Microplastic particles in sediments and waters, south of Caspian Sea: frequency, distribution, characteristics, and chemical composition

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Abstract

This study assesses the occurrence of microplastics (MPs) in coastal and sea surface sediments, as well as water samples, collected from the coastal region of the southern Caspian Sea, Mazandaran province, Iran. A total of 32 sediment and 10 water samples were studied. The mean concentration of MPs was 15 units kg^{-1} in the sediments and 710 units m^{-3} in the coastal water. Fibers constituted by far the dominant MPs in both media, accounting for 97% of the MPs in both sediment and water samples. The MPs were mainly black in color. The dominant size of MP particles in sediment samples was between 250 and 500 µm, while the fraction >1000 µm dominated in the water samples. Polyethylene terephthalate (PET), polystyrene (PS), and nylon (NYL) were the main polymers and/or copolymers composing MPs in both sediment and water samples. The MP particles had a relatively smooth surface morphology, although signs of weathering were observed. The number of MP particles in sediment and water samples showed a general decrease from west to east in the study area. This may be reflecting the spreading of MP loading from the outlets of Sefidrud, Tonekabon, Chalus, the major rivers entering the Caspian Sea just west of the study area, and the overall decrease in the spatial distribution of touristic and fishery activity. The main sources of MP particles could be local emissions from a large number of domestic wastewater effluents and urban surface runoff due to high population density, and industrial and fishing activities in this region. This study indicated that MP particles, based on their characteristics and chemical composition, are circulated between coastal waters, and shore and sea surface sediments of the Caspian Sea, leading to their uneven distribution in the different depths. To the best of our knowledge, this is the first work studying the distribution of MP particles in sea surface sediments and also the most comprehensive on MPs in shoreline sediments and coastal waters in the southern Caspian Sea.

Keywords:
Microplastic; Fiber; Sediment; Water; Caspian Sea

1. Introduction

Coastal regions are very productive ecosystems and provide habitat for large biodiversity of aquatic organisms. These coastal environments are strongly and constantly affected by anthropogenic activities worldwide (Vasconcelos et al., 2007, Bi et al., 2017, Rebeiro et al., 2018). Increased levels of contaminants have led to the rapid degradation of our shore and sea ecosystems. Marine litter has become a major environmental challenge.

Global production of plastic has increased exponentially over the past decades, from only 1.7 Mt yr^{-1} in the 1950s to 359 Mt yr^{-1} in 2018 (UNEP, 2015; EuropePlastics, 2019). Yearly, about 5 to 13 million tons of plastics thereby enter the oceans (Dhiman and Marques, 2016). Plastic litter is composed of various polymers. The main polymers are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyethylene terephthalate (PET), and nylon (NYL) (Avio et al., 2017, Martellini et al., 2018). Due to their inert structural composition, the plastics are very slowly degraded, and thus tend to persist in the environment (Yoshida et al., 2016; Martellini et al., 2018).

Their ingestion by fauna is understood as detrimental although there is still a scarcity of long term studies (Ma et al., 2020). Plastics can adsorb persistent organic micro-pollutants (Fossi et al. 2016, Auta et al. 2017). This suggests that ingested MP particles can act as a source and as a carrier for contaminants into the organism. However, contaminants sorbed to MP particles can be less bioavailable than dissolved contaminants (Sørensenet et al., 2020).

Persistent pollutants are ultimately stored in marine sediments (van der Oost et al., 2003; Ruiz-Compean et al., 2017) and for that reason, sediments are used to evaluate the state of the environment (Moore et al., 2015). The spatial distribution of the contaminants in sediments is subsequently used to assess sources of toxicants and their dispersion pathways (Salomons, 2012; Ruiz-Compean et al., 2017).

Previous works have identified MP particles in the shore sediments and or waters of the Caspian Sea (Masoudnik et al., 2017; Mataji et al., 2019; Mehdinia et al., 2020) but there is lack of knowledge regarding their transport, distribution, and fate in brackish water in the shoreline, and sea surface sediments. Besides, this is the first research on the status of MPs in sea surface sediments from south of the Caspian Sea.

Major rivers and numerous discharge pipes from wastewater treatment plants entering the Caspian Sea are hypothesized as the main point sources of MP particles to the sea environment. In addition, there is a wide range of shipping and fishing activities in the area that could comprise significant diffuse sources of MP particles to the Caspian Sea. Once in the sea, buoyancy, as well as hydro-physical factors, affect transport and fate of MP particles. The salinity of the brackish water and the density of the plastic material govern buoyancy. Hence, this research seeks to provide an insight into the type, distribution, and fate of the MP particles as well as assessing their sources in this coastal environment.

2. Materials and methods

2.1. Study area

The Caspian Sea is the largest inland water body in the world with an average depth of 180 m (Kosarev, 1994), 1200 km in length, and an average width of 330 km, constituting a surface area of about 436000 km^2 (Agah et al., 2007). It is bordered by the five littoral countries of Iran, Azerbaijan, Federation of Russia, Kazakhstan, and Turkmenistan. Its average salinity in the south part of the brackish sea is 12-13 ppt (Clark, 1992). The water chemistry is of sodium chloride type with a pH of 8.5 (Tuzhilkin et al., 2005). A total of 130 rivers supply about 300 km^3 y^{-1} water to the Caspian Sea, whereas most are provided by the Volga river (CEP, 2002). The south coast of the Caspian Sea stretches about 820 km through to the Guilan, Mazandaran and Golestan provinces in Iran. The contribution of Iranian rivers accounts for approximately 5% of the discharge. Sediment loading to the Iranian Caspian coasts is mainly from 61 rivers (Afshin, 1994; Alizadeh et al., 2018).

The southern Caspian region has a subtropical climate (Alizadeh et al., 2018). Precipitation gradually increases from the eastern coasts towards the west (Khaleghizavareh, 2005; Alizadeh et al., 2018). Traditionally, tourism, agriculture, and fishery are the main economic activities in the region, though since the 1960s the industrial activities have increased (Alizadeh et al., 2018), all leading to high population density in the Mazandaran coastal areas. Mazandaran is well-known in the middle east for its textile industry, producing artificial fibers and cotton fabrics.

The study area is located in Mazandaran province, which comprises the longest Iranian shoreline Sea (487 km) (Fig. 1). The Iranian Caspian shore is characterized by the dominance of siliciclastic deposits (Alizadeh et al., 2018). The prevailing water current in the south of the Caspian Sea (Kostianoy et al., 2019) flows from west to east (Sharbaty, 2012; Bohluly et al., 2018). Coastal sediments and waters in the Caspian Sea receive contaminants from the two major rivers Kura in southern Azerbaijan and Sefidrud in Iran (Voropaev et al., 1998), both entering the Caspian Sea west of the study area. In addition, there are several important local streams, such as Tonekabon, Sardabrud, Chalus, Haraz, Babolrud, Tajan, and Nekarud, from west to east, respectively, as well as numerous outlets from sewage treatment plants and local surface runoff.

2.2. Sampling and sample preparation

Sampling sites were selected to include expected hotspots of MP pollution, as well as control sites, along the Mazandaran shoreline. This was based on the spatial distribution of potential major sources of MPs, including outlets of rivers draining watersheds with high population density, along with commercial, industrial, fishing, farming, and touristic activities (Supplementary material 1). Sediments and water sampling were carried out during the dry season in May 2019. Shore sediment samples were collected from 17 coastal sites. At each site, a total of 1 kg of sediment samples

(made as a composite from five subsamples) were collected and mixed. To assess the distribution of MP particles out into the sea sediments, 15 surface sediment samples were collected along five coastal transects (Fig. 2a). From each of the coastal transects, sea surface sediment samples were collected where the water depth was 5, 10, and 20 m. Transects are located where at least one major potential source of MPs (supplementary material 1) is pervasive. Therefore, transects reflect the influence of tourism/fishing (T-T/F), tourism/fishing/population (T-T/F/P), tourism/commercial (T-T/C), industrial/population (T-I/P), and commercial/industrial (T-C/I) (Fig. 2a). Shore sediment samples were collected at low tide using a pre-cleaned stainless-steel shovel. A Van Veen grab sampler was used to collect surface sea sediments. Sediment samples were collected in pre-clean glass jars. A total of 10 composite water samples were collected from the surface until around 20 cm below the surface to include both light and heavier MP particles. Each sample was constituted by ten 1-liter subsamples, collected at 100-meter intervals along the coast and homogeneously mixed to make a composite sample. Water samples were then stored in clean glass bottles, and sealed by thick aluminum foils and glass stoppers. Water samples were transferred to the lab and kept at room temperature for further experiments. Therefore, a total of 32 sediment samples, including 17 coastal and 15 sea samples, along with 10 coastal water samples were collected.

2.3. Sample treatment and MP particle extraction

Sediment samples were transferred to glass beakers, using prewashed steel spoon, and dried in a sand bath at 80° C for 10 days. Dried samples were passed through a 5 mm cut-off metallic sieve and were stored in glass beakers at room temperature. To avoid misidentifying organic matter as MP, which could cause an overestimation of the number of MP particles or their size, a digestion step was carried out to improve the determination of MP particles. This step was carried out following previous work (Prata et al., 2019). Therefore, organic matter in the sediment samples was removed by oxidation with 30 % hydrogen peroxide (Nuelle et al. 2014) for 10 days until bubble formation stopped, implying that there was no more organic matter to oxidize to CO_2 left in the sample. The remaining H_2O_2 was decanted and the sediment was washed with deionized water to remove remaining H_2O_2 and to collect adhered particles on the wall of the beaker. Prior to decanting the solutions were allowed to rest for minutes in order to allow suspended particles, including MP > 100 µm, to settle. The mineral sediment samples were finally dried in a sand bath at 80 °C for 8 h (Dehghani et al., 2017).

The MP particles were separated out of the sediment samples by flotation in a saturated $ZnCl_2$ salt solution with a density of 1.6 g cm^{-3} (Löder and Gerdts, 2015). This allowed the light MP particles to separate from mineralogical particles based on their greater density. The supernatant from treated sediment and the water samples were centrifuged (4000 rpm) and filtered (2 µm pore size S&S

filter papers (blue band, grade 589/3)). The filters containing the MP particles were left to dry and transferred to Petri dishes for further analyses.

2.4. MP particle identification

MP particles on each filter were visually identified. Diagnostic features, such as size, shape, and color were determined using a binocular optical microscope (Carl-Zeiss, Weet Germany) with up to ×200 magnification (Stolte et al., 2015; Dris et al., 2017). The identification was aided with ImageJ software. MP particles were classified according to length (L) into four fractions (100≤L<250 µm, 250≤ L< 500 µm, 500≤ L< 1000 µm, 1000≤ L< 5000 µm). MP particles smaller than 100 µm were disregarded due to the limit of identification with the microscopy used. The particles were classified by shape into three types (fiber, fragment, and pellet), and by color into 5 classes (black-grey, blue-green, yellow-orange, red-pink, and white-transparent). Detailed surface morphological characteristics, along with the elemental composition of MP particles, were examined using Scanning Electron Microscope (SEM, TESCAN Vega 3, Czech Republic) with a resolution of 2 nm at 20 kV equipped with an energy-dispersive X-ray microanalyzer (EDS). MP particles were mounted onto double-sided copper adhesive tapes and coated with gold for SEM-EDS analysis. The polymeric constituents of MPs were determined using a confocal Raman microscope (Lab Ram HR Evolution, Horiba Japan) equipped with a 785 nm laser and detection between the range 400-1800 cm^{-1}. To identify polymer type, the spectra obtained for each MP particle by Raman were compared with the reference spectra of each polymer. Raman is commonly used in the characterization of polymeric constituents (Anger et al., 2018; Araujo et al., 2018; Kniggendorf et al., 2019).

2.5. Quality control

The same analyst studied all samples. To prevent sample contamination laboratory glassware was washed with phosphate-free soap; double rinsed with distilled water; left in 10 % HNO_3 for 24 h and then rinsed twice with double-distilled water and left semi-closed to dry at room temperature. Prior to any analysis, the laboratory benches were thoroughly cleaned using ethanol. All reagents and solutions were filtered through S&S blue band filters (grade 589/3, 2 µm pore size). All plastic laboratory equipment and synthetic garments were banned in the labs involved in this study. Cotton gloves and laboratory coats were used. To check that no contamination of airborne MP particles occurred in the lab, a blank control dish was left open on the lab bench during the experiments. The analysis confirmed no MP contamination in the control dish. To achieve a measure for the precision and accuracy of the analytical instruments, blind samples were introduced in each batch of samples for analysis. Replicate spectroscopic measurements were carried out (n=4). All control measures gave acceptable and satisfactory results. The precision and accuracy of the analytical instruments are also seasonally checked using reference and blank materials by the laboratory.

2.6. Statistical analyses

The spatial distribution of sampling sites was assessed using ArcGIS version 10.3. The normality of the data was checked using the statistical Shapiro-Wilk (S-W) and Kolmogorov-Smirnov (K-S) tests. Non-parametric Mann-Whitney (M-W) U-test was applied to assess significant differences ($p < 0.05$) in the amount of MP particles between sediment and water samples, as two independent groups. Non-parametric Kruskal-Wallis (K-W) H-test was used to evaluate significant differences ($p < 0.05$) between the amount of MP particles in shoreline sediments, sea surface sediments, and water samples, as three independent groups. These statistical analyses were performed using SPSS version 22.

3. Results and discussion

3.1. MP particles concentration and distribution

The number of MP particles found in sediments and water samples are presented in Supplementary material 2. The frequency of MP particles was non-normally distributed ($p < 0.05$) in the shoreline sediments, and normally distributed ($p > 0.05$) in the sea sediment and water samples (Supplementary material 2). Overall, a total of 480 MP particles were found in the 32 sediment samples, and 7100 MP particles were found in the 10 water samples. In the sediment samples, the number of MP particles ranged from non-detected to 48 items kg^{-1}. In the water samples, the numbers varied between 200 and 1500 items m^{-3}. The mean and median of the number of MP particles detected was 15 and 12 items kg^{-1} in the sediments, and 710 and 650 items m^{-3} in the water, respectively.

The distribution of the MP (in the number of detected plastic items < 5mm) between water and sediment samples was significantly different ($p < 0.05$). However, no significant difference ($p > 0.05$) was found between the number of MP particles in shoreline and sea surface sediments. Nevertheless, this may be different during the rainy season in the autumn, due to the increased overland flow and of the intermittent streams. The number of MP particles in shoreline and sea surface sediment samples ranged from 4 to 48 particles kg^{-1}, and from 0 to 32 particles kg^{-1}, with a mean frequency of 18 and 11 particles kg^{-1}, respectively. The relatively similar level of MP particles found in shoreline and sea sediments indicates that similar hydrogeochemical processes and sources could be governing the spatial distribution of MP particles in these sediments. These transport processes are mainly governed by buoyancy, determined by the differences in relative density of water and the MPs, sediment-water interactions, and governed by the physical resuspension of particles. Comparing this brackish coastal environment with freshwater systems, the relative level of the MP pollution found in the sediments here is generally lower than that found in sediments from freshwater systems (Li et al., 2020). This is in line with a greater buoyancy of the MP particles in this denser brackish water.

The number of MP particles in the sediment samples from this study displayed an uneven and non-uniform spatial distribution, both along the shoreline and throughout the transects into the sea surface sediments. The spatial variations in the number of MP particles were lower in water compared to the sediment samples. This can be because water allows for greater mixing of pollution. Specifically, the spatial distribution of MP particles in shore and sea surface sediments showed that most contaminated sites lie in the western part of the studied area (Fig. 2a) while the highest levels of MP particles in water samples occur both in the far west and in the east of the study area (Fig. 2b). This is likely due to differences in local and more regional pollution sources, as well as differences in governing factors working on the transportation processes distributing the MP particles in this environment. The greater concentration of MP particles in the west could be due to the abundant touristic and fishing activities in the zone (Fig. 1). In addition, there are large inputs from major rivers, such as Sefidrud, Tonekabon, and Chalus, entering the Caspian Sea in the far west of the study area. MP particles are, upon entering the Caspian sea, transported towards the east by the prevailing sea current. In the east, there are a large number of industrial and commercial activities that can release MPs. These, together with domestic wastewater effluents from a large population, as well as urban surface runoff could reach the coastal areas (Fig. 1). Previous studies of MP in this region pointed out that municipal waste disposal by shallow burial and open stockpiles, with no leachate treatment system, exists in many coastal sites of Mazandaran (Abduli et al., 2007, 2015, Mehdinia et al., 2020). Therefore, they indicated that MP particles from disposal wastes, untreated wastewaters, and landfill leachates may be transported to the Caspian Sea via a large number of rivers.

3.2. Size of MP particles

The dominant MP particle size class was between 250 and 500 μm in all shoreline, and sea surface sediments, constituting 36, 33, and 43% of the total MP particles found at the sampling sites, respectively (Fig.3). In contrast, in the water samples, the fraction > 1000 μm (42%) dominated (Fig. 3). Also, a higher percentage of MP particle size class greater than 1000 μm was found in shoreline sediments (31%) compared with sea sediments (19%). This implies that a larger percentage of the primary (i.e. as released to the environment) MP particles may be present in the shoreline sediments, while MP particles present in sea surface sediments may be more influenced by weathering, and changed to the secondary MP particles. Larger particles are more readily transported by convection than smaller particles in the water. Larger MP particles are thus more likely to be washed up on the beach. It is also worth mentioning that the coastal water samples were collected directly in contact with shoreline sediments, and thus they are directly interconnected. The physical wave erosion on the coastal areas leads to flushing of also dense MPs from shore sediments into the coastal waters. A variety of MP particles in size and shape can thereby be flushed to the water. This may explain why a greater variety of MP particles are found

in the coastal waters than in sediments. These results reflect that the size of the MP particles is an explanatory factor for the distribution between sediment and water samples.

3.3. Shape of MP particles

Fiber, fragment, and pellet shapes of MP particles were found in sediment samples. Fibers and fragments were also found in the water samples (Fig. 4). Fibers constituted by far the dominant MP particles shape, accounting for 97% of the MP particles in both sediment and water samples. This high frequency of fibers is in agreement with the previously reported values (e.g. Thompson et al., 2004; Claessens et al., 2011; Dai at al., 2018; Sathish et al., 2019; Mu et al., 2019; Yuan et al., 2019; Firdaus et al., 2019; Zhang et al., 2019; McEachern et al., 2019; Aslam et al., 2020; Zhang et al., 2020). Fibers and pellets constituted 99 and 1 % of the shoreline sediments (Fig. 4c), while fibers and fragments accounted for 95 and 5 % of the sea surface sediments (Fig. 4d), respectively. The predominance of fibers suggests that the main source of MP particles in the study area could be from domestic wastewater effluents from laundry washing. In addition, the use of various types of commercial fishing gear, such as demersal or bottom trawls, gillnets, purse seines, poles and lines, and pots and traps, are potentially other sources of these MP fibers.

3.4. Color of the MP particles

MP particles in all sediment and water samples were mainly black-grey (40 and 49 %, respectively), though a significant amount of blue-green (26 and 17 %) and yellow-orange (20 and 16 %) particles were also found (Fig. 5). Shoreline sediments contained a higher percentage (37 %) of blue-green, and yellow-orange (20 %) MP particles compared with sea sediments, which were mainly comprised of black-grey (45 %), red-pink (14 %), and white-transparent (5 %) MP particles (Fig. 5). These colors point to various colored plastic products, such as textiles, packaging materials, and fishing gear, as potential sources of the fibers. Exposure to sunlight during their transportation in the environment causes photobleaching of their color (Kalogerakis et al., 2017; Wu et al., 2018; Firdaus et al., 2019). This may be why there are much lighter colors such as white/transparent and yellow/orange in sediments compared to the water as the MP particles in the sediments are assumed to be older, based on their smaller sizes that could have resulted from prolonged plastic degradation.

3.5. Chemical composition of MP particles

The polymer types and elemental composition of a set of 16 selected MP particles, comprising a range of sample matrixes, MP particle shape, and color, as well as abundance, are presented in Table 1. PET, PS, and NYL are the most abundant polymers in the MP particles in both sediment and water samples. In the sediment samples, black MP fibers were mostly made of PET and polyamide, more known as NYL, constituting PET-NYL copolymers, while blue and white MP

fibers constitute PET and Polystyrene (PS), comprising PET-PS copolymers (Supplementary material 3a-d).

In the water samples, black fibers are made of PETs and PET-PS copolymers, and blue fibers contain NYL and PET-PS copolymers (Supplementary material 3e-h). Yellow fibers in the water samples were made of PET (Supplementary material 3i). We hypothesize that the NYL MPs may come from ropes in fishing gear and that PET MPs could mainly from textile fibers. This is assuming that these are mainly primary MPs in the water and is based on 1) the abundance of fibers; 2) the most common use of the polymers; and 3) the predominant types of anthropogenic activities in the region. The discharge of treated domestic and textile industry wastewater effluents and fishing is thus likely a main source for MP contamination in the study area, although further research in relation to potential sources and hotspots is necessary.

MP particles with low density, and thus only slightly negative buoyancy, are expected to remain longer suspended in the upper water column. These particles thereby tend to end up more in the shore sediments, while those with high density and more negative buoyancy will sink rapidly down the water column over time, and are expected to accumulate in the sea surface sediments. Buoyancy, governed by the relative density difference between the density of MPs and the brackish water, is thus a key parameter governing MP particles within seawater. The brackish Caspian seawater has a density of around 1.01 g cm^{-3}, while PS, NYL, and PET have the density values of 1.05, 1.15, and 1.38, respectively (Lusher, 2015). Hence, all MP particles are negatively buoyant in the Caspian seawater. They are therefore at the onset expected to sink down the water column over time and be accumulated in the sea surface sediments. On the other hand, the density of the plastic materials differs due to the inclusion of air. This is especially the case for the styrofoam (PS) which floats. Nevertheless, our study shows that the MPs in the sea and shore sediments of the Caspian Sea are made of a narrow range of polymers. Although these polymeric materials per se have different buoyancy in the brackish water, the size and shape of the MP particles will likely have a greater effect due to their different hydrodynamic properties. Convection due to waves, as well as water current velocity, affects buoyancy and thereby residence time of the MP particles in the water, and thus where they settle. Vertical mixing of the water column may re-suspend and redistribute MP particles between sediment and water. Fibers become easily transported, are light, and are thus expected to have a lower settling rate than pellets. Additives used in the plastic material may also influence the density and thereby buoyancy. Biofouling, i.e. organisms' colonization on the polymers of MP particles (Ye and Andrady, 1991; Lobelle and Cunliffe, 2011), is another factor that will decrease buoyancy by increasing the density of MPs.

The surface morphology of 6 selected MP items, illustrated in Fig. 6, shows a relatively smooth surface texture, although fingerprints of chemical and mechanical weathering as grooves, fractures, pits, and cracks are found. The relatively soft morphology of the MP particles indicates that most

of the MPs were derived from primary sources, although due to weathering, some may degrade further into smaller MP particles.

Carbon (C) and Oxygen (O), along with minor amounts of inorganic elements, were detected on the surface of the 16 of the MP specimens that were studied in detail with SEM-EDS (Table 1). SEM-EDX has limed sensitivity for the analysis of metals. Inherently, C and O constitute the major elemental composition of MPs, though, despite large differences in the relative amounts of carbon and oxygen content between PET, NYL, and PS polymers, the content of C and O on their surfaces did not differ significantly. Trace amounts of Cu, Al, Si, Fe, Cl, Zn, Cd were also detected in some of the analyzed MP particles. Several of these elements are applied as additives in the plastic polymers in order to achieve special properties. For instance, inorganic additives, such as silica, are commonly incorporated into thermoplastics in order to improve their deformation resistance and physical strength. Fe and Cd, are commonly used as inorganic pigments to produce red and yellow colors (Bolgar et al., 2015). Al and Si are antioxidant elements that may have been used as additives to defer the oxidation of the plastic material. On the other hand, the inorganic elements may be found in minerals adsorbed on the surface of the MP particles. Sediment samples contain clay and silt minerals, which are composed of Al- and Si oxides. The presence of these elements may, therefore, be due to these minerals adsorbed to the surface of the MP particles. It is worth noting that some chemical treatments during sample preparation can influence subsequent characterization results. For instance, using $ZnCl_2$ and or NaI to extract MPs can affect the results of the elemental composition of MPs regarding the concentration of Zn, Cl, Na, and I, as already reported by Abbasi et al., (2019), and Mehdinia et al., (2020), and the treatment with H_2O_2 could affect the weathering observed in MPs from sediments.

3.6. Status of the MPs in the southern Caspian Sea relative to other localities

This research is the first to report the occurrence of MPs in the coastal waters, as well as sea surface sediments, of the southern Caspian Sea. In addition, the major sources of the MP particles have been assessed based on a comprehensive characterization of the MP particles. A compilation of literature data on MP particles frequency and characteristics in sediment and water of the southern Caspian Sea and other coastal and marine localities is presented in Supplementary material 4. Although a direct detailed comparison between our monitoring study cannot be done due to different sampling protocols and sample treatment it is clear that the concentration of MP particles in the seawater is lower in the studied area than what is commonly found for sites with a similar level of anthropogenic activity. The frequency of MP particles in the sediments of the southern Caspian Sea is also relatively lower than what is found in most of the other sites. The major polymers of MP particles in the sediment and water of the present study are somewhat similar to what is found elsewhere. Similarly, fiber is the major shape of MP particles in most of the other coastal sites.

4. Conclusion

MPs contamination in the coastal environment of the Caspian Sea along the whole Mazandaran province coasts has been found to be diverse in terms of the number of MP particles, with also strong heterogeneity in size and color of MP particles in both sediment and water samples. In contrast, the shape, morphology, and composition of the MPs were more homogeneously distributed. The spatial distribution pattern appears to be governed by major point sources, and hotspots of more diffuse anthropogenic sources, as well as transport processes. This study indicated that MP particles circulated between coastal waters, shore, and sea surface sediments of the Caspian Sea, leading to their uneven distribution in the different depths. Identifying conceptual mechanisms governing the transport of MP particles in the aquatic environment is challenging due to the variety of diffuse and point sources. Fibers, made of NYL, PET, and PS, are the main type of MP particles. Their source could be greywater from wastewater plants, treating domestic sewage and effluent from the textile industry, and the fishing industry activities. For abatement actions, there is an opportunity to address the sources of fibers and achieve a major impact in the coastal area of the Caspian Sea. A variety of elemental compositions were found in the MP particles. Hence, determining different kinds of additives and/or adsorbed chemicals (e.g., heavy metals and polycyclic aromatic hydrocarbons) in the MP particles and matching it with plastic products may be an area of focus in future research to pinpoint the main sources of the MP material.

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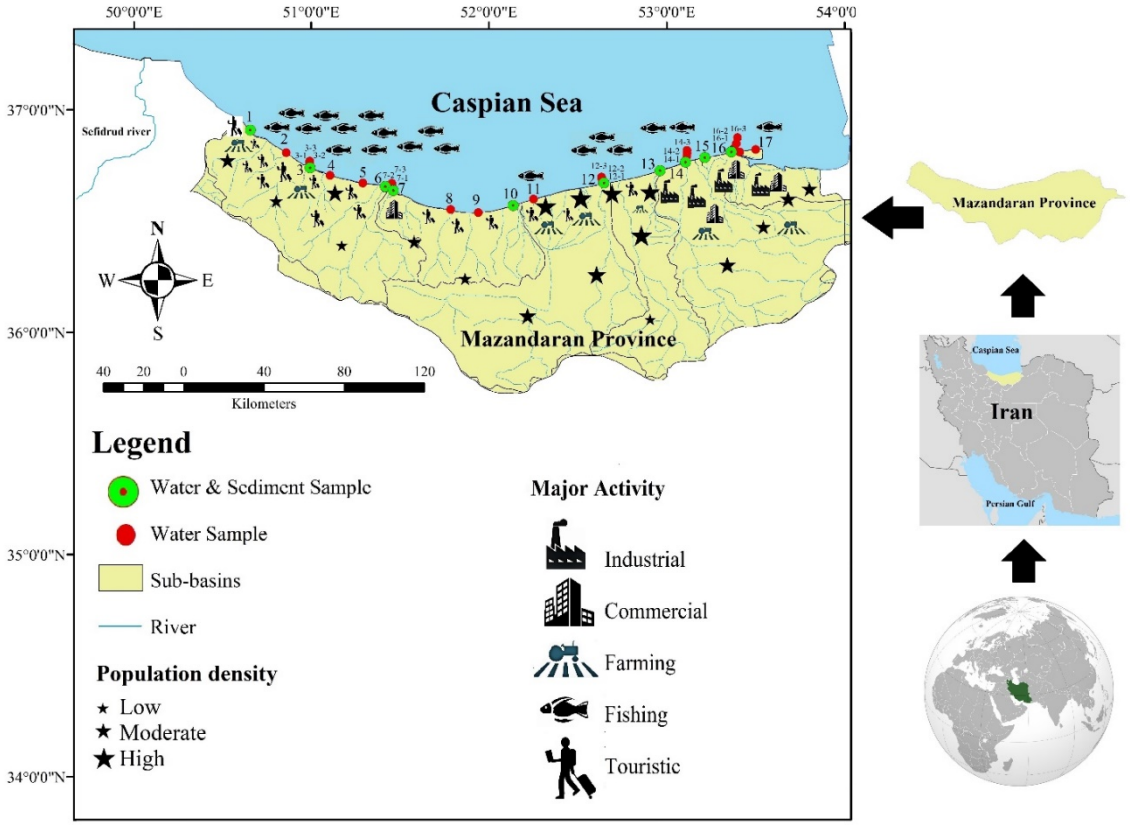


Fig. 1 Study area, local rivers, and their watersheds, sampling point location, and some potential major sources for the plastic pollution in each area

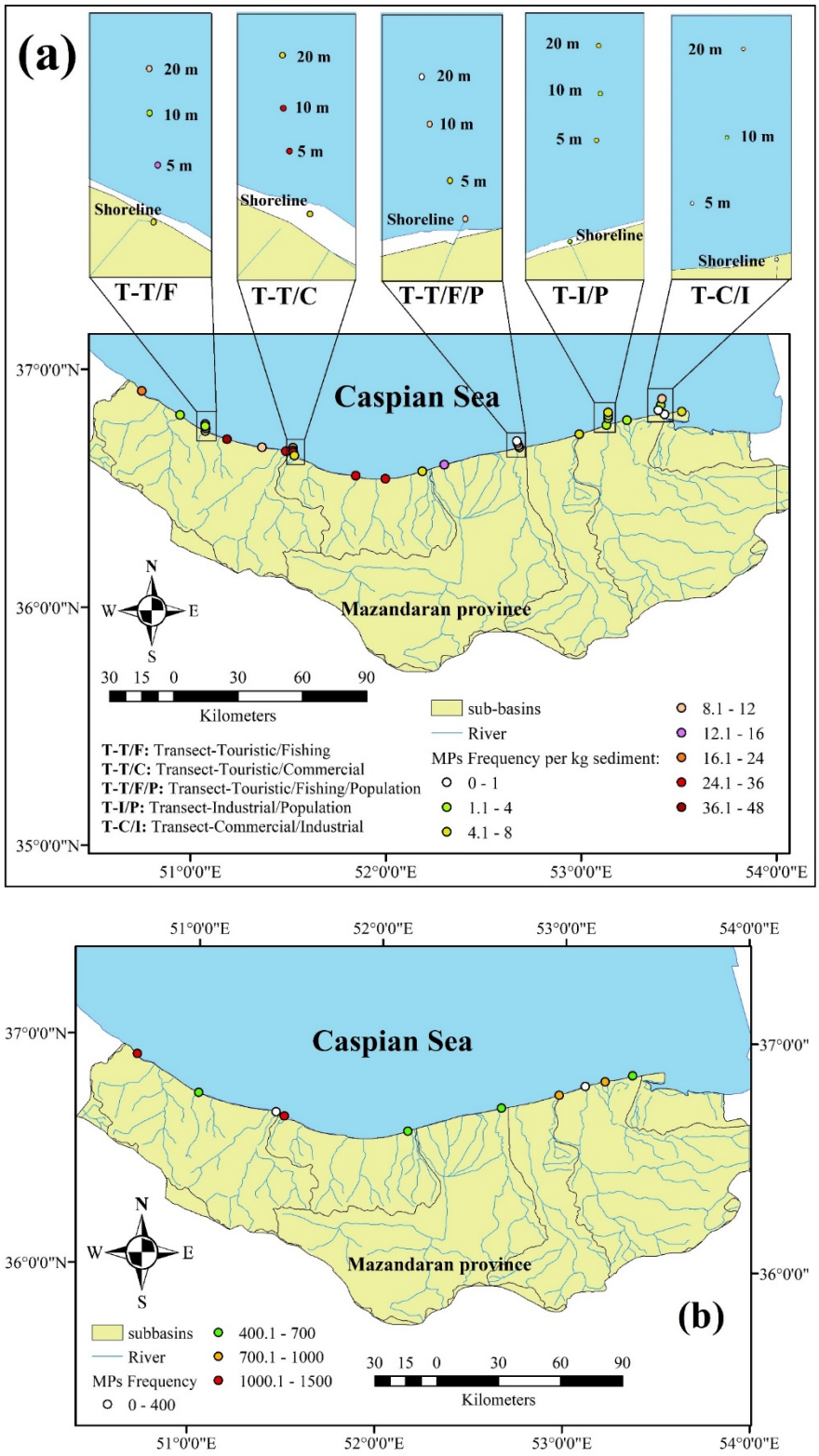


Fig. 2 Spatial MP items distribution in **a)** shoreline and sea sediments (including 5 transects), and **b)** coastal waters in the south of the Caspian Sea

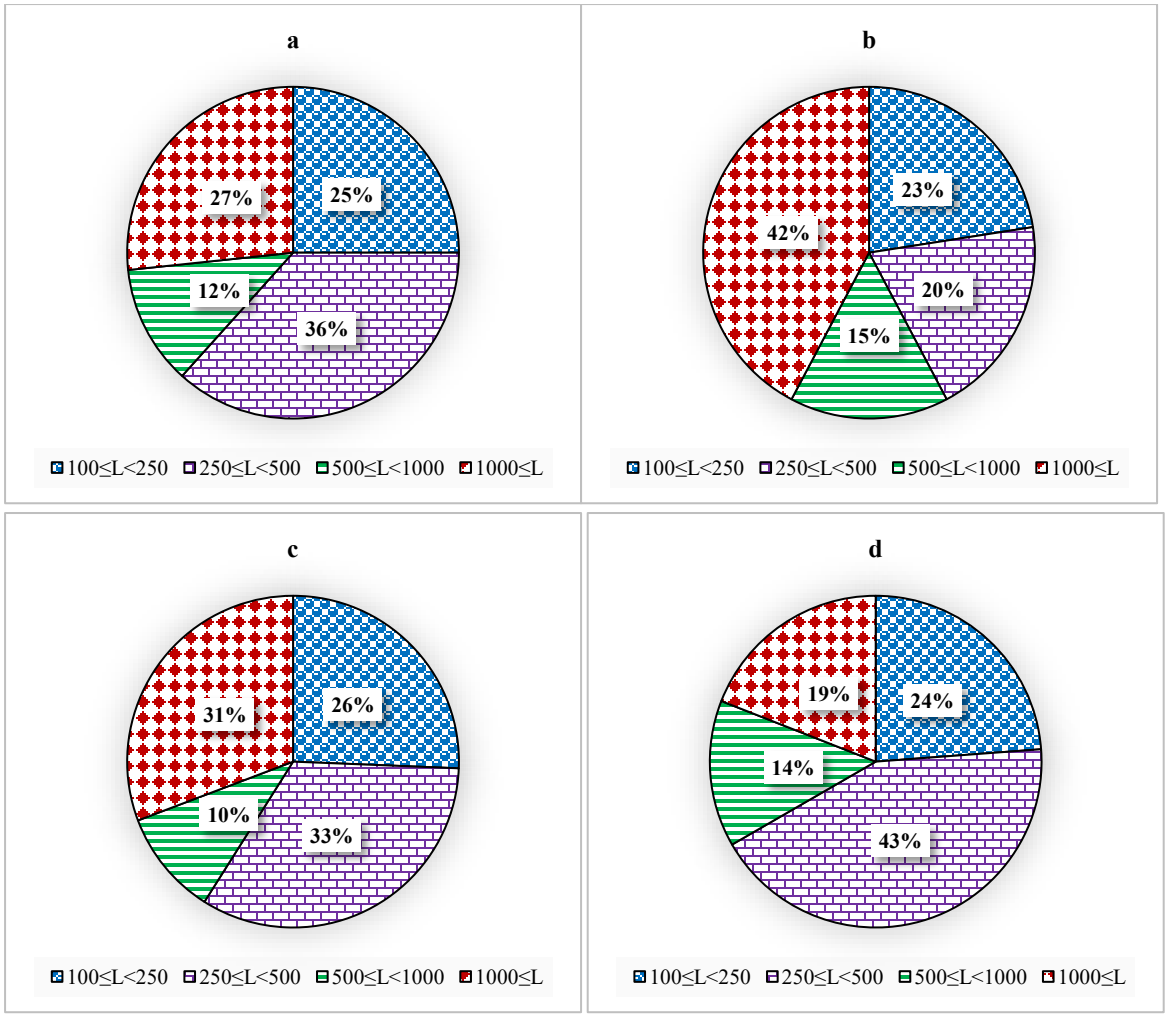


Fig. 3 Percent distribution of size classes (in µm) of MP particles in **a)** all sediment, **b)** water samples, **c)** shoreline sediments, and **d)** sea surface sediment

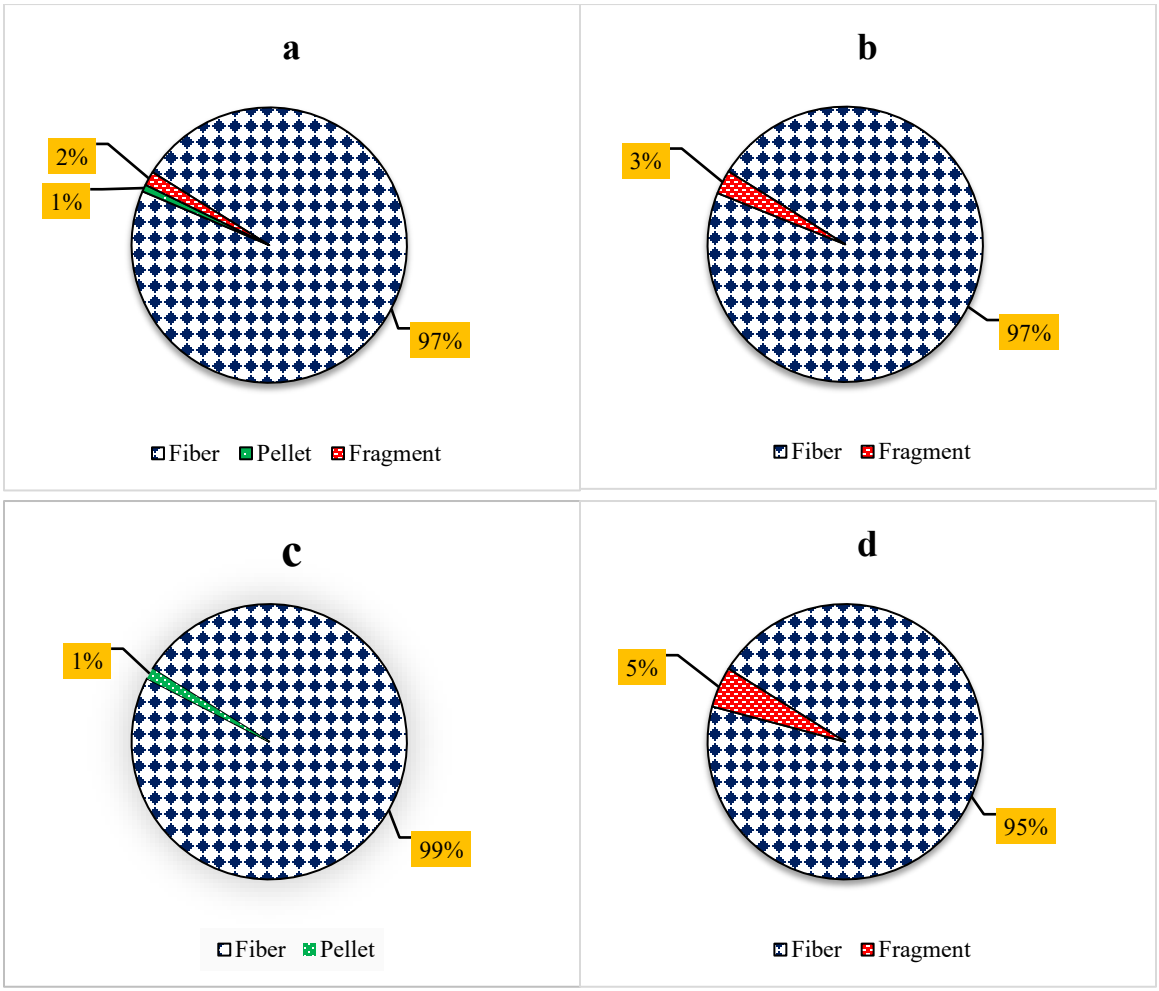


Fig. 4 Percent distribution of MPs shape types in **a)** all sediment samples, **b)** water samples, **c)** shoreline sediments, and **d)** sea sediments

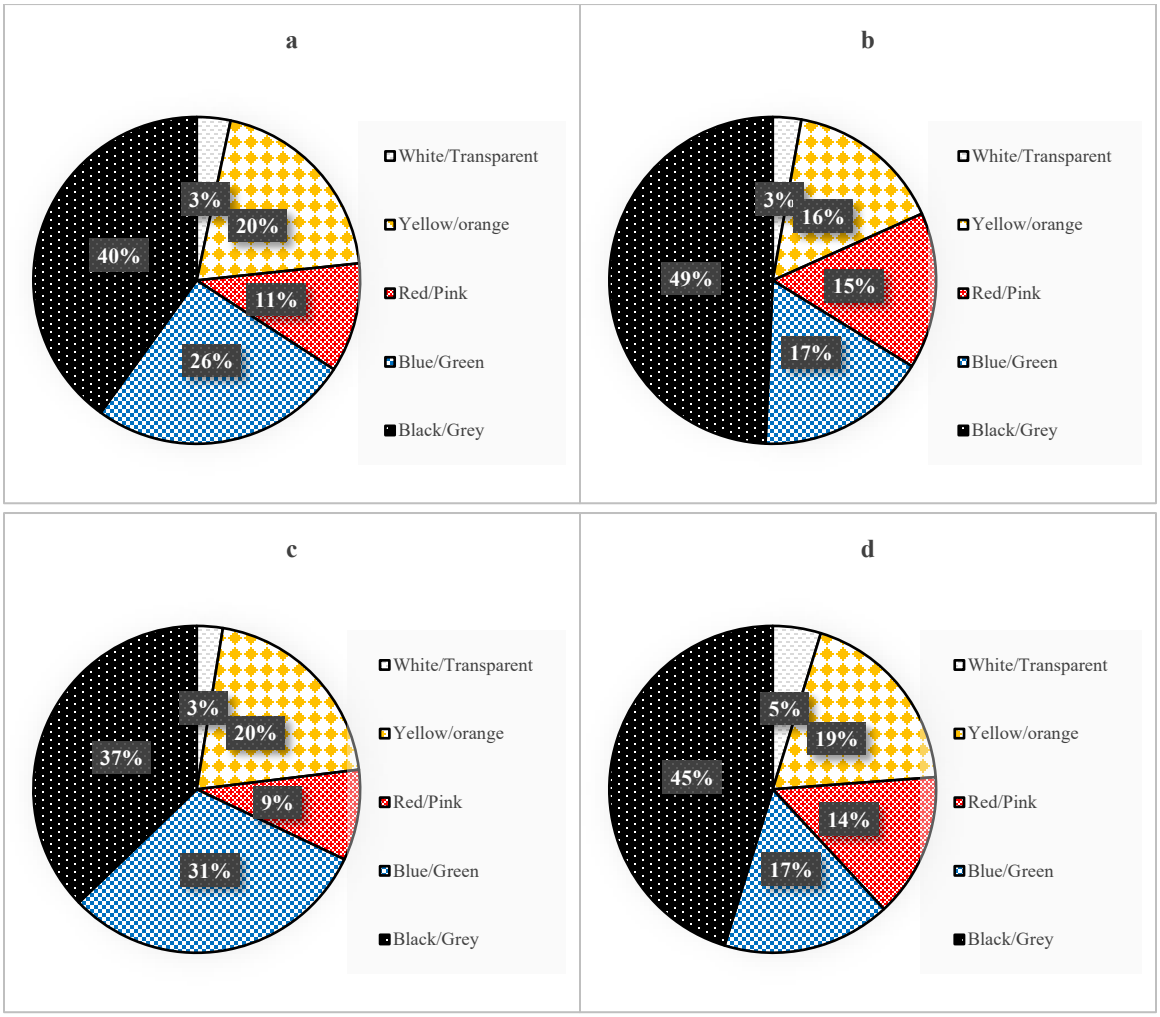


Fig. 5 Percentage of MPs color classes in **a)** all sediments, **b)** water samples, **c)** shoreline sediments, and **d)** sea sediments

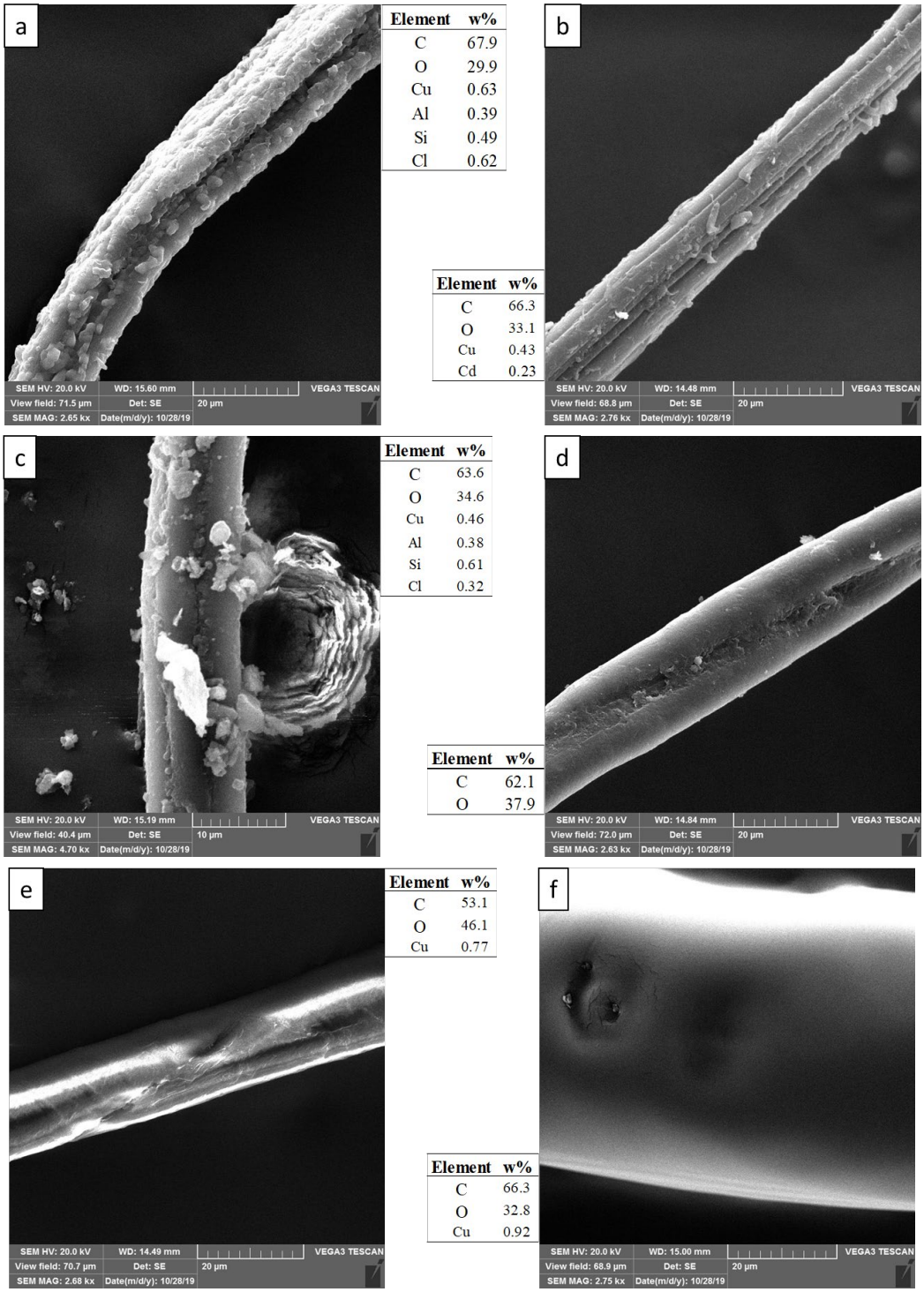


Fig. 6 SEM images showing the surface morphology along with chemical composition obtained by EDS of the selected MP fibers from sediment (a, b, and e) and water (c, d, and f) samples

Table 1 Physical and chemical features of selected MPs in sediment and seawater samples

Sample type	Sample Id	Shape	Color	Polymer type	Elemental composition (w%)								
					C	O	Cu	Al	Si	Fe	Cl	Zn	Cd
Sediment	15	Fiber	Black	PET-NYL	73.7	25.4	1	0	0	0	0	0	0
Sediment	12	Fiber	Black	PET-NYL	71	21.8	0.8	0	0	0	4.7	2	0
Sediment	14	Fiber	Black	PET-NYL	67.9	29.9	0.6	0.4	0.5	0	0.6	0	0
Sediment	7	Fiber	Blue	PET-PS	66.3	33.1	0.4	0	0	0	0	0	0.2
Sediment	4	Fiber	Blue	PET-PS	67.9	31.1	0.7	0	0	0	0.4	0	0
Sediment	8	Fiber	Blue	PET-PS	53.1	46.1	0.8	0	0	0	0	0	0
Sediment	1	Fiber	White	PET									
Sediment	16	Fiber	White	PS									
Water	1	Fiber	Black	PET	66.3	32.8	0.9	0	0	0	0	0	0
Water	16	Fiber	Black	PET	71.3	27.8	0.9	0	0	0	0	0	0
Water	13	Fiber	Black	PET-PS	65.3	31.9	0.6	0.6	1.4	0.2	0	0	0
Water	14	Fiber	Black	PET-PS	63.6	34.6	0.5	0.4	0.6	0	0.3	0	0
Water	7	Fiber	Blue	NYL	70	30	0	0	0	0	0	0	0
Water	6	Fiber	Blue	NYL	70.7	29.3	0	0	0	0	0	0	0
Water	12	Fiber	Blue	PET-PS	62.1	37.9	0	0	0	0	0	0	0
Water	7	Fiber	Yellow	PET									

Supplementary material 1: Number of MPs found in shoreline and sea sediment (per kg), and water (per m^3) samples, along with major sites features. Site number refers to the numbering in Figure 1.

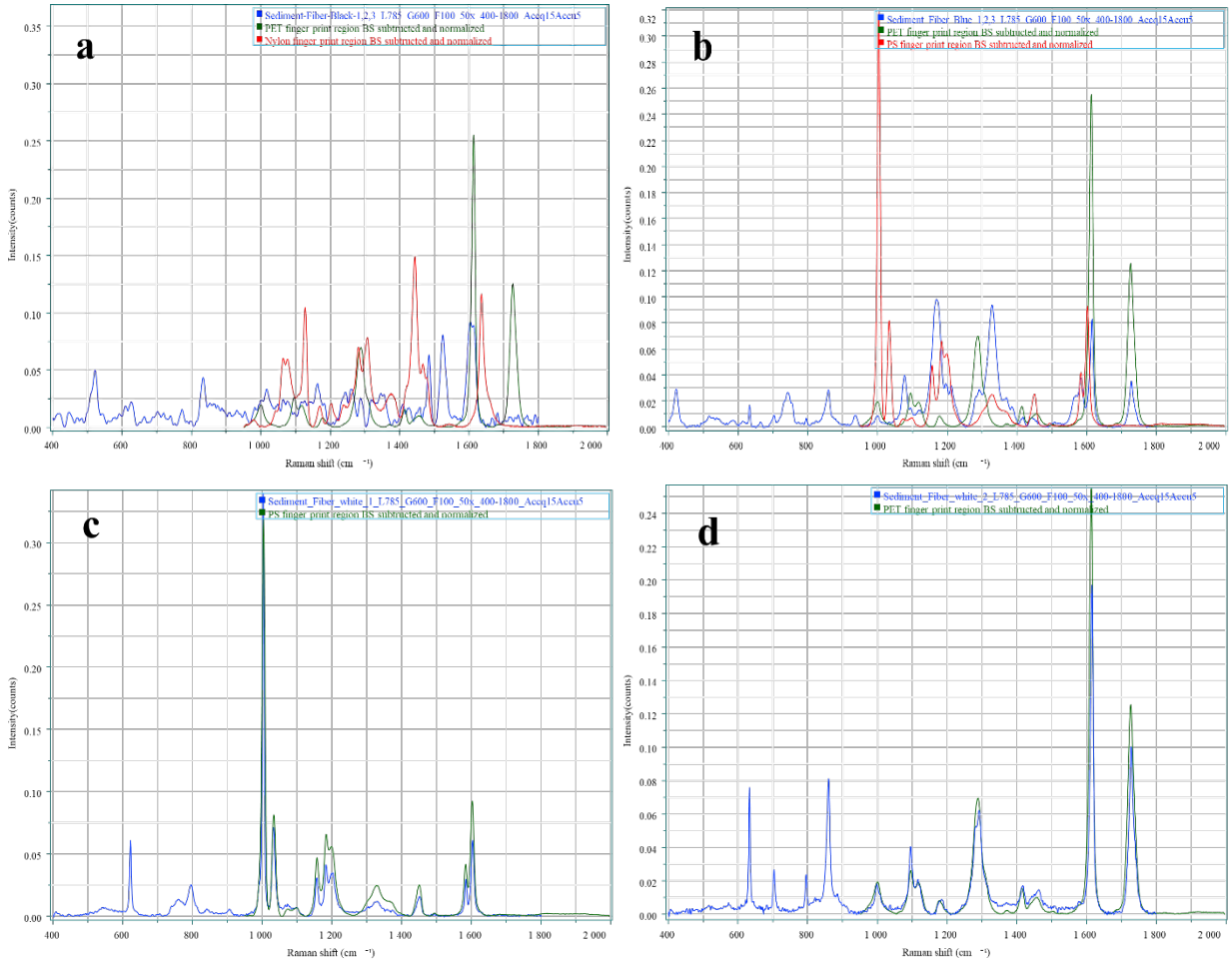
Site N.	Sample type	Water Depth (m)	MP items frequency		Major site features				
			Sediment	Water	Touristic	Farming	Commercial	Industrial	Fishing
1	Shoreline sediment & coastal water	-	36	1500	☒	☒			
2	Shoreline sediment	-	8	-	☒				
3(T-T/F)	Shoreline sediment & coastal water	-	36	600	☒				☒
3-1	Sea sediment	5	8	-	☒				☒
3-2	Sea sediment	10	16	-	☒				☒
3-3	Sea sediment	20	32	-	☒				☒
4	Shoreline sediment	-	36	-	☒				☒
5	Shoreline sediment	-	4	-	☒	☒			☒
6	Shoreline sediment & coastal water	-	8	200	☒				☒
7(T-T/C)	Shoreline sediment & coastal water	-	4	1100	☒		☒		
7-1	Sea sediment	5	12	-	☒		☒		
7-2	Sea sediment	10	4	-	☒		☒		
7-3	Sea sediment	20	4	-	☒		☒		
8	Shoreline sediment	-	12	-	☒				
9	Shoreline sediment	-	16	-	☒				
10	Shoreline sediment & coastal water	-	20	600	☒				
11	Shoreline sediment	-	48	-	☒	☒			☒
12(T-T/F/P)	Shoreline sediment & coastal water	-	16	500	☒				☒
12-1	Sea sediment	5	16	-	☒				☒
12-2	Sea sediment	10	0	-	☒				☒
12-3	Sea sediment	20	20	-	☒				☒
13	Shoreline sediment & coastal water	-	4	800		☒			☒
14(T-I/P)	Shoreline sediment & coastal water	-	8	300		☒		☒	☒
14-1	Sea sediment	5	0	-		☒		☒	☒
14-2	Sea sediment	10	12	-		☒		☒	☒
14-3	Sea sediment	20	0	-		☒		☒	☒
15	Shoreline sediment & coastal water	-	32	800		☒	☒	☒	
16(T-C/I)	Shoreline sediment & coastal water	-	12	700			☒	☒	
16-1	Sea sediment	5	24	-			☒	☒	
16-2	Sea sediment	10	16	-			☒	☒	
16-3	Sea sediment	20	4	-			☒	☒	
17	Shoreline sediment	-	12	-			☒		☒

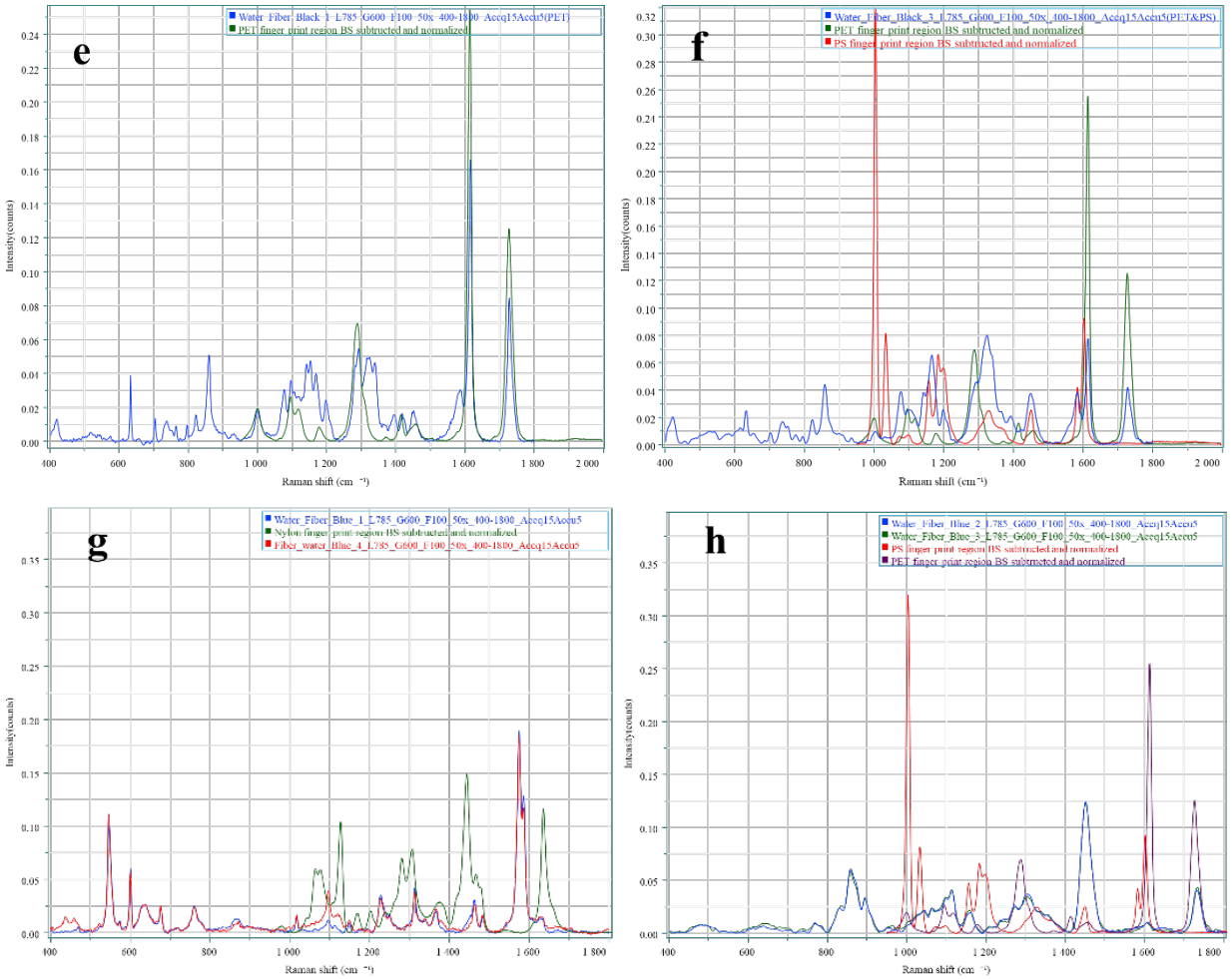
T-T/F: transect-tourism/fishing; **T-T/F/P**: transect- tourism/fishing/population; **T-T/C**: transect- tourism/commercial; **T-I/P**: transect-industrial/population; **T-C/I**: transect-commercial/industrial.

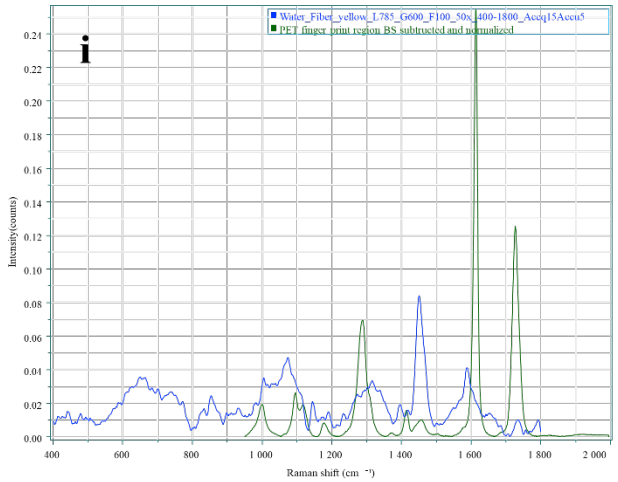
Supplementary material 2 Statistical summary of MPs found (N kg^{-1}) in sediment and water

Medium	Total	Min.	Max.	Mean	Median	S.D.	C.V (%).	S.W.	K.S.
All sediments	480	0	48	15	12	12.4	1.88	0.00	0.01
Shore sediment	312	4	48	18	12	13.9	1.32	0.02	0.04
Sea sediment	168	0	32	11	12	9.59	1.17	0.24	0.20
Water	7100	200	1500	710	650	378	1.21	0.57	0.20

N: number. SD: standard deviation. CV: coefficient of variance. S.W: Shapiro-Wilk test. K.S: Kolmogorov–Smirnov test


a
Sediment-Fiber-Black-1,2,3_1.785_G600_F100_50x_400-1800_Accq15Accu5
PET finger print region BS subtracted and normalized
Nylon finger print region BS subtracted and normalized
b
Sediment_Fiber_Blue_1,2,3_1.785_G600_F100_50x_400-1800_Accq15Accu5
PET finger print region BS subtracted and normalized
PS finger print region BS subtracted and normalized
c
Sediment_Fiber_white_1_1.785_G600_F100_50x_400-1800_Accq15Accu5
PS finger print region BS subtracted and normalized
d
Sediment_Fiber_white_2_1.785_G600_F100_50x_400-1800_Accq15Accu5
PET finger print region BS subtracted and normalized
Intensity(counts)
Raman shift (cm⁻¹)


e
Water_Fiber_Black_1_L785_G600_F100_50x_400-1800_Accq15Accu5(PET)
PET finger print region BS subtructed and normalized
Intensity(counts)
Raman shift (cm⁻¹)
f
Water_Fiber_Black_3_L785_G600_F100_50x_400-1800_Accq15Accu5(PET&PS)
PET finger print region BS subtructed and normalized
PS finger print region BS subtructed and normalized
Intensity(counts)
Raman shift (cm⁻¹)
g
Water_Fiber_Blue_1_L785_G600_F100_50x_400-1800_Accq15Accu5
Nylon finger print region BS subtructed and normalized
Fiber_water_Blue_4_L785_G600_F100_50x_400-1800_Accq15Accu5
Intensity(counts)
Raman shift (cm⁻¹)
h
Water_Fiber_Blue_2_L785_G600_F100_50x_400-1800_Accq15Accu5
Water_Fiber_Blue_3_L785_G600_F100_50x_400-1800_Accq15Accu5
PS finger print region BS subtructed and normalized
PET finger print region BS subtructed and normalized
Intensity(counts)
Raman shift (cm⁻¹)



Supplementary material 3 Raman spectra for the selected MPs with specific shape and color in sediment (a-d) and water (e-i) samples

Supplementary material 4 MPs frequency in sediment (kg^{-1} DW) and water (m^{-3}), along with their major polymeric and shape types in the study area and other localities

ND: Non-detected; **a:** Mean ± SD; **b:** range; **c:** total abundance

Region	Medium	MPs frequency	Major Polymer	major shapes	Reference
South of the Caspian Sea, Iran	Sediment	15±12.40[a] (ND-48)[b], 480[c]	PET, PS, NYL	fiber	Present study
	Water	710± 378.45[a] (200-1500)[b]	PET, PS, NYL	fiber	
Southern coasts of the Caspian Sea, Iran	Sediment	25-330[b]	PS, PE	fiber	Mehdinia et al., 2020
Southern coasts of the Caspian Sea, Iran	Sediment	210±81[a]	PS, PE, PP	Foam, fragment	Mataji et al., 2019
	Water	34.49/km^2	PS, PE, PP	Foam, fragment	
Poyang Lake, China	Sediment	54-506[b]	PP, PE	fiber	Yuan et al., 2019
	Water	5000-34000[b]	PP, PE	fiber & film	
Lebanese coast, Lebanon	Sediment	2433±2000[a]	PP	fragment, fiber, pellet	Kazour et al., 2019
	Water	4.3±2.2[a]	PE	fragment, microbeads, fiber	
Tampa Bay, Florida	Sediment	280±290a (30-790)[b]		fiber	McEachern et al., 2019
	Water	940±502[a]		fiber	
Ma'an Archipelago, Shengsi, China	Sediment	30±0.0 - 80±14.1[a]	Cellophane	fiber	Zhang et al., 2020
	Water	200±100 - 600±200[a]	PE-PP, PP, PE	fiber	
Sidi Mansour Harbor, Tunisia	Sediment	252-5332[b]	PE, PP	fragment, granule	Chouchene et al., 2019
Banten bay, Indonesia	Sediment	267±98[a] (101–431)[b]	PS	foam, fragment, granules, fiber	Falahudin et al., 2020
Jagir Estuary, Indonesia	Sediment	92-590[b]	PES	fiber	Firdaus at al., 2019
Coast of Dubai, UAE	Sediment	59.71[c]	PE	fiber	Aslam et al., 2019
Coastal areas of Bandar Abbas, Iran	Sediment	38-367[b]	PE, NYL, PET	filament, fragment	Yazdani Foshtomia et al., 2019
Coastal sediments of Khark Island, Persian Gulf	Sediment	59-217[b]		fragment	Akhbarizadeh et al., 2017
Coastal areas of Tamil Nadu, India	Sediment	33±30 - 439±172[a]	PE	fiber	Sathish et al., 2019
Northern Bering and Chukchi Seas, China	Sediment	ND-68.88[b]	PP	fiber	Mu et al., 2019
Kaliningrad beach, Russia	Sediment	1.3-36.2[b]	PS	foam	Esiukova, 2017