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Floating plastics in Adriatic waters (Mediterranean Sea): From the macro- to the micro-scale

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ABSTRACT

Macro- and microplastics abundances were determined in the Adriatic Sea following the MSFD TG10 protocol. The studied areas included populated gulfs, river outlets and offshore waters in five Adriatic countries. The use of small ships enabled us to detect small sized plastics (2.5–5 cm) and record average macroplastics densities of 251 ± 601 items km^{-2}, one order of magnitude higher than previously considered. Results from manta net tows for microplastics revealed an average abundance of 315,009 ± 568,578 items km^{-2} (217 ± 575 g km^{-2}). We found significantly higher microplastics abundances in nearshore (≤4 km) than in offshore waters (> 4 km) and this trend seems to affect also the small sized macro plastic fragments (2.5–5 cm). The dominant polymers were polyethylene and polypropylene while the presence of some rare polymers and waxes used in food and dentistry indicated waste water treatment plants as potential sources of microplastics.

1. Introduction

In the past years, the increasing awareness regarding the ubiquitous presence of plastics, including microplastics, in the marine environment has alerted both the scientific and policy-makers' communities towards the study and mitigation of this type of pollution (UNEP, 2005; Rochman et al., 2013a; Galgani et al., 2015). Plastics production has risen from 1.5 million tons in 1950 to > 335 million tons today (Plastics Europe, 2017). The dependence of the modern society on plastics - in particular packaging and single-use products - is increasing exponentially as is consequentially also the production of plastic waste. As a result, the waste management and recycling sectors are lagging behind and plastics are accumulating in the environment. It has been estimated that between 5 and 13 million tons of plastics have thus ended up in our oceans in 2010, a figure expected to rise in the near future (Jambeck et al., 2015).

Growing scientific evidence documents the presence of plastics and microplastics in both populated coastal areas (Browne et al., 2011; Thiel et al., 2011; Ryan, 2013; Zhou et al., 2016; Suaria et al., 2016) and remote parts of the world such as the polar seas and deep abyssal environments (Lusher et al., 2015; Eriksson et al., 2013; Goldstein et al., 2013; Bergmann et al., 2017; Courtene-Jones et al., 2017) and plastic pollution has been recognized a global environmental problem of our times. Plastic pollution has several adverse effects in the marine environment including alterations in biodiversity and ecosystem health (Gall and Thompson, 2015; Rochman et al., 2016), entanglement and ingestion by marine biota (Allen et al., 2012; Foekema et al., 2013; Fossi et al., 2018), leaching of chemicals (Rochman et al., 2013b; Kwon et al., 2014). Plastic pollution has also socio-economic consequences as it is directly related to tourism, shipping, fishing and aquaculture

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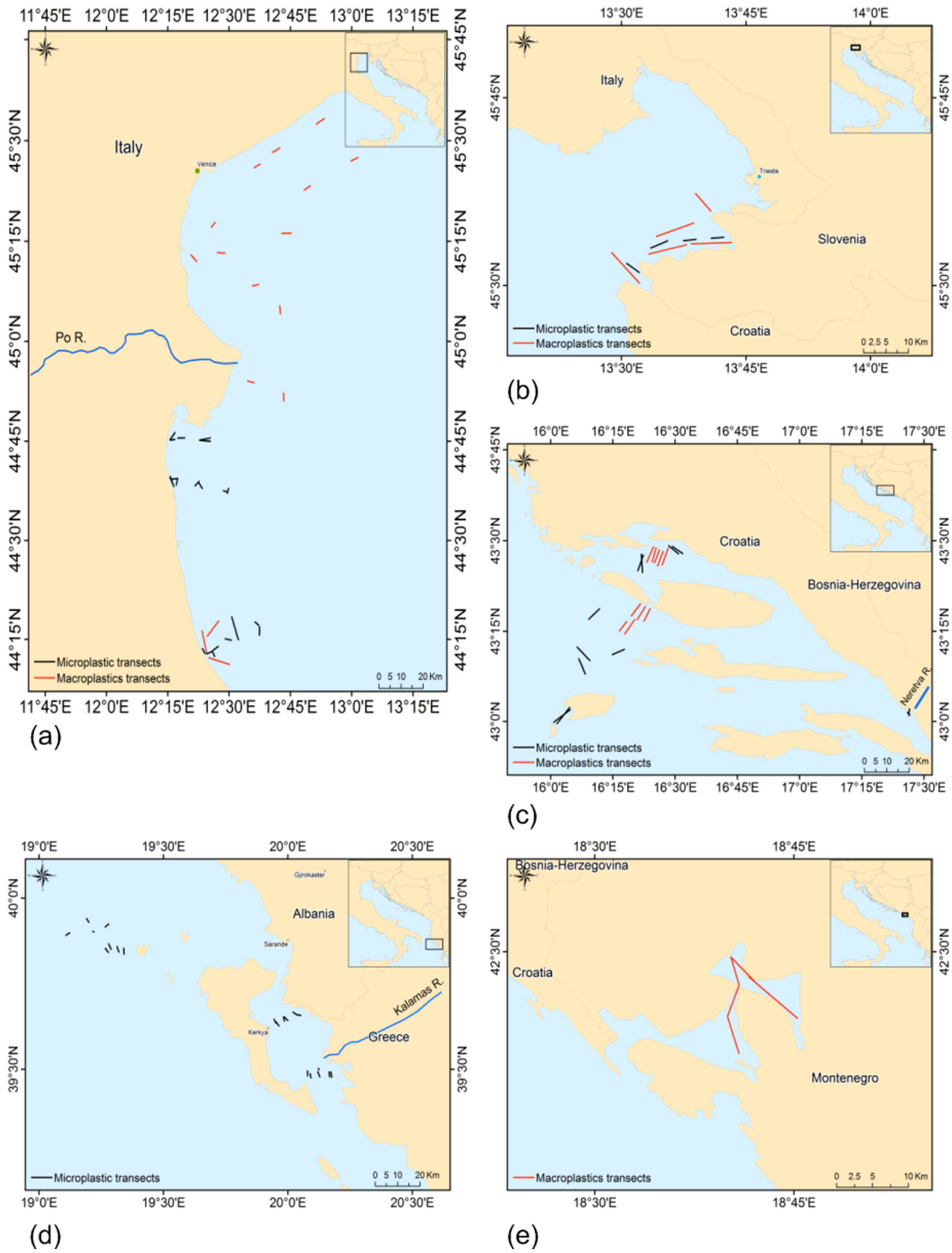

Fig. 1. Observational transects for macroplastics (red lines) and manta-net sampling transects for microplastics (black lines) in: (a) Gulf of Venice and Cesenatico waters; (b) Gulf of Trieste; (c) Gulf of Split and Croatian channel waters; (d) Gulf of Corfu and South Adriatic waters; (e) Gulf of Kotor. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

activities and human well-being at large (Jang et al., 2014; Brouwer et al., 2017; Vlachogianni, 2017). Despite the acquired knowledge, many questions still remain open regarding the abundance and transport mechanisms of plastics, potential preference to specific marine organisms, long-term effects and their risks to the ecosystems and human health.

Recent studies provide evidence that plastic pollution in the Mediterranean Sea is significant (Aliani et al., 2003; Collignon et al., 2012, 2014; de Lucia et al., 2014; Fossi et al., 2012; Pedrotti et al., 2016; Suaria and Aliani, 2014; Faure et al., 2015; UNEP/MAP, 2015; Gajšt et al., 2016; Ruiz-Orejón et al., 2016, 2018; Suaria et al., 2016; Arcangeli et al., 2017; Di-Méglio and Campana, 2017; Gündoğdu and Çevik, 2017; van der Hal et al., 2017; Baini et al., 2018) with floating microplastics concentrations comparable to those of oceanic accumulation zones (Cózar et al., 2015). The coastal population (~160 million residents in 2012) and tourism (~350 million overnight stays per year estimated in 2012) (UNEP/MAP, 2012) in combination to the enclosed character of this sea and the specific thermohaline and mesoscale surface circulation features (Millot and Taupier-Letage, 2005; Robinson et al., 2001) are considered the major causes of the high amounts of litter and plastics recorded. In particular the Adriatic Sea is a narrow elongated sub-basin with a high land to sea ratio (1.80) (Ludwig et al.,

Table 1
Conditions during observational surveys for macroplastics and manta-net trawlings for microplastics at the various sub-areas studied.

	Area	Vessel Speed (knots)	Obs. Height (m)	Wind Speed (Beaufort)	No of transects (seasonal replicates)	Distance from coast (km)	Total distance covered (km)
Macroplastics	Gulf of Kotor (Montenegro)	2	2.5		(1 × 3) + 1 = 4	1–2	146
	Gulf of Split Channel waters (Croatia)	2.8 ± 0.17	3	1.4 ± 1.1	(10 × 2) = 20	2–10	101
	Gulf of Trieste (Slovenia)	2.9 ± 0.44	2.5	2 ± 1.4	(5 × 3) = 15	4–7	74
	Gulf of Venice (Italy)	2.4 ± 0.15	3.2	1 ± 1.3	(8 × 2) + 5 = 21	4–33	56
	Cesenatico (Italy)	3	1	2 ± 0.9	(3 × 2) = 6	4–5	39
	Total				66		415
Microplastics	Gulf of Split Channel waters (Croatia)	2.5 ± 0.4	–	0.8 ± 0.6	(5 × 2) + 5 = 15	0.9–11	32
	Gulf of Trieste (Slovenia)	2.5 ± 0.3	–		4 × 2 = 8	0.8–2	18
	Cesenatico (Italy)	2.4 ± 0.3	–	1.5 ± 0.7	11 × 2 = 22	0.52–19	37
	Gulf of Corfu S. Adriatic (Greece)	1.9 ± 0.1	–	1.1 ± 0.6	10 × 2 = 20	1.58–21	39
	Total				66		126

2009) surrounded by seven countries (~3.5 million inhabitants in the coastal zone) and many tourist centers (e.g. Venice, Split, Dubrovnik, Corfu). Other anthropogenic activities include extensive maritime traffic, fishing, aquaculture and offshore exploitation. About 1/3 of the Mediterranean freshwater discharges flow into the Adriatic Sea and the river Po ranks second in terms of Mediterranean rivers water discharge (1570 $m^3 s^{-1}$) (Ludwig et al., 2009). These conditions lead to a substantial accumulation of marine litter and plastics, as it has been reported for beaches (Laglbauer et al., 2014; Munari et al., 2016; Poeta et al., 2016; Prevenios et al., 2018; Vlachogianni et al., 2018), seafloor (Strafella et al., 2015; Pasquini et al., 2016; Mistri et al., 2017; Melli et al., 2017) and sea surface (Suaria and Aliani, 2014; Suaria et al., 2016;, Gajšt et al., 2016; Arcangeli et al., 2017). At the same time the impact of plastics on marine biota has been reported regarding ingestion (Anastasopoulou et al., 2018; Digka et al., 2018; Pellini et al., 2018), rafting (Tutman et al., 2017) or as pathogen vector (Kovač Viršek et al., 2017). Efforts on modeling the transport of floating plastics in the Adriatic Sea have shown that the prevailing cyclonic circulation with two alongshore surface currents is critical in determining the distribution and residence time of plastics in this basin (Liubartseva et al., 2016; Carlson et al., 2017, implemented in Gajšt et al., 2016).

Our study aims to describe the distribution, abundance, sizes and composition of floating plastics including both macroplastics (2.5 cm–100 cm) and microplastics (330 μm–5 mm) on a transnational level along the Adriatic coasts. The work was conducted in line with the EU Marine Strategy Framework Directive (MSFD, 2008/58/EC) and common harmonized methodologies have been used in five Adriatic countries: Italy, Slovenia, Croatia, Montenegro and Greece. To our knowledge this is the first time, at least for the Adriatic Sea, that two approaches have been used to report floating plastic debris from the macro to the micro size for the same spatiotemporal coverage. In addition, this work is one of the few (Suaria et al., 2016; Pedrotti et al., 2016; Vianello et al., 2018) providing information on the polymeric composition of floating microplastics in the Mediterranean Sea. The results of the present work are useful for the setup of monitoring programmes and provide additional scientific understanding towards the assessment of plastic pollution on a regional level.

2. Materials and methods

2.1. Study areas

Surveys for floating plastics were conducted in five gulfs of the Adriatic Sea covering the most important population and touristic centers and harbours, namely: Gulf of Venice (Italy); Gulf of Trieste (Slovenian waters); Gulf of Split (Croatia); Gulf of Kotor (Montenegro). Three more study areas include Italian waters off Cesenatico (south of the Po River delta and Cesenatico River); Croatian channel waters of the Middle Adriatic; Neretva river outlet and southern Adriatic waters close to the Otranto strait. We have also studied the Gulf of Corfu and Kalamas river outlet (Greece) at the southern boundary of the Adriatic with the Ionian Sea (Fig. 1). In the Venice and Kotor gulfs data were collected only for macro plastics, while in Neretva river outlet, the Gulf of Corfu and southern Adriatic waters only for microplastics. Overall, the waters studied were at distances ranging from ~100 m to ~35 km from the closest coast. All areas were visited 2 to 3 times during the years 2014–2015 on a seasonal basis (autumn–winter 2014–15; spring–summer 2015).

2.2. Visual Observations for macroplastics (> 2.5 cm)

Visual observations for macro plastics (> 2.5 cm) were conducted following the protocol proposed by the MSFD TG10 Guidance on Monitoring of Marine Litter in European Seas (Galgani et al., 2013). Small-sized ships (observation height < 3.2 m) were used and in total 66 transects were conducted, some of them were repeated 2 or 3 times on a seasonal basis (Oct.–Dec. 2014; Apr.–July 2015) at the studied areas (Fig. 1). A distance of 415 km was covered corresponding to 89 h of observations. The length of each transect was measured according to start-end GPS points. Detailed information on the observations made is presented in Table 1. Observations were conducted always from one side of the ship, by two observers which rotated in order to avoid fatigue and without the use of binoculars. All surveys were performed under low wind speed conditions recorded with a portable anemometer (< 4 $m s^{-1}$).

All litter items were identified according to their type (G) and size as described in the MSFD TG10 Master List for floating litter. Six size classes were recorded (2.5–5 cm; 5–10 cm; 10–20 cm; 20–30 cm; 30–50 cm; > 50 cm). The total surface of the surveyed area was estimated by multiplying the transect distance by the observation width and then litter density (items km^{-2}) was calculated by dividing the items count with the surveyed area surface. No specific methodology (Buckland et al., 1993) or correction factors (Ryan, 2013) regarding the effective strip width were applied. It was assumed that the detection efficiency for all items larger than 2.5 cm was highest at a distance of 10 m from the side of the boat. Observations from heights similar to the ones reported in this paper have been conducted also by Thiel et al. (2003) and Suaria et al. (2015) (1 m and 4 m respectively). These surveyors conducted width measurements and estimated that 10 m was their observation width.

2.3. Sampling of microplastics (≤5 mm)

Sampling and analysis of floating microplastics was carried out by four different laboratories (Institute for Oceanography and Fisheries (IOF) – Croatia; Institute for water of the Republic of Slovenia (IWRS) – Slovenia; Regional Agency for Environmental Protection in the Emilia-Romagna region (ARPAE) –Italy; Hellenic Centre for Marine Research (HCMR) –Greece) using a common methodology (Ryan et al., 2009; Eriksen et al., 2014; Kovač Viršek et al., 2016). A total of 65 tows using manta-nets were conducted along the Adriatic coasts (Fig. 1; Table 1). All manta-nets used had the same dimensions (W 60 cm × H 24 cm rectangular frame opening; 3 m length) and net opening of 330 μm. Samplings were carried out using small vessels at low wind conditions recorded by a portable anemometer or by ship's instruments. As suggested for manta net samplings (Kukulka et al., 2012), wind velocity was always $< 0.4\ m\ s^{-1}$ and water friction velocity $< 0.6\ cm\ s^{-1}$. The water friction velocity (u*) ($cm\ s^{-1}$) was calculated as $u^* = \sqrt{\tau/\rho_w}$ (where τ is the wind stress during samplings and $\rho_w = 1.026\ g\ cm^{-3}$, the average density of Adriatic surface water (Giorgetti, 1999)). The manta-net was towed for 30 min and the vessel speed was kept < 3 knots. Start-end position points were recorded from the ship's GPS. All toes were conducted from the ship's side and beyond the ship's wake. After completion of each tow the net was washed thoroughly with seawater in order to collect all particles in the cod end. The sample collected in the cod end was then rinsed with seawater on a 300 μm metallic sieve and transferred in glass jars in 70% ethanol solution for further analysis.

2.4. Laboratory analysis of microplastics (< 5 mm)

In the laboratory samples were dried at 40 °C and any natural litter objects of a size > 5 mm were removed from the sample, dried and weighed, then samples were weighed again to obtain the mass of potential microplastics. Samples were examined under a stereomicroscope (OLYMPUS SZX10, OLYMPUS SZX12, NIKON SMZ800N, Carl Zeiss Stereodiscovery V8) and microplastics were recognized, removed and counted on the basis of their shape, cuts, texture, and colour. Six different types of particles were recorded: fragments, filaments, films, foam, pellets, and granules. Non-plastic particles were also recorded. Only for samples rich in organic gelatinous material, a step of H_2O_2 digestion at 60 °C followed by filtration was included before examination. Weight measurements and size classification of microplastics were conducted only by three laboratories (ARPAE, IWRS, HCMR). Data were pooled into three size classes: Small microplastics (SMP): 330 μm < 1 mm; Large microplastics (LMP):1 mm- ≤ 5 mm; Mesoplastics: > 5 mm. In order to check for airborne filament contamination, a blank filter was left open in the laboratory during all stages of the analyses and cross examined. Filaments present in samples with features similar to those collected on the blanks were not taken into account.

2.5. Fourier transform infrared spectroscopy

A subset of particles (n = 1306 particles), accounting for 7% of total counted particles, was spectroscopically examined for its polymer composition using ATR-FTIR spectroscopy (Varian Cary 630). Only particles falling in the 1 mm- ≤ 5 mm size range were analyzed. Polymer identification was made possible using a combination of instrument's and in-house libraries. Spectral range was 4000–650 cm^{-1} with a resolution of 4 cm^{-1}. The threshold for % spectra similarity was set to 80% and the integration time to 8 s.

2.6. Statistical treatment

For checking the non-normal distribution of the data the Shapiro-Wilk test was used. Significant differences in plastic densities (for macro and microplastics) for all studied areas were tested using the non-parametric Kruskal-Wallis test. Mann-Whitney *U* test was used for pairwise comparisons of plastics densities between inshore and offshore waters. Spearman's non-parametric correlation coefficient was used to identify significant correlation between macroplastics densities and coastal population. The level of statistical significance was set at $p < 0.05$. The IBM SPSS statistics 20 software package was used for all statistical analyses performed.

3. Results

All data presented in this work are included in the EMODnet - chemistry database (www.emodnet.eu).

3.1. Macro plastics abundance, size-classes and composition

All anthropogenic litter objects were recorded but for the purpose of this study we present results only for plastics items (artificial polymer materials) which accounted for 91.4% of total litter items recorded. A total of 658 plastic items were visually counted. Floating macroplastic densities ranged from 0 to 4480 items km^{-2} (251 ± 601 items km^{-2}; median 99 items km^{-2}) (2.6 ± 5.6 items km^{-1}) and were highly variable in the small geographical scale of each sub-area studied (CV% range: 65% - 225%). Out of the 66 transects, 13 were litter- free (~20%) and were included in the statistical analyses. Nevertheless, among the various areas sampled the densities were found comparable ($p = 0.074$) (Kruskal-Wallis significance test for $p < 0.05$). When separating macro plastic concentrations in inshore waters (≤4 km from coast) from those found in offshore waters (> 4 km from the coast), then concentrations were found significantly higher in offshore waters (Mann-Whitney *U* test, $p = 0.036$) (Fig. 2). This difference however is due to two outlier values. Upon exclusion of the two outliers, no significant difference exists. The two extreme values were recorded in offshore waters of Croatia (1834 items km^{-2}) and Venice (4480 items km^{-2}). A positive but not significant correlation could be established with the respective population densities for Kotor, Split, Trieste and Venice gulfs ($R = 0.7$, $p = 0.18$, Spearman correlation) (Fig. 2). For Cesenatico, the increased variability of plastic densities could be related to the vicinity of these waters to the Po and Cesenatico rivers.

The majority of plastic items (90%) were smaller than 20 cm in length, in accordance to the size of the most common packaging materials used for consumer products and the fragmentation of plastics in the environment. The percentage contribution of small-sized items, in the range of 2.5–5 cm, was highest (49%) and diminished progressively to 27% and 13% for the 5–10 cm and 10–20 cm size ranges respectively (Fig. 3a). The percentage contribution of small-sized items (2.5–5 cm) diminishes in offshore waters in favor of items in the 5–10 cm size range (Fig. 3b).

The 658 plastic litter items identified in the Adriatic waters were attributed to 11 out of the 19 floating plastic type categories (artificial polymer materials) as described in the MSFD TG10 guidance document (Galgani et al., 2013) (Fig. 4a). Plastic bags (G2) hold the highest share (29%), followed by plastic pieces (G79) (22%) and sheets (G67) (15%). Fish boxes of expanded polystyrene (G58) hold a 13%. Other categories with significant contribution were: cover/packaging (G38) (8.8%),

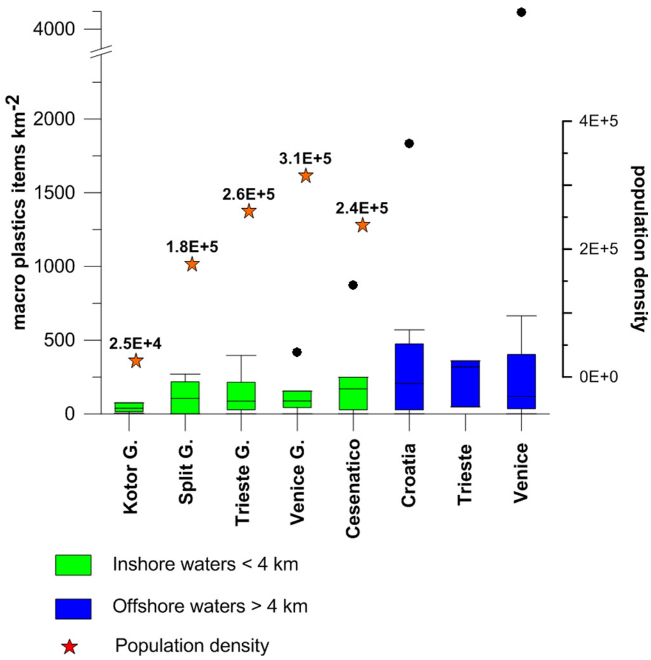

Fig. 2. Box-plots of macroplastic densities (items km^{-2}) for all transects and seasons in each area studied (inshore waters: green boxes; offshore waters: blue boxes). The boundaries of the boxes indicate the 25th and 75th percentiles, the whiskers above and below the boxes the 95th and 5th percentiles. Outliers are indicated as black dots. The horizontal line denotes the median value. Right hand Y-axis corresponds to population density shown with red stars on the plot. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

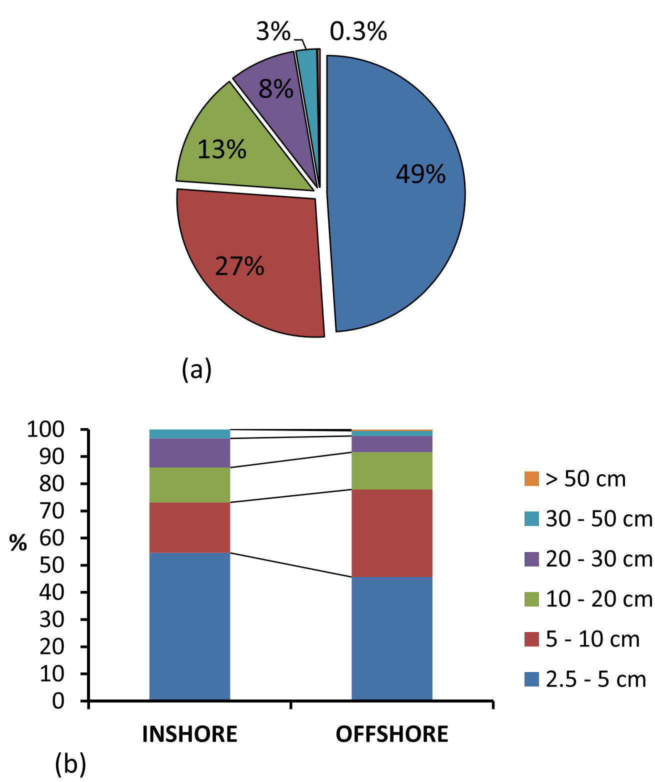

Fig. 3. Percentage (%) contribution of the macroplastics size classes detected (a) in all studied areas; (b) in inshore (≤ 4 km) and offshore (> 4 km) transects.

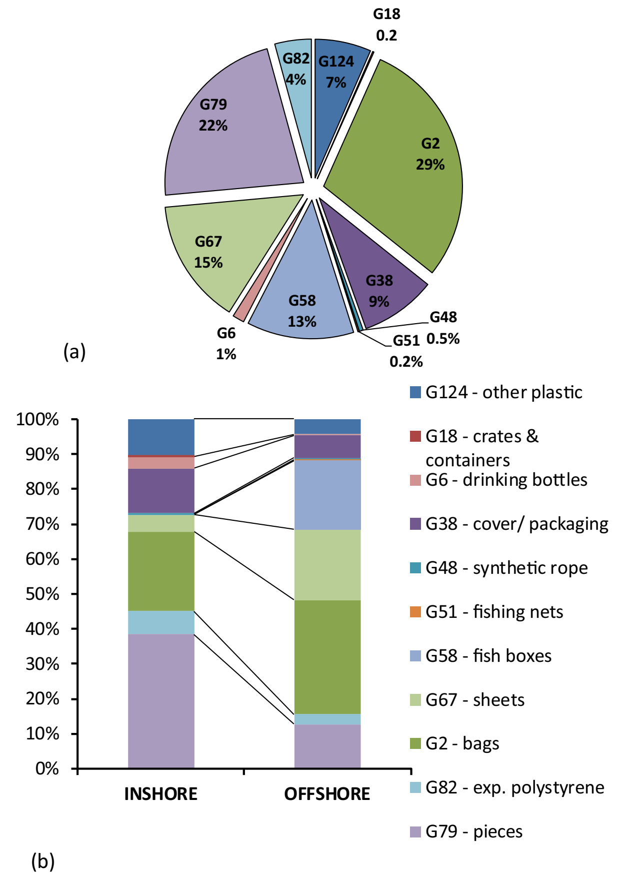

Fig. 4. Floating macroplastic types expressed as % contribution of total items recorded (a) in all study areas; (b) in inshore and offshore transects separately.

other plastic items (G124) (6.5%), polystyrene pieces (G82) (4.3%) and plastic bottles (G6) (1.4%). These 8 plastic litter categories hold 98.9% of all plastics identified. The distribution of the plastic types was not similar between inshore and offshore transects (Fig. 4b). Polystyrene fish boxes (G58) and fishing nets (G51) were recorded only during offshore transects, while crates and containers (G18) only close to the coast. Plastic pieces (G79) and polystyrene pieces (G82) hold 45% of floating plastics in inshore waters and 15% in offshore waters; plastic bags (G2) and sheets (G67) contribute by 28% in inshore waters and rise to 53% in offshore ones.

3.2. Microplastics abundance, size-classes and polymer composition

Microplastic particles were present in all tows except one carried out at the South Adriatic in October 2014; i.e. 1.5% of tows were plastic free. In total, 22,245 microplastic particles were counted, resulting in an average density of floating microplastics of 315,009 ± 568,578 items km^{-2} (median: 80,990 items km^{-2}) (Fig. 5a). As with macroplastics, the distribution of microplastics abundances was found highly variable in each sub-area studied (CV%: 180%). Inshore waters (≤ 4 km) and river outlets showed comparable concentrations of floating microplastics (Mann-Whitney *U* test, $p > 0.05$). In offshore waters (> 4 km) two outlier samples were recorded, at Cesenatico (3,234,330 items km^{-2}) in April 2015 and in the South Adriatic Sea (1,619,658 items km^{-2}) in October 2014. With the exclusion of these two samples, differences in microplastics abundance were found significant between nearshore (≤ 4 km) and offshore waters (> 4 km), being lower in offshore samples (Mann-Whitney *U* test, $p = 0.043$) (Fig. 5a). Microplastic concentrations by weight were on average 217 ± 575 g km^{-2} (median: 41 g km^{-2}; max: 3430 g km^{-2}; CV%:

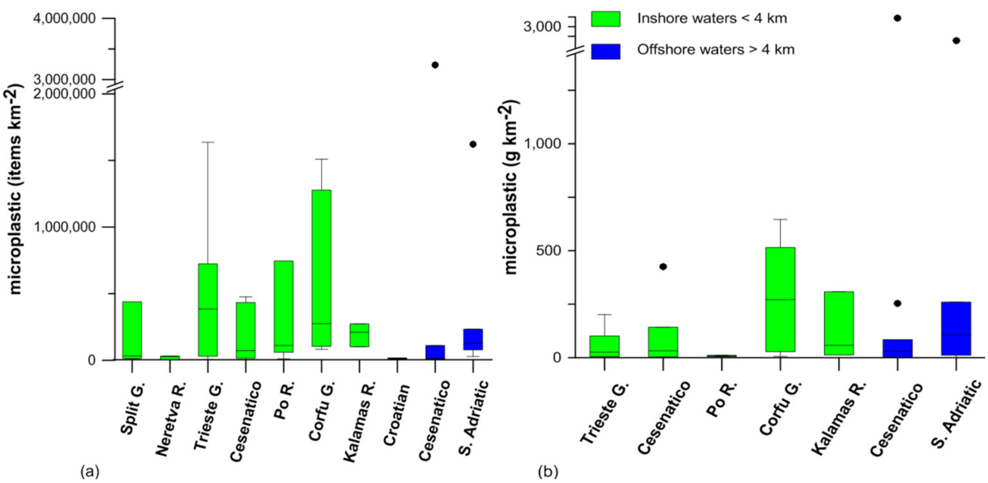

Fig. 5. Box-plots of: (a) microplastic densities (items km^{-2}), (b) microplastic densities by weight ($g\,km^{-2}$), for all transects and seasons in each sub area studied. The boundaries of the boxes indicate the 25th and 75th percentiles, the whiskers above and below the boxes the 95th and 5th percentiles. Outliers are indicated as black dots.

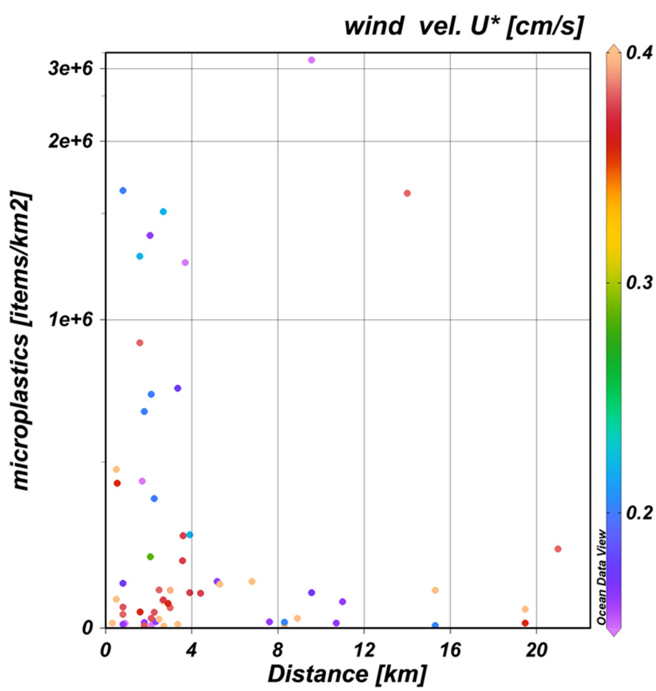

Fig. 6. Microplastics abundance (items km^{-2}) vs distance (km) from the closest coast. Z-axis colour bar denotes the variation in water friction velocity u* ($cm\,s^{-1}$).

265%) (Fig. 5b). In this case no significant differences were found among sub areas or distance from the coast. Although all manta net samplings were conducted under low wind regime (Kukulka et al., 2012), we further checked if the distribution of microplastics was affected by weather conditions during our samplings. In Fig. 6 we have added the water friction velocity as the Z axis of the graph of the distribution of the abundance vs distance from coasts. No consistent pattern between the variability of u* and microplastics abundance (items km^{-2}) was observed, suggesting that the observed distribution was independent from the variation in the weather conditions.

Out of the six different types of particles identified, fragments had the highest share (77%), followed by films (9%) and filaments (7%), while the rest of the types were present in low percentages (< 3%). Size distribution of microplastic particles into three size classes, SMP: 330 μm < 1 mm; LMP: 1 mm- ≤ 5 mm; meso-plastics: > 5 mm, revealed that 34% of particles were in the range of SMPs, 64% in the LMP range and only 2% were larger than 5 mm.

Polymer identification by ATR-FTIR spectroscopy revealed that 66.5% of the analyzed particles were polyethylene (PE) (LDPE and HDPE), followed by polypropylene (PP) (17.9%). Expanded polystyrene (EPS) ranked third holding a 4.2% of the characterized particles. A contribution of 3.1% was attributed to synthetic fibers such as nylon, polyamides (PA) and polyacrylonitrile (PAN) and of 1.6% to polyester (PES). Polyethylene terephthalate (PET) accounted only for 1%. Less frequent polymers present in our subset of particles were polyvinylchloride (PVC) (0.2%) and cellulose acetate (0.1%). Five more polymer types accounted for 0.4%, namely: ethylene vilyl acetate (EVA); polyvinyl siloxane (PVS) and polyvinyl acetate (PVA) and Poly4methyl1pentene (Fig. 7). About 5% of the analyzed particles were not synthetic polymers and corresponded to natural materials (2.8%), while wax materials identified as carnauba wax and calcium stearate accounted for 2.1%. This percentage is in accordance to the findings by Suaria et al. (2016), Vianello et al. (2018) and Palatinus et al. (2018; in prep.) for the Mediterranean surface waters.

4. Discussion

Table 2 shows information regarding floating anthropogenic litter densities and observation conditions as reported in the literature for the Mediterranean Sea. It appears that density data obtained during the present study are one to two orders of magnitude higher than most of those previously reported for the Mediterranean and Adriatic Seas. Most published works have been conducted with oceanographic vessels

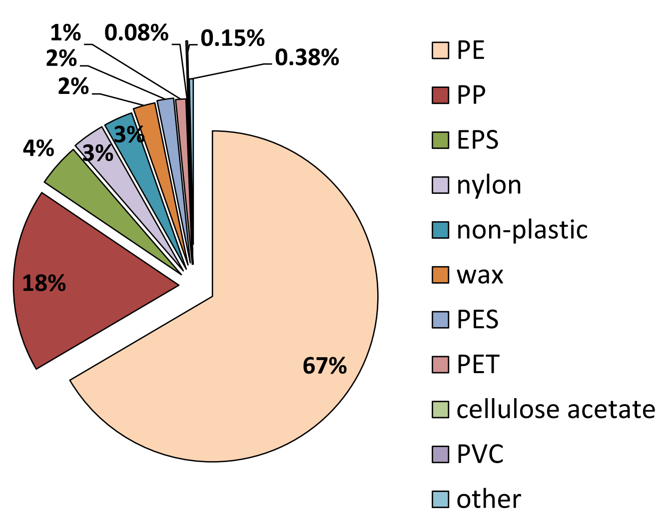

Fig. 7. Polymeric composition (%) of floating microplastics (n = 1306) based on ATR – FTIR spectroscopy.

Table 2
Literature data on floating macro litter densities and observation conditions for the Mediterranean Sea.

	Year	Vessel speed (Knots)	Obs. height (m)	Obs. width (m)	Distance travelled (km)	Density items/km^2	Minimum detectable size class	Source
Mediterranean Sea	1979	–	12	10	–	2000	> 1.5 cm	Morris, 1980
Ligurian Sea	1996	3.2–11.5	top deck	50	176	14–25	–	Aliani et al., 2003
	2000	6	top deck	50	252	1.5–3.0	–	
Western Mediterranean	2006–2015	6	3		5171.57	15 ± 23	> 1 cm	Di-Méglio and Campana, 2017
Western Mediterranean	2013	10	5	30	1538	0–162 (24.9 ± 2.5)	2–10 cm	Suaria and Aliani, 2014
Adriatic					277	55 ± 11		
Western Mediterranean	2013–2016	19–25	17–25	100	18,113	2.3 ± 0.4	> 20 cm	Arcangeli et al., 2017
Adriatic					6733	4.7 ± 5		
Adriatic	2014–2015	2–3	1–3	10	415	260 ± 596[a]	2.5–5 cm	Present work

[a] Plastics only: 251 ± 601 items km^{-2}.

travelling at > 6 knots and from an observation height ranging from 3 m to 25 m; equally variable was the observation width of each survey. The variability in observational conditions leads to variable detection of the small macro- litter sizes (2.5 cm to 5 cm) as has been already acknowledged (Galgani et al. 2013; Ryan, 2013) and it is now advisable to report data on floating litter along with the minimum size detected. Until now, only some studies report the minimum size class detected and even less have reported the size classes of the litter items observed (Morris, 1980; Ryan, 2013; Suaria and Aliani, 2014; Shiomoto and Kameda, 2005; Sá et al., 2016; Arcangeli et al., 2017; Di-Méglio and Campana, 2017). The implementation of a common methodology will definitely improve the accuracy and comparability of reported marine litter densities. Nevertheless, the small-sized items (2.5 cm–5 cm) will still be prone to underestimation when medium or large size vessels travelling at high speed are used. In contrast, when smaller ships are used, it is expected that the ability to detect small-sized items will increase and also that each item size class is homogeneously distributed within the effective strip (i.e. the observer can detect all items present within the strip) (Thiel et al., 2003; Suaria et al., 2016). This is the case of our surveys during which the observation height varied from 1 m to 3.2 m and the vessel speed was always kept between 2 and 3 knots. Given the aforementioned differences in the minimum litter size detected, the litter densities presented in Table 2 cannot be considered directly comparable. It is interesting to note, however, that for the Mediterranean Sea the highest litter density of 2000 items km^{-2} was reported by Morris (1980). Although his observations were made from an elevation of 12 m, this author clearly states that observation conditions enabled him to detect small-sized items > 1.5 cm. Our results on macroplastics densities, obtained using low height vessels, clearly suggest that the amount of floating small-sized plastics is significant; they show that a considerable amount of macroplastics afloat on the Adriatic Sea than previously considered. This result has important implications for any quantification and mass balance attempts for plastics.

The most important factors affecting the distribution of floating plastics in marine waters are the vicinity to marine litter sources (i.e. urban and touristic centers, shipping lanes, fishing areas) and pathways (i.e. rivers, wastewater treatment plants - WWTPs), with high uncertainties regarding their fluxes, as well as the specific oceanographic conditions prevailing. For our study area, some of the expected plastic sources include (i) shipping (www.marinetraffic.com) and fishing activities (FAO, 2016), (ii) populated urban and tourist centers (e.g. Venice, Bari, Trieste, Split, Ravenna and Corfu); while potential pathways of plastic from land to sea are considered the Adriatic rivers (Po and Adige Rivers flowing at the west coast, Buna/Bojana, Neretva and Kalamas rivers flowing at the east coast). Our data on macroplastics densities showed increased variability in the different times and areas sampled and no clear relationship could be established with the respective population density (www.worldpopulationreview.com) (Fig. 2). A similar variability in data presented by Gajšt et al. (2016) was successfully explained by the temporal and geographic variation of wind and current conditions. The vicinity of the surveyed area of Cesenatico to the Po river plume is the likely explanation for the elevated macroplastics densities observed there. Analysis of floating plastics composition shows that bags and sheets, plastic fragments and expanded polystyrene (Styrofoam) boxes dominate the assortment of floating plastics (Fig. 4a), in accordance with the findings of Di-Méglio and Campana (2017), and Suaria and Aliani (2014) for the NW Mediterranean and Adriatic seas. Styrofoam fish boxes (G58) were recorded only in the gulf of Venice during the spring surveys, along with fishing nets (G51) (0.2%). The intense fishing and aquaculture activities in the gulf of Venice (Pasquini et al., 2016) relatively to the other areas sampled, may explain the presence of styrofoam fish boxes and nets only there. The relative increase of bags and sheets (53% of total plastics) in the assortment of plastics in offshore waters could be related to the intense marine traffic in the area. In this case, we would expect that other types of litter indicative of tourism would be present, such as drinking bottles and food packaging. In contrast, bottles (G6) were found only in inshore waters and cover/packaging plastics (G38) diminished by 50% in offshore relatively to inshore waters (Fig. 4b). The relative absence of bottles (G6) and the reduced presence of cover/packaging plastics (G38) in offshore waters may indicate that they are removed rapidly, either to the shore and/or to the seafloor via stranding or sinking mechanisms. Drinking bottles are made of PET which has a relatively higher density (1.38–1.45 g cm^{-3}) than seawater (1.28 g cm^{-3}) and in addition uncapped bottles can easily fill with water and sink. On the other hand, bags and sheets are made of PE with density of 0.90–0.99 g cm^{-3} and can stay afloat longer on the sea surface. It is possible that bags and sheets are carried away by surface currents, due to their film-like shape, and trapped in the prevailing surface circulation of the Adriatic basin. The observed distribution of bags and sheets contrasts the one reported for South Atlantic waters off Africa (Ryan, 2015). In that case, the fast sinking of bags was explained by the increased effect of fouling on buoyancy loss of flexible items with high surface to volume ratio. This discrepancy is related to the fact that in the work of Ryan (2015) shelf and oceanic waters were sampled at distances > 20 km from the coasts, much farther than our transects, where longer travelling time of plastics is expected.

Plastic pieces and fragments (G79) seem to follow a land-ocean gradient with higher abundances in inshore waters (Fig. 4b). Plastic pieces and fragments (G79) (2.5 - < 50 cm) that can be produced either on beaches (Corcoran et al., 2009; Kalogerakis et al., 2017) or transported via rivers and urban runoff from land, correspond to about 1/4 of total floating macroplastics in our study area. They are actually fragmented plastics and therefore can be considered 'old' – 'aged' plastic which re-circulates at the sea surface and probably is being exchanged between the shore and the sea several times before being deposited on the seafloor.

As with macroplastics, microplastics densities were also found

Table 3
Literature data on floating microplastic densities for the Mediterranean Sea.

Region	Year	Net mesh (μm)	Items km^{-2} ± SD	$g\ km^{-2}$ ± SD	Source
Cretan Sea	1997	500		119 ± 250	Kornilios et al., 1998
NW Mediterranean Sea	2010	333	116,000	202	Collignon et al., 2012
Ligurian/Sardinian Sea	2011	200	310,000 ± 100,000		Fossi et al., 2012
Bay of Calvi (Corsica)	2011–'12	200	62,000		Collignon et al., 2014
W. Mediterranean Sea	2011–'12	333	135,000	187	Faure et al., 2015
Ligurian Sea	2013	333	125,930 ± 132,485		Pedrotti et al., 2016
Mediterranean Sea	2013	200	243,853	423	Cózar et al., 2015
Western & Central Mediterranean Sea	2011–'13	333	147,500 ± 25,051	579 ± 156	Ruiz-Orejón et al., 2016
W. Mediterranean & Adriatic Seas	2013	200	400,000 ± 740,000	672 ± 1544	Suaria et al., 2016
Tyrrhenian Sea	2013–'14	330	69,161	41	Baini et al., 2018
W. Mediterranean Sea	2014	330	82,000 ± 79,000		Fossi et al., 2017
Israeli coastal waters	2013–'15	333	1,518,340		van der Hal et al., 2017
Adriatic Sea	2014	300	472,000 ± 201,000		Gajšt et al., 2016
Adriatic Sea	2014–'15	330	315,009 ± 568,578	217 ± 575	Present work

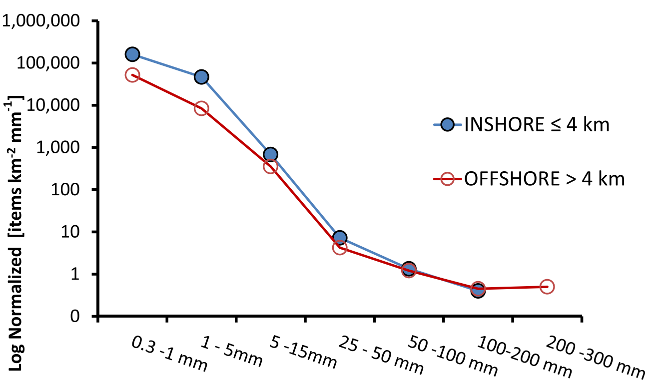

Fig. 8. Log normalized plastics densities (items $km^{-2}\ mm^{-1}$) according to the respective size range for inshore (≤4 km) and offshore waters (> 4 km).

highly variable among seasons and areas sampled but were comparable with those previously reported for Adriatic and Mediterranean waters (Table 3) (Collignon et al., 2012; Fossi et al., 2012; Cózar et al., 2015; Suaria et al., 2016; Ruiz-Orejón et al., 2016; Gajšt et al., 2016). The only consistent distribution pattern observed was that densities were significantly elevated in enclosed gulfs and closer to the coasts following the distribution of plastic fragments (G79). In order to get some insights on the size distribution of plastics in our study area, we have plotted for inshore and offshore transects separately, the size normalized densities (items $km^{-2}\ mm^{-1}$) of all plastic fragments in the macro size ranges (G79); of *meso* plastics caught in manta nets (approx. ~5–15 mm) and of the two microplastics size classes (LMP, SMP) (Fig. 8). Normalization of plastics size densities (items km^{-2}) by the corresponding size range length (mm) was made in order to facilitate comparisons. It appears that, indeed, large plastic fragments (> 5 cm) are equally distributed between inshore and offshore waters, while smaller fragments (< 5 cm), meso- and microplastics increase in inshore waters as size range decreases. Higher concentrations of microplastics close to the coasts have been previously observed in the Mediterranean (Pedrotti et al., 2016; Ruiz-Orejón et al., 2016, 2018; Baini et al., 2018) and in other enclosed seas (Gewert et al., 2017). This feature has been attributed to the input of microplastics from the coasts or to retention close to them due to coastal currents, in combination to their fast sedimentation due to buoyancy loss relatively to larger plastic items. The effect of fouling is more important on small-sized plastics with relatively increased surface to volume ratios. Our data show that the decoupling in plastics' densities between inshore and offshore waters starts from the 2.5–5 cm size class. In addition, both distributions (in inshore and offshore waters) do not follow a linear or exponential increase as size class decreases, but show a drop in the concentrations of small-sized plastics (< 1 mm) which is more obvious in inshore waters (Fig. 8). The loss of small sized plastics from the sea surface has been reported for oceanic and Mediterranean waters based on detailed microplastic size classes (Morét-Ferguson et al., 2010; Cózar et al. 2014, 2015; Pedrotti et al., 2016; ter Halle et al., 2017; Ruiz-Orejón et al., 2018). Kooi et al. (2016), using a vertical array of nets have experimentally demonstrated that small-sized plastics (< 1 mm) with elongated shapes tend to suspend deeper in the water column, due to water friction effects, a finding which explains the observed distributions. At the same time processes such as the ingestion of small-sized plastics by marine organisms may also affect the observed distribution pattern.

Some information about the sources of microplastics can be inferred by their polymer types. The predominance of PE and PP in our samples is in agreement with previous works in the Mediterranean Sea (Pedrotti et al., 2016; Suaria et al., 2016; Vianello et al., 2018) and elsewhere (Enders et al., 2015; Gewert et al., 2017) and reflects the increased production and use of polyolefins (PE, PP) relatively to other plastic materials, mostly in packaging and single use products. In Europe the production of PE and PP correspond to ~50% of the total plastics demand, while PVC and PET to 10% and 7.4% respectively (Plastics Europe, 2017). As quoted previously, PE and PP are low density polymers (0.90–0.99 $g\ cm^{-3}$; 0.85–0.92 $g\ cm^{-3}$) and hence have longer residence time at the sea surface, while heavier polymers such as PVC (1.38–1.41 $g\ cm^{-3}$) and PET (1.38–1.45 $g\ cm^{-3}$) are prone to rapid sinking. Of the least abundant materials, EVA has heat sealing properties and is used in packaging; PVA is used as a coating; while polyvinyl siloxane (PVS) is a silicon elastomer widely used in dentistry as a moulding material, previously recorded also in Mediterranean waters by Suaria et al. (2016). Both waxes (calcium stearate a non-plastic and carnauba wax a natural wax) present in our samples have a variety of applications including food products, coatings, plastics' colorants and additives. The polymers identified in our study areas, which include coastal waters and enclosed gulfs of the Adriatic Sea, point to land sources of microplastics including WWTPs. Several installations of WWTPs are situated along the Adriatic coasts, while some coastal cities still lack sewage treatment plants (UNEP/MAP, 2012).

5. Conclusions

Our results demonstrate that the amount of small-sized macroplastics (2.5–5 cm) is significant in the Adriatic waters and result to at least one order of magnitude higher plastics densities than previously reported for this sea. This is of particular importance with regards to the environmental status of marine waters, as these small-sized items cannot be controlled and prevented and by no means can they be removed in significant amounts. The abundance of macroplastics in the enclosed gulfs (Kotor, Split, Trieste, Venice) of the Adriatic Sea shows some increasing trend in parallel to the population density, albeit not significant; nor is it higher than the macroplastics abundance observed in offshore waters. Possible explanations are considered the sea-based sources of macro plastics as well as the different buoyancy features of various macroplastic items most probably related to their polymer types and shape. With the exception of fisheries, no other apparent sea-based source of floating plastics could be discerned. In addition, macroplastics' compositional differences between inshore and offshore waters infer that items' properties (polymer type and shape) are also important in determining their distribution. The only consistent land-ocean gradient was observed for small-sized floating plastic fragments (2.5 cm–5 cm), meso- (> 5 mm) and microplastics (≤ 5 mm) densities. Our data show elevated densities in inshore waters not only for microplastics, as previously documented, but for all fragments smaller than 5 cm. The polymeric identification of microplastic particles revealed that the two most commonly used and highly buoyant materials, PE and PP, were dominant. At the same time the presence of particles with sole uses in dentistry provide evidence that WWTPs are sources of microplastics in Adriatic waters. These results highlight the importance of the chemical characterization of polymers not only for the assessment of microplastics pollution but also for the development of targeted and effective measures. Recent policy advances highlight the need to address popular misconceptions with regards to marine litter measures and the abundance of small- sized macroplastics. One of these misconceptions is related to the use of oxo-degradable plastics as a more 'environmentally-friendly' option over traditional plastics, when in fact oxo-degradable plastics break down into small fragments and become harmful small- sized macroplastics, mesoplastic and microplastic pollution. This is why the Environment Committee of the European Parliament has recently voted for a set of amendments to the European Commission's strategy to tackle plastic pollution, where a complete ban on oxo-degradable plastics by 2020 has been recommended. Other targeted measures such as banning single-use items and or setting up better management schemes for styrofoam fish box boxes are expected to have a direct effect on minimizing floating plastics in the Adriatic sea. Efforts towards minimizing mismanaged waste on land should be also reinforced, while at the same time, continuous awareness raising campaigns targeted to citizens, mainly in their professional capacity but also as individuals are an imperative need in order to prevent and reduce this kind of pollution.

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