Plastic debris in the Mediterranean Sea: types, occurrence and distribution along Adriatic shorelines

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Abstract

Small plastic debris in sediments from five beaches were investigated to evaluate their occurrence and abundance in the Northern Adriatic coast for the first time. Plastic debris extracted from sediments were counted, weighted and identified by Fourier-transform infrared spectroscopy (FT-IR). A total of 1345 items of debris (13.491 g) were recorded, with a mean density of 12.1 items kg^{-1} d.w. and 0.12 g kg^{-1} d.w. Fragments were the most frequent type of small plastics debris detected. In terms of abundance, microplastics (<5 mm) accounted for 61% of debris, showing their wide distribution on Adriatic coasts, even far-away from densely populated areas. The majority of the polymers found were polyolefins: there were greater quantities of polyethylene and polypropylene compared to other types of plastic. Primary microplastics accounted for only 5.6% of the total plastic debris. There were greater quantities of microplastics at sites subjected to stronger riverine runoff. The results will provide useful background information for further investigations to understand the sink and sources of this emergent and priority contaminant.

Keywords

Microplastics; Polymer composition; Beaches; Adriatic Sea; FT-IR spectroscopy

1. Introduction

Plastics are essential in our everyday lives. World production of plastics has strongly expanded, from 1.7 million tonnes in 1950 to 322 million tonnes in 2015 (Plastic Europe, 2016). Whether deliberately or accidently, when plastics waste is not properly disposed it may end up as litter in the environment, seas and rivers and harming wild life, fisheries and tourism. Through a combination of photodegradation, oxidation and mechanical abrasion, the degradation rate of plastics in the environment is slow and results in production of small fragments and microplastics (Barnes et al., 2009). The existence of microplastics (plastic particulates < 5 mm; Ivar do Sul and Costa, 2014) in the marine environment has been known for nearly half a century (Carpenter and Smith 1972). While pictures of macroplastic debris in ocean gyres (Moore at al. 2001) and of the excessive accumulation of litter on beaches in the most remote locations worldwide (e.g. Convey et al., 2002; Foster-Smith et al., 2007) have fostered the awareness of plastic pollution, microplastics have emerged as a an imminent source of plastic contamination in the marine environment only recently as a consequence of their eluding presence in sediments and seawater (Claessens et al., 2011; Ivar do Sul and Costa, 2014).

The most widely used plastics are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS) and polyethylene terephthalate (PET), which represent grossly 90% of the total world production (Andrady and Neal, 2009). Millions of tonnes of plastic waste (4.8 - 12.7 million tonnes in 2010) end up the marine environment (Jambeck et al., 2015). Certain plastics are expected to occur in greater abundances than others due to the relative proportions that are manufactured, used, and discarded. For instance, half of all the plastics that are produced annually are polyolefins, i.e. PE and PP (Plastic Europe, 2016), which are principally used to make packaging that is used once and then discarded. It is, however, not known whether polyolefins occur in greater abundance as items of debris compared to other polymers. The most prominent types of microplastics identified in the marine environment include pellets, irregular fragments, films and fibres (Wright et al., 2013) of which can be classified as primary or secondary microplastics. Primary microplastics are intentionally produced as precursors to other products, while secondary microplastics result from the degradation of macroplastics due to chemical, mechanical and photolytic degradation processes in the marine environment (Mathalon and Hill, 2014). The sources of primary microplastics are usually plastic pellet processing facilities at petrochemical plants, and specific trading activities such as oceanic shipping routes (Thompson et al., 2009). Small sized primary microplastics granules are also present in cosmetics products and used as abrasives in a wide range of applications (Browne, 2015).

The Marine Strategy Framework Directive, MSFD (2008/56/EC; European Commission, 2008) establishes a framework for each Member State to take action to achieve or maintain Good Environmental Status (GES) for the marine environment by 2020. The MSFD follows a holistic functional approach identifying a set of 11 Descriptors, which collectively represent the state and functioning of the whole system (Borja et al., 2010). Descriptor 10 (D10) is identified as "Properties and quantities of marine litter do not cause harm to the coastal and marine environment" (European Commission, 2008). Microplastics are considered specifically in descriptor 10 of the MSFD (10.1.3 “*Trends in the amount, distribution and, where possible, composition of micro-particles (in particular micro-plastics)*”), and implicitly in the indicator related with impacts of litter on marine life. According to the MSDF, microplastics should be categorized according to their physical characteristics including size and shape. It is also important to obtain information on polymer type (Gago et al., 2016).

The Adriatic Sea is characterized by one of the greatest seafloor litter pollution among Mediterranean regions (Pasquini et al. 2016). The north-western Adriatic coast is thus vulnerable to plastic accumulation on beaches from land sources due to river discharges, marine sources due to aquaculture, fishing and recreational maritime activities, as well as being an important route for commercial vessels and cruise ships. Abundant scientific literature has extensively explored the various anthropogenic impacts affecting this fragile coastal ecosystem (Munari et al., 2011; Torresan et al., 2012; Romano and Zullo, 2014), but the presence and diffusion of microplastics as contaminants have not yet been investigated in any environmental compartment.

With the present study we wanted to assess, for the first time in the north-western Adriatic coast, the quality and quantity of small plastic debris occurring in beach sediments to address the gap in knowledge and to serve as a baseline for future comparisons. Further hypotheses tested were that: (1) microplastics will be found in greater numerical abundance than macroplastic debris; (2) PE and PP will be more abundant than other polymers due to differences in levels of production; (3) the amount of primary microplastics will be prevalent respect to secondary microplastics because of nearby petrochemical industrial parks (Marghera, Ferrara, Ravenna); (4) there will be differences in microplastics abundance between beaches with strong riverine inputs and those with weak riverine inputs. We considered beach sediments at the high water line, since they reflect the amount of microplastics washed towards the coastlines with the tidal flows (Martins and Sobral, 2011).

2. Methods

2.1 Study area

Along the north-western Adriatic coast a large number of rivers discharge into the sea, being the Po River the most relevant, followed by the Adige. Five beaches (Fig. 1), differently affected by riverine runoff, were sampled. The considered area is subjected to intense marine traffic from supplier vessels for offshore activities (gas platforms), trawl-fishing vessels, and recreational boats. It is also an area of intense aquaculture, with offshore mussel farms, and coastal clam cultivations. Inland, a few dozen kilometers away from the sampled beaches, there are three important petrochemical industrial parks: Porto Marghera, Ferrara, and Ravenna.

2.2 Sampling and analysis

Beach surveys were conducted at the 5 beaches in May 2015. At each beach there were two replicate sites separated by 200 m. Each site consisted of a 10 m stretch of linear shoreline. At each site, sampling was performed by using quadrats placed along the last high tide mark, as plastic is preferably accumulated in this zone (Martins and Sobral, 2011). Three replicate samples were collected at each site by scraping the first 5 cm of sand from 50 × 50 cm quadrats (Galgani et al., 2011; Martins and Sobral, 2011; Jayasiri et al., 2013). Replicates of the same site were separated by 5 m. All samples were obtained during calm conditions with low wave activity. Samples were placed in labelled bags and transferred to the laboratory, where all replicates were analyzed separately.

In the laboratory, sediment samples were dried at 50ºC during 48 hours. Each sediment sample was then divided into subsamples and the plastic debris were removed under a dissection microscope (Nikon SMZ45T, magnification 3.35-300x), counted and weighted to the nearest 0.0001 g. The identified plastics were measured at their largest cross-section using calipers and classified into four groups: micro (≤5 mm), meso (>5–20 mm), macro (>20–100 mm) and mega (>100 mm) (Jayasiri et al., 2013). Plastic debris were also categorized according to shape (i.e., fibre, film, fragment or pellet).

Fourier-transform infrared spectroscopy (FT-IR) analysis of 20 plastic debris for each shape type was carried out with a CARY 600 FT-IR (Agilent Technologies) instrument. Measurements were carried out in attenuated total reflectance (ATR) configuration, with a Pike Miracle diamond cell. Tests were carried out at 25°C in dry air. Particles were identified by comparing FT-IR absorbance spectra of the microplastics to those in a self-collected, polymer reference library.

Differences in abundances of plastic debris (categorized by shape and dimension) were analyzed through permutational analysis of variance (PERMANOVA). The similarity matrix was calculated using the Bray-Curtis index and abundance data were log (x + 1) transformed. The experimental design incorporated two factors: "Location" (fixed) with 5 levels: Rosolina (ROS), Volano (VOL), Bellocchio (BEL), Casalborsetti (CAS) and Bevano (BEV), and "Site" (random and nested within the factor "Location") with 10 levels: ROS1, ROS2, VOL1, VOL2, etc. Similarity percentage (SIMPER) analysis was used to explore differences in plastics distribution (categorized by dimension) within and between beaches. All statistical analyses were performed using PRIMER v.6 and its add-on package PERMANOVA+ (Anderson et al., 2008).

Data of river runoff was obtained by Regional Agencies Annual Reports (ARPAV, 2014; ARPA, 2015).

3. Results

Thirty quadrats were sampled at the five beaches. Some examples of plastic debris collected during the study are shown in Fig. 2. The smallest debris collected was 0.8 mm of length. All sediment samples collected on the beaches contained plastics. A total of 1345 items of debris (13.491 g) were recorded from the 30 samples of sediment, with a mean density of 12.1 items kg^{-1} d.w. and 0.12 g kg^{-1} d.w. The greatest plastic abundance by number and weight was observed at Volano (21.6 ± 12.8 items kg^{-1} d.w., and 0.28± 0.29 g kg^{-1} d.w., respectively). In contrast, the lowest mean values by number and weight were 5.99 ± 3.25 items kg^{-1} d.w. and 0.013 ± 0.01 g kg^{-1} d.w. at Bellocchio.

As predicted there was greater abundance of smaller debris (micro and meso) compared to macro and mega plastic debris (Hypothesis 1). This was reflected in the frequency distribution of different sizes of debris, which were skewed toward smaller debris (Fig. 3). In terms of numerical abundance, microplastic accounted for 61% of the total amount found. Small plastic debris (micro and meso plastics) made up 89.9% of total amount, while larger debris (macro and mega plastics) accounted for 10.1%.

Identification through FT-IR spectroscopy evidenced that at all beaches the majority of the polymers found were polyolefins (Fig.4). As predicted by Hypothesis 2, there were greater quantities of PE (37.7% in weight) and PP (34.5% in weight) compared to other types of plastic (Nylon: 12.2%; PS: 9.4%; PET: 3.9%; PVC: 1.8%; and thermoplastic polyhuretane, TPU: 0.6%). At all beaches the majority of plastic debris were PE, except at Bevano where it was PP. The composition in weight by polymer type of plastic debris at each of the five beaches is shown in Fig. 5. The primary shape types (by number) were fragments (60.6%), followed by film (23.6%), and fibres (10.3%). Contrary to what expected, pellets made up only 5.6% of all plastic shape types (Hypothesis 3). In Table 1 the average abundance of shape type of beach plastics collected is shown. Fragments were identified as PE, PP, PVC, PS and TPU. Fibres were identified as PE, PP and Nylon, and the film polymers were PE and PP. Pellets were composed of PE or PP.

Rosolina is subjected to the Adige River runoff (average flow: 235 $m^3 s^{-1}$), Volano to the Po River runoff (1540 $m^3 s^{-1}$), Bellocchio to the Reno River (96 $m^3 s^{-1}$), Casalborsetti to the Lamone River (11 $m^3 s^{-1}$), and Bevano to the Fiumi Uniti (10 $m^3 s^{-1}$) and Bevano creek (1.5 $m^3 s^{-1}$) runoff. So we have two beaches subject to strong riverine runoff (Volano and Rosolina), and the other three (Bellocchio, Casalborsetti and Bevano) to weak riverine runoff. According to PERMANOVA, significant differences were found between locations (Tab. 2): there were differences in plastic abundances categorized by shape and dimension between beaches subjected to strong riverine runoff (Rosolina and Volano) and the others (Tab. 3; Supplementary Materials). As the number of unique values under permutations was very low, P-values were obtained using Monte Carlo samples from the asymptotic permutation distribution (Anderson and Robinson, 2003). These results were corroborated by SIMPER analysis (Tab. 4, Supplementary Materials). Finally, a significant relationship ($r=0.91$, $P<0.001$; Hypothesis 4) between the average abundance of microplastics (< 5 mm) and riverine runoff was found (Fig. 6).

4. Discussion

Biodegradation of plastic litter entering the environment from land- or sea-based sources is extremely slow (Thompson et al., 2004). As almost all main Italian rivers flow into the Adriatic Sea (Po, Adige, Brenta, Tagliamento, Isonzo, etc.), the Adriatic and its beaches provide a large sink for undegraded synthetic polymers (Munari et al. 2016; Pasquini et al. 2016). The results of this study demonstrated the presence of small plastic debris at all of the sampled Adriatic beaches. Quantified microplastic concentrations in this study are comparable to other studies (e.g. Van Cauwenberghe et al., 2015), although the wide array of existing techniques and quantification units limits the comparison of results.

As predicted by Hypothesis 1, at all the 5 beaches microplastics comprised the majority of the plastic debris (61%), with a declining plastic size with increasing plastic debris abundance. Barnes et al. (2009) reported a generalized decrease in the mean size of plastic debris in the global environment, along with the increasing abundance of such particles due to continuous degradation. Because of weathering degradation, beaches are better settings than other natural environments for the breakdown of plastic debris (Andrady, 2011), so it is extremely likely that the plastic debris present in the 5 beaches will continue to fragment into smaller particles: this may facilitate dispersion by wind or wave action, and thus the entry of microparticles into food webs. As shown in Fig. 2, most of microplastics were colored. The colors of plastic debris, especially of the microplastics, causes them to resemble natural food that is likely ingested by the biota (Andrady, 2011). Microplastics comprise a frequently reported size category in ingestion studies (Thompson et al., 2004), and for this reason they must be regarded as a real threat to marine life.

Results of FT-IR spectroscopy analysis indicated that most plastics were polyolefins, and as predicted by Hypothesis 2, we found greater abundances of PE and PP compared to other polymers. These are plastic resins with specific gravity less than one, permitting them to be positively buoyant and easily deposited on beaches (Andrady 2011). Our finding is in agreement with previous studies of macroplastic debris in which packaging was the most abundant type of debris found in coastal habitats (Jayasiri et al., 2013; Zaho et al., 2015; Munari et al. 2016). This is not surprising, since PE, with an annual global production of around 80 million tonnes, is mainly used to manufacture packaging (plastic bags, plastic films, containers including bottles), and PP, with an annual global production of around 55 million tonnes, is mainly used for packaging, reusable containers, stationery, textiles, ropes, etc. (Thompson et al., 2004).

Fragments by number and weight were the most frequent type of small plastics debris detected, and were identified as PE, PP, PVC, PS and TPU. The main source of fragments was attributed to the breaking down of larger items. Fragmentation of larger items is mainly driven by photo-oxidative, thermal- and biodegradation (Andrady, 2011), but rates and mechanisms may vary among polymer types: PE, for example, is more readily fragmented by weathering events, while PP is more subject to mechanical degradation (Cooper and Corcoran, 2010). At our beaches, fragments had all sorts of shapes, but the majority were jagged fragments of larger plastic items. Films were the second most common type of plastic debris, and were composed by PE and PP. Plastic film is mainly used for single-use packaging for food. Fibres were the third most common type of debris: PE, PP and Nylon fibres are used to produce bags and ropes, which are widely used in the local aquaculture and fishing industry. Contrary to Hypothesis 3, primary microplastics (i.e. virgin plastic pellets) accounted for only 5.6% of the 1345 sampled plastic debris. The presence of these virgin plastic pellets does not imply long-range marine transports since there are large petrochemical industrial parks with pellet-producing and pellet-processing plants (e.g. Lyondell-Basell at Ferrara, Eni-Versalis at Ravenna and Porto Marghera) nearby the sampled beaches. These results show that north-western Adriatic beach sediments are more contaminated by secondary microplastics than by virgin plastic pellets. These results may also mean that, at least in northeastern industrial area, petrochemical companies have become sensitive to environmental issues, and their policies to prevent accidental spilling of virgin plastic pellets during production and transport phases seem real and effective.

Most studies report high microplastic concentrations in sediments close to densely populated areas (Barnes et al., 2009; Costa et al., 2010; Claessens et al., 2011; Jayasiri et al., 2013; Zhao et al., 2015). In this study, higher values ranging from 21.6 ± 12.8 items kg^{-1} d.w. (Volano) to 16.6 ± 2.3 items kg^{-1} d.w. (Rosolina), were obtained in natural zones far-away from densely populated areas, being all beaches considered in this study included in the Po River Delta Parks and in the Natura 2000 Italian network. The comparison of our results with those from the Lagoon of Venice (Vianello et al., 2013; plastics ranging from 2175 to 672 items kg^{-1} d.w.) shows that the level of contamination from small plastics in our beaches is lower than in the Lagoon. In addition to the different method of microplastics extraction (there is no standardized procedure for microplastics analysis; Morét-Ferguson et al., 2010) this is probably because of the different local hydrodynamic regimes since microplastics tend to accumulate in low-dynamic areas. Small plastic debris, discharged into the sea indirectly via wastewaters, sewage pipelines and terrestrial runoff (Derraik, 2002), would be expected in higher quantities in beaches subjected to stronger riverine runoff, since higher discharge might positively impact the higher plastic density. As a matter of fact, the highest microplastic concentrations in this study were encountered at Volano and Rosolina, two beaches subjected to Po and Adige River runoff respectively, rather than Bellocchio or Bevano, which are affected by a much weaker riverine runoff (Hypothesis 4). Trivially, this study provides evidence indicating that natural areas are not excluded from microplastic contamination. This causes concern since the presence of microplastics in beach sediments may result in changes in their physical characteristics, such as sediment permeability and thermal insulation properties, that can have a variety of potential impacts on beach organisms (Carson et al., 2011).

5. Conclusions

This study represents a baseline for microplastic research in the coastal sediment compartment in the Mediterranean Sea. We sampled 5 north-western Adriatic beaches for small plastic debris and we found all beaches to be contaminated. Microplastics (<5 mm) resulted in greater abundance than other plastic debris. Seven polymer types were found, but at all beaches the majority of plastic debris were polyethilene and polypropylene. Secondary microplastics were dominant, resulting more abundant in beaches with strong riverine inputs. The cleaning of these beaches is promoted by local NGOs like Legambiente or WWF, it is occasional (grossly once a year) and carried out by citizens and school groups on a voluntary basis. However, beach cleaning only concerns medium-large sized litter, as bottles and bags. As there are no cleaning possibility available for such a small items, the only option is to prevent and combat the presence of larger plastic items in the environment. The ubiquitous prevalence of microplastics in north-western Adriatic beaches (and thus in the marine environment) indicates the need of more research to understand the sink and sources of this emergent and priority contaminant in the marine environment and biota.

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Figure Legend

Fig. 1. Location of the study beaches (ROS: Rosolina; VOL: Volano; BEL: Bellocchio; CAS: Casalborsetti; BEV: Bevano). Main petrochemical industrial parks are also indicated.

Fig. 2. Examples of the collected plastic debris.

Fig. 3. Composition of plastic debris collected at the five beaches according to size: Micro (≤5 mm), Meso (>5–20 mm), Macro (>20–100 mm) and Mega (>100 mm).

Fig. 4. FT-IR spectroscopy spectra of the plastics collected in this study.

Fig. 5. Weight composition of plastic debris collected at the five beaches according to type of polymer.

Fig. 6. Relationship between average riverine flows and abundance of microplastics (< 5 mm).

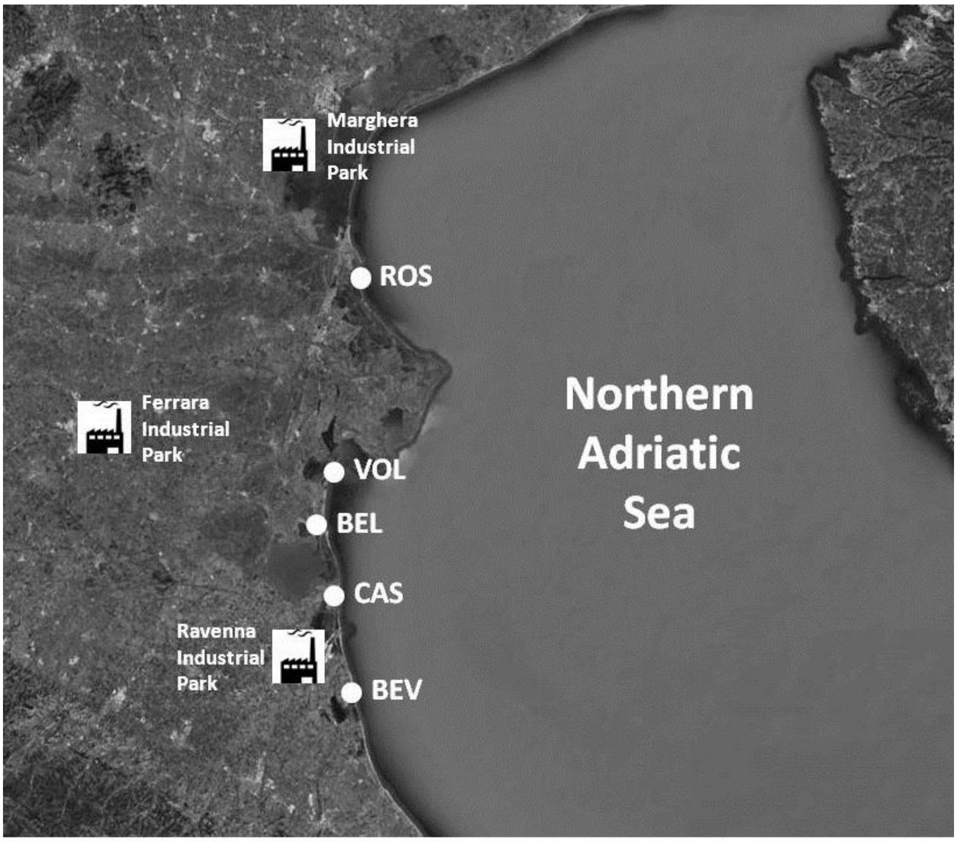

Marghera Industrial Park
ROS
Ferrara Industrial Park
VOL
BEL
CAS
Ravenna Industrial Park
BEV
Northern Adriatic Sea

Fig. 1

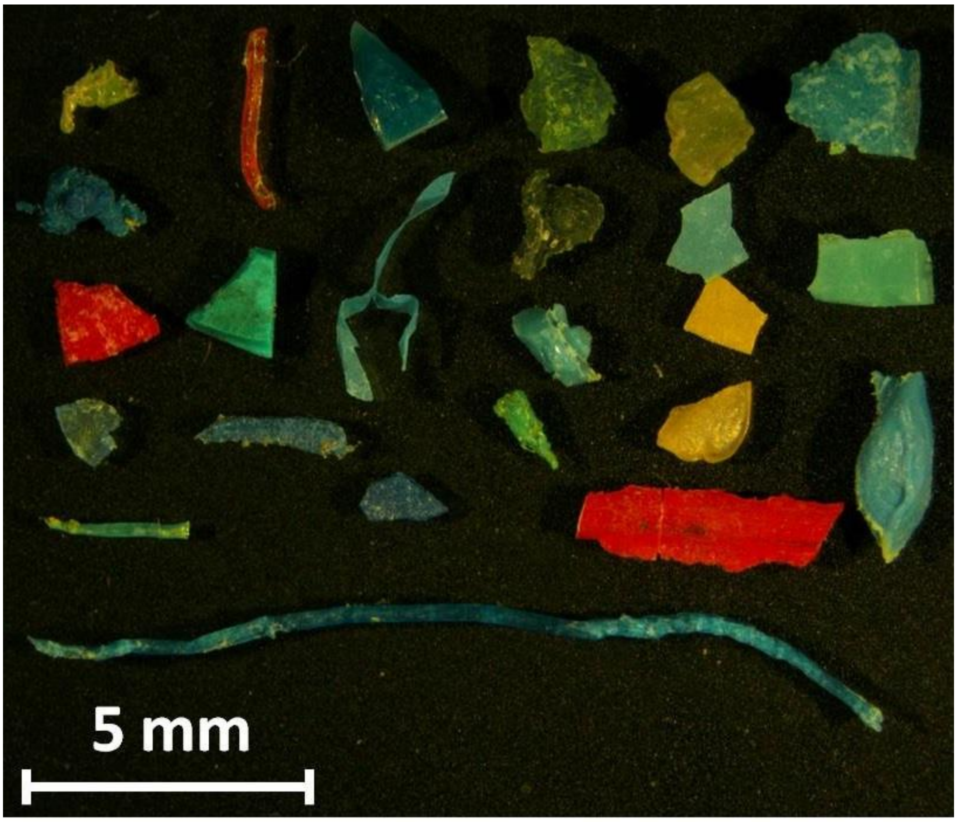


Fig. 2

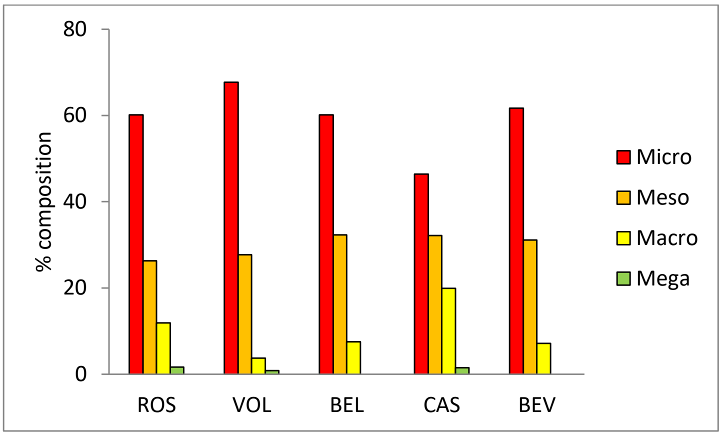

80
60
40
20
0
% composition
ROS
VOL
BEL
CAS
BEV
Micro
Meso
Macro
Mega

Fig. 3

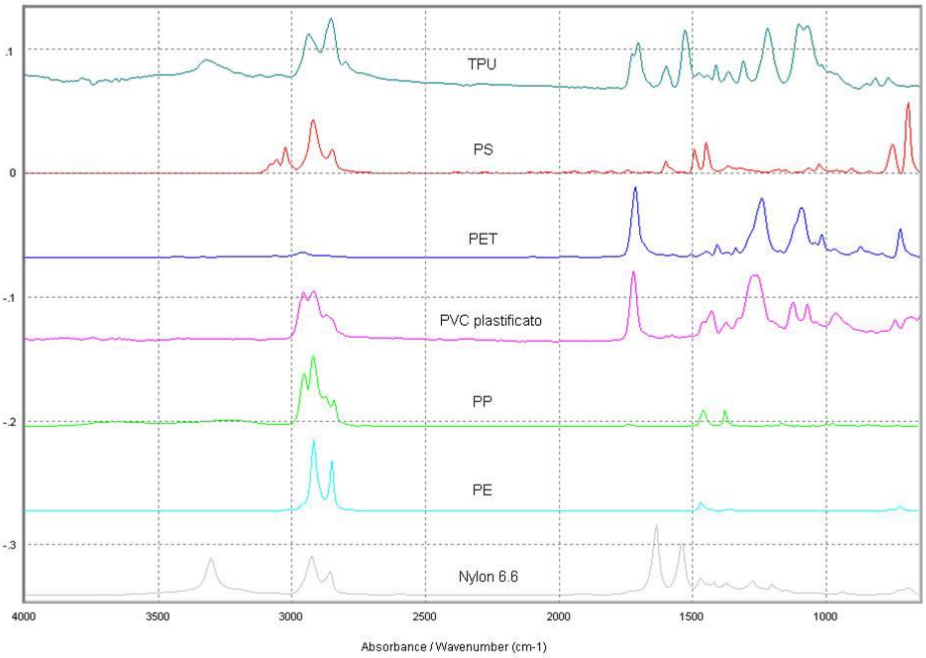
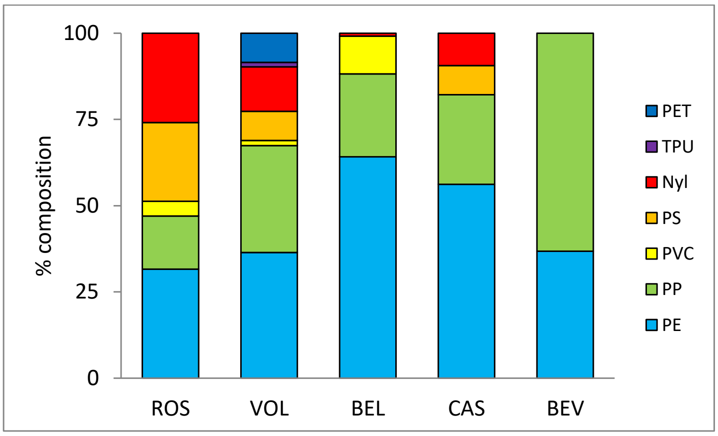

TPU
PS
PET
PVC plastificato
PP
PE
Nylon 6.6
.1
0
-.1
-.2
-.3
4000
3500
3000
2500
2000
1500
1000
Absorbance / Wavenumber (cm-1)

Fig. 4.

404



405

406 Fig. 5

407

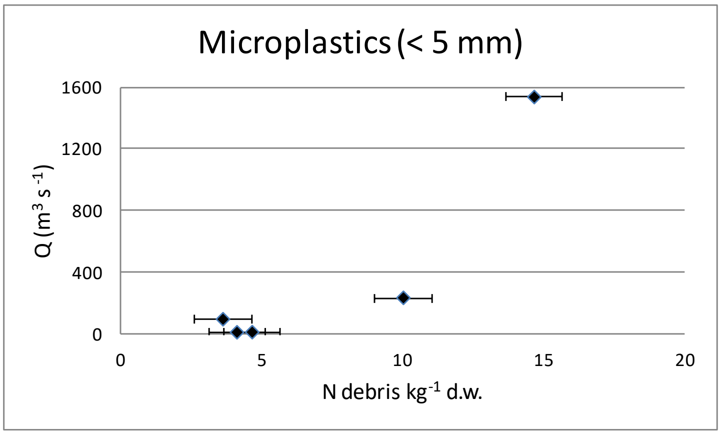

Microplastics (< 5 mm)
Q (m³ s⁻¹)
1600
1200
800
400
0
0
5
10
15
20
N debris kg⁻¹ d.w.

Fig. 6

Table 1.

Shape type of beach plastics collected. Values represent average abundance (pcs *per* kg of dry sediment) and standard deviation,~~ (~~in italics (for comparison with other studies, use a conversion constant of 14.8 to obtain pcs *per* m^{-2}).

Location	**Site**	**Fibre**	**Film**	**Pellet**	**Fragment**
Rosolina	ROS1	0.99	4.77	1.35	10.36
		0.87	*0.95*	*0.47*	*3.53*
	ROS2	1.80	4.68	1.44	7.84
		0.41	*1.84*	*0.68*	*3.01*
Volano	VOL1	1.17	5.41	0.90	10.81
		0.31	*6.58*	*0.78*	*9.87*
	VOL2	1.08	5.14	1.26	17.48
		0.27	*1.77*	*0.41*	*7.20*
Bellocchio	BEL1	0.63	1.53	0.27	4.23
		0.68	*1.28*	*0.27*	*2.92*
	BEL2	0.90	0.63	0.54	3.24
		0.41	*0.41*	*0.27*	*0.81*
Casalborsetti	CAS1	3.51	2.16	0.36	3.51
		1.89	*1.43*	*0.31*	*1.64*
	CAS2	2.16	1.62	0.36	3.96
		0.54	*0.47*	*0.16*	*1.80*
Bevano	BEV1	0.09	1.17	0.09	7.12
		0.16	*0.83*	*0.16*	*3.20*
	BEV2	0.09	1.44	0.18	4.86
		0.16	*1.13*	*0.16*	*2.15*

Table 2.
Main tests from PERMANOVA on unrestricted permutation of log(x+1) shape and dimension data of plastic debris. Significant P-values are in bold

Shape	*df*	*SS*	*MS*	*Pseudo-F*	*P(perm)*	*P(MC)*
Location = Lo	4	5191.7	1297.9	14.169	**0.0023**	**0.0003**
Site = Si(Lo)	5	458	91.6	0.435	0.9353	0.9248
Residual	20	4213	210.7			
Total	29	9862.6				
Dimension	*df*	*SS*	*MS*	*Pseudo-F*	*P(perm)*	*P(MC)*
Location = Lo	4	2746.9	686.7	3.112	**0.0285**	**0.0388**
Site = Si(Lo)	5	1103.5	220.7	1.203	0.3162	0.3163
Residual	20	3669.6	183.5			
Total	29	7519.9				