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Baseline characterisation of microlitter in the sediment of torrents and the sea bottom in the Gulf of Tigullio (NW Italy)

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ABSTRACT

Microlitter (<5 mm) distribution was investigated in the bottom sediments of the Entella Torrent and its tributaries as well as in the sea area in front of its mouth (Gulf of Tigullio; NW Italy). Microlitter was extracted from sediment using a concentrated hypersaline solution (1.2 g cm^{-3}), filtered with 0.45-—m porosity and black grid membranes and examined using a binocular microscope. Items were counted, dimensionally measured and classified according to type, shape, colour and appearance. A total of 56 torrent and sea sediment samples revealed the presence of 4,302 items. The mean concentrations were 1.5 items $cm^{-3} \pm$ 1.3 standard deviation (SD) and 1.6 items $cm^{-3} \pm$ 1.3 SD for torrent samples and sea sediment samples, respectively. Transparent fibres predominated, followed by fragments. The most common colour was white-cream. In total, 1.8% of items were analysed with Fourier transform infrared spectroscopy (FT-IR) 2D imaging, using a focal plane array (FPA) detector to identify their composition. Nylon was the most common plastic polymer type; however, approximately 50% of the analysed samples consisted of cotton transparent fibres. The influences of human activities (cities, port and discharges) on microlitter composition were highlighted and torrents were confirmed as a vector of microlitter from land to sea.

1. Introduction

Environmental pollution is a global phenomenon, with plastic debris playing an important role worldwide because it is present at every latitude from the Equator (Syakti et al., 2017) to the Poles (Bergmann and Klages, 2012; Munari et al., 2017), in every kind of environment from remote mountains (Free et al., 2014) to the sea bottom (Van Cauwenberghe et al., 2013) and in every environmental matrix from water (Lusher et al., 2014; Moore et al., 2011) to sediment (Blumenröder et al., 2017; Guerranti et al., 2017) and from air (Dris et al., 2016) to biota (Romeo et al., 2015).

In marine environments, the distribution and abundance of marine litter are influenced both by anthropogenic and environmental factors, including current and wave dynamics, precipitation and geomorphology (Galgani et al., 2013). Marine litter is more concentrated in coastal areas and within mid-ocean gyres than on continental shelves or at the deep-sea bottom (Cózar et al., 2014; Galgani et al., 2013; Mathalon and Hill, 2014). This is due to the presence of densely populated cities that are commonly located close to water courses and that are responsible for waste discharge into the environment (Miller et al., 2017).

Plastic debris represents 60%–80% of marine litter and is thus one of the most abundant contaminants in the environment (Lusher et al., 2014; Singh, 2016). Moreover, processes of plastic degradation are slow, rendering plastic a persistent material in marine environments, where it represents a threat to ecosystems and marine organisms (Hidalgo-Ruz et al., 2012). Plastic enters marine environments through various sources, such as urban run-off, fishing activities, illegal releases into the sea, the natural run-off of rivers and torrents (small rivers characterised by a periodic stream of fast-moving water; Bertolotto et al., 2005; Capello et al., 2017), environmental disasters and atmospheric fallout (Dris et al., 2016; Miller et al., 2017). Plastics introduced into marine environments vary in size from micrometres to metres (Hidalgo-Ruz et al., 2012). Plastic items smaller than 5 mm are widely regarded as microplastics, even though there is not a universal definition that accurately includes all the criteria that might be used to describe what microplastics are Frias and Nash (2019). Microplastics may be specifically produced for various uses (primary sources), such as pellets for cosmetics (microbeads) and exfoliants, or be derived from the breakage or deterioration of

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larger plastic pieces (secondary sources), due to ultraviolet (UV) radiation, photodegradation, oxidative processes, hydrolytic seawater properties and abrasion caused by wave action and contact with bottom sediment (Claessens et al., 2013; Hidalgo-Ruz et al., 2012). An important source of microplastic fibres in particular is represented by the domestic or industrial washing of synthetic clothes (De Falco et al., 2017; Zubris and Richards, 2005). Indeed, microplastics contained in domestic wastewaters are transported to sewage treatment plants or are released untreated into water bodies (Siegfried et al., 2017; Karbalaei et al., 2018).

Rivers and torrents play an important role in microplastic transport into marine environments (Moore et al., 2011). Certainly, it is believed that the majority of microplastics originate in land-based sources (Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection - GESAMP, 2015; Guerranti et al., 2017) and that watercourses constitute the main transport vectors of microplastics to the sea (Siegfried et al., 2017). Lebreton et al. (2017) have estimated that between 1.15 and 2.41 million tons of plastic waste annually reach the sea via watercourses. Population density, level of urbanisation and industrialisation near watercourses, precipitation rates and the presence of artificial barriers or dams all affect overall microplastic input into seas and oceans (Lebreton et al., 2017). Once they have reached marine environments, microplastics tend to accumulate in sediments due to vertical transport mechanisms and organic matter accumulation, conveying them to the sea bottom (Cózar et al., 2014; Guerranti et al., 2017), in turn causing sediments to become microplastic accumulators worldwide (MEPEX, 2014).

Marine litter (including microplastics) is descriptor number 10 considered by the European Marine Strategy Framework Directive (MSFD; Directive 2008/56/EC), which sets as a goal for the European states the achievement of Good Environmental Status (GES) for their marine waters. Therefore, numerous studies and projects on microplastic sources (De Falco et al., 2017; Dris et al., 2016; Siegfried et al., 2017) and distribution (Sherman and van Sebille, 2016; Zhang, 2017) have been implemented in recent decades or are currently ongoing. In addition, many efforts are underway to find a common methodology for microplastic sampling, treatment and analysis, as one does not presently exist. Consequently, some guides have been published since 2013 and include "Guidance on monitoring of marine litter in European seas" (Galgani et al., 2013), "Guidelines for the monitoring and assessment of plastic litter and microplastics in the ocean" (GESAMP, 2019) and "Standardised protocol for monitoring microplastics in seawater" of the Joint Programming Initiative Healthy and Productive Seas and Oceans - JPI Oceans (Gago et al., 2018). The ultimate objective of these efforts to investigate microplastic pollution is to understand the sources, fate and transport of such microlitter in order to improve their management (Prata et al., 2019).

In the present study, sediments from the catchment area of the Entella Torrent and its tributaries as well as from the sea bottom in front of its mouth (Gulf of Tigullio, Ligurian Sea; NW Italy) are examined. Torrent and sea bottom sediments are analysed to characterise their microlitter content. Specifically, the shape, appearance, colour and size of the items found in the sediment samples are analysed in order to determine the spatial distribution of microlitter and to assess the contribution of each torrent as a microlitter vector to the marine environment off the mouth of the Entella Torrent. A preliminary investigation into the microlitter component is performed, focusing on some of the items found in sediment samples with Fourier transform infrared spectroscopy (FT-IR) 2D imaging, using a focal plane array (FPA) detector to identify microplastics and their polymer composition. The results presented in this study will help facilitate deeper investigation into microlitter and microplastic pollution in the study area, as these baseline data will support the development of a specific monitoring plan for this purpose.

2. Study area

The study area includes the basin of the Entella Torrent (catchment area of 372 km^2) and its tributaries (Lavagna, Sturla and Graveglia torrents) as well as the sea bottom of the Gulf of Tigullio in front of the mouth of the Entella Torrent (Fig. 1). The study area has a total of 120,000 inhabitants and a very low population density, with only the territory of the municipality of Chiavari exceeding 2000 inhabitants km^{-2} (Fig. 1; www.istat.it) The most densely inhabited and greatly urbanised areas are located along the coast and by the banks of the Entella Torrent. Nevertheless, the inland population density is $<$100 inhabitant km^{-2}, with woodland representing 75% of the territory and only 2% dedicated to urban and infrastructural destination. Inland – especially along the Lavagna Torrent – are some productive activities (e.g. production and processing of thermoplastic material, vegetable oils, pinwheels, electrical or electronic equipment, zinc oxide, junkyards), several quarries (limestone and slate) and a landfill of solid urban waste (Fig. 1).

The coastal area of Chiavari and Lavagna is characterised by low and linear coasts, produced by the alluvial deposits of the Entella Torrent (Regione Liguria, 2011). The sea bottom is mainly sandy, with bathymetries parallel to the coast. In recent decades the eastern coastal zone has suffered from chaotic urbanisation, characterised by a low-quality modification of the territory and uncontrolled urban growth due to building speculation (Brandolini et al., 2017). The study area is affected by three sewage discharges (Fig. 1): one is in the municipality of Cicagna along the Lavagna Torrent (2500 inhabitants, treatment by Imhoff tanks), while the other two are off the coast of Chiavari and Lavagna (33,000 and 40,000 inhabitants, respectively, with treatment by biological oxidation tank).

The Gulf of Tigullio sees a general north-westward circulation parallel to the coast, characterised by brief periods of inversion and counter-currents with a south-eastward drift close to the coast (Fig. 1; Doglioli et al., 2004). The study area is characterised by a typical Mediterranean climate, with mean annual precipitation of 1763.5 mm and a mean precipitation trend marked by a maximum of 213 mm in October and November and a summer minimum of 65 mm in July. The Entella Torrent basin has a mean annual flow of 14.5 m^3 s^{-1} (Provincia di Genova, 2009).

3. Materials and methods

The materials and methods used were based on the guidelines of the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP), which advises the United Nations (UN) system on the scientific aspects of marine environmental protection. The study drew on the following literature: "Guidance on monitoring of marine litter in European seas" (Galgani et al., 2013) and "Guidelines for the monitoring and assessment of plastic litter and microplastics in the ocean" (GESAMP, 2019).

3.1. Sampling and measurements

Sediment sampling and microlitter analysis were undertaken following the recommendations of Galgani et al. (2013) and Marine & Environmental Research Institute - (MERI, 2012). Bottom sediments (250 mL of sediment at each station) were sampled along the banks of the Lavagna, Sturla, Graveglia and Entella torrents (24 sampling stations) with a steel spoon (2–3 cm of surface sediments) and on the sea bottom off the mouth of the Entella Torrent (32 sampling stations) between 5 and 50 m depth, with a Van Veen grab (2–3 cm of surface sediments). The sediment samples were stored in glass jars with Al-foil film between the

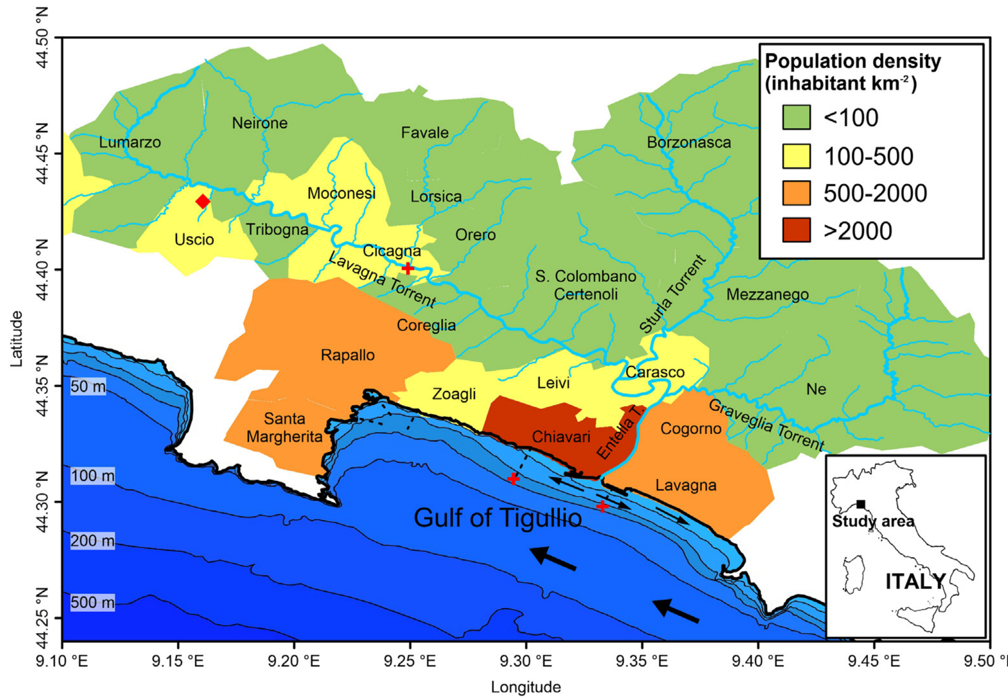

Fig. 1. Study area (NW Italy) and population density (in different colours; inhabitant km^{-2}). Red rhombus highlights the localisation of the landfill of solid urban waste; red plus-signs show sewage discharges in Lavagna Torrent and Gulf of Tigullio; black arrows show the dynamics near the coast and offshore.

cap and the sediment to avoid sample contamination. The distribution of the 56 sampling stations is shown in Fig. 2. Sampling was carried out along torrents in the dry period (May–June) and at sea in early summer (June).

Environmental parameters such as temperature and density were measured at every sampling station using a conductivity, temperature, depth (CTD) multiparametric probe. Current velocity and direction data (expressed in E and N components) were measured with an over-the-side-mounted current metre (vertical acoustic Doppler current profile, V-ADCP).

3.2. Laboratory treatment and analysis

In the laboratory, for each sediment sample, 50 mL were used for microlitter analysis, while the remaining 200 ml were used for granulometric analyses. Grain size analysis of the coarse fraction (Ø > 63 μm) was performed by dry sieving as indicated by Capello et al. (2016), whereas analysis of the fine fraction (Ø < 63 μm) was carried out using a Coulter Counter® Multisizer 3 (Beckman Coulter, Inc.). For microlitter analysis, 200 mL of a pre-filtered and concentrated hypersaline solution (1.2 g cm^{-3}) were added to 50 mL of sediment and stirred for 2 min. The mixture was then allowed to settle for 1 h; subsequently, supernatant was collected on cellulose acetate membrane with 0.45-μm porosity and a black grid. This extraction process was performed three times for each sample.

Each membrane had been conserved in a glass Petri dish and oven dried at 60 °C. Afterwards, the particles collected on the membranes were examined using a binocular microscope (Leica Z16 apochromatic microscope equipped with Leica DFC290 digital camera with 0.57x–9.2x zoom range). The items were counted, dimensionally measured and classified into different categories according to type (fragments, pellets, fibres, films, foamed items, granules, styrofoam and "others"), shape (cylindrical, discs, flat, ovoid, spheruloid, rounded, subrounded, subangular, angular and "others") and colour (white, cream, red, orange, blue, black, grey, brown, green, pink, tan, yellow and "others") (Galgani et al., 2013). Item appearance (lucid, opaque, crystalline, transparent) was divided from the colours. The number of items was referred to cm^{-3} of sediments.

For the dimensional aspect, the items were divided according to sediment grain size classes: fine sediments (Ø < 63 μm), very fine and fine sand (63 < Ø < 250 μm), medium sand (250 < Ø < 500 μm), coarse and very coarse sand (500 < Ø < 2000 μm) and gravel (Ø > 2000 μm). The correlation between the percentage of items for each dimensional class and the percentage of sediment of the same dimensional class found in each specific area was statistically tested.

Using a scanning electron microscope (SEM, Vega3, Tescan) equipped with an energy dispersive spectroscopy platform (Apollo X Silicon Drift Detector (SDD) Series, EDAX), some filaments were photographed to characterise their surface structure.

After recognition, all items collected at every station of each compartment (sea sediments, Lavagna Torrent sediments, Sturla Torrent sediments, Graveglia Torrent sediments) were separated from the membranes using a needle mounted on a wooden stick and divided into four types (fragments, spheres, white/transparent filaments, coloured filaments) and collected in four different glass jars containing milliQ water (a jar for each type for each study compartment), for 20 jars in total. These were then filtered again using Wathman GF/F Ø 47 mm filters for the next micro FT-IR chemical analysis. The samples from the Entella Torrent were not included in this analysis because this torrent collects material from its tributaries, hence any analysis would risk being a useless repetition.

3.3. Procedure for the selection/grouping of sampling stations and polymer determination

The 2D imaging-FTIR analysis of the samples was undertaken directly on the dry fibreglass filters (with no further sample preparation) using a Cary 620–670 FTIR microscope, equipped with an FPA 128 × 128 detector (Agilent Technologies). Spectra were recorded directly on the sample surface (or on that of the

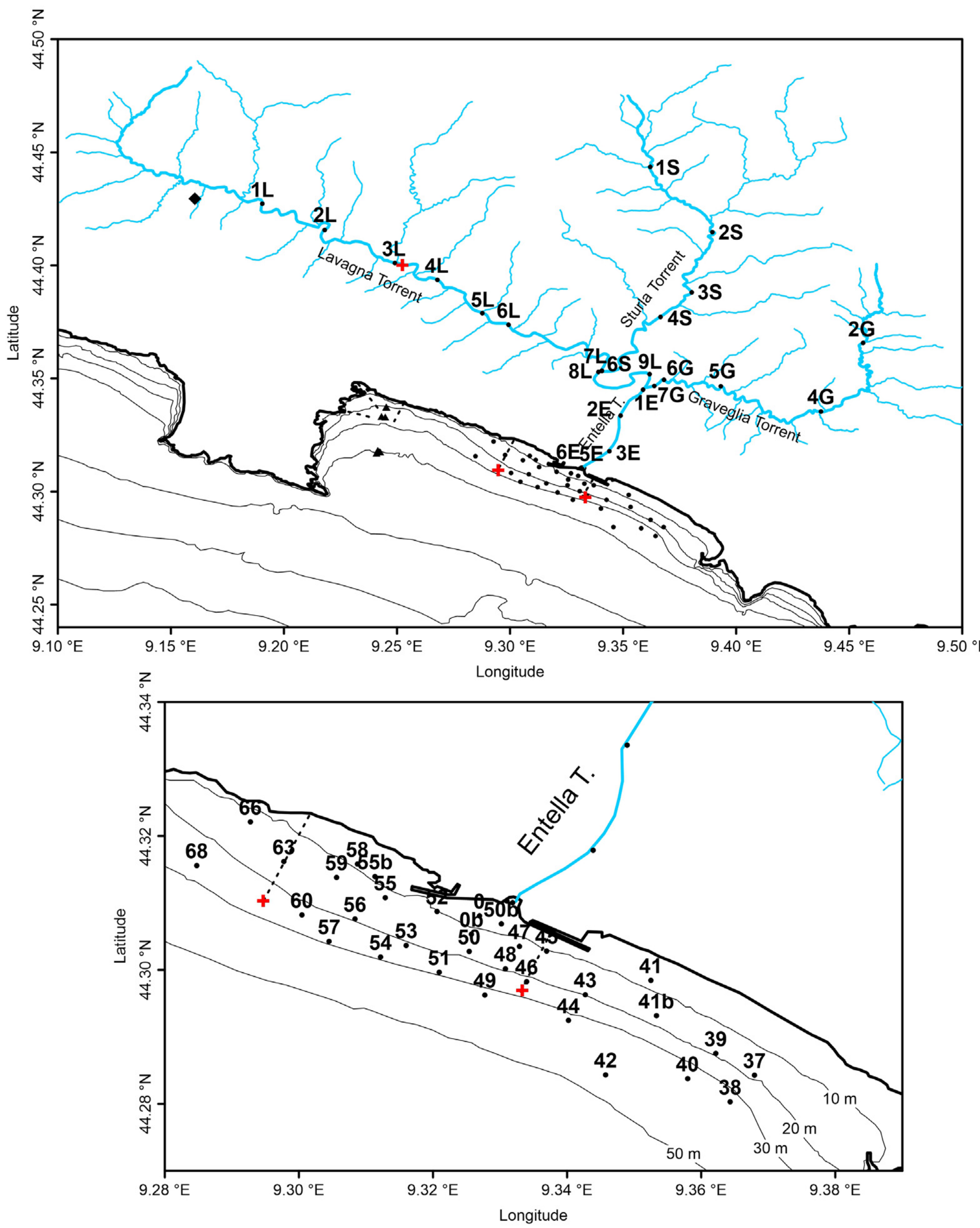

Fig. 2. Sediment sampling station distribution (black circles): (a) Entella Torrent and tributaries water basin; (b) Mouth of the Entella Torrent. Black rhombus shows localisation of the landfill of solid urban waste; red plus signs show sewage discharges on the Lavagna Torrent and in the Gulf of Tigullio.

Au background) in reflectance mode, with an open aperture and a spectral resolution of 8 cm^{-1}, acquiring 128 scans for each spectrum. A "single-tile" analysis resulted in a map of 700 × 700 μm^2 (128 × 128 pixels); the spatial resolution of each imaging map was 5.5 μm (i.e. each pixel had dimensions of 5.5 × 5.5 μm^2). Overall, 100 samples were analysed with this method. In each 2D map, the intensity of the characteristic bands of the plastic polymer investigated was imaged. The chromatic scale of the maps exhibited increasing absorbance of the bands as follows: blue<green<yellow<red. In some cases, spectra were collected in attenuated total reflectance (ATR) mode to grant higher signal-to-noise ratios (S/N) and to permit easier identification than is feasible in reflectance mode. In such cases, a GeATR crystal (Agilent Technologies) was mounted on the FT-IR microscope. In ATR mode, a 64 × 64 FPA grid was used, with a spatial resolution of 1.1 μm and a field of view of 70 × 70 μm^2.

4. Results and discussion

A total of 4302 items were found in 56 sediment samples, with mean concentrations of 1.5 items cm^{-3} ± 1.3 standard deviation (SD) and 1.6 items cm^{-3} ± 1.3 SD in torrent and sea sediment samples, respectively. The concentrations found were higher than those reported for example by Norén (2007) and Thompson et al. (2004) in Sweden (coastal areas) and the United Kingdom (beaches and estuarine and subtidal sediments), respectively. These were the only studies with microlitter concentrations comparable to the present study.

The results of the sediment granulometric analysis (shown in Table 1) highlight the predominance of coarse sediment (Ø > 63 μm) both in torrents and sea samples. Indeed, all torrent samples were mainly characterised by coarse and very coarse sand (500 < Ø < 2000 μm; from 49.3% to 58.8%), whereas in the

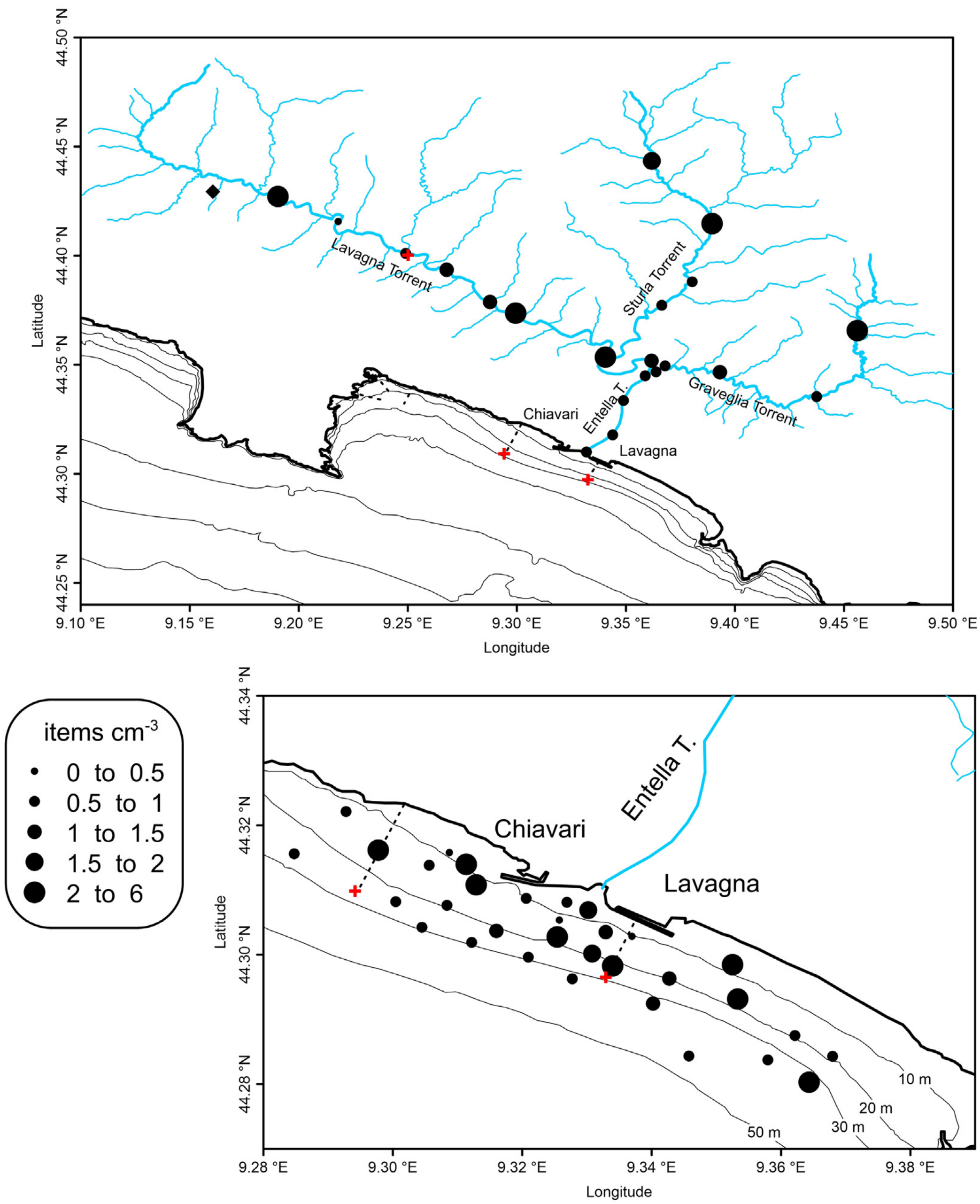

Fig. 3. Concentration of items (black circles) in torrent and sea sediments. (a) Entella Torrent and tributaries water basin; (b) Mouth of the Entella Torrent. Black rhombus shows the landfill of solid urban waste; red plus-signs show sewage discharges.

sea a prevalence of medium sand ($250 < \emptyset < 500$ μm; 46.4%) was found.

The item distribution, graphically shown in Fig. 3, indicates that the highest concentrations of items were found in the upper part of torrents, at torrent confluences and off the mouth of the Entella Torrent within the bathymetry of 20 m. In particular, the maximum concentration of items (6.0 items cm^{-3}) found in the torrent samples was centred on station 1L (along the Lavagna Torrent), which is situated within a town and crucially is immediately downstream of a landfill of solid urban waste. After this point (station 1L), the samples (from 2L to 5L) showed a relatively constant number of items, without a progressive decrease from upstream to downstream areas of the torrent, contrary to the findings of Rodrigues et al. (2018) in the Antuã River (Portugal), for instance. Such findings likely owe to the presence of other unknown sources of waste along the waterway (e.g. the landfill of solid urban waste present along the Lavagna Torrent); furthermore, despite the considerable distances between sampling stations, no decrease in the quantities of items was noted. Regarding the Sturla Torrent, the samples that proved the richest in items found were 1S and 2S, positioned upstream of a small dam in the torrent (a site of possible microplastic accumulation) and in the town of Mezzanego, respectively. In the Graveglia Torrent, the richest sample, positioned downstream of some small villages and quarries, was 2G. These peculiar distributions seem to confirm the presence of direct inputs (such as the landfill of solid urban waste) and the influence of human activities and populated areas that contributed in various ways to the item distribution seen in the study area (Peng et al., 2018). At sea, the influence of the port input seems to be clear. Certainly, ports have been shown to manifest the highest levels of microlitter contamination in coastal studies (Claessens et al., 2011; Tsang

Table 1
Mean grain size concentrations (%) and standard deviations (std dev) of both torrent and sea sediment samples.

Sediment grain size	Lavagna Torrent (% ± std dev)	Sturla Torrent (% ± std dev)	Graveglia Torrent (% ± std dev)	Entella Torrent (% ± std dev)	Sea (% ± std dev)
Silt and clay (<63 μm)	4.5 ± 3.1	4.0 ± 2.0	2.6 ± 1.4	2.8 ± 3.7	25.8 ± 20.7
Very fine and fine sand (63–250 μm)	9.4 ± 12.6	6.1 ± 5.2	3.1 ± 2.3	5.2 ± 7.0	23.5 ± 10.4
Medium sand (250–500 μm)	26.6 ± 21.7	20.5 ± 13.8	11.3 ± 9.4	21.5 ± 22.2	46.4 ± 24.6
Coarse and very coarse sand (500–2000 μm)	49.3 ± 26.3	51.6 ± 17.2	58.8 ± 10.6	55.8 ± 33.6	4.2 ± 13.2
Gravel (>2000 μm)	10.2 ± 15.6	18.0 ± 11.9	24.2 ± 14.1	14.7 ± 15.9	0.1 ± 0.1

et al., 2017) due to activities such as boating, fishing and shipyards. The item distribution is linked to the laboratory technique used, hence the employment of a concentrated hypersaline NaCl solution (solution density 1.2 g cm^{-3}) may have caused the total number of items to be underestimated due to the sinking of the denser (heavy) and thus uncollected objects (Graca et al., 2017; Hidalgo-Ruz et al., 2012).

The items in the sediment samples generally assumed two shapes (Fig. 4a): fibres (54%) and fragments (26.2%) [with rounded or subrounded fragments (15.5%) and angular or subangular fragments (10.7%)], as found by various other authors (Abidli et al., 2018; Guerranti et al., 2017). In particular, fibres are the most common type found across the world (Abidli et al., 2018; Claessens et al., 2011; Gewert et al., 2017; Graca et al., 2017; Guerranti et al., 2017; Peng et al., 2017). In the present study, fibres were more often seen in sediment torrent samples (63.3%) than at the sea bottom (47.2%); in fact, the number of fragments and spheruloid pellets increased in marine sediment samples (20.2% and 13.3% for rounded and angular fragments, respectively, and 6.9% for spheruloid pellets). This may have owed to particles different sedimentation behaviours, related not only to their density but also their shape. Indeed, fibre sedimentation may be much higher than that of spheruloid particles with the same mass due to the fibre larger surface and consequently to their faster time of fouling and the related density increase (Chubarenko et al., 2016).

A white-cream colour characterised 50.8% of the items found in the sediment samples (Fig. 4b), 66.7% being fibres. Other significant colours in the samples were black and blue, with 13.1% and 10.7%, respectively. Moreover, fibres were the main items displaying these colours. Red and orange (13.2% of total items) were the most represented colours in sub- or rounded and sub- or angular fragments reaching 30%. A transparent appearance (Fig. 4c) was linked to the white colours (fibres and ovoid pellets), whereas an opaque appearance was mainly linked to red–orange and blue–black colours (fragments and fibres). Furthermore, a transparent appearance was mainly linked to fibres, as also found by Graca et al. (2017). The samples mainly comprised items with dimensions of 63 < Ø < 250 μm (36.1%; Fig. 4d), followed by items with dimensions of 500 < Ø < 2000 μm (26%) and 250 < Ø < 500 μm (20.6%). Items with dimensions of Ø > 2000 μm were poorly represented (only 5.2%) – as additionally noted by Graca et al. (2017) and Fastelli et al. (2016) – and consisted of long, transparent fibres of different colours (93%), an item type that can reach very high dimensions, as reported by Gewert et al. (2017). Statistical analysis showed no significant correlation (R^2 = 0.1632) between the percentage of items in each dimensional class and the percentage of sediment in the same dimensional class found in each specific area (Fig. 5).

Focusing on the detailed distribution of item types and shapes in the study area (Fig. 6), it is possible to note changes in composition close to both town and torrent confluences. Certainly, along the Lavagna Torrent (Fig. 6a) from station 1L to station 3L was a decrease in type variability up to the net prevalence of fibres (93.9%), while from 4L (after the sewage discharge of Cicagna) to 9L a considerable increase in variability accompanied the presence of fragments, pellets and granules. After the

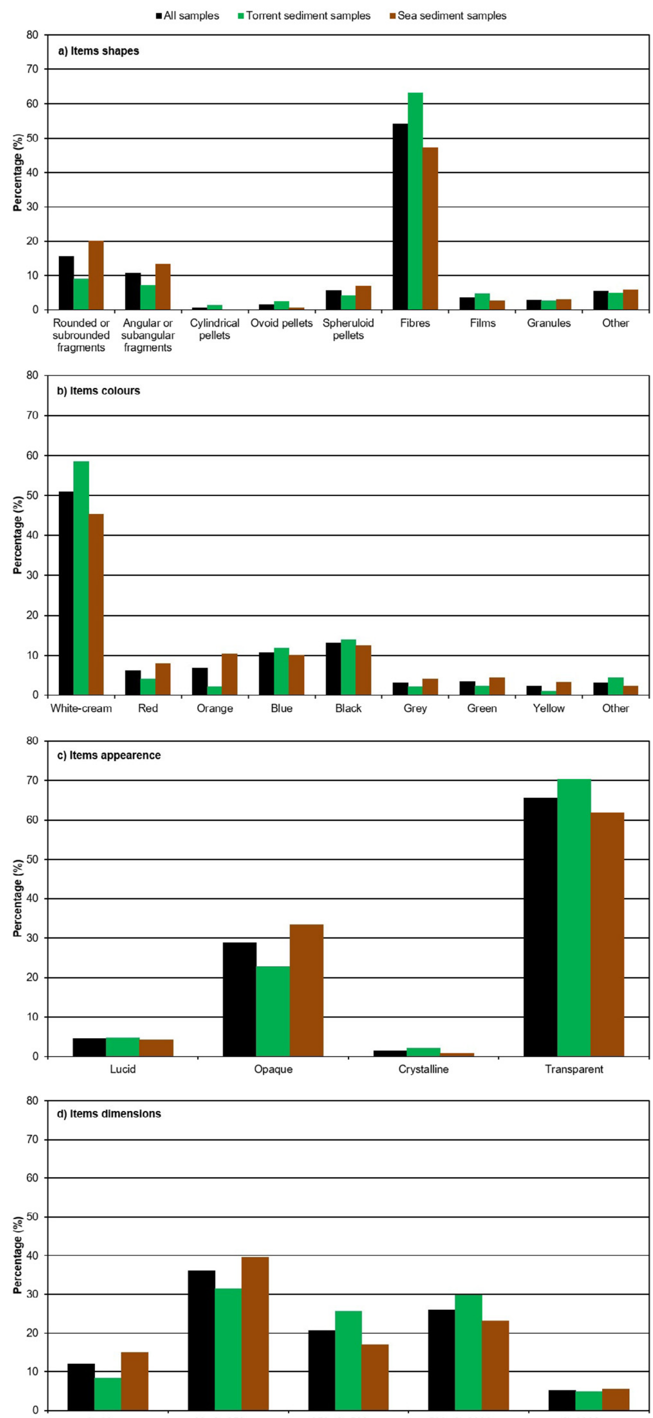

Fig. 4. Percentage distribution of items types and shapes (a), colours (b), appearance (c), and dimensions (d).

Table 2
Assignments for each specific sampling area and type of polymer found.

Site	Polyamide (nylon)	Polyurethane/PET blend	Acrylonitrile-acrylate	Polystyrene
Graveglia Torrent	1 (sphere)	1 (fibre)	1 (fibre)	
Sturla Torrent	3 (1 sphere + 2 fragments)	2 (fragments)	1 (fibre)	
Lavagna Torrent	5 (1 sphere + 3 fragments + 1 fibre)		1 (fibre)	
Marine sediment	7 (4 spheres + 3 fragments)			1 (fragment)

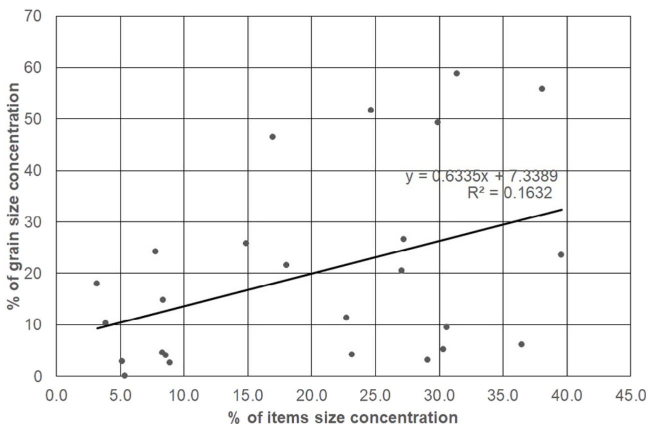

Fig. 5. Correlation between the percentage of items for each dimensional class and the percentage of sediment of the same dimensional class found in each specific area.

confluence of the Sturla Torrent downstream of the town of Carasco, items of "others" types increased to 26.8% at station 8L. Moreover, along the Sturla Torrent (Fig. 6b), station 1S was characterised by pellets, fragments and fibres, from 2S (localised downstream of the town of Borzonasca) there was an increase in fibres (more 79.4%), whereas from 3S fragments increased again (39.5%). Lastly, in the upper part of the Graveglia Torrent (Fig. 6c) was a general prevalence of fibres (>66.0%), although all item types were well represented (granules excluded) at station 5G in the town of Conscenti, where a local waste separation and recycling unit is located. Furthermore, in this case it is possible to highlight the influence of cities and human activities in terms of changes in item composition. Changes along the waterway have also been observed in other cases across larger geographical areas (e.g. the case of the Rhine River; Mani et al., 2015) due to the presence of cities and metropolitan areas, industries, wastewater treatment plants, tributaries and weirs. Nevertheless, it is difficult to relate the observed item types and concentrations to specific local human activities, as already indicated by Claessens et al. (2011).

At the sea bottom (Fig. 6e) immediately off the mouth of the Entella Torrent, the sampled items mainly consisted of fibres, these being the most represented type in the torrent sediments. This finding confirms the influence of torrents as a vector of microlitter from land to sea. The coastline sea bottom samples also comprised numerous fragments, especially in the samples off the ports of Chiavari and Lavagna. A substantial number of different items were expected in front of port areas, as such environments present various anthropogenic activities; in addition, ports are usually characterised by a particular morphology, for example due to the presence of breakwaters, which create

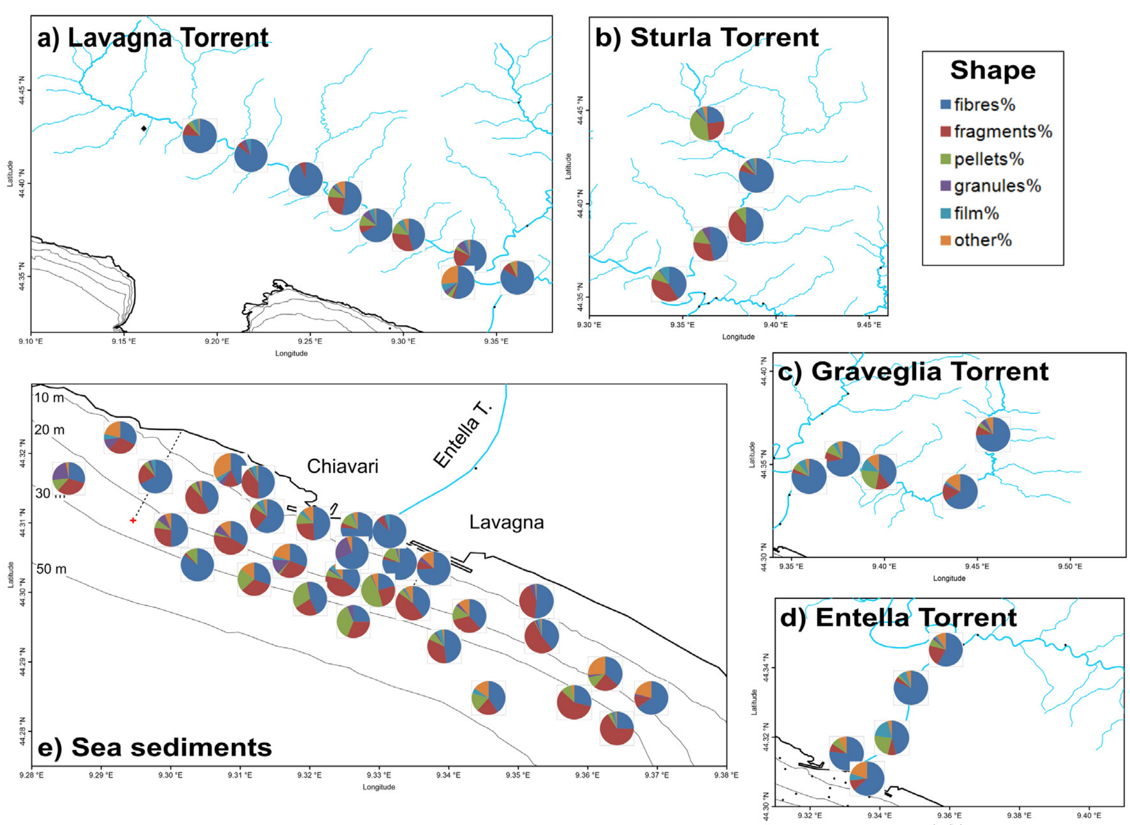

Fig. 6. Distribution of item types in sampling stations: (a) Lavagna Torrent, (b) Sturla Torrent, (c) Graveglia Torrent, (d) Entella Torrent, (e) sea bottom sediments.

distinctive current conditions that can trap items inside the port basin (Claessens et al., 2011; Tsang et al., 2017). For a deeper analysis, the sea sediment stations were divided into three groups based on bathymetry: depth <10 m (11 stations), 10–20 m deep (11 stations) and depth >20 m (10 stations). The abundance of different item types at each depth was then calculated (Fig. 7). As shown in Fig. 7, fibres decrease from the coast to the open sea. This is probably due to the dynamics of the coastal area (front area between fresh and sea waters) that allow the deposition of these light and abundant types of items, whereas counter-currents lead to their transportation along the coast. In fact, in the open sea, fibre deposition is counteracted by the current system. However, fragments, pellets and "others" item types increased at the 10–20 m depth where, despite the currents, they could be deposited on the sea bottom due to their weight. This suggests that some fragments, pellets and "others" item types originated in neighbouring areas and were consequently carried by currents in the study area. In terms of pellets, the richest samples were located west of the sewer discharge of Lavagna and they mainly comprised spheruloid pellets and microbeads (Fig. 6e). Different studies have already found microlitter pellets (and in particular microbeads) in marine environments in both water column and sediments (Cheung and Fok, 2017; Tsang et al., 2017), with some indicating that such forms of microlitter are often released from sewage because they are not retained by the sewage treatment plants (Cheung and Fok, 2017; Duis and Coors, 2016; Kalčíková et al., 2017). Granules and film decreased at depths greater than 20 m. The reduction in item abundance from the coast to the most distant stations highlights that the influence of the Entella Torrent and the anthropogenic impact was concentrated near the shore. In addition, Zobkov and Esiukova (2017) have found differences in microlitter type distribution related to sea dynamics, with the maximum concentration of fibres near the shore and a steady decrease moving away from the coast, as true in the present study. However, these authors have also highlighted the presence of fragments only near the coastline (<10 m from the coast). From this, it can be deduced that microlitter distribution is site-specific and depends on both the distinctive dynamics of the area and the specific microlitter input (De Carvalho and Neto, 2016; Mathalon and Hill, 2014; Van Cauwenberghe et al., 2013).

Some fibre images were taken with SEM to highlight their surface morphology. The details of the fibre surfaces are reported in Fig. 8: the presence of fractures, protrusions and other irregularities indicated that the fibre had been damaged during transportation, as additionally indicated by Zhou et al. (2018).

Regarding the dynamics of the study area, obtained using an ADCP, two different currents could be observed in the surface layer in the western part of the gulf: one current to the north, the other to the south, but both exhibiting the same direction (SE-NW) and intensity (11.6 cm s^{-1}). The current in the northern part close to Rapallo and Zoagli tends to turn back to the south in the form of a coastal current, whereas the current in the southern part turns towards the open sea, intercepting the main Ligurian current, also known as the Ligurian-Provençal Current or Northern Current. In the sea area off the mouth of the Entella Torrent, two flows could instead be observed along the coast: one directed eastward and one westward with respect to the mouth.

Among all the fibres/fragments analysed with 2D imaging-FTIR (77 samples), 46 (60%) were identified as cotton fibres, 16 (21%) as polyamide (nylon), 3 (4%) as acrylonitrile-acrylate fibres, 3 (4%) as polyurethane/PET blends, 1 (1%) as polystyrene and 8 (10%) were unassigned/unknown or non-plastic samples. Table 2 shows assignments for each specific sampling area and type of polymer found.

Despite cotton never being identified as a pollutant, the consistent presence of cotton fibres may be alarming, as such items

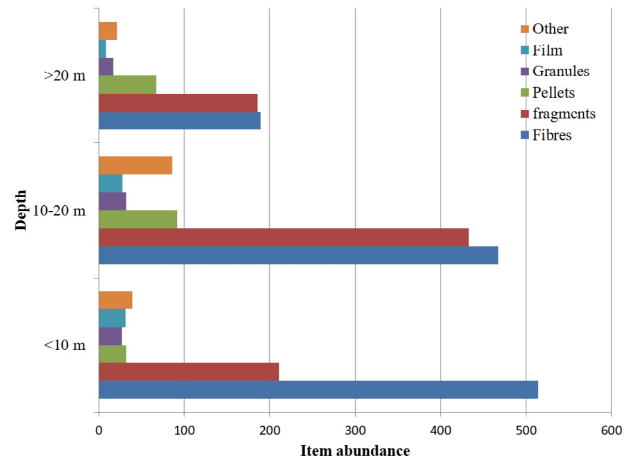

Fig. 7. Distribution of abundance of items divided by types, in the sea sediment sampling stations, at <10-m, 10–20-m and >20-m depth.

are often treated with various colour additives, including azoic pigments that are used in the textile industry (Christie, 2001). These colourants can be dangerous if they are ingested or dispersed into the environment, for example dinitroaniline orange and ortho-nitroaniline orange (Gestis substance database; http://gestis.itrust.de/nxt/gateway.dll/gestis_en). As is true of microplastics, cotton fibres can be affected by processes of bioaccumulation and biomagnification in marine organisms, as observed by Remy et al. (2015). It is necessary to avoid underestimating the presence of cellulose fibres, as these may prove problematic in the future. The micro-FTIR for the polymer analysis revealed that nylon and acrylonitrile/acrylate were the most common polymer types. The presence of these types of contaminants is very worrying because they can convey pollutants through the food chain; indeed, microplastics such as nylon and acrylonitrile as well as PET and polystyrene have been found in organisms that are consumed in large proportions by humans, such as fish (e.g. sea bass, Atlantic cod, common carp), oysters and mussels (Hanachi et al., 2019; Karbalaei et al., 2018). The high percentage of non-plastic materials found here highlights how it is necessary to undertake further analysis in order to identify and characterise polymers, such as through FT-IR spectroscopy. Simple visual identification can lead to an under- or over-evaluation of microplastic particles (Song et al., 2015). In the present study, not all items were analysed using FT-IR spectroscopy, because the purpose was simply to collect preliminary data to develop a specific monitoring plan of the area, in order to focus on the sites with the most intense concentration of microlitter and to improve the characterisation of microplastics in those places where plastic litter is most abundant.

5. Conclusions

The distribution of the items found in the sediments was characterised by considerable variability in both torrent and sea samples. The highest item concentrations were found in stations located close to towns and urban waste landfill sites, in torrent confluences and near the mouth of the principal torrent of the area and port entrances. The implication of such findings is that different human activities had different impacts on item distribution.

Fibres predominated as well as the transparent appearance, mainly linked to the fibre shape. The second-most common shapes were fragments, which especially characterised the samples off the ports along the coast. Fibres were predominant in torrent sediments and in sea sediments close to the mouth of the

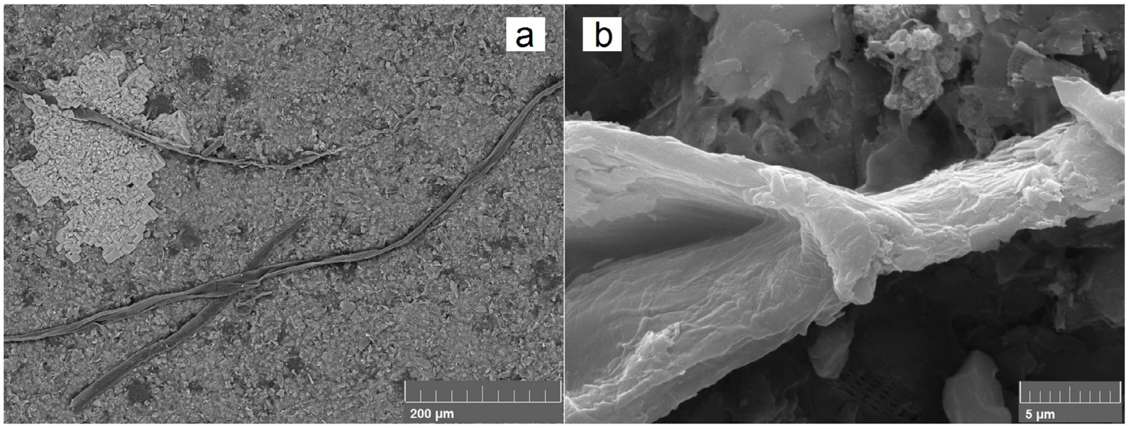

Fig. 8. Details of fibres captured with the Scanning Electron Microscope (SEM, Tescan Vega 3 LMU).

Entella Torrent, hence torrents can be confirmed as a vector of microlitter from land to sea. White-cream was the most common colour and was mostly associated with fibres, whereas red and orange were primarily linked to fragments. Items with larger dimensions (Ø > 2000 μm) were poorly represented (4.4%) and were constituted by fibres, while items with smaller dimensions (Ø < 250 μm) were far more abundant (75.6%).

The sea current measurements allowed to evaluate the effect of dynamics on items. It is probably true that the main currents can intercept the marine litter that comes from inland areas through the torrents investigated, transporting it in the eastern part of the gulf.

The prevalence of non-plastic materials found through chemical analysis suggests that visual analysis of samples alone is not enough to characterise microlitter and identify microplastics. Standardisation of the methods and spectral analysis used for microlitter sampling and analysis is necessary in order to facilitate comparison with the results reported by different studies.

This study has only aimed to be a baseline investigation on microlitter pollution and composition in the study area. This is why we decided to analyse the chemical composition of merely a percentage of all the items found in the sediment samples, while also defining priorities based on cost-effectiveness. The purpose has been to deeply analyse all items using visual analysis and then to focus only on those items that might reveal interesting information about different item composition as well as the various polymer types that constitute them. This baseline study should prove useful in developing a plan in the future to monitor those areas that manifested a higher concentration of microplastics, enabling more thorough information to be collected to characterise microplastics. In addition, further studies should consider seasonal differences in microlitter and microplastic composition in the study area and characterise the areas neighbouring the hotspots in order to develop a more complete picture of microplastic transportation and distribution. All of this information is necessary to improve the management of microplastic pollution in the area.

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.

CRediT authorship contribution statement

Laura Cutroneo: Conceptualization, Data curation, Writing - original draft, Writing - review & editing. **Alessandra Cincinelli:** Methodology, Data curation, Validation, Writing - original draft. **David Chelazzi:** Data curation, Writing - original draft. **Alessia Fortunati:** Investigation, Formal analysis. **Anna Reboa:** Data curation, Formal analysis, Writing - review & editing. **Sara Spadoni:** Data curation, Writing - original draft. **Enrico Vena:** Investigation, Writing - original draft. **Marco Capello:** Supervision, Methodology, Validation, Project administration, Funding acquisition.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.rsma.2020.101119.

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