Egos, Santanyí, insufficient bathing water quality | Extension of the port of Palma |
| Melilla | Melilla | Extension of the desalination plant without emissary | Malfunctioning treatment plant |
| Valencia | Alicante | Discharges from San Gabriel beach treatment plant | Coastal erosion on Babylon beach, Guardamar del Segura |
| | Castellon | Les Fonts Beach (Alcosebre) | New jetty on the beaches of Fortí-Fora Forat-Cervol |
| | Valencia | Medicalia beach chlorination | Extension north port of Valencia |
| Murcia | Murcia | Portman Bay and Sierra Minera: industrial pollution | Mar Menor: agricultural pollution and eutrophication |

in concentration over the years in the first three cases, while in La Llana concentrations remain constant throughout the years. In autumn, the same tendency occurs in Las Azucenas, Marenys and Cal Francés. It is remarkable the case of Famara (in Canary Islands) in this season, as it experiences a decrease in concentration over the years. In contrast to that, the beach of Lambra is the most contaminated of the 14 analyzed in 2019. Also, there were two campaigns in Rodas with no MPs found, during spring 2018 and autumn 2019.

Analyzing the abundance of MPs by coastal area in Spain in 2019, when the 14 beaches were analyzed and significant comparisons can be made, it can be observed that the beaches of the Canary Islands (Famara and Lambra) are those with a significantly higher concentration than the others (819 and 8861 particles/m² in Famara and Lambra during spring, respectively, and 67 and 1968 particles/m² during autumn). The beaches of the Strait of Gibraltar-Alboran and the Levantine-Balearic coast (Las Azucenas, San Miguel, La Llana, Marenys, La Pineda and Cal Francés) are in an intermediate situation, with similar concentrations between the two areas (between 86 and 810 particles/m² in spring and 114–1136 particles/m² in autumn). Finally, beaches on the Cantabrian Sea coast (Covas, Rodas and Oyambre) and the Atlantic Ocean (Doñana and Castilnovo) have the lowest concentrations on average (between 12 and 147 particles/m² during spring and 0–200 particles/m² in autumn).

Other studies have been published that evaluate the concentrations of MPs in Spanish beaches. Table 5 represent the concentrations of MPs found along with the extraction and identification methods, in order to observe possible influence on the results of the studies. In the case of the Canary Islands, Herrera et al. (2018) carried out a study on the beaches of Lambra, Famara and Las Canteras. The reported concentrations of MPs are quite consistent with that provided by CEDEX. However, the authors detected a greater concentration of MPs in autumn on Lambra beach and in spring-summer on the beaches of Famara and Las Canteras, which is not the case with the CEDEX results, where Lambra is always more contaminated. The high concentrations of MPs in these beaches are usually associated with periods of strong winds and waves, and also the influence of the oceanic gyre and the Canary Current, which attract pollution from other parts of the world. Another study (Álvarez-Hernández et al., 2019) analyzed seven beaches in Tenerife, finding

much lower concentrations of MPs than those reported by the CEDEX (2019) and Herrera et al. (2018), despite using the same extraction method and a better identification method (FTIR Spectrometer). Specifically, concentrations varied between 2.0 and 115.5 particles/m² on beaches that are periodically cleaned because they are considered suitable for tourism. When measuring the concentration of MPs on Playa Grande, a beach considered unsuitable for bathing and not cleaned, up to 2971.5 particles/m² were found. However, doing a single sampling like in Álvarez-Hernández et al. (2019) might not be representative, as it can be a day with no ideal current, wind and wave conditions to accumulate MPs, so in this type of studies it is advisable to sample with a certain periodicity. Other recent studies conducted in beaches of Canary Islands confirmed the great variability of plastic abundance depending on the location, the sampling dates and the identification method (Rapp et al., 2020; Reinold et al., 2020). Rapp et al. (2020) found approximately twice the concentration of MPs on Gran Canaria beaches than Reinold et al. (2020) found on Tenerife beaches. These differences are due, among other factors, to the extraction and identification methods used, which were much more precise in the first study (Rapp et al., 2020). In addition, the not authorized discharged of wastewater to the sea without receiving any treatment or only primary treatment can be the cause of MPs contamination in some of these Gran Canaria beaches.

The Levantine-Balearic coast has also been analyzed by other authors. Bayo et al. (2019) carried out a study on the presence of MPs along the coast of the Mar Menor, an area seriously affected by intense urban development and tourism. The results of the study revealed concentrations of MPs ranging from 8.2 to 166.3 particles/kg of sediment, with higher concentrations in samples collected in the intertidal zone. However, these concentrations are much higher than those reported by CEDEX for La Llana, the beach closest to that area. The main difference is that Bayo et al. (2019) analyzed sediments from the interior part of the lagoon, while La Llana is an open beach located in the exterior part. The geomorphological characteristics and the maritime and climatic factors which affect the lagoon are different to those of open beaches, but as occurs in many other studies, there is finally a tendency to find higher concentrations of MPs in areas of important water run-off, intense tourism or fishing activities. Also, Bayo et al. (2019) used an extraction



Fig. 3. Geographical location of the beaches sampled annually by CEDEX in Spain.

Table 3

Average concentration of MPs found in sediments of Spanish beaches during spring sampling campaigns.

| Beach | Spring | | | | | |
|-------------------------|--------------------------|--------------|--------------------------|--------------|--------------------------|--------------|
| | 2017 | | 2018 | | 2019 | |
| | Particles/m ² | Particles/kg | Particles/m ² | Particles/kg | Particles/m ² | Particles/kg |
| Covas | 42.66 | 4.08 | 173.09 | 22 | 12.61 | 1.7 |
| Rodas | 72.45 | 7 | 0 | 0 | 71.67 | 6 |
| Oyambre | 96.28 | 6.82 | 9.45 | 1 | 75.26 | 7.89 |
| La Llana | 59.06 | 5.82 | 58.44 | 5 | 86 | 12.73 |
| Castilnovo | 51.74 | 5.51 | 196.66 | 17.31 | 89.38 | 10.18 |
| Doñana | – | – | – | – | 146.9 | 16.54 |
| Marenys | 12.96 | 1 | 107.93 | 11 | 225.5 | 15.66 |
| Cal Francés | 55.35 | 4 | 152.19 | 13 | 285.9 | 22.51 |
| La Pineda | 433.49 | 46.2 | 39.55 | 4.34 | 337.4 | 31.26 |
| Las Azucenas | – | – | 206.33 | 28 | 429.6 | 37.19 |
| Itzurun | – | – | – | – | 494.5 | 48.51 |
| San Miguel Cabo de Gata | – | – | – | – | 810.2 | 71.03 |
| Famara | 1312.83 | 127.92 | 247.58 | 22.4 | 818.9 | 72.29 |
| Lambra | – | – | – | – | 8861.1 | 881.3 |

Source: CEDEX, 2017, 2019; CEDEX, 2018.

method with filters with a very little pore size (0.45 μm) which can contribute to a higher finding of MPs. Another study carried out on beaches on the islands of Mallorca and Cabrera (Alomar et al., 2016), found concentrations between 100 and 900 particles/kg of sediment. According to the authors, although higher concentrations could be expected in densely populated and touristic areas, the fact is that the largest amounts of MPs (244.01 and 897.35 particles/kg) were located in virgin areas further away and controlled by the local authorities. This suggests that there may be a transport of particles from distant areas to other deposition areas, related to complex hydrodynamic conditions, as this area is dominated by the Almeria-Oran thermohaline current, in which macroplastics are usually floating in the water. Currently, CEDEX does not analyze any beach in Balearic Islands, but the results found by Alomar et al. (2016) are much higher than those reported by CEDEX for

the beaches located in the Levantine-Balearic coast (San Miguel, La Llana, Marenys, La Pineda and Cal Francés), which range from 1 to 114 MPs/kg in autumn, the same season in which the study by Alomar et al. (2016) was carried out. However, again it can be not representative a study that has only carried out one sampling campaign and too many years ago. More recent is the study of Simon-Sánchez et al. (2019), who found great MPs concentrations in sediments from the Ebro River estuary. This area is close to La Pineda beach, in which CEDEX found much less MPs (65 MPs/kg). However, the conditions in an estuary are very different than in an open beach. Specifically, the authors attribute the concentration of MPs to the continuous discharge from the river, which is causing the accumulation of MPs in an area with low hydrodynamic conditions, as the river is close to urban areas where dumping can occur, as well as to some waste water treatment plants. Also, Simon-Sánchez

Table 4
Average concentration of MPs found in sediments of Spanish beaches during spring sampling campaigns.

| Beach | Autumn | | | | | |
|-------------------------|--------------------------|--------------|--------------------------|--------------|--------------------------|--------------|
| | 2017 | | 2018 | | 2019 | |
| | Particles/m ² | Particles/kg | Particles/m ² | Particles/kg | Particles/m ² | Particles/kg |
| Rodas | 437.15 | 43.94 | 487.13 | 41.99 | 0 | 0 |
| Famara | 343.67 | 37.72 | 124.18 | 11.76 | 67.4 | 7.17 |
| Oyambre | 132.23 | 18 | 73.46 | 7.79 | 73.71 | 7.5 |
| Castilnovo | 221.79 | 12.39 | 12.49 | 1.07 | 82.49 | 6.82 |
| La Llana | 187.09 | 16.14 | 45.89 | 3.93 | 113.99 | 10.02 |
| Covas | 126.22 | 23 | 63.87 | 7.37 | 120.9 | 14 |
| Las Azucenas | – | – | 94.72 | 10 | 158.9 | 15.65 |
| Doñana | – | – | – | – | 199.7 | 20 |
| La Pineda | – | – | – | – | 457.8 | 65.08 |
| Itzurun | – | – | – | – | 554.2 | 55.32 |
| San Miguel Cabo de Gata | – | – | – | – | 972.6 | 79.99 |
| Cal Francés | 201.8 | 27.17 | 398.22 | 47.04 | 1049.8 | 114.1 |
| Marenys | 8.14 | 1 | 86.98 | 6 | 1135.7 | 87.17 |
| Lambra | – | – | – | – | 1968.2 | 194 |

Source: CEDEX, 2017, 2019; CEDEX, 2018.

Table 5
Comparison between MPs concentrations in different coastal areas of Spain and characteristics of the study carried out in each case.

| Beach | Average MPs concentration | Extraction method | Filter pore size (µm) | Identification method | Number of campaigns | Reference |
|--------------------------------|---------------------------------|--------------------------------------|-----------------------|------------------------------|-----------------------------|----------------------------------|
| All Spanish coast | ^a See Tables 3 and 4 | Density separation (NaCl) | – | Nile Red staining | Twice a year during 3 years | CEDEX (2017, 2019); CEDEX (2018) |
| Andratx (Balearic Islands) | 120–160 MPs/kg | Density separation (distilled water) | 63 | Stereomicroscope | One - October 2013 | Alomar et al., 2016 |
| Santa Maria (Balearic Islands) | 240–900 MPs/kg | Density separation (distilled water) | 63 | Stereomicroscope | One - October 2013 | Alomar et al., 2016 |
| Es Port (Balearic Islands) | 100 MPs/kg | Density separation (distilled water) | 63 | Stereomicroscope | One - October 2013 | Alomar et al., 2016 |
| Mar Menor lagoon | 8.2–166.3 MPs/kg | Density separation (NaCl) | 0,45 | Trinocular microscope + FTIR | Two - winter 2017 and 2018 | Bayo et al., 2019 |
| La Herradura (Granada) | 45.0 MPs/kg | Density separation (NaCl) | 7–9 | Nile Red staining | One - autumn 2018 | Godoy et al., 2020 |
| La Rábida (Granada) | 22.0 MPs/kg | Density separation (NaCl) | 7–9 | Nile Red staining | One - autumn 2018 | Godoy et al., 2020 |
| Motril (Granada) | 31.5 MPs/kg | Density separation (NaCl) | 7–9 | Nile Red staining | One - autumn 2018 | Godoy et al., 2020 |
| Ebro River estuary | 422 MPs/kg | Density separation (NaCl) | 0,7 | Stereomicroscope + µm-FTIR | One - winter 2017 | Simon-Sánchez et al., 2019 |
| Lambra (Canary Islands) | 1657.6 MPs/m ^{2a} | Density separation (ethanol) | – | Visual inspection | Twice a month during 1 year | Herrera et al., 2018 |
| Famara (Canary Islands) | 875.5 MPs/m ^{2a} | Density separation (ethanol) | – | Visual inspection | Twice a month during 1 year | Herrera et al., 2018 |
| Las Canteras (Canary Islands) | 430.8 MPs/m ^{2a} | Density separation (ethanol) | – | Visual inspection | Twice a month during 1 year | Herrera et al., 2018 |
| Gran Canaria coast | 36.3–4900 MPs/m ² | Density separation (ethanol + NaCl) | 10 | Stereomicroscope | Four during 2018 | Rapp et al., 2020 |
| Tenerife coast | 1,50–2509.66 MPs/m ² | – | – | Visual inspection | Every 5 weeks during 1 year | Reinold et al., 2020 |
| Tenerife coast | 2.0–2971.5 MPs/m ² | Density separation (NaCl) | – | FTIR Spectrometer | One - autumn 2018 | Álvarez-Hernández et al., 2019 |

^a These concentrations include microplastics (1–5 mm) and mesoplastics (5–25 mm), with a higher proportion of the latter than the former in Lambra.

et al. (2019) used an extraction method with very little pore size (0.7 µm) which can contribute to a higher detection of MPs than CEDEX.

Finally, also some beaches from the Strait-Alboran coast were analyzed by Godoy et al. (2020). The results obtained (Table 5) are much lower than those reported by other authors for Spanish beaches, but are twice the concentrations provided by CEDEX for Las Azucenas in autumn (between 10 and 16 MPs/kg), which is very close to Motril beach. Although being the same sampling season between the two studies, this area is very affected by the complex geomorphology of the coast of the province of Granada. The granulometry of the sediment is gravel and coarse sand type due to the proximity of the mountain to the coast, which makes it difficult for small and low-density particles to be retained, as they move too easily between the pores and voids of the sediment. In addition, the current system is complex due to the proximity of the Strait of Gibraltar, where water from the Atlantic enters and

mixes with the Mediterranean.

Regarding the sizes of the MPs, CEDEX focuses on quantifying for each beach, the proportion of MPs between 1 and 5 mm and between 0.05 and 1 mm in both autumn and spring. Fig. 4 shows the results obtained for all the beaches except Lambra, as the quantities of MPs obtained here were much greater than on any other beach and did not allow the data to be displayed correctly on the graph. In addition, only the results from the most recent campaign (2019) have been represented in order to make the interpretation easier. In spring, the beach of Lambra obtained 759.89 MPs/kg between 0.05 and 1 mm and 121.4 MPs/kg between 1 and 5 mm. In autumn, the concentration of MPs was lower and 170.9 MPs/kg were found between 0.05 and 1 mm and 23.02 MPs/kg between 1 and 5 mm.

On all the beaches, except for Itzurun, Oyambre and La Pineda in the spring season, MPs smaller than 1 mm are more abundant, a fact that

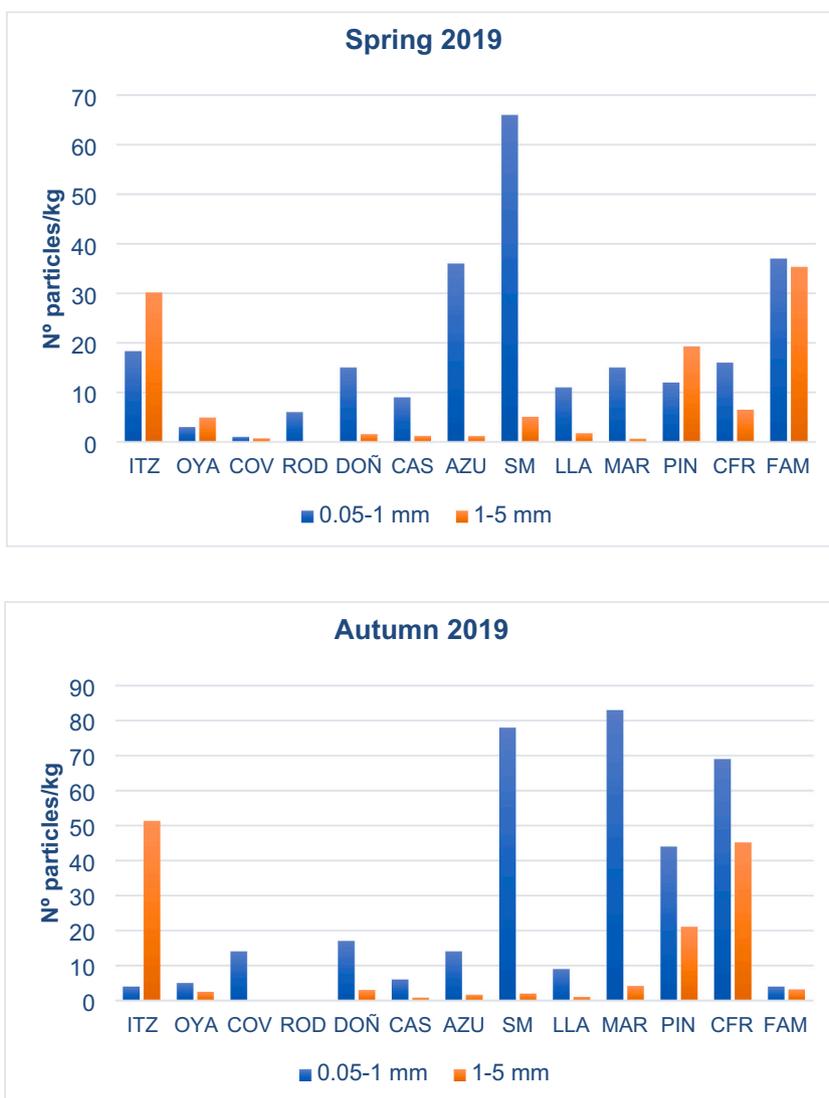


Fig. 4. Classification of MPs by size on the different Spanish beaches in spring and autumn. Source: modified from CEDEX, 2019.

coincides with that provided by many other studies in both salt and freshwater environments. According to Kooi and Koelmans (2019), the most common and probable distribution of MPs sizes in the studies carried out is usually between 20 μ m and 5 mm, with an inverse relationship between particle size and concentration, which means that the more the size increases the more the concentration decreases. In comparison with previous years, in 2017 it was remarkable the case of Famara, which accounted for 99% of MPs bigger than 1 mm, both in spring and autumn. However, in the rest of the beaches analyzed, almost no MPs of this size were detected. The same situation occurred in 2018 except for La Pineda in autumn, when half of the MPs found were between 1 and 5 mm. In general, it can be concluded that the main tendency over the years and in almost all beaches is to find more MPs smaller than 1 mm. Furthermore, the results of CEDEX are in accordance with other studies for Spanish beaches (Bayo et al., 2019; Herrera et al., 2018; Simon-Sánchez et al., 2019). In the case of Itzurun, where more MPs bigger than 1 mm were found, a report from OSPAR (Convention for the Protection of the Marine Environment of the North-East Atlantic) (2015) analyzed some macrolitter from beaches close to Itzurun and concluded that plastics bigger than 2.5 cm were found among the main objects collected. Consequently, if these objects do not degrade enough, can produce MPs of great sizes.

Finally, with regard to the most common forms of MPs found on the Spanish beaches, Fig. 5 shows clear differences between the autumn and spring campaigns, and also between years. In spring, there is a clear increase in the proportion of pellets over the years, from 7% of the total in 2017 to 50% in 2019. This type of particles appeared in the spring campaign from 2019 on 12 of the 14 beaches (all except the beaches of Rodas and Castilnovo) while in autumn were detected in 10 beaches (all except the beaches of Covas, Rodas, Doñana and Castilnovo). Concentrations were particularly high in Famara and Lambra from Canary Islands, and in Cal Francés and La Pineda, from Catalonia. On the contrary, the autumn campaigns do not exhibit a clear trend, being the most significant findings the high proportion of expanded polystyrene (EPS) in 2017 and 2019 (49% and 25%, respectively) and the high proportion of pellets in 2018 (68%), most of them found in La Pineda. In 2017, EPS were detected in almost all beaches in high proportions.

On the one hand, the presence of pre-production pellets on the beaches from Catalonia might be related to losses from the plastics industries, which are close to the coast, an event that has already been demonstrated by other authors (Karlsson et al., 2018). In the case of the beaches sampled in Canary Islands, thousands of kilometres away from any direct source of input, is related to the oceanic gyre circulation and the Canary Current (Rapp et al., 2020). The results obtained by CEDEX

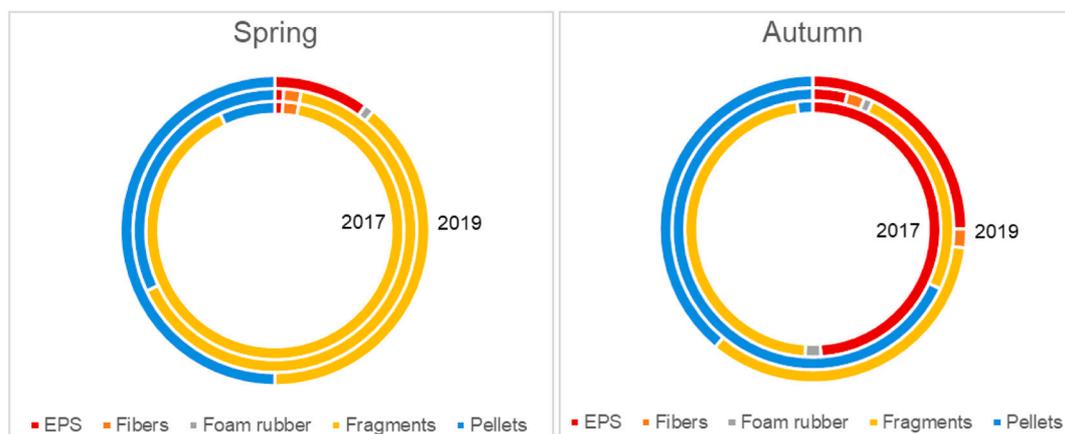


Fig. 5. Distribution of MPs found on Spanish beaches by morphology, from 2017 to 2019. Source: CEDEX, 2017, 2019; CEDEX, 2018.

in Canarian beaches are similar to those presented by Herrera et al. (2018), who found that 11.7% and 44.3% of MPs in Lambra and Famara, respectively, were pellets. Also, Rapp et al. (2020) reported that 13.7% of MPs found in beaches from Gran Canaria were pellets. On the other hand, Bayo et al. (2019) found a higher proportion of fragments (59.4%) than pellets (5%) in sediments from Menor Sea, probably because of the distances from plastic industries. In addition, Menor Sea is a highly touristic area with great anthropogenic pressure, so it is common to find fragments of MPs from the various leisure and commercial activities that take place there. Also, Simon-Sánchez et al. (2019) found that approximately 90% of MPs found in riverbed sediments and sandy beaches from the estuary of the Ebro River were fibers, and the remained percentage was mainly divided in fragments and films. The authors explained this fact by the proximity of two wastewater treatment plants located 27 and 14 km from the Ebro estuary, that discharge directly into the river. Both plants use a treatment that is not able to remove all the textile fibers from laundry.

Finally, the presence of foam elevated in some cases in the beaches analyzed by CEDEX could be explained by the fact that most of the floating macroplastic in the seas and oceans are polystyrene foam from diverse food and commercial packaging, and swimming kickboards that split into MPs (Eriksen et al., 2014).

3.2. Microplastic pollution in sea surfaces

Some works have evaluated the concentrations of floating plastic in sea surfaces in Spain. Cozar et al. (2015) provided the first study about the magnitude of the plastic pollution in the surface waters of the Mediterranean Sea in Spain. A total of 28 sites were sampled using neuston net for periods of 15 min. Authors found MPs in all surface net tows, with the majority of items being millimeter-sized fragments of larger rigid objects. The average concentration measured was around 0.25 items by m^2 . The biological richness and concentration of economic activities in the Mediterranean Sea, the effects of plastic pollution on marine and human life are expected to be particularly frequent in this plastic accumulation region (Cozar et al., 2015). Other paper that examined the plastic particles in sea surfaces of Spain was the work of Gago et al. (2015). These authors analyzed the presence and distribution of plastic particles in waters of the Cantabrian Sea and in the NW Spanish Atlantic coast during spring of 2013 and 2014. A total of 1463 plastic microparticles were counted in the 41 neuston samples that were collected. The average concentration measured was 0.03 and 0.18 items by m^2 for samples collected in 2013 and 2014, respectively. Finally, Herrera et al. (2020) performed the first evaluation of MPs in the Macaronesian region. Authors found very variable results in function of sampling area, for example, they found average concentration of MPs of

0.02 items by m^2 in Los Gigantes (Tenerife, Canary Islands) and 1.00 item by m^2 in Las Canteras (Gran Canaria, Canary Islands).

3.3. Microplastic pollution in wastewater treatment plants (WWTP)

Wastewater treatment plants receive millions of cubic meters of water per day through the sewage system, which come from domestic wastewater and storm water, with these plants being one of the main sources of environmental pollution (Prata, 2018; Mahon et al., 2016; Browne et al., 2011). The treatment plants receive water with a high content of MPs, which source is mainly from the plastic industry, cleaning products, personal care products, tyres, paints, cosmetics or synthetic clothing (Kay et al., 2018).

Currently, the wastewater treatment plants normally comprise a primary, secondary and sometimes tertiary treatment to prevent the emission of a high number of pollutants along with the final effluent to the environment quite effectively (Gatidou et al., 2019). Without focusing on MPs, it is estimated that approximately 90–95% of these pollutants can be retained after treatment (Michielssen et al., 2016; Blair et al., 2019; Blasing and Amelung, 2018). In the case of MPs, in primary treatments about 72% of MPs are removed, mostly larger MPs. Secondary and tertiary treatments removed 88% and 94% of MPs presented in the influent, respectively (Iyare et al., 2020). But despite these high retention values, the remaining fraction still represents a huge number of MPs that are discharged into rivers and oceans, causing a major environmental problem (Conley et al., 2019; Sun et al., 2019). Most of the MPs removed during the wastewater treatment are retained in the sludge (Ngo et al., 2019). This large amount of MPs in the sludge represents another environmental problem, since this sludge is often used as fertilizer in agriculture due to its high organic and inorganic content (Murphy et al., 2016; Gherghel et al., 2019; Singh and Agrawal, 2008), and the MPs remain in the soil longer than the nutrients, being a threat to soil ecosystems (De Souza Machado et al., 2018). In short, both the reuse of the final effluent and the sludge causes the reintroduction of MPs into the environment, constituting an environmental threat (Gatidou et al., 2019). Despite this problem, few studies have been carried out on MPs in effluents, influents and sludge from wastewater treatment plants (Wang et al., 2020; Liu et al., 2019), as is the case with industrial wastewater treatment plants, being even more alarming due to their large source of pollution (Lechner et al., 2014).

Although wastewater treatment plants can retain a high amount of MPs, they are not designed to remove these materials. In addition, little information is available for the actual removal of MPs in different stages of the wastewater treatment plants and there are no specific studies that analyze new processes to extract and characterize MPs in this type of installations. Despite the little information, the studies that exist on the

analysis of MPs removal of the different technologies in wastewater treatment plants, have showed that one of the most effective treatments is the membrane bioreactor that produces a lower amount of MPs in the final effluent, with a removal rate between 82.1 and 99.9% (Iyare et al., 2020). In any case, in order to reduce the number of MPs in the final liquid effluent and sludge, it would be necessary to improve the conventional technology of wastewater treatment plants and design optimal operations for MPs removal (Katyal et al., 2020).

3.3.1. Effluents

The wastewater treatment plants, as mentioned above, have a large capacity for the removal of MPs, but as a large amount of water is discharged every day, a large amount of MPs is discharged at the same time. Table 6 presents some of the urban and industrial wastewater treatment plants located in Spain, together with their treatment capacity and the concentration of MPs in both primary and secondary effluents. The MPs abundance gradually decreased from primary treatment to secondary treatment. Regards the abundance, it is different in function of a variety of complex factors such as the situation of the treatment plant, since it influences the type of population served, the purification process, the wastewater sources (municipal or industrial), the season of the year or the rainy season (Liu et al., 2021; Long et al., 2019; Sol et al., 2020). The concentrations or abundance was also affected by the sampling and analysis methods (Liu et al., 2021). Therefore, direct comparison of information available for the different wastewater treatment plants is very difficult.

In Spain, the plant located in Valencia can treat about 40,000 m³/day, and it releases about 3.0 · 10⁸ microparticles/day along with the secondary effluent, so that the removal efficiency, from primary effluent to secondary effluent, was 75% (Bretas et al., 2020). Other wastewater treatment plant located near Madrid and that discharges effluent to the Henares River was estimated to discharge 1.1 · 10¹⁰ microparticles/day (Edo et al., 2020). Finally, the Cartagena wastewater treatment plant, discharging to Mediterranean Sea was calculated discharges around 6.7 · 10⁶ microparticles/day (Bayo et al., 2020). This large amount of microparticles discharged per day, around 10⁶–10¹⁰, was also reported by other researchers for similar plants (Lares et al., 2018; Liu et al., 2019; Magni et al., 2019; Mason et al., 2016).

The abundance of the MPs in influent and effluent ranged from 3.2 to 1567 MP/L and from 0.31 to 328 MP/L, respectively. It results in a decrease of abundance in the range of 46–98%. It agrees with results reported previously in the literature (Liu et al., 2019; Talvitie et al., 2017).

Comparison of the concentration of MPs in treatment plants in different countries can be found in the review by the authors Habib et al. (2020). The concentration of the different plants located in different countries varies significantly, depending on numerous variables such as

the concentration of MPs in the influent, the concentration of MPs that end up in the sludge, the different treatments (primary, secondary and tertiary), etc. For example, in Finland a concentration range of 0.005–13 MP/L was found, in the USA 0.02–90 MP/L, in England 0.25–3 MP/L, in Australia 0.28–1.54 MP/L, in Germany 0.1–59 MP/L, in the Netherlands 9–91 MP/L, in France 14–50 MP/L, in Russia an average of 70 MP/L, in Turkey 4.11–7.02 MP/L, in Denmark an average of 54 MP/L, in Canada 0.5 MP/L, in Poland 0.028–0.96 MP/L and in China 0.59–9.04 MP/L.

Regards shape of MPs found in Spanish wastewaters plants, fibers and fragments were the most dominant fractions. Microfibers are mainly originated from domestic washings. It has been estimated that a single garment when washed can produce around 1900 fibers (Browne et al., 2011). The fragments can be originated from personal care products (Liu et al., 2021). The study of Bayo et al. (2020) also showed a high abundance of film shape perhaps due to the proximity of the Cartagena's plant to agriculture greenhouses.

Fig. 6 shows the average size of the microparticles found in the primary and secondary effluent and in the sludge. In the primary effluent 42 ± 12% corresponds to the size range 150–500 µm, while only 6 ± 10% corresponds to the size range 2–5 mm. In the secondary effluent 43 ± 15% corresponds to the same size range 150–500 µm, while only 4 ± 6% corresponds to the size range 2–5 mm. Finally, in the sludge the majority fraction with 48 ± 15% corresponds to the size range 150–500 µm. Similar results were obtained by other authors globally (Lares et al., 2018; Magni et al., 2019).

Bayo et al. (2020) also studied the colours of the microparticles found in the wastewater treatment plant of Cartagena, being beige the most common colour of these microparticles (36.9%), followed by white with 23.6%, blue with 7% and finally green with 3.9%. Other authors (Bretas et al., 2020) took samples from the wastewater treatment plant in Valencia, where the predominant particles colour was blue with 48%, followed by black with 25% and finally red with 16%. These big differences are due to the manufacturing processes of the different textile fibers mainly, as different chemical compounds are applied.

The most abundant polymers in the fragments are mainly polyethylene and polypropylene, while in the fibers are polyesters and polyethylene terephthalate (Bretas et al., 2020; Bayo et al., 2020).

The five forms detected in those particles were fragments, fibers, film, beads and foam. Fragments and fibers were the predominant groups, 36 ± 9.5% of fragments and 29 ± 18.8% of fibers were found, whereas the least abundant forms were film with 19 ± 13%, followed by beads with 16 ± 3.5% and finally foam with 1 ± 0.9%, as can be seen in Fig. 7. Van den Berg et al. (2020) estimated an 83% of fragments and 14% of fibers in the Valencia wastewater treatment plant. Other authors (Edo et al., 2020), determined a 60% of fragments and 28% of fibers in the wastewater treatment plant of Madrid, while Sol et al. (2020)

Table 6

Information of the WWTPS and industrial wastewater treatment plants (IWWTPs) in Spain and MPs abundance in effluents.

| | Facility capacity (m ³ /day) | Population | Treatment processes | Influent (MP/L) | Effluent (MP/L) | Reference |
|--------------|---|------------|-----------------------------|------------------|-----------------|----------------------|
| WWTP | | | | | | |
| Valencia | 40,000 | – | Pri, Sec | 11.1 | 2.8 | Bretas et al. (2020) |
| Madrid | 28,400 | 300,000 | Pri, Sec (A2O) | 171 ^a | 10.7 | Edo et al. (2020) |
| Cartagena | 35,000 | 21,000 | Pri, Sec (activated sludge) | 3.2 | 0.31 | Bayo et al. (2020) |
| Cádiz-1 | 50,000 | 280,274 | Pri, Sec, Ter | 378 | 7 | Franco et al. (2020) |
| Cádiz-2 | 111,790 | 223,580 | Pri, Sec | 586 | 18 | Franco et al. (2020) |
| Cádiz-3 | 36,363 | 72,726 | Pri, Sec, Ter | 275 | 8 | Franco et al. (2020) |
| Cádiz-4 | 1399 | 11,029 | Sec | 613 | 328 | Franco et al. (2020) |
| Cádiz-5 | 1306 | 6002 | Pri, Sec | 1567 | 278 | Franco et al. (2020) |
| IWWTP | | | | | | |
| Cádiz-1 | 203 | – | Sec | 87 | 24 | Franco et al. (2020) |
| Cádiz-2 | 83 | – | Sec | 119 | 16 | Franco et al. (2020) |

Pri, Sec, Ter refer to primary treatment (based on physical mechanisms), secondary treatment (based on biological mechanisms) and tertiary treatment.

A2O refers to anaerobic, anoxic, oxic biotreatment.

^a Primary effluent.

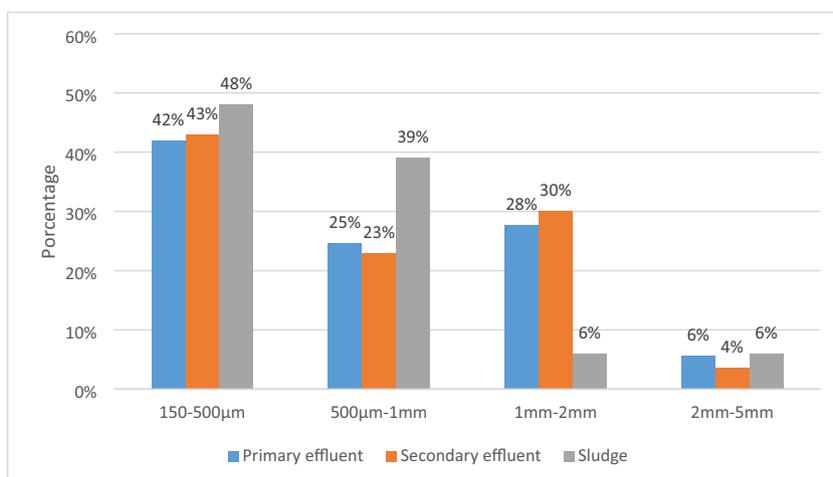


Fig. 6. Average microparticles size distribution for primary effluent, secondary effluent and sludge samples. Source: Bretas et al., 2020; Franco et al., 2020.

estimated a 34% of fragments and 57% of fibers in the same plant. These big differences are mainly due to the season in which the sample is taken, since in winter a greater use of the washing machines is made causing a greater amount of fibers in the winter seasons than in the summer ones (Browne et al., 2011).

3.3.2. Sludge

The MPs removed from the treated water are mostly accumulated in the sludge. The sludge can be managed by incineration, deposition in landfills or in soil application, being the latter the most used (Habib et al., 2020). In Spain, there is an extensive use of fertilizers and about 65% of the sludge from sewage treatment plants is used as fertilizer in Spain, while in Europe only 40% is used (Roig et al., 2012). Thus, in Spain there are studies on plastic contamination of agricultural soils (Zubris and Richards, 2005), but there is little research quantifying the power of MPs from sludge in soil contamination (Van den Berg et al., 2020).

Table 7 shows the number of MPs and microfibres found in the sludge of urban wastewater treatment plants in Spain. The highest concentration was found in the treatment plant from Madrid with 183 MPs/g of

dry sludge (Edo et al., 2020), while the lowest amount was found in a treatment plant in Cadiz with 5.2 MPs/g of dry sludge. Similar values are found in international studies, for example a water treatment plant in China was found to have a concentration of 22.7 MPs/g of dry sludge (Li et al., 2018) or in Chile an estimated concentration of 34.1 MPs/g of dry sludge (Corradini et al., 2019). In addition, in the reviews of Habib et al. (2020) and Iyare et al. (2020), a comparison of the microplastic concentrations in sewage sludge of different treatment plants in different countries can be found. For example, a microplastic abundance between 0.72 and 149 MPs/g (in wet weight) and between 1 and 170.9 MPs/g (in dry weight) has been reported from sewage sludge in the recompilation of studies of the interesting review of Iyare et al. (2020).

The application of sewage sludge causes an increase in the presence of MPs in agricultural soils. Van den Berg et al. (2020) analyzed agricultural soils without sewage sludge application and found 930 ± 740 MPs light density per kg and 1100 ± 570 MPs heavy density per kg, whereas soils with added duration sludge contained 2130 ± 950 MPs light density MPs per kg and 3060 ± 1680 MPs heavy density per kg. But this increase in concentration may be due to other non-quantifiable factors such as atmospheric transport (Allen et al., 2019).

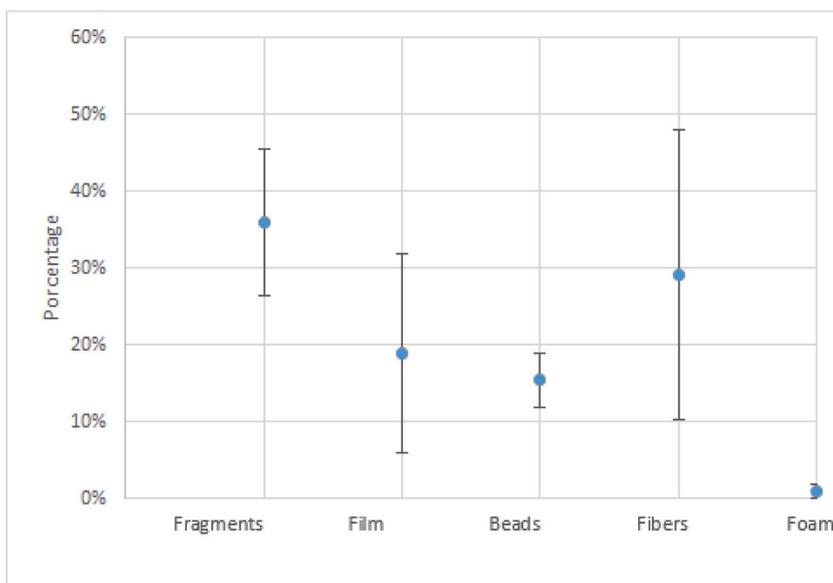


Fig. 7. Shape categories across the WWTP. Source: Bayo et al., 2020; Franco et al., 2020.

Table 7
Number of MPs and microfibers in sludge samples.

| Sludge sample | MPs concentration | Microfibers concentration | Reference |
|---------------------------|-------------------|---------------------------|----------------------------|
| | MPs/g dry weight | MFs/g dry weight | |
| Valencia | 112 | 105.6 | Bretas et al. (2020) |
| Madrid | 183 | 133 | Edo et al. (2020) |
| Valencia-Canet d'En Beren | 73 | – | Van den Berg et al. (2020) |
| Valencia-Sagunto | 4.83 | – | Van den Berg et al. (2020) |
| Valencia-Ontinyent | 21.86 | – | Van den Berg et al. (2020) |
| Valencia-Albaida | 22.65 | – | Van den Berg et al. (2020) |
| Cádiz | 5.2 | – | Franco et al. (2020) |

3.4. Microplastic pollution in freshwater environments and transitional waters

Much of the plastic waste that reaches the sea comes from the land, which is transported through the rivers. Overall, land-based sources contribute with about 80% of the waste that ends up in the sea, while activities such as fishing and shipping contribute with 20% (Kershaw, 2016). Rivers can transport between 1.15 and 2.41 million tonnes of plastic to the world's oceans each year (Lebreton et al., 2017). Because of this high contribution, rivers, lakes or estuaries are key areas to avoid pollution from plastics.

3.4.1. River

Simon-Sánchez et al. (2019) studied the MPs found in the surface waters of the Ebro river delta, this river discharges an average of 464 m³/s, so that the waters of the Ebro river contribute 2.14 × 10⁹ MPs/year to the Mediterranean Sea. In this investigation a concentration of 3.5 ± 1.4 MPs/m³ was estimated. Non-synthetic particles such as cotton or coal and different polymers were identified. The most abundant polymers were polyamide (24%), followed by polyethylene (16%), poly (methyl methacrylate) (acrylic, 12%), polyester (12%), polypropylene (8%) and polyacrylate (4%). In terms of morphology, mainly fibers, fragments and films were found. Regarding size, the most abundant size range was 200–500 µm with 25.5%, followed by 100–200 µm with 23%, 500–1000 µm with 16% and 50–100 µm with 14%. Coloured MPs were the most abundant, followed by white, black and finally transparent.

3.4.2. Estuaries

Estuaries can accumulate sediments, nutrients and pollutants for so long time, due to the low water speed in those zones, being a potential source of pollution for MPs both on land and at sea (Eisma, 2012).

Díez-Minguito et al. (2020) studied MPs and their transport to the estuaries in the Ría de Vigo whose most important river is the Miño. This river discharges an average of 100 m³/s in summer and 800 m³/s in winter (Sousa et al., 2014). The concentration of MPs in the estuary was measured estimating an average value of 0.64 MPs/m³. The concentration of MPs in the estuaries depends mainly on the meteorological and oceanographic conditions. The most abundant forms were fibers with a 56% presence, followed by paint sheets (15%), filaments (7%) and fragments (6%). In terms of colour, the most predominant was blue with 44%, followed by black (14%), transparent (14%) and red (12%). Finally, in this same research, the average size of the plastics found in Ría de Vigo was determined, with a presence of 38% in the range 2–5 mm, followed by 1–2 mm (21%), 1–0.5 mm (13%) and 0.5–0.3 (4%), while approximately 24% of the plastics identified were larger than 5 mm (Díez-Minguito et al., 2020).

Simon-Sánchez et al. (2019) studied the MPs found in the estuary of

the Ebro river delta and estimated a concentration of MPs of 2052 ± 746 MPs/kg of sediment. The most predominant size range was 200–500 µm with 30% abundance, followed by 500–1000 µm and 1000–2000 µm with 19% abundance in the two size ranges. Coloured MPs were the most abundant, followed by black, white and finally transparent.

3.5. Plastic pollution in biota

Investigations about ingestion of MPs by marine organisms have been extensively reported (Wright et al., 2013; Guzzetti et al., 2018; Fangzhu et al., 2019). The range of affected organisms including fish, turtles, seabirds, marine mammals and others is increasingly wide. MPs are often mistaken with food, causing impact on organisms that ingest them, for example, ingestion of MPs can produce stomach disruptions or altering other functions of organisms such as reproduction (Wright et al., 2013; Setälä et al., 2014). In addition, it is now believed that smaller plastic particles (nanoplastics) could even penetrate cells and tissues, where their effects could be much harder to predict. Currently, there is no Spanish or European legislation that regulates the presence of microplastics and nanoplastics (even smaller than the former) as contaminants in food. At the moment, there is no data on effects of MPs can cause when they enter in the food chain and are consumed by humans. How the body can or cannot absorb these elements and what is the effect of these by-products on respiratory, intestinal or digestive function, is not been established. This is, there is no information about what extent they accumulate or not in the tissues or if there is a safe level above which to start worrying. However, the World Health Organization (WHO) believes that this is an issue that must be studied in depth and that it is necessary to investigate not only the presence of microplastics in the environment around us, the water we drink or food that we take. The Spanish health authorities have already announced that once the investigation stage is completed and more solid conclusions are reached, they will implement all the risk management measures necessary to rule out any risk to consumer health. It should be also highlighted that the European Commission's Rapid Alert System for Food and Feed (RASFF)'s and the European Food Safety Authority's (EFSA) report data about the presence of these plastic fragments in a wide variety of human food items (Antao Barboza et al., 2018).

Following, in this section, a review of studies performed in Spain including ingestion of MPs by Spanish biota is reported.

3.5.1. Commercial fish

Ingestion by fish has been intensely confirmed. New works should focus on releasing of chemicals derived from MPs into organisms, the drive of MPs from the stomach to other organs and mainly on health effects. It is also important to explore the incidence of MPs linked to other factors such as the prevalence of prey and parasites (Collard et al., 2017).

Table 8 shows a summary of works published about MPs ingestion by fish in Spain.

One of the fish species most studied in Spain is bogues (*Boops boops*), a demersal to semi-pelagic fish species living in a variety of habitats from 0 to 350 m depth (Tsangaris et al., 2020). In Spain, the first study that reported the presence of MPs in the gastrointestinal tracts of bogue was that of Nadal et al. (2016). The authors analyzed a total of 337 fishes in the Balearic Islands (Mediterranean Sea) and reported that high percentage of them (average value of 68%) had MPs with an average abundance of 3.75 microplastic items per fish. They are high values if data are compared with those found previously in the English Channel by Lusher et al. (2013) or in Portuguese coast in the North Atlantic by Neves et al. (2015). These findings suggest that variations in MPs ingestion values potentially reflect environmental differences in sampling locations regards to different levels of waste discharges, legal protection regulations, maritime routes or anthropogenic pressure. According to study of Nadal et al. (2016) the filaments-type MPs were the higher MPs found in samples, with values ranging from 42% to 80% in

Table 8
Literature review of MPs ingestion by fish in Spain.

| Fish species | Frequency, % | Items per individual | Sampling location | Reference | |
|---|---|----------------------|---|--------------------------------|---------------------------|
| Bogues (<i>Boops boops</i>) | 60 | 4.89 | Cala Ratjada (Mallorca) | Nadal et al. (2016) | |
| | 42.22 | 4.68 | Cao Blanc (Mallorca) | | |
| | 80.43 | 3.69 | Espardell (Eivissa) | | |
| | 77.27 | 2.47 | Cala Tarida (Eivissa) | | |
| | 67.7 | 3.75 | Balearic Islands (mean values) | | |
| | 64.71 | 2.59 | Highly anthropized area (city of Barcelona) | Garcia-Garin et al. (2019) | |
| | 35.29 | 1.42 | Intermediate anthropized area (Blanes) | | |
| | 38.24 | 1.38 | Cap of Creus | | |
| | 46 | NA | Catalan coast (mean values) | | |
| | 16.67 | 0.33 | Balearic islands | | |
| | Galeus melastomus | 37.5 | 0.58 | Peninsular coast South | Rios-Fuster et al. (2019) |
| | | 46.1 | 1.96 | Central region | Tsangaris et al. (2020) |
| | | 47.0 | 1.77 | Northern coast of Catalonia | |
| | | 16.05 | 0.35 | Palma | Alomar and Deudero (2017) |
| | | 18.18 | 0.32 | Soller | |
| Striped red mullet (<i>Mullus surmuletus</i>) | | 16.80 | 0.34 | Balearic Islands (mean values) | Alomar et al. (2017) |
| | | NA | 0.92 | Palma | |
| | | NA | 0.39 | Port d'Alcúdia | |
| | | NA | 0.34 | Cala Ratjada | |
| | | NA | 0.28 | Santanyí | |
| | NA | 0.04 | Port d'Andratx | | |
| | 27.30 | 0.42 | Mallorca Island (mean values) | | |
| Dogfishes (<i>Scyliorhinus canicula</i>) | 4.2 | 1.0 | Galician coast | Bellas et al. (2016) | |
| | 20.8 | 1.20 | Cantabrian coast | | |
| | 20.8 | 1.20 | Gulf of Cádiz | | |
| Hakes (<i>Merluccius merluccius</i>) | 16.7 | NA | Gulf of Cádiz | | |
| Red mullets (<i>Mullus barbatus</i>) | 33.3 | 1.75 | Barcelona (Mediterranean coast) | Rodríguez-Romeu et al. (2020) | |
| | 11.1 | NA | Cartagena (Mediterranean coast) | | |
| | 13.9 | NA | Málaga (Mediterranean coast) | | |
| | 10.0 | NA | Mahón (Mediterranean coast) | | |
| | 20.0 | NA | Ciudadella (Mediterranean coast) | | |
| | 18.8 | 1.9 | Mediterranean coast (mean values) | | |
| | 50 | 1.48 | Catalan coast (NW Mediterranean) | | |
| | 15.24 | 0.21 | Western Mediterranean Sea | | |
| Sardine (<i>Sardina pilchardus</i>) | 58 | 1–2 | Northwestern Mediterranean Sea | Compa et al. (2018) | |
| | 14.29 | 0.14 | Balearic Islands | Pennino et al. (2020) | |
| | 23.08 | 0.44 | Peninsular Mediterranean coast | Rios-Fuster et al. (2019) | |
| | Anchovy (<i>Engraulis encrasicolus</i>) | 14.28 | 0.18 | Western Mediterranean Sea | Compa et al. (2018) |
| | | 60 | 1–2 | Northwestern Mediterranean Sea | Pennino et al. (2020) |
| 0.0 | | 0.0 | Balearic Islands | Rios-Fuster et al. (2019) | |
| Mackerel | 6.67 | 0.07 | Peninsular Mediterranean coast | | |
| | 78.3 | 2.17 | Canary Islands | Herrera et al. (2019) | |
| | <i>Trachurus mediterraneus</i> | 30.0 | 0.40 | Balearic Islands | Rios-Fuster et al. (2019) |
| 44.16 | | 1.22 | Peninsular Mediterranean coast | | |

function of sampling location. The highest occurrence of microplastic ingestion was observed in Espardell (80.43%) in Eivissa Island despite its strict ecological protection regime. Some years later, Garcia-Garin et al. (2019) collected 102 bogues from three areas of the coast of Catalonia (Spain) that present different industrialization degrees. MPs were discovered in 46% of the samples tested. The presence of MPs was greater in Barcelona (the area with a higher anthropogenic pressure). Most of the MPs ingested ranged from 0.1 to 0.5 mm, and the most common type of polymer ingested was polypropylene. Rios-Fuster et al. (2019) also analyzed bogues in Balearic Islands and the peninsular coast (South). These authors found lower frequency and items per individual in Balearic Islands. These results are very different according to previous study of Nadal et al. (2016), sampling locations. The distance to the coast in the sampling can be the reason to these differences (Rios-Fuster et al., 2019). More recently, Tsangaris et al. (2020) captured and analyzed a total of 884 bogues in different geographical areas of the Mediterranean Sea (Spain, France, Italy and Greece). MPs were found in 46.8% of fish samples, with an average number of items per individual of 1.17. Filaments were of the predominant form type, while polyethylene and polypropylene were the most common types of polymers. Again, significant differences were found between geographical areas of the diverse countries analyzed in terms of the frequency of occurrence, size, shape, colour and composition of the ingested MPs. These differences were mainly related to the degree of anthropogenic development in the

sampling sites. However, in the two sampling locations of Spain no differences were found in MPs frequency and items per individual among the different geographical areas analyzed. In addition, the results are in agreement with those of Garcia-Garin et al. (2019).

In Spain, the ingestion of MPs was also investigated in other fishes by different researchers. For example, Alomar and Deudero (2017) also analyzed a total of 125 *Galeus melastomus* individuals for microplastic ingestion around the two different locations of Balearic Islands (Mediterranean Sea). Results showed that 16.80% of the individuals had ingested MPs with an occurrence of 0.34 MPs items by individual and without significant differences between locations. With regards to type of microplastic items, the filament-type MPs of cellophane and PET composition were the most predominant. Frequency of microplastic ingestion of *Galeus melastomus* individuals in this study was higher than that previously reported by Anastasopoulou et al. (2013) in the Eastern Ionian Sea with a value of 3.2%. These results are in agreement with the general distribution and density of marine debris in the European seas since higher values have been reported in western seas than in central and eastern seas in the study of Pham et al. (2014). In the article of Alomar et al. (2017) a total of 417 striped red mullet (*Mullus surmuletus*) were analyzed with two main objectives, first quantifying the quantity and typology of ingested MPs and secondly analyzing the effect of ingested MPs on the enzyme response and cellular oxidative damage. The authors found that 27.30% of individuals presented MPs in their

stomachs and that the abundance of MPs ingested (mean of 0.42 MPs by individual) increased with the size of the fish and did not depend on the ground distance of the sampling. According to these authors this result showed the ubiquity of MPs in the marine environment. Also, [Bellás et al. \(2016\)](#) reported the microplastic ingestion in three fish species that were collected in the Galician coast, the Cantabrian coast, the Gulf of Cadiz and different points of the Spanish Mediterranean coast (Barcelona, Cartagena, Málaga, Mahón and Ciutadella). A total of 72 dogfishes (*Scyliorhinus canicula*), 12 hakes (*Merluccius merluccius*) and 128 red mullets (*Mullus barbatus*) were analyzed. The abundance of MPs in the different fish species was 15.3% for dogfish samples, 18.8% for red mullet samples and 16.7% of hake samples, with an average concentration of 1.56 MPs per individual. Also, important differences were observed between the locations of the sampling. The highest levels of MPs were found in red mullets from Barcelona (33.3%; 1.75 MPs by individual) while the lowest abundance was observed in dogfishes from the Galician coast (4.2%; 1.0 MPs by individual). As, it was indicated before, the area sampled in Barcelona is close to an important urbanized area with a very relevant commercial port, which may explain the observed differences in the ingestion values, as compared to other areas with less urban impact. [Rodríguez-Romeu et al. \(2020\)](#) analyzed the ingestion of plastic fibers in red mullet (*Mullus barbatus*), both on a temporary and geographical scale. Fibers were present in 50% of the digestive pathways of fish, with an average occurrence of 1.48 fibers per individual. With regard to temporal developments, in Barcelona, there was a 46% increase in fiber intake when comparing the data published in this work (year 2018) with the data published in 2007. In terms of the characterization of fibers by typologies, 57% were cellulose fibers and 31% PET fibers. If results of frequency are compared with those commented of [Bellás et al. \(2016\)](#) in the same area, higher data were found in the work of [Rodríguez-Romeu et al. \(2020\)](#), although similar items per individual were reported in both studies. In general, comparisons must be considered with caution because results can greatly depend in the methodology used in each case.

Commercial fishes such as sardine (*Sardina pilchardus*), anchovy (*Engraulis encrasicolus*), mackerel (*Scomber colias*) or horse mackerel (*Trachurus mediterraneus*) were also investigated. Two of the most captured and consumed pelagic species (those living near the surface) in the waters of the western Mediterranean are sardines and anchovy, two fish representing up to 39% of catches from this region of Mare Nostrum and whose biological importance is also crucial for the conservation of marine ecosystems, as they are among the prey most coveted by tuna, seabirds and cetaceans. Authors as [Compa et al. \(2018\)](#) evaluated the ingestion of MPs in sardine and anchovy in the western Mediterranean Sea. The authors examined the gastrointestinal tract of 210 individuals and found that 14.28 and 15.24% of sardine and anchovy, respectively, had ingested MPs and natural fibers. The authors also indicated that larger individuals with better physical condition are less likely to ingest MPs and natural fibers. PET fibers were the most common type of polymer found. Also, the recent research of [Pennino et al. \(2020\)](#) showed that 58% of sardines and 60% of anchovies caught in fishing grounds in the western Mediterranean had ingested MPs (the same area of the study of [Compa et al., 2018](#)). The differences between results of studies of [Compa et al. \(2018\)](#) and [Pennino et al. \(2020\)](#) could suggest that the distribution of MPs abundance in the water column is heterogeneous since different sampling methods were used ([Choy et al., 2019](#)). Also, the collection was performed in different seasons and years. In any case, anchovies had lower frequencies of ingested MPs than those caught in the Eastern and Central Mediterranean areas. For instance, [Kazour et al. \(2019\)](#) found that the 83.3% of European anchovies analyzed had ingested MPs in the Lebanese coast (Eastern Mediterranean Basin) and [Renzi et al. \(2019\)](#) found a 90% in the Adriatic Sea (90%). In the case of sardines, [Pennino et al. \(2020\)](#) reported that the individuals in worse bodily conditions are more likely to ingest these residues contaminated, while in the case of anchovies, the most likely specimens were those with a higher gonad somatic index (an indicator that measures the

weight of gonads, the sex glands, relative to the percentage of total weight, and which is often used to measure the sexual maturity of individuals) and were smaller in size. According to this research, the geographical areas where fish are most likely to ingest MPs are the Gulf of Alicante in the case of sardines and the Gulf of León and Ebro Delta in that of anchovies. In both species there is a positive relationship between the presence of parasites and the ingestion of MPs, which shows that these factors can affect both the health of marine species and that of human consumers. Other authors reported ingestion on MPs on mackerel. Particularly, [Herrera et al. \(2019\)](#) analyzed the presence of MPs in the gastrointestinal content of mackerel caught in the coastal waters of the Canary Islands. Of the 120 gastrointestinal fish tracts examined, 78.3% contained some type of MPs with most of them synthetic fibers with an average number of items per individual of 2.17. According to these authors, the high frequency of MPs in the gastrointestinal content of mackerel could be attributed to the fishing areas are close to urban areas in Canary Islands. Also, the previously mentioned work of [Rios-Fuster et al. \(2019\)](#) studied 197 gastrointestinal tracts of horse mackerel, sardine, anchovy and bogue. A total of 127 anthropogenic particles were identified in the gastrointestinal tract of 28% of the samples. Significant differences were found in the ingestion of anthropogenic particles between different species. Thus, for example, 43% of horse mackerels had MPs with mean values of 1.13 to 0.16 particles/individual, on the other hand, only 2.56% of anchovies had MPs and also at a concentration varying from 0.03 to 0.16 particles/individual. The authors also analyzed the differences between the capture areas and in this case the presence of MPs was similar in the two areas studied (Balearic Islands and the Peninsular Mediterranean coast). Particularly remarkable is the low ingestion of MPs in both areas by anchovies (*Engraulis encrasicolus*) especially if results are compared to those published by [Pennino et al. \(2020\)](#).

The high proportion of fish with MPs in their stomachs (for example, 33% in red mullets from Barcelona in the study of [Bellás et al., 2016](#)), is similar to the levels observed in pelagic fish from the North Sea and Baltic Sea (36.5%, [Lusher et al., 2013](#)) or the North Pacific (35%, [Boerger et al., 2010](#)) or the Central Mediterranean Sea (32.4% in Bluefin tuna, [Romeo et al., 2015](#)), and higher than the levels observed in pelagic and demersal commercial fish of the Portuguese coast (19.8%, [Neves et al., 2015](#)). On the other hand, lowest values reported (for example, 4.2% in from Galician coast in the study of [Bellás et al., 2016](#)) are in the same range as those reported by [Rummel et al. \(2016\)](#) for demersal fish from the North Sea and Baltic Sea (3.4%).

Although MPs ingestion by fish has been demonstrated in Spain, at present, it is difficult to understand the physiological effects of plastics in fish due to the lack of evidence on ingested MPs translocation from gastrointestinal tracts to other organs and to the absence of knowledge on MPs residence time in tissues ([Boerger et al., 2010](#); [Lusher et al., 2013](#)). Further studies are necessary to provide an appropriate understand of the magnitude of MPs pollution, its role as vector for pollutants, the trophic transfer and, consequently, its derived biological consequences.

Regards a possible explanation for the difference in microplastic ingestion in different species from the same area could be linked to feeding strategies and traits since previous studies have related them to microplastic ingestion ([Anastasopoulou et al., 2013](#); [Romeo et al., 2015](#); [Setälä et al., 2018](#)).

3.5.2. Birds

Seabirds have been usually evaluated as biomonitors of plastic pollution in Spain by means of the measurement of plastic ingestion. In order to obtain the first data on the ingestion of plastics by different species of seabirds, authors like [Franco et al. \(2019\)](#) analyzed a total of 159 seabirds of fifteen different species. A total of 26 birds (16% of individuals) and a total of 9 species (60% of total species) contained plastics in their organism. The frequency of appearance of plastics ranged from 0% (Razorbill) to 100% (species of the family

Procellariidae). The authors, based on different criteria, proposed the Common Guillemot and the Atlantic Puffin as more promising candidates as biomonitors of plastic waste in the ocean. Previously, in the study of Rodríguez et al. (2012), the Cory's shearwaters (*Calonectris diomedea*) were analyzed. Eighty-three percent of the birds contained MPs with an average 2.97 mg distributed in 8.0 pieces per bird. The authors found no significant relationships between body size or condition and plastic concentration. Also, Gil-Delgado et al. (2017) analyzed plastic residues in the faeces of European coot (*Fulica atra*), mallard (*Anas platyrhynchos*) and shelduck (*Tadorna tadorna*), different species of waterfowl, in five wetlands in central Spain. A percentage of 43.8% of the 89 samples of shelduck, 60% of the 10 samples of European coot and 45% of the 40 samples of mallard had plastic residues that were of two types, fragments and threads. These residues were attributed to traces of plastic objects used in agricultural fields surrounding the lakes where sampling was carried out. Differences between the abundance, the size and the typology of plastic residues found in the different species of waterfowl were not statistically significant. Finally, Delgado et al. (2020) analyzed the presence of plastic in nests of patiamarillas gulls (*Larus michahellis*) in the southeast of the Bay of Biscay and found that plastic residues in the nests of seabird colonies were abundant. In Ulia, 40% of the nests had some type of artificial material, although in all cases these wastes accounted for less than 5% of the nest surface. Of these nests, one had a piece of cloth, five had ropes and 20 had flexible packaging plastics.

3.5.3. Marine mammals

Hernández-González et al. (2018) was the first and exclusive record of the presence of MPs in the digestive tract of marine mammals of the Iberian Peninsula. This study analyzed a total of 35 samples of the stomach content of dolphin to quantify and describe MPs. The 100% of samples contained MPs with an average concentration of 12 MPs per sample. Most plastic items were microplastic fibers but authors manifested that they cannot guarantee possible MPs contamination during the collecting protocols. Also, some higher fragments and a cord were found.

3.5.4. Mussels

The work of Reguera et al. (2019) quantified the MPs present in mussels (*Mytilus* spp.) collected in two different areas of the coast of northern Spain. In addition, a comparison of two widely used methods was carried out to digest organic matter, acid digestion with nitric acid and basic digestion with potassium hydroxide. The average abundance of MPs observed in mussels digested with potassium hydroxide was significantly higher than that detected in mussels digested with nitric acid. Meanly, the average MPs present in the mussel samples collected in the Cantabrian Sea (2.55–2.80 MPs by gram of fresh weight) was slightly greater than that observed in the mussel samples collected in the Ria of Vigo (1.59–1.28 MPs by gram of fresh weight).

4. European and Spanish legislation about plastics and marine pollution

4.1. European strategies

4.1.1. The circular economy of plastics in Europe

In 2015 a package of measures on circular economy was published that indicated plastics as one of the priority areas to be developed and later, in 2018 the “European Strategy for Plastics in a Circular Economy” was published, focused on promoting the recovery of plastics to be able to reintroduce them in the production process in a profitable way. At the beginning of 2020, the European Commission presented the new action plan framed within the European Green Deal (The European Green Deal), a roadmap with different actions that should lead Europe to become the first climate-neutral continent in 2050 (European External Action Service, 2020). The strategy presents a vision for a smart,

innovative and sustainable plastics industry that creates growth and jobs in Europe and helps reduce greenhouse gas emissions and dependence on imported fossil fuels.

The European strategy sets out three main challenges to achieve the circular economy of plastics:

- Incorporate 10 million tons of recycled plastic in Europe by 2025;
- Make all plastic packaging on the European market reusable or recyclable by 2030;
- Achieve complete elimination of plastic waste abandoned in nature.

Although there are currently very valuable initiatives to promote innovation and cooperation between companies and sectors such as the Circular Plastics Alliance, it is the power of society whose demand from large corporations can precipitate the end of plastic as we know it, because today it has earned a negative image, especially among the younger generations.

4.1.2. Proposal to restrict the use of microplastics of European Chemicals Agency (ECHA)

In addition to the MPs that can be generated after the degradation of larger plastic materials in different ecosystems (secondary MPs), we must take into account MPs that are intentionally added to consumer products (primary MPs). In this regard, in January 2019 the European Chemicals Agency (ECHA) proposed a far-reaching restriction on the intentional use of MPs in concentrations above 0.01% by weight for products marketed in the EU/EEA (European Chemicals Agency, 2020). With this proposal, it is estimated to reduce emissions by 90% and avoid the release of 500,000 tons of MPs in the 20 years following its approval (European Chemicals Agency, 2020).

After the public consultation period, the Risk Assessment (RAC) and Socioeconomic Analysis (SEAC) Committees have adopted their draft opinions on the proposal. The consolidated opinion of RAC was adopted in June 2020 and of SEAC in December 2020. Both supported ECHA's proposal but made some recommendations for the European Commission to consider in the decision-making phase.

4.1.2.1. RAC's opinion. RAC's most important recommendations in this regard are:

- Biodegradable polymers: Although they are excluded from the restriction, the RAC believes it is necessary to establish more stringent criteria to have greater evidence of their possible biodegradation in the environment.
- Use of MPs as infill material in artificial grass pitches: Due to the lack of information on the effectiveness of risk management measures, the RAC proposes a complete ban after a transition period of 6 years.
- Definition of ‘microplastic’: ECHA proposes a lower size limit of 100 nm until more stringent analytical detection methods have been developed and validated. RAC recommends that this limit not be modified in the future as potential restrictions may be made in other ways, such as reviewing raw materials throughout the supply chain.

4.1.2.2. SEAC opinion. In this sense, the SEAC agrees with the costs and benefits that this proposal can generate in society since they consider the transition periods sufficient for companies. Although these vary according to the end use, in most cases they are between 4 and 10 years, except in the case of cosmetic products and detergents with microbeads, where no transition period is contemplated due to the voluntary cessation of their use by the industry. The SEAC emphasizes the irreversibility of contamination by MPs, since once released into the environment it is very difficult to eliminate them, so early action can be very beneficial for society.

SEAC also recommended a lower size limit of 1 nm for restricting MPs. It also considered that a temporary lower size limit of 100 nm may

be necessary to ensure that the restriction can be enforced by detecting MPs in products.

To control the release to the environment of infill material from artificial turf pitches, SEAC did not prefer any of the risk management options proposed by ECHA over the others.

From there, the Commission will consider the opinions and whether the conditions for the restriction are met. It will then prepare a proposal that the Member States can put to a vote in the REACH Committee, which will be followed by a scrutiny period from the European Parliament and the Council before the restriction measure can be adopted (European Chemicals Agency, 2020).

4.2. Spanish strategies

4.2.1. New normative proposal in Spanish legislation

4.2.1.1. Introduction. Last June, the Council of Ministers approved the Spanish Circular Economy Strategy (EEEC), in addition to the draft Law on Waste and Contaminated Soils, whose main objective is to offer an effective solution to the growing volume of waste and reduce the consumption of plastics. All this to respond to the European directives EU218/851 on waste and the EU219/904 on single-use plastics and which are aimed at complying with the objectives of the Circular Economy Action Plan of the European Commission (European Parliament, 2019).

The draft Law on Waste and Contaminated Soils includes for the first time in Spanish legislation limitations to single-use plastics, and also includes restrictions on their introduction into the market and information obligations to the consumer. In addition, and to reduce single-use plastic containers, a tax is established on them (MITECO, 2020a).

In order to promote a circular and low-carbon economy in Spain, this text reviews the current regulations on waste and contaminated soils to comply with the new objectives established in the community directives of the Circular Economy Package, as well as those derived from the single-use plastics directive. The text pursues two fundamental objectives: a general one of establishing measures aimed at protecting the environment and human health, by preventing and reducing the generation of waste and its adverse impacts on the environment, and by reducing the global impact the use of resources and the improvement of their efficiency; and another specific one, applicable to certain plastic products to prevent and reduce their impact on the environment, in particular the aquatic environment, and on human health (Directive (EU) 2018/851; MITECO, 2020a).

Together with this draft law, the Spanish Government has approved the Spanish Circular Economy Strategy (EEEC), "Spain Circular 2030", and a royal decree that improves the traceability and control of waste shipments, three key elements of the Framework Circular Economy that the Government wants to use as a lever for economic recovery after the health crisis of COVID-19.

In relation to single-use plastics, for the first time Spanish legislation includes limitations to certain single-use plastics, restricting the introduction into the market of some of them and establishing a tax to advance in the reduction of packaging of non-reusable plastic (MITECO, 2020a). It is a tax similar to the one that is intended to be implemented in other neighboring countries such as the United Kingdom or Italy.

Single-use plastic products subject to reduction include beverage cups, including their lids and stoppers, and food containers, such as boxes, with or without lids, used to contain food that is intended for consumption immediate, on-site or to go (Directive (EU) 2018/851; European Council, 2020).

For these products, a 50% reduction in their commercialization must be achieved in 2026, compared to 2022 and in 2030, that reduction should be 70%, also with respect to 2022. To meet these objectives, all the agents involved in the commercialization will promote the use of reusable alternatives or other non-plastic material.

Likewise, in many countries the production of single-use plastics is being restricted, but many times the legislation simply considers single-use plastics to those that have a thickness below a certain value. What this produces is that single-use objects continue to be produced, but with a greater thickness (and price). It does not contribute to reducing the amount of plastic waste.

On the other hand, the normative text reinforces the hierarchy of waste or, what is the same, the order of priority in the waste management options: prevention; preparation for reuse; recycling; another type of recovery, including energy recovery, and, as a last option, disposal. And it does so by urging the adoption of economic instruments and other incentive measures by the administrations as the fees on landfilling or incineration, the promotion of payment systems for generation and the use, within the framework of contracting, of public purchases to promote the use of products reusable and repairable and made of easily recyclable materials.

Complying with this order of priority, when managing waste, is key to turning the current system around. Thus, in 2017, the last year for which data are available, in Spain only 46.1% of municipal waste was recycled, in line with the European average (46.9%), which means that both Spain and Europe, there is room for improvement for the optimization of resources, especially in a context in which raw materials are increasingly scarce and expensive. In addition, with this, the generation of activity and employment linked to the circular economy is favored.

However, despite all the measures proposed and in accordance with information published by various environmental organizations and various media, in Spain most of the plastic packaging continues to end up in landfills, incinerated or disposed of in the environment. In fact, these media assure that between 70 and 80% of the waste found on the beaches is plastic, making Spain the second European country that dumps the most plastic into the Mediterranean. This indicates that the measures taken have not been sufficient being necessary to take more drastic measures beyond recycling or reuse.

4.2.2. Join to European Plastics Pact

Spain has also joined the European Plastics Pact, an initiative whose objective is to accelerate the transition towards the circular economy in the field of plastics, eliminate plastic waste from the environment, reduce the unnecessary use of plastic and bet on innovation in the reuse and recycling of plastic. At the moment, 12 other European countries have joined this initiative, in addition to Spain, the Netherlands, France, Denmark, Germany, Italy, Greece, Sweden, Portugal, Finland, Slovenia, Lithuania, Latvia as well as one 90 multinationals and associations, and it is expected that there will be more accessions.

This initiative sets four measurable objectives for the year 2025 and will act on all plastics used in packaging and single-use products placed on the market in the European Economic Area (European Plastics Pact, 2020; MITECO, 2020b). These four objectives are (MITECO, 2020b):

- Design all plastic packaging and single-use plastic products placed on the market to be reusable as much as possible, and in all cases to be recyclable by 2025.
- Move towards a more responsible use of plastic packaging and single-use plastic products, with the goal of reducing virgin plastic products and packaging by at least 20% (by weight) by 2025, with half of this reduction coming from an absolute reduction of plastics.
- Increase the collection, sorting and recycling capacity of all plastics used in packaging and single-use plastic products by at least 25% by 2025 and achieve a quality standard for the entire recycling process appropriate to the demand of recycled plastic.
- Boost the use of recycled plastics in new products and packaging as much as possible by 2025, and that companies using plastics achieve an average of at least 30% recycled plastics (by weight) in their range of products and packaging.

The Pact has been promoted by France and the Netherlands and is

inspired by the Ellen MacArthur Foundation's work on the New Plastics Economy.

The signatories undertake to support the Pact, recognizing their different roles in the value chain and their different responsibilities as plastic production and recycling companies; plastic consuming companies; collection and sorting companies; national governments of countries within the European Economic Area or other support organizations (civil society organizations, regional and local authorities, business associations, technology service providers, knowledge institutions, or Extended Responsibility Systems). The European Commission will participate as an observer (European Plastics Pact, 2020; MITECO, 2020b).

The Pact is linked to a global network of Plastics Pacts (including South Africa, Malaysia and Chile), led by the Ellen MacArthur Foundation in collaboration with WRAP (Waste and Resources Action Programme), helping to advance global efforts to combat pollution by plastic (European Plastics Pact, 2020; MITECO, 2020b).

5. Final considerations and conclusions

This review aims to provide an overview of the marine plastic pollution situation in Spain from different perspectives: sources of pollution, presence of MPs on beaches and in sewage and ingestion of these particles by organisms.

With regard to microplastic contamination on beaches in Spain, annual monitoring by the Spanish government shows contamination along the entire coast of the country, with particularly high concentrations in the Canary Islands and the Levantine-Balearic coast over the years. The percentage of pellets is increasing every year in general, whereas the presence of EPS fragments is significantly high in autumn campaigns. The main factors postulated for the distribution of these particles are marine currents and the geomorphological characteristics of the site studied.

Despite the effectiveness of MPs removal in wastewater treatment plants, a large amount of these pollutant particles is still being reintroduced into the environment through aquatic and terrestrial routes, due to the sludge produced in these plants. Average estimations of $6.7 \cdot 10^6$ and $1.1 \cdot 10^{10}$ MPs/day release with the effluent of Spanish wastewater treatment plants were reported. Also, rivers are other of the main transport routes for MPs.

The work also has confirmed that plastic contamination is causing stress in marine ecosystems and affecting fishery and aquaculture resources. Numerous works have found ingestion of MPs by Spanish marine organisms.

There are factors that promote the accumulation of marine litter in certain areas of Spain. In the first place, we can mention the high population density in coastal areas, a third of Spaniards live on the coast or in nearby areas, which makes these areas susceptible to the accumulation and dumping of waste in the sea. Mainly the Mediterranean coast stands out, where some of the main Spanish cities and the mouths of rivers such as the Ebro, the Turia, the Júcar or the Segura (another source of plastic discharges into the sea) are located. In addition, Spain was the second touristiest country in the world before the COVID-19 pandemic and there are studies that point to tourism as one of the main sources of marine litter. The topography and diversity of the seabed are the second factor that explains why Spain is conducive to the accumulation of plastics in its depths. Spain is the second deepest country in Europe, after Portugal, and is particularly worrying due to factors such as the large number of marine geological formations present and the high depth of its seabeds (more than 3000 m on average). In this way, canyons, escarpments and seamounts, conducive to the retention of plastics and other human waste, accumulate and channel the waste generated inland or on the coast to great depths. Especially vulnerable areas stand out due to their great ecological value, such as the area around the Balearic archipelago, where the largest marine national park in the Western Mediterranean (Parque Nacional de Cabrera) is located,

as well as the areas of Murcia and the Alboran Sea, very vulnerable due to its high biodiversity, the result of the confluence of Atlantic, Mediterranean and North African species. Other examples are the area of the escarpment that delimits the Cantabrian continental shelf (Galicia escarpment) and the surroundings of the island of Tenerife, due to the concentration of urban centers and the great slope that generates the rapid disappearance of the continental shelf, furrowed in turn by several canyons. These characteristics, combined with the great biodiversity that these habitats present, make Spain especially vulnerable to the accumulation of plastics and that its ecosystems may be threatened.

On the other hand, plastic pollution can affect key economic sectors in Spain, especially fishing and tourism. Polluted beaches can also discourage visitors, leading to reduced jobs and income and increased costs for cleaning beaches and ports. In some coastal areas, such as Almeria and Granada, there is a very significant presence of greenhouses with plastics used for agriculture.

The plastic pollution is a very important problem that requires the collaboration of everyone: governments, companies and individuals. At following, a set of actions and recommendations that would significantly reduce plastic pollution in marine environments are proposed.

In our opinion, the solution goes beyond measures such as recycling or reuse, and unquestionably involves limiting the use of plastics, mainly single-use plastics, and also drastically reducing their production globally. Regarding necessary actions at the international level, a legally binding agreement could be approved to eliminate plastic dumping in the oceans, including a monitoring and evaluation framework and a financial mechanism to support implementation. Similarly, international trade standards should be adopted for plastic waste.

In the specific case of the actions necessary at national level, national objectives should be established to eliminate the use of single-use plastics, for example, recently, single-use plastic has already been banned in the Balearic Islands, to use recycled materials and review the extended producer responsibility, for example, lower rates for recyclable packaging or for the use of recycled materials. Similarly, in Spain it is necessary to establish/improve an integrated waste management system that includes a more efficient classification of plastic waste. Also, education is a key part of starting to deal with this problem, as well as increasing the awareness campaigns addressing MPs.

Regarding the actions required by the industry, it should invest in innovation in recyclable alternatives, redesign the infrastructure of production processes and supply chains that allow the use of recycled materials and adopt a zero-waste policy to stop single-use plastic products.

For consumers it is recommended to avoid disposable products, store food in glass containers, avoid cosmetic products that contain MPs, buy products in bulk, this is without packaging (fruits, vegetables, cheeses, meats, fish, etc.), pay attention to the city's deposit and recycling procedures, in conclusion, to be responsible citizens avoiding single-use plastic items and disposing of all plastic waste properly.

CRedit authorship contribution statement

M.A. Martín-Lara: Conceptualization, Formal analysis, Investigation, Methodology, Supervision, Writing – original draft, Writing – review & editing. **V. Godoy:** Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **L. Quesada:** Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **E.J. Lozano:** Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **M. Calero:** Formal analysis, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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