



Plastic debris accumulation in the seabed derived from coastal fish farming[☆]

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ABSTRACT

In this study, we assessed plastic accumulation in marine sediments due to finfish aquaculture using floating net-pens. We studied plastic concentrations around three fish farms located at the Mediterranean coastline of Spain. The macroplastic categories and abundances were determined by video monitoring, detecting the majority of elements (78%), including ropes, nets and fibres, a basket trap and a cable tie, close to the facilities, which were not exclusively linked to fish farming but also to fishing activities. Concentrations of microplastics (<5 mm) ranged from 0 to 213 particles/kg dry weight sediment with higher values in sites directly under the influence of the fish farms. Most particles (27.8%) were within the size fraction from 1.1 to 2.0 mm and fibre was the most common shape with 62.2%. The Infrared spectroscopy analysis showed that PE and PP were the predominant types of polymers analysed. In addition, changes in the enthalpy of melting (ΔH_m (J/g)) and the degree of crystallinity indicate degradation of the microplastics analysed. This study shows that, in the studied fish farms, levels of microplastic pollution can be one order of magnitude lower compared to other areas suffering other anthropogenic pressures from the same or similar regions. Nevertheless, more research effort is needed to get concluding results.

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1. Introduction

Plastics have contributed to societal prosperity over the last decades, due to their advantageous properties and inexpensive way of production (Andrady and Neal, 2009; Avio et al., 2017). From 1950 to 2015, the global plastic production increased from 1.5 to 322 million tons per year (PlasticsEurope, 2015). However, their inappropriate handling along with their very low biodegradability is making plastic pollution become ubiquitous.

From 1950 to 2015, 6.300 million Mt of plastic waste have been produced, of which only 9% have been recycled, 12% incinerated, and 79% accumulated in landfill or the natural environment (Geyer et al., 2017). According to Jambeck et al. (2015), between 4.8 and

12.7 million Mt of plastic enter the seas and oceans yearly. The degradation of plastics in water is lower than on land, because of reduced UV exposure and lower temperatures (Gregory and Andrady, 2003). About 80% of plastics in the ocean come from land based sources (UNEP, 2016). Sea-based sources of plastic debris constitute activities like fisheries, maritime transport and marine industries, such as aquaculture (Cole et al., 2011). Marine habitats are especially sensitive to plastic pollution. Plastics can transport alien species via floating plastics (Gregory, 2009) or cause the death of wildlife as a result of plastic entanglement, ingestion or toxicity (Cole et al., 2011), and accumulate in organisms leading to trophic transfer (Wright et al., 2013).

Aquaculture is the fastest growing industry in food production (SOFIA, 2016). The development of this activity has been accompanied by the assessment of the interactions with the environment (Grigorakis and Rigos, 2011). Specifically, fish farming produces a large amount of organic waste (Sanz-Lázaro et al., 2011; Sanz-Lázaro et al., 2011). This extra input of food is leading to changes in

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biological communities (Sanz-Lázaro et al., 2015; Sanz-Lázaro and Marín, 2011), largely increasing the amount and biomass of fish in the vicinity to fish farms (Dempster et al., 2004). As a result, the fishing pressure close to the farms by artisanal and recreational fisheries is enhanced (Arechavala-Lopez et al., 2011).

Studies dealing with plastic debris derived from aquaculture activities in either the water column or the seabed are relatively scarce (Andréfouët et al., 2014; Hinojosa and Thiel, 2009). Plastic quantification, characterization and identification of its sources, are needed to establish specific measures to reduce plastic inputs to the environment (Veiga et al., 2016). However, this assessment is missing for many human activities, including aquaculture. This information will enable us to develop mitigation management strategies to promote the sustainability of aquaculture.

Microplastics (defined as particles below 5 mm, Arthur et al., 2009) are an emerging issue with increasing concerns about their impact in the environment and wildlife (UNEP, 2016; Lusher et al., 2017). When characterizing microplastics, approaches vary greatly and no standardized procedures have been accepted amongst scientists so far (see e.g. Rochman et al., 2017). However, following reviews of identification and quantification of microplastics, the current consensus focuses on the following criteria: size, morphological and physical characterization and the chemical identification (see e.g. Hanvey et al., 2017; Hidalgo-Ruz et al., 2012; Shim et al., 2017; Gago et al., 2016). In general, chemical identification is strongly recommended, as visually identified microplastics are often not confirmed to be plastics (Hidalgo-Ruz et al., 2012; Silva et al., 2018).

To date most studies about plastic debris and aquaculture concentrate on microplastics ingested by cultured animals (see recent review by Lusher et al., 2017). Only very recently, the contribution of aquaculture as a source of plastic debris has been studied in China (Chen et al., 2018). Thus, there is a lack of knowledge regarding the accumulation of plastics on the seabed originating from fish farming activities.

This study aims to improve this knowledge by: (1) quantifying the concentration of macro- and microplastics in marine sediments at three fish farms in the Western Mediterranean of Spain, and (2) identifying the type of polymer of the plastics to infer their sources. Following the above-mentioned rationale, we characterized microplastics into colour, shape and size classes. For a number of suitable particles, we used attenuated total reflectance (ATR) FTIR spectroscopy. To evaluate the degradation status of microplastics, we additionally performed a Differential Scanning Calorimetry (DSC). The performed analyses support our visual inspection and present a first approach on microplastic present in the fish farm sediments and their possible origin.

2. Material and methods

2.1. Study area

We sampled three fish farms culturing gilthead sea bream (*Sparus aurata* L.) and European sea bass (*Dicentrarchus labrax* L.) at the Western Mediterranean coastline of Spain (Fig. S1 of Supporting Information) that aimed to embrace the diversity of fish farms in this region of the Mediterranean. The first fish farm (FF1) was the oldest farm of the investigation (~21 years); it is located nearly 3 km away from the coastline with a depth of ~31 m and a production of nearly 300 t of fish per year. The second fish farm (FF2) has been active for 16 years, around 2.8 km away from the coast with a depth of 34 m and a production of ~1200 t per year. The third fish farm (FF3) has been 13 years in operation, located 6 km away from the coast, and had many other fish farming facilities in their proximity

separated by a distance of at least 1 km. The farm had a depth of ~37 m and a production of approx. 800 t per year. The main polymers used in the fish farms studied are polyethylene (for the nets) and polysteel (a mixture of polyethylene and polypropylene that floats in the water).

2.2. Experimental design

Each location (FF1, FF2, FF3) comprised each fish farm and its corresponding area of influence. At each location, four zones were sampled: the "Allowable Zone of Effects" (AZE), the Zone of Influence (ZI), and two different reference zones (RE1 and RE2). AZE represents an area in the immediate proximity of the fish farm cages. At AZE some exceedance of environmental standards is accepted, but not beyond a certain threshold so the ecological status is not irreversibly compromised (Sanchez-Jerez and Karakassis, 2012). ZI is a nearby zone inside the lease, located approx. 40–60 m from the closest cage. Each of the two different reference zones was approximately 300–500 m away from the concession limits. In each zone, three sites were chosen and, in each site, three replicates were taken. A scheme of the chosen sampling sites can be found in Fig. S3 of the Supporting Information.

2.3. Sampling methodology

For the macroplastics, a GoPro Hero 3 Camera® (San Mateo, USA; video resolution 1080p; minimum focal field 30 cm) attached to a metal structure with a frame of 1 × 1 m was gently lowered to the seabed and pulled up for approximately 1 min to minimize sediment resuspension. The camera was in video mode at all times and pictures were extracted later on for the analyses. Our experimental unit contained a sampling area of 1 m². This procedure was replicated 3 times on each site, so in each zone of every location an area of 9 m² was sampled. To determine plastic concentration in the sediment, a superficial sediment sample (approx. 10 cm depth; since the first cm of the surface is where most of the plastics are accumulated (Martin et al., 2017)) of approximately 20 × 20 cm was taken by a Van-Veen grab. Three random replicates per site were collected, obtaining 9 replicates per zone and in total, 36 replicates per location. For the extraction of microplastics, a sample of 100 mL of sediment of each replicate was frozen immediately. A subsample of 1 g of each replicate was used to estimate the water content. Another 300 mL from each zone (AZE, ZI, RE1, and RE2) of each farm (location) were taken to determine the grain size. To avoid contamination by other plastic materials during the sampling and storage process, only glass, wood and aluminium materials were used.

2.4. Sediment characterization

In the sediment of FF1, all grain size fractions were present and rather evenly distributed within the zones. In FF2, finer sand was present with grain sizes <0.25 mm (>90%). The sediment of FF3 showed mainly coarser sandy sediments with grain sizes between 0.5 and > 2.0 mm (>70%). An exception was reference zone 2 with less than 30% of coarser fractions and a predominant size fraction of <0.063 mm. For a detailed depiction of grain size, see Fig. S2 of Supporting Information. The sediment in the sampled locations are carbonated sediments with total organic carbon, total nitrogen and total phosphorous mean contents that range between 0.8 and 2.2, 0.035–0.17 and 0.008–0.9% of dry weight, respectively (Aguado-Giménez and García-García, 2004; Holmer et al., 2007). In many cases, the sampled sediments generally contained debris of *Posidonia oceanica*, which are a highly recalcitrant form of organic

matter that can be confounded with conventional plastic fragments.

2.5. Sample treatment

The 1 g subsample of sediment was used to estimate the water content by drying it during 24 h at $\sim 80^\circ\text{C}$. This was used to calculate the actual dry weight of each subsample of sediment for the extraction of microplastics (ISO11465, 1993). To extract the microplastics from the wet sediment a slightly modified density separation method (Klein et al., 2015) was used. For this, the 100 mL, which represent a sample size similar to other studies (Hidalgo-Ruz et al., 2012), of wet sediment were mixed with 250 mL of saturated NaCl solution (~ 365 g of NaCl per litre), stirred for 3 min and left untouched overnight. The next day, the supernatant water was vacuum-filtered through a glass fibre filter GC (nominal pore size 1.2 μm , diameter 47 mm). Then, the filters were placed in a mixture (1:3) of hydrogen peroxide (30%) and concentrated sulphuric acid (Merck, Darmstadt, Germany) and incubated overnight to destroy other, natural materials attached to the filters (Klein et al., 2015). The filter was checked visually so no traces of plastics remained attached to it. The solution was poured into 150 mL of distilled water, filtered again through a new filter and rinsed by adding another 150 mL of distilled water.

During the lab procedure contamination was minimized by covering all openings containing samples or filters with aluminium foil, working with materials made of metal and glass and wearing 100% cotton coats. Brand new filters (either wet or dry, depending on the corresponding state of the processed filter carrying the sample) were exposed to the air while working with the samples. These filters were used as quality controls to evaluate possible cross-contamination during the processing of filters in the laboratory (Woodall et al., 2015).

2.6. Analyses

The macroplastics recorded by the camera and caught by the Van-Veen grab were categorized by their shape in the following five categories: rope, net, cable tie, basket trap and plastic bag. The microplastics were identified following the visual inspection criteria of the Marine & Environmental Research Institute, Main (Marine and Environmental Institute, 2015). Microplastics were separated from the filter with tweezers and stored in glass Petri dishes, pictures were taken, and the particles were categorized by colour, size and shape using a zoom stereomicroscope (7–45 \times). Each filter was visualized during approx. 15–20 min. To determine sizes of macro- and microplastics the longest edge of the particles/item were measured, pictures were analysed with the image-editing program ImageJ (Rasband, 2015) to accurately measure the size of each particle/item.

The composition and degradation status of the visually identified microplastics were assessed through Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR) and Differential Scanning Calorimetry (DSC). Due to the size restrictions, only the plastics that had a minimum size of at least 2 mm could be analysed constituting a relatively low percentage out of all the plastics sampled. Samples of polyethylene (PE) (CAS 9002-88-4), low density (LDPE), film, thickness 0.05 mm, L 1 m and polypropylene (PP) (CAS 9003-07-0), film, thickness 0.05 mm, L 1 m, grade heat sealable were obtained from Sigma Aldrich (Saint Louis, MO, USA) as reference materials.

2.6.1. ATR-FTIR analysis

Plastic particles were identified according to their polymeric-specific IR spectra and compared with reference materials. The

ATR-FTIR spectra of the samples were collected by using a Bruker Analytik IFS 66 FTIR spectrometer (Ettlingen, Germany) equipped with a DTGS KBr detector and a Golden Gate Single Reflection Diamond ATR accessory (incident angle of 45°). Spectra were recorded in the absorbance mode from 4.000 to 600 cm^{-1} ; using 64 scans and 4 cm^{-1} resolution and corrected against the background spectrum of air. Two independent spectra were obtained for each sample. To ensure that the acid treatment, which was performed during the plastic separation, did not influence the IR absorption bands of the studied polymers, spectra of control polymers were recorded.

To evaluate the degradation status of the samples, the carbonyl index (I_{co}) was calculated as the absorbance of the carbonyl group (band in the 1780–1600 cm^{-1} region) relative to the absorbance of the methylene group (band in the 1490–1420 cm^{-1} region). The absorbance of the carbonyl group increases with the ageing of the material, due to the effect of UV radiation on ketone, carboxylic acid and ester functional groups of the material (ter Halle et al., 2017).

2.6.2. Differential Scanning Calorimetry (DSC) analysis

DSC tests were conducted by using a TA DSC Q-100 (New Castle, DE, USA) under inert nitrogen atmosphere. The samples were introduced in aluminium pans (40 μL) and were heated from 25 $^\circ\text{C}$ to 220 $^\circ\text{C}$, then cooled to -90°C and finally heated again to 220 $^\circ\text{C}$, being always the heating/cooling speed of 10 $^\circ\text{C}/\text{min}$. The crystallization and melting parameters were determined from the first heating event. Melting temperature (T_m) was used to identify the material samples and the % of crystallinity was used to evaluate the degradation status, based on the equation described in the supplementary material.

2.7. Data analysis

An asymmetrical analysis of variance (ANOVA) was performed. We tested whether the microplastic concentration differed among locations and zones and if interactions existed between locations and zones. The experimental design integrated three factors: *zone* (AZE, ZI, RE1, RE2), *location* (FF1, FF2, FF3) and *site* (three different sites per zone). The factor *zone* was considered fixed, while the *location* and *site* were considered random. Zone was orthogonal with location and site nested within the interaction *location* \times *zone*. The following *a priori* comparisons were made: RE's vs AZE, RE's vs ZI and RE1 vs. RE2.

To test the homogeneity of variances, Cochran's test was used, and when necessary, data were transformed by square root to meet this parametric assumption. When the main effects or the interactions among effects showed significant differences, a Student-Newman-Keuls (SNK) pair-wise test was performed to determine which level/s were different within each factor. All statistical tests were conducted with the software R (v. 2.15.0) using the statistical package GAD (Sandrini-Neto and Camargo, 2015) and a significance level of $\alpha = 0.05$. Data were reported as mean \pm standard error (SE) unless stated otherwise.

3. Results

3.1. Macroplastic accumulation

In total, 9 items of macroplastics have been identified. The macroplastics found can be classified into five categories: rope, net, basket trap, plastic bag and cable tie. Ropes were the most frequent, taking up over 30% of all found items. The largest item found, was a net measuring 127.66 cm within the observed frame. The majority (78%) of macroplastics occurred in AZE and ZI, representing the following categories: rope, net, octopus cage and cable tie. In the

Table 1
Location, type and size (cm) of all detected macroplastics along the study area.

Zone	Location	Type of plastic	Size (cm) ^a
Macroplastics identified by picture or caught with grab			
AZE	FF1	rope	69.17
AZE	FF1	basket trap	26.36
AZE	FF3	rope	66.89
ZI	FF2	rope	34.53
ZI	FF2	net	127.66
ZI	FF2	net fibre	11.59
ZI	FF3	cable tie	5.36
RE2	FF1	plastic bag	11.87
RE2	FF2	plastic bag	22.15

^a Longest edge of found item was measured.

reference zones, only plastic bags were found (Table 1).

3.2. Assessing cross-contamination of microplastics

The filters, which were exposed to the laboratory environment as quality control to evaluate the level of contamination, showed 0.54 particles per filter on average. Based on these relatively low values, due to the measures to avoid cross contamination in the laboratory, we assumed no substantial contamination derived from the processing in the laboratory and decided not to include these values for the calculations of microplastic concentrations in sediments.

3.3. Microplastic concentrations and properties

Microplastic abundance in sediments ranged from 0 to 213 particles/kg dry weight sediment. A higher concentration of microplastics was found in AZE (68 ± 9.6 particles/kg) compared to both, ZI (32 ± 5.3 particles/kg) and the reference zones RE1 and RE2 (13 ± 1.3 particles/kg) (Fig. 1, Table 2). There was no significant difference between the zones ZI and RE1+RE2, as well as between the reference zones RE1 and RE2. The different locations showed similar values for microplastic. But, FF1 and FF2 showed statistical differences for the interactions of location and zones, showing a difference between ZI and the reference zones (Table 2).

The size distribution of microplastic particles showed similar patterns for all farms and locations (Fig. 2). In general, the highest

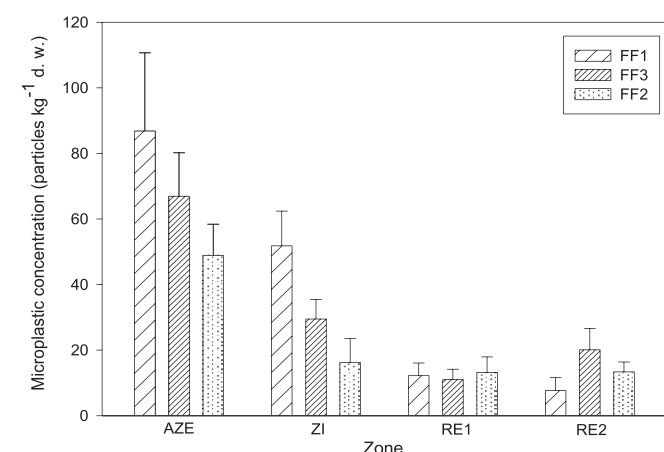


Fig. 1. Concentration of microplastics in the sediment of the four zones (Allowable Zone of Effects, AZE; Zone of Influence ZI; Reference zones; RE1 and RE2) from the different Fish Farms (FF1, FF2, FF3). Values indicate the mean +SE, each mean was calculated by pooling three sites of which each had three replicates (n = 9).

Table 2
Results of the analysis of variance (ANOVA) with the three factors, location = loc (FF1, FF2, FF3), zone (AZE, ZI, RE's) and site for the concentration of microplastics. Significant effects are shown in bold.

Source of variation	df	MS	F	P
RE's vs. AZE	1	338.20	102.48	0.01
RE's vs. ZI	1	87.70	2.47	0.26
RE1 vs. RE2	1	0.01	0.00	0.97
Location = Loc	2	13.20	1.71	0.20
(RE's vs. AZE) x Loc	2	3.30	0.43	0.66
(RE's vs. ZI) x Loc	2	35.50	4.61	0.02
(RE1 vs. RE2) x Loc	2	5.20	0.68	0.52
Site (Zone x Loc)	24	7.70	1.22	0.25
Residuals	72	6.30		
Total	107			
Cochran's C test		C = 0.1264,		
		P = 0.3173		
Transformation		\sqrt{x}		

SNK test.

(RE's vs. ZI) x Loc (p<0.05 for comparison between FF1 and FF2).

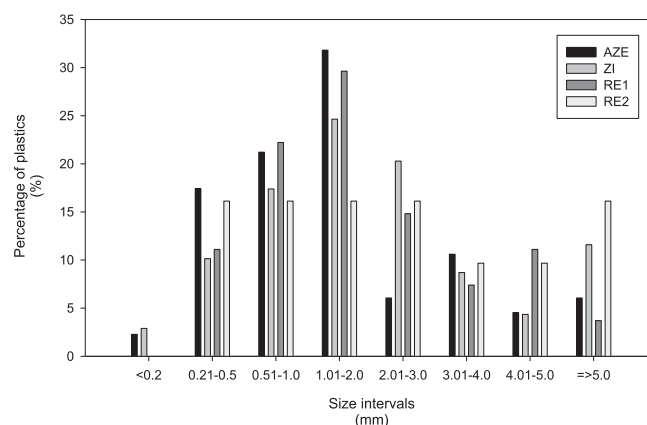


Fig. 2. Size distribution of plastics in the sediment sampled with Van-Veen grab within the four zones (Allowable Zone of Effects, AZE; Zone of Influence ZI; Reference zones; RE1 and RE2) for all fish farms. Values indicate the mean +SE, each mean was calculated by pooling three sites of which each had three replicates for three fish farms (n = 27).

percentage of particles (27.8%) was found in the size fraction of 1.1–2.0 mm. Just a very few pieces were smaller than 0.2 mm, all of those were found in AZE and ZI. The smallest particle observed under the microscope was 0.128 and the largest 13.405 mm.

Microplastic particles were categorized according to their shape into fibre, fragment and pellet. The most common type was fibre (AZE: 62%, ZI: 68%, References: 55%) and the least present type was pellet (AZE: 7%, ZI: 4%, References: 7%). Regarding shape distribution, AZE and ZI were more similar to each other compared to the reference zones, with a higher portion of fibres and a smaller percentage of fragments (Fig. S5 of Supporting Information).

The highest variety of colours was found in AZE, whereas ZI and the references had a similar colour variation; however, the colours of the collected samples were not consistent. The most common colours were black and transparent, representing approximately 50% in each zone. Other commonly used colours in the facilities were blue and yellow, which were found in all zones. Red was the only colour that was just observed in AZE (Fig. S4 of Supporting Information).

3.4. Polymer identification

The FTIR data of PE and PP before and after the acid treatment

Table 3

Carbonyl indices (I_{CO}) and DSC parameters obtained for the studied PE and PP samples.

DSC (First Heating scan)				FTIR
Samples	ΔH_m (J/g)	T_m (°C)	% Crystallinity	I_{CO}
PE_FF3	65.3	109	22.5	1.15
PE_FF2	66.8	109	23.0	1.03
PE_AT	88.5	108	30.5	0.01
PE	110.0	109	37.9	0.02
PP_FF3	36.9	160	26.7	1.20
PP_FF1	61.8	158	44.8	0.50
PP_AT	44.3	163	32.1	0.06
PP	49.5	162	35.8	0.03

ΔH°_m : 290 J/g (100% crystalline PE).

ΔH°_m : 138 J/g (100% crystalline PP).

Reference samples: PE, PP.

Reference samples after acid treatment: PE_AT, PP_AT.

(samples PE_AT and PP_AT, respectively) showed no significant differences indicating that the acid treatment in our methodology did not influence the surface of the studied polymers (Fig. S6 of Supporting Information). Due to the reduced size of the particles, only six particles of plastic could be analysed resulting in four different types of plastic polymers: polyethylene (PE), polypropylene (PP), a commercial nylon fibre and a cellulose derivative. For detailed explanations on the identification of the materials, see Supporting Information.

3.5. Degradation status of plastics

I_{CO} , of PE and PP reference samples (PE; PE_AT; PP and PP_AT) was very close to zero. Contrastingly, the plastics identified as PE and PP presented an absorption band in the carbonyl region (Fig. S7). I_{CO} values were 1.03 and 1.15, for PE and 0.50 and 1.20, for PP, indicating degradation. Changes in the enthalpy of melting (ΔH_m (J/g)) and the degree of crystallinity are shown in Table 3.

4. Discussion

4.1. Macroplastic assessment

The zones close to the source (AZE + ZI) accounted for 78% of the macroplastics observed. Most of the items found in close proximity such as ropes, nets and cable ties, are commonly used in aquaculture and most probably originated from the fish farms. In the reference zones, only plastic bags were observed, which might have had different sources (for a more detailed explanation see subsection 4.5). In conclusion, only 9 items of macroplastic have been identified out of 108 pictures of the seabed. The capacity of static pictures to quantify macroplastic abundance could be more limited than seabed video transect (Debrot et al., 2014).

4.2. Microplastic assessment

As for the macroplastics, our results showed a higher concentration of microplastics in sediments close to the fish farm cages compared to the reference zones. The difference of particle sizes between the zones can be explained by physical and ecological differences, such as currents, depth, temperature or the effect of biota, which either could accelerate or decelerate the breakdown of microplastics (Andrady, 2011; Lusher et al., 2017). The predominance of fibres coincides with previous findings of microplastics in coastal sediments (Claessens et al., 2011; Gago et al., 2018; Guerranti et al., 2017; Maes et al., 2017). An explanation for the high percentage of fibres in marine sediments could be the polymer

structure of which the fibres consist: many of them are polystyrenes, polyacrylamides or polyamides (nylon), which have a higher density than seawater and therefore, sink to the seabed (Maes et al., 2017).

The high amount of black particles found in AZE is consistent with the fact that most structure of fish farms are made of black plastic. Additionally, ropes and nets used in the farms were usually of green colour, which is represented in AZE and ZI, but not in the reference zones. However, since only the larger polymers could be analysed through FTIR and DSC, we can only suggest that these particles could have derived from the installations.

Out of the plastics that could be analysed through FTIR and DSC, we found a relative high diversity of polymers: polyethylene (PE), polypropylene (PP), a commercial nylon fibre and a cellulose derivative. PE and PP are dominant polymers in aquaculture and fishing tools, used as fibres in ropes and fishing nets (Oxvig and Hansen, 2007) or as buoyancy materials (Park et al., 2016). These findings are consistent with other studies on microplastic identification in marine sediments, where PE and PP were predominant (Frias et al., 2016; Vianello et al., 2013). Both PE and PP polymers have versatile applicability and a high industrial usage, covering nearly 50% of European demand for plastics in 2014 (PlasticsEurope, 2015). Thus, there is a high probability that PE and PP are widely distributed in the environment.

The found nylon fibre was expected to be a fishing line (Fig. S7 of Supporting Information). Several studies directly related nylon fibres found in marine sediments (Reddy et al., 2006) and animal guts (Dantas et al., 2012; Possatto et al., 2011) to fishery activities. In addition, a cellulose derivative in the form of a recyclable plastic bag was found. Rayon and viscose, semi-synthetic cellulose-based polymers, have been observed in coastal sediments (Frias et al., 2016) and animals (Lusher et al., 2013; Remy et al., 2015). Other polymers, for example polyvinylchloride (PVC) and polyethylene terephthalates (PET), which are also highly produced plastics, were not detected, probably due to their high specific density compared to sodium chloride (Klein et al., 2015). Another reason could have been, that the acid treatment performed to destroy the natural debris influenced or even dissolved this materials so that they could not have been detected. However, we have validated our methodology for the found plastic materials (see subsection 2.6.1) and methodologies using acid digestion with sediment samples have been used previously (Claessens et al., 2013; De Witte et al., 2014; Klein et al., 2015).

The presence of a carbonyl region as well as the changes in the enthalpy of melting and the degree of crystallinity in the polymers identified as PE and PP, compared to their respective pristine polymers, suggest a certain degradation of the analysed polymers (ter Halle et al., 2017). The degradation of the polymer occurs mainly in the surface layer, which can be eroded, leaving a non-degraded layer. Accordingly, the degradation status cannot be accurately estimated after a certain stage of oxidation (Andrady, 2017).

The performed analyses provide us with a first approach on microplastic present in the fish farm sediments. However, the reduced size of most microplastics prevented us from applying FTIR and DSC techniques to all particles, resulting in a considerably small sample size to analyse and therefore, does not allow us to draw conclusions on the abundances of different types of polymers.

4.3. Fish farm comparison

When comparing microplastic abundance among fish farms, the oldest one showed the highest concentrations. Therefore, it seems that age is a relevant factor for microplastic accumulation on the sediment. Additionally, slightly more macroplastics were found at

Table 4
Comparison of microplastic concentrations found in coastal areas affected by anthropogenic activities. Values show the range of the mean values between the sampled zone with the highest and lowest value on each study. In our study the value was obtained by taking the highest and lowest mean value of the Allowable Zone of Effects and the Zone of Influence from each fish farm.

Area	Location	Concentration of microplastics (plastics/kg dry sediment)	Applied method	Size fractions (mm)	References
North Sea	Harbour	126–213	Density separation NaCl, supernatant sieved (38 µm mesh) and directly examined	0.038–1	Claessens et al. (2011)
Mediterranean	Industrial, agriculture and urban discharges	672 - 2175	Density separation NaCl, supernatant sieved (32 mm mesh) and consequently filtered; pore size 0.7 µm, diam. 47 mm	<0.1, 0.1–0.5, >0.5, smallest particles observed 0.015	Vianello et al. (2013)
Mediterranean	Sewage and harbour influence	123–164	First sieving sediment (2, 1, 0.5, 0.25, 0.125 and 0.063 mm mesh), density separation distilled water and directly examined	0.063–0.125, 0.1251–0.25, 0.251–0.5, 0.51–1, 1.1–2, >2	Alomar et al. (2016)
Mediterranean	Discharges by rivers	108–291	First sieving sediments (4, 2, 1, 0.063 mm mesh), direct examination and density separation for particles < 0.063 mm with NaCl; pore size 10 µm, diam. 47 mm	<5, 5.1–25, >25, smallest particles observed 0.5	Guerranti et al. (2017)
Mediterranean	Aquaculture	32–69	Density separation NaCl, supernatant filtered; pore size 1.2 µm, diam. 47 mm	<0.2, 0.21–0.5, 0.51–1.0, 1.1–5.0, >5.0, smallest particles observed 0.128	Present study

the farm with the highest production. This hypothesis seems coherent with the fact that most of the microplastics found were secondary microplastics, which are the result of larger pieces of plastic breaking down into smaller pieces (Andrady, 2011). Thus, fish farm production could be also a relevant factor, indirectly favouring microplastic pollution over the long term.

4.4. Pollution by fish farming compared to other activities

Plastic pollution studies use a large diversity of methods for estimating plastic pollution and different size classifications for plastics, hindering exact comparisons among studies. Our study showed a plastic pollution level one order of magnitude lower compared to other studies on other anthropogenic pressures, such as agriculture or marine traffic that used comparable methods (Table 4).

We assume the reason why our obtained values are rather low is because European fish farms (including the ones sampled), generally have a waste management plan since marine aquaculture is a highly regulated activity (Rosenthal et al., 2000). However, plastic wastes have a further spatial reach than the one studied (Lavers and Bond, 2017), which was not assessed in this study. Nevertheless, the low occurrence of plastics found in the close vicinity of the fish farms suggests that the waste management plans of the sampled fish farms are effective.

4.5. Other sources of plastic pollution

Two plastic bags were found in the studied area; both occurred in the reference zones. This might suggest that they originated from another source of human activities than aquaculture such as recreational sailing or from a coastal source, since most marine debris originates from land-based sources (Andrady, 2011). However, plastic bags could have also been used by farm workers and be transported into the reference zones (Veiga et al., 2016). It is especially difficult to identify the source of some items, such as plastic bags. This fact illustrates the difficulty of finding reference zones in areas that are subjected to many anthropogenic pressures such as the Mediterranean.

The high use of the coastal areas favour interactions among the activities that take place there. In the case of fish farms, they attract

wild fish and consequently, artisanal and recreational fishers (Dempster et al., 2002; Sanz-Lázaro and Marín, 2011; Arechavala-Lopez et al., 2011).

Artisanal fisheries from the study area use fishing nets (Marengo et al., 2015) and basket traps for octopus among other species (Lloret and Font, 2013). Recreational fishery is also an intense activity along the Western Mediterranean coast, mainly comprising spearfishing and angling (Seytre et al., 2013). This type of fishery is responsible for a considerable amount of debris entering the marine environment (Derraik, 2002; Seytre et al., 2013). Thus, the found debris in the AZE and ZI, a basket trap and a fishing line suggest that fisheries could interact with fish farming contributing to plastic pollution. If so, artisanal and recreational fisheries could be a potential additional source of plastic pollution within and near the lease of the fish farm facilities. Nevertheless, more research effort is needed to get concluding results.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2019.113336>.

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