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Effect of land use on microplastic pollution in a major boundary waterway: the Arvand River

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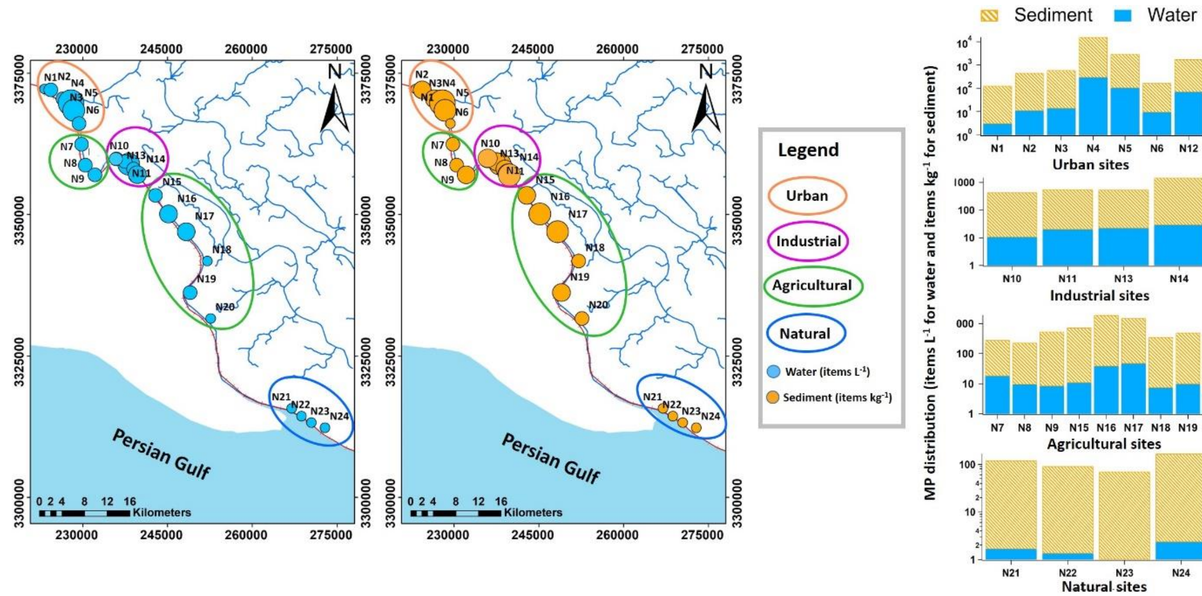
Graphical Abstract



Highlights

The first microplastic (MP) assessment in water/ sediments of the Arvand River

MP contamination was greater in urban than in industrial/agricultural/ natural areas

Fibres and 200-500 µm, >1000 µm dominant in Arvand river. No predominant colour

Urban effluents could be responsible for MP hotspots in water and sediment

Clay and organic matter can control MPs distribution in sediment

Abstract

The occurrence of microplastics (MPs) was investigated in the Arvand River (Iran). The Arvand River (200 Km) is a major water body that flows through land with diverse use and it meets the Persian Gulf. The MPs abundance measured in 24 stations located along the river, in water and sediment, ranged between 1 and 291 items L^{-1} and 70 to 15620 items kg^{-1} (dw). The majority of MPs were fibres, black/gray and yellow/orange in color, and mainly 250 - 500 µm and > 1000 µm in size. Polyethylene terephthalate (PET), polypropylene (PP), nylon (NYL), high-density polyethylene (HDPE), and polystyrene (PS) were found in sediment samples. All the mentioned polymers except HDPE are also identified in the water samples. The dominant polymers were polyethylene terephthalate (PET) and polypropylene (PP) in water; and PET and polystyrene (PS) in sediment. The vicinity of urban wastewater effluents could be behind MP pollution in both water and sediment. Significant differences ($p < 0.05$) of MP concentrations were affected by different land uses when comparing undisturbed natural area with urban, industrial and agricultural areas reflecting contribution of activities inland as the release of MPs into the river. A strong correlation between MP fibers and fragments carried out by PCA biplots revealed similar distribution of fibers and fragments in the water. In the sediment samples, fiber and fragment MP particles are significantly correlated with colloidal particles (e.g., clay and OM) suggesting the significant role of the colloidal particles in aquatic ecosystem of Arvand River in transport and fate of MPs. This study contributes to the better understanding MP pollution in a major river system.

Keywords: Microfibre, microbead, water, sediment, land use, Persian Gulf

Introduction

Marine plastic litter as emerging environmental pollution is a global challenge (Isobe et al., 2021). Rivers are a major route of plastic litter to the ocean. Each year, between 1.15 and 2.41 million tons of plastic waste are estimated to reach the oceans from the rivers (Lebreton et al., 2017). Sewage discharge and runoff from urban areas are major contributors of plastic pollution to rivers worldwide (Strokal et al., 2021). Moreover, effluents from industries with improper treatment, and agricultural drainage strongly impact the plastic load in riverine systems (Gallagher et al., 2016; Piehl et al., 2018). See https://theoceancleanup.com/sources/.

Important amounts of plastic particles appear to be entering aquatic ecosystems. As plastics are designed to last, plastic waste accumulate in the marine environment for a notably long time (Delacuvellerie et al., 2021; Wang et al., 2016). Under the influence of a number of factors such as UV irradiation, oxidants, hydrolysis and mechanical abrasion, large plastic pieces disintegrate into smaller fragments (< 5 mm) (Song et al., 2017) generally referred to as microplastics or MPs. Furthermore, some plastics directly enter the environment as primary MPs which are manufactured for a number of purposes such as personal care products, industrial abrasives (Lehtiniemi et al., 2018) or as precursors for the production of polymers. Following the entrance of MPs to surface water, they distribute in the water column (surface water and subsurface water) to superficial sediments, (Hidalgo-Ruz et al., 2012). In particular, low-density MPs can be re-suspended to surface and subsurface water, while denser MP particles remain in the lower part of the water column and become entrapped and accumulate in surface sediments (Defontaine et al., 2020; Lenaker et al., 2019). The fate of the sedimentated MP in superficial sediments will be influenced by the sediment structure, grain size, its organic matter content and the bioturbation but knowledges of these processes are relatively scarce (Pohl et al., 2020).

Due to small size, their olfactory trap, MPs are easily mistaken for food and hence they can be ingested or uptaken through skin and gill by a wide range of marine and freshwater organism (O'Connor et al., 2020; Savoca et al., 2016). The release of plastic additives and adsorbed toxic elements, persistent organics and substances of concern such as antibiotics and even microorganisms (Delacuvellerie et al., 2022) may occur during digestion of MPs in the digestive tracts of marine species (Caruso, 2019; Yu et al., 2021). Thus, MPs can be potential vectors of toxic chemicals and their intake could result in toxicity to aquatic life (Pannetier et al., 2020). Contaminated fish and seafood can result in MPs ingested by humans and affect our health directly (Blackburn and Green, 2021). Recent studies indicates that human exposure to MPs and nanoplastics (NPs) might cause oxidative stress, cytotoxicity, neurotoxicity, reproductive toxicity, carcinogenicity, and translocation to other organs (Anbumani and Kakkar, 2018; Meng et al., 2022; Rahman et al., 2021).

The Arvand River constitutes part of the southern border between Iraq and Iran. The total length of the Arvand River is 200 km (Patiris et al., 2016). Its width varies between 232 and 800 m and its depth varies between 7 and 20 m (Rahimi Moazampour et al., 2021). The river is the result of confluence of Tigris and Euphrates rivers (in Iraq), and the Karkheh and Karun rivers (in Iran). Arvand is a wide navigable river that flows through three major cities including Basra (1,381,731inhabitants) in Iraq, and Abadan (231,476 inhabitants) and Khorramshahr (133,097 inhabitants) in Iran (Hosseini et al., 2013). Most of the aquatic species especially fish caught in this river is consumed by local inhabitants. Furthermore, some aquaculture products are exported to gulf emirates and Southeast Asian countries (Soltani et al., 2019). Arvand River plays an important role in supplying potable water and it supports agriculture and industries such as Abadan oil refinery and Abadan Petrochemical complex. The area where the Arvand River flows has hot humid summers with mild winters, typically. According to internal report of Meteorological Department of Khuzestan Province, the annual average precipitation of the area is 154.5 mm (year 1971-2021). The minimum and maximum temperatures are -4 and 53 °C with the annual average of 25.3 °C, respectively (year 1971-2021).

This study will focus on the assessment of MP pollution in the Arvand River. It is flowing North East the Persian Gulf, it is the most important inland freshwater body in Iran (Zahed et al., 2008) and it has associated variety of activities that could result in plastic pollution and this river is source of fish for locals and exportation. This study is aimed to assess the effect of different land use on MPs distribution in water and sediment and this knowledge will inform pathways and fate of MP in large rivers located in areas of comparable societies.

Materials and methods

Sample collection and preparation

Surface water and sediment samples were collected considering various land uses within the river basin (i.e., urban, industrial, agricultural and natural areas). Detailed information of 24 sampling sites (N1 - N24) is presented in the Table S1 (Supplementary Information). Briefly, seven sites were located within the urban areas of the cities of Abadan and Khorramshahr (N1-N6, N12); four sites were near industrial units including the oil refinery and petrochemical complex in Abadan (N10, N11, N13, N14); nine sites were near agricultural farms (N7-N9, N15-N20); and four sites were natural areas (N21-N24) far from populated areas in the estuary where the river meets the Persian Gulf (Fig. 1). At each station, 3 L water samples were collected in pre-cleaned amber glass bottles with aluminum foil -lined caps. Specifically, sample bottles were rinsed at least twice with surface water at each station and then fully submerged till the mouth of bottle was approximately

20 cm below the surface then capped immediately after filling. Sediment samples (3 kg, top 5 cm) were simultaneously collected in each station using cleaned stainless-steel Van Veen grab sampler and placed in wide mouth amber glass jars. All samples were transported to the laboratory and stored at 4°C until further analysis. Sampling took place in January 2018.

Physico-chemical parameters of sediment samples

Directly after picking, each sediment sample was divided into two subsamples. The first part was stored at room temperature in the laboratory and sieved through a 2-mm sieve for physico-chemical characterisation. The second part of the sample was used for monitoring MPs. The physico-chemical characterisation included sediment pH, electrical conductivity (EC), organic matter and cation exchange capacity (CEC). pH and EC were determined according to Ryan et al. (2001): after 10 min stirring of a 50 g dry sediment in 50 ml distilled water pH was measured using a pH-meter (Eutech instrument, Waterproof CyberScan PCD 650, Singapore). EC was determined using 1:5 water extracts (w:v) with a EC-meter (CyberScan PCD 650). Organic matter content was determined using the loss-on-ignition method (Miyazawa et al., 2000) while the hydrometer method was used for particle size distribution (Gee and Bauder, 1986). The ammonium acetate method (Kahr and Madsen, 1995) was employed to determine cation exchange capacity (CEC) in the soil.

Extraction and analysis of MPs

Water samples with recorded volume were filtered using a 2 µm pore size BOECO filter paper (grade 391, Germany) assisted with vacuum. Each filter paper was placed in a watch glass dish, covered with aluminum foil and dried in an oven for 24 h at 50 °C. Then, the wet sediment samples were oven-dried at 50 °C for three days, weighed, and sieved through a 5-mm stainless steel sieve. The MPs were purified using density separation technique (Dekiff et al., 2014; Nuelle et al., 2014). Briefly, dry sediment samples (200 g) were treated with 35% (v/v) hydrogen peroxide (H_2O_2) for 21 days allowing wet peroxide oxidation to remove organic matter that impedes MP detection (Prata et al., 2019). Subsequently, the samples were vacuum filtered through BOECO filter paper and rinsed with filtered deionized water to remove remaining H_2O_2. After, samples were brought to complete dryness on a sand bath at 60°C. Sodium iodide (NaI) solution (1.6 g/cm^3) was added and shaken for 5 min at 350 rpm and left to settle for 1.5 h. The cleared solution supernatant was centrifuged (for 5 min at 4000 rpm) and then filtered. Flotation (n=3) was carried out to separate MP. Supernatants were transferred to the same filter. Finally, filters were left to dry at room temperature and kept in a petri dish for further MPs visual assessment.

A binocular optical microscope with × 200 magnification (Carl-Zeiss, Oberkochen/West Germany) was used to observe the filter membrane. Color, size and shape of MP fragments were recorded. The number of MPs in water and sediment are reported as items/m^3 and items/kg, respectively.

Surface morphology and elemental composition of MPs were characterized by Scanning Electron Microscope (SEM, TESCAN Vega 3, Czech Republic) coupled with an energy-dispersive X-ray detector (EDX). MPs were coated with gold previous to the analysis.

Chemical composition of MPs was determined using a high-resolution Raman spectroscopy (Lab RAM HR Evolution, Horiba, Japan) with a 785 nm (max power = 17 mW) laser. MPs type was recognized via comparing the obtained MPs spectrums with standard database.

Prevention of contamination

Before sampling, tools and containers to be used were washed with deionized water pre-filtered with 2 µm BOECO filters (grade 391, Germany). Prior analysis, lab utensils and glassware were rinsed with the pre-filtered water and with ethanol. The work surfaces used for MPs extraction and counting were wiped with cotton cloth impregnated with ethanol. Cotton lab coats and clothing and nitrile gloves were the only ones worn while the MP study was ongoing. All containers used for the sampling and analysis were made of glass or stainless steel. To minimize contamination by airborne MP particles and fibres, reagents and solutions were filtered throughout the procedure using 2 µm BOECO filters (grade 391, Germany). Laboratory blanks (consisting of material collected onto open clear glass petri dishes left near the working area during the MP purification) were analysed in parallel to water and sediment samples. A total of 3 laboratory blanks were carried out (No MPs were detected in controls).

Data analysis

Statistical tests were conducted on log-normalized data using SPSS 23 software. The independent Mann-Whitney U test and t-test were applied to compare means of MP concentrations and discriminate significant differences between water and sediment. The homogeneity of variances for the t-test was checked using Levene's statistic test. Kruskal-Wallis test was carried out to reveal significant differences ($p < 0.05$) of pH in sediment samples among the four investigated land uses. Furthermore, to compare the means of MP concentrations between different land-uses, a one-way analysis of variance (ANOVA) was used. To check the equality of means and verify significant differences between the means, the robust Welch and Brown-Forsythe tests based on p-values < 0.05 were employed. The Levene statistic test was used to check the homogeneity of variances and select the most efficient way to carry out one-way ANOVA. Dunnett's T3 method was used to carry out ANOVA analysis. To find interrelations between variables, the two-dimensional principal component analysis (PCA) was applied using Minitab 16, based on eigenvalues greater than 1, and the varimax rotation method. To obtain optimum component numbers and adequacy of samples, the Kaiser normalization and Kaiser–Meyer–Olkin (KMO) methods were applied, respectively.

Results and discussion

Distribution of MPs in water and sediment

The physicochemical characteristics of the sediment samples is given in Table S2 and section S1. Microplastics were found in all surface water and sediment samples from the 24 sampling sites distributed along the Arvand River (Fig. 1). A total of 2218 and 6199 MP items were collected in water and sediment samples, respectively, from a total of 3 L and 0.2 kg of sediment. Compared to other urban rivers, MP items in surface water of Arvand River is higher than Coyote Creek, USA (41 items m^3) and San Gabriel River, USA (170 items m^3) and lower than Los Angeles River, USA (3473 items m^3) (Moore et al., 2011). The distribution of MPs in areas with contrasting land use on the banks of the river are compiled in Table 1. The MPs distribution in the water and sediment samples indicated that sediments could accumulate greater levels of MP pollution ($p < 0.05$): it ranged from 1 to 291 items L^{-1} in water and from 70 to 15620 items kg^{-1} dw in sediment. The number of MPs in surface water is 42 times lower (Mann-Whitney U test, $p < 0.05$) than in the sediment samples. Scherer et al. (2020) also found higher MP particles in sediment compared to water in the Elbe River. Thus, sediments could be long-term sink of MPs (Ding et al., 2019; Scherer et al., 2020).

Spatial heterogeneity in MPs distribution was observed in the study area in both water and sediment samples. The MP concentrations in different land uses showed the following decreasing trend: urban > industrial > agricultural > natural areas in surface water and sediment. The highest MPs concentrations were found in samples from the urban area representing 68% (in water) and 69% (in sediment) of all the items found, followed by industrial and agricultural land uses. In particular, N4, which was collected near an urban wastewater discharge to the Arvand River, had the highest MP concentration in water and sediment. Wastewater effluents from Khorramshahr were discharged into the river without any treatment, and this led to levels found at N4. These results agree with greater MP pollution in urban soils than in industrial soils in a study in Ahvaz (Iran) (Nematollahi et al., 2021). The average abundance of MPs in surface water and sediment in the urban area was about 45 and 28 times higher than natural area. Tibbetts et al. (2018) also reported greater MPs abundance in urban section of the Tame River compared with rural area. In urban areas MPs can originate from different sources such as building sector, manufacturing industries, road litter, overflow from sewer, and vehicles traffic (Kawecki and Nowack, 2020). At industrial area, the Arvand River is impacted by receiving industrial effluents from Abadan oil refinery and Abadan petrochemical complex, which could be a major source for MPs inputs to the river. Another industrial unit in the area is Khorramshahr soap factory, where microbeads were used to manufacture soap and MPs could be discharged to the Karoon River and eventually reach the Arvand River. Although, this factory has not been active for the last 10 years MPs pollution could persist in the sediment. Alam et al. (2019) showed that, in a densely populated region with an industrial complex, MP contamination occurs in river sediments.

Agricultural input is also an important source of MPs pollution at the Arvand River area. In agricultural sector of the Arvand River, application of plastic mulches in vegetable cultivation result in MPs release. Moreover, in the vicinity of Arvand River, plastics are frequently used in greenhouse farming, winter cultivation and covering small date palm trees in different stages of growth. Plastic liners are also used in ponds to increase irrigation water velocity and prevent water loss. Previous investigations have also shown that a number of farming activities contribute to MPs pollution (Cao et al., 2021; Chen et al., 2020). In natural area, fishery activities and marine transportation will affect the contamination of water and sediment with MPs (Bringer et al., 2021).

Characterization of MPs

The identified MPs in water and sediment samples varied in shape, color and size, and this diversity is illustrated in Fig. 2. In both surface water and sediment samples, fibers were the dominant MP shape in different land uses including urban (79% of water samples and 85% of sediment samples), industrial (74% of water samples and 88% of sediment samples), agricultural (80% of water samples and 88% of sediment samples) and natural (95% of water samples and 93% sediment samples). Fragment items were also relatively abundant in water and sediment samples compared to other MP types. Importantly, spherules were least abundant MP in water and sediment samples (Fig. S1). The high number of MP fibres and fragments may derive from fragmentation or weathering of discarded plastics transported over long distances by rivers (Andrady, 2017; Mani et al., 2015). Domestic washings, farm equipment, fishing nets and gear, atmospheric deposition and urban runoff, which includes debris from tyres among other types of plastics, are also potential sources of plastic fibers and fragments (Cesa et al., 2017; de Jesus Piñon-Colin et al., 2020; Dris et al., 2016).

Black/grey and yellow/orange were the dominant MPs colors in the surface water and sediments (Fig. S2). In general, MPs vary greatly in color depending on the parent material, but yellow/orange were the most common colors among the MPs found in the urban, industrial and agricultural sectors, while black/grey were the dominant MPs color in natural area. Different MP colors originate from various sources (Pan et al., 2019b; Xu et al., 2018). Moreover, environmental weathering especially ultraviolet oxidation (Wang et al., 2020) and MP extraction treatment (Karami et al., 2017; Nuelle et al., 2014) may cause MP color change.

In surface water the 250 –500 µm size fraction included higher proportion of MPs in most sampling sites. In parallel, 250 - 500 µm and > 1000 µm MPs prevailed in the sediment samples (Fig. S3). The > 1000 µm size is the prevalent fraction in sediment samples within urban and industrial areas. Similarly, > 1000 µm is the most dominant size in water samples collected from urban areas. In the sediment samples of the Arvand River, larger particles (> 1000 µm) were found to be more abundant than small MPs. This can be explained by higher densities of larger particles, causing large particles to settle more quickly (Di and Wang, 2018). The distribution, fate and retention of MPs is significantly affected by the size of MP particle, regardless of the polymer type (Sagawa et al., 2018).

Morphology of MPs

SEM - EDX analyses with the elemental composition at the surface of MP are illustrated in Fig. 3. The EDX spectra of most particles exhibited a strong C signal, followed by a smaller O signal. The high percentage of C and O show that the detected MPs are organic substances that could be plastics or natural polymers. Moreover, EDX analysis of the studied MPs showed that in addition to large amounts of C and O, minor amounts of other elements such as Na, Mg, Si, Cl, and Ca, Ba, S, Fe, and I also occur as polymer additives, adhered materials on the MP surfaces and MPs chemicals used in the procedure (e.g., NaI used in MP floatation during sample treatment).

Morphological features of MP surfaces with various degree of weathering were also examined by SEM analysis (Fig. 3). Compared to some fiber particles separated from water samples with relatively smooth surfaces (Fig. 3 a), most extracted MPs from sediment samples showed uneven surfaces with damaged and broken edges (Fig. 3 d, e and f). Spherule particles show weathering features as pits (Fig. 3 c). The cracks, flakes, holes and small grooves were determined in some fiber and fragments in sediment samples (Fig. S4). The relatively rough surfaces of most MPs particles indicate that the particles probably have undergone different levels of weathering and fragmentation processes. In aquatic environments, MP particles may be affected by weathering under chemical, mechanical and biological degradation resulting in changes in MPs surface morphology (Corcoran, 2020). The surface roughness and cracks probably generate during particle transportation and prolonged residence in aquatic environment (Pan et al., 2019a) but some could be generated during the SEM imaging of the sample. Weathering of MPs can be determined by linear cracks, pits and flakes and adhering materials (Wang et al., 2017) which affect their sorption capacity towards other materials such as organic and inorganic chemicals (Dong et al., 2020). Cai et al. (2018) used SEM images to demonstrate that the surfaces of the pristine MPs become rougher under UV irradiation and it makes plastic susceptible to fragmentation (Wang et al., 2021).

The EDX results of two weathered particle revealed a decrease in C (W%) in the weathered MP particles with rough surfaces (Fig. S4 a, b) compared to the pristine MPs with smooth surfaces (Fig. 3 a, b, and e). During weathering processes physicochemical properties of MPs (e.g. color, size, structure, mechanical characteristic and oxygen functional groups) may result in release of toxic additives and absorbed chemicals (Liu et al., 2020). The toxic leachates from weathered MPs cause potential adverse effects on organisms (Campanale et al., 2020; Simon et al., 2021). Using

SEM/EDX, Fries et al. (2013) showed that the presence of Al and Zn could be related to the ability of MP particles to attract these elements while titanium-dioxide nanoparticles (TiO_2-NPs) could be leached from polymer as additives. MPs have different resistance to weathering, depending on their properties and polymer additives (Andrady, 2017).

Polymer composition

Raman spectroscopy is widely used to characterize MPs due to their non-destructible and rich spectrum (Araujo et al., 2018). Raman spectroscopy was used for the determination of the MP polymer composition. Fig. 4 shows the spectra of isolated MPs from the sediment and water samples. Overall, five polymers including polyethylene terephthalate (PET), polypropylene (PP), nylon (NYL), high-density polyethylene (HDPE), and polystyrene (PS) were detected in sediment samples. The same polymers except for HDPE were also detected in water samples. PET (25%), PS (22%) and HDPE (18%) were the prevalent polymers in sediment samples, while most MP particles in the water samples were made of PP (31%) and PET (24%). Most fiber particles in the water samples were identified as NYL (33%) followed by PET (27%). Similarly, fragment MPs comprised mainly PP, accounting for 36% of the total fragment MPs. In the sediment samples, fibers were mostly made of PET (34%) and PS (23%) and fragments were made of HDPE (35%). In the water samples, PET had the largest share of MPs (39%) in urban area, followed by PS comprising 31% of the detected polymer (Fig. 5 and Table S3). In industrial sites, PET (44%) was detected as a dominant type of MP. PP was the dominant polymer type in agricultural and natural areas with percentages of 42 and 75%, respectively. In sediment samples, PET was the dominant polymer in the urban (37%) area, while HDPE (39%) was the prevalent polymer in industrial sectors. PS, PP and HDPE were major polymer types in sediment in agricultural sites. Similar to water samples, PP (56%) was the dominant polymer in natural area of the river.

The composition of MPs found in this study included PET, PP, PS, NYL, and HDPE, which also represent the most commonly encountered polymers in the aquatic ecosystem (Andrady, 2011). The proportion of high-density polymers including PET, PS and NYL appeared higher but not significantly ($p > 0.05$) in the sediment samples (66%) compared to the water samples (33%). The results show that PP (31%) is the most common polymer in the water samples, while PET (25%) and PS (22%) are the most abundant polymers in the sediment samples. Due to lower density, PP (density: $0.895 - 0.92$ g/cm^3) is found suspended in the surface water (Erni-Cassola et al., 2019; Issac and Kandasubramanian, 2021). PET with the density of $1.38 - 1.41$ g/cm^3 is classified as dense MPs and hence prone to settle after entering the marine environment (Driedger et al., 2015). Denser MPs such as PET, NYL and PS, with density ranging from 1.04 to 1.41 g/cm^3, were identified in water samples while lightweight MPs including PP was extracted from sediment samples. The presence of light density particles in sediment can be explained by several processes that increase MP density and increase its hydrophobic nature including weathering, biofouling, hetero-aggregation or biomolecule adsorption that enhances deposition of MPs in to the sediments (Chubarenko et al., 2016; Kaiser et al., 2017). Adsorption or incorporation of foreign substances such as clay minerals or quartz grains in surficial pores and cracks of weathered MPs may change sinking behavior of MPs (Dai et al., 2018; Zhou et al., 2018). Conversely, resuspension of bottom sediments result in re-entrance of denser MPs into the water column (Lambert and Wagner, 2018). The identification of chemical composition of the MPs may reveal the origin of the released MPs (Pan et al., 2019a). The results showed that different polymer types probably were released from different land uses. For instance, PET and PS were the main type of polymers in surface water and sediment of the urban sites. PET is commonly used in household cleaning products, clothing industry, cable lining, plastic beverage containers, and packaging (Gong et al., 2018; Guerranti et al., 2019). PS is a thermoplastic polymer with a wide variety of applications such as packaging, household appliances, medical items, construction, and electronics (Lynwood, 2014). The predominant HDPE MPs observed in sediment samples collected near industrial units may originate from industrial effluent mostly from Abadan oil refinery and Abadan petrochemical complex. Wastewater from industries with improper treatment is also a main relevant plastic source (Gallagher et al., 2016). HDPE, a commonly used polymer, is resistant to a broad range of chemicals (Osborne, 2008) and has variable applications in pipe systems, bulk containers for industrial use, cables and wires cover, processing equipment, industrial chemicals such as detergents, bleach and acids (Peacock, 2000; Sam et al., 2014). The predominance of PS, PP and HDPE was observed in water and sediment of agricultural areas. Different types of MPs are released from agricultural soils into water bodies as a result of applying sewage sludge and compost for soil amendment and also plasticulture in agricultural practices (Rochman, 2018; Yang et al., 2021). Ding et al. (2020) also reported PS and PP in agricultural soil of Shaanxi Province, China. Similarly, Corradini et al. (2021) observed that PS and PP were predominant polymers in crop lands (Chile). Agricultural HDPE pipes are generally used to transport chemical fertilizers through irrigation water to the farmlands (Gaj and Madramootoo, 2021). PP was the most abundant MPs in water and sediment of natural area where fishing is a common practice in this area due to proximity to the Persian Gulf. Thus, PP used in fishing ropes and nets are probably the main source of MP pollution. Jang et al. (2020) also found higher proportion of PP from the rural site of Geoje, South Korea and related it to the widespread use of PP rope in fishing.

Interrelations of MP concentrations in the environment

Interrelations of MP concentrations between water and sediment indicated significant differences between the two media ($p < 0.05$) (Table 2). This can result from a range of factors governing MP distribution in water and sediment, suggesting that MP distribution in each medium should be evaluated separately. Furthermore, physicochemical properties of sediments are seemingly important parameters in controlling MP distribution. The mean MP concentrations in each land use indicated significant differences between MP concentrations in the undisturbed natural environment and other areas in both water and sediment (p < or close to 0.05), despite the fact that there is no significant co-relationship between MP concentrations in other land uses ($p > 0.05$). This implies that urban, industrial and agricultural sectors constitute major sources of MPs pollution towards the Arvand River.

PCA biplots, including two principal components (PCs), revealed the interrelations between types of MPs, sediment parameters, sampling sites and land uses in both media (Fig. 6). In water, PC1 and PC2, accounted for 68 and 17 % of total variances, respectively (Fig. 6 a). PC1 was greatly responsible for MP distribution in water and MP fibers and fragments fell in PC1 and had a strong correlation, indicating that they comply with similar distribution in the water sampling sites. Scores of water samples in the PCA indicated that N4 (sampling site near an urban wastewater effluent) had a very strong positive score in both PCs, reflecting that N4 greatly controls the distribution of different types of MPs. However, positive sample scores of N4, N12, N5, N14, N16, N17, N13, N11, and N7 in PC1 are can be affected by the distribution of fibers and fragments in water. The distribution of MP films and spherules is mainly governed by positive scores of N4, N7, and N10, and N4 in PC2, respectively. The PCA revealed that the highest and lowest concentration of MPs emitted into the water from urban and natural land uses, respectively.

In sediment, PC1 and PC2 explained 33 and 26 % of total variances and MPs contributed to both PCs. Fiber, fragment and clay, had greater influence in PC1; and film, spherule and OM carbonate in PC2 were co-correlated and have high positive factor loadings, respectively. Sand was inversely correlated with the MPs and it could indicate that it is related with low levels of them due to the effect of sand filtration. N4 had the highest sample score in PC1 (Fig.6 b) followed by N5, N3, N16, N12, N17, N2, N14, N15, N19, and N13, respectively, while in PC2, the highest sample scores were N11, N9, N17, N7 N12, N10, N6, N13, N5 and N14. Based on sample scores, the degree of contamination (PC1) was mainly affected by the fiber and fragment release from urban sources and are associated with or controlled by clay and OM, whereas film and spherule MPs (PC2) are mainly influenced by industrial and agricultural land uses and are mostly associated with carbonate and sand in sediment. Similar to water, natural land use released the lowest concentration of MPs into the sediment.

Sample score of sediment implied that clay and OM are good explanatory factors in distributing fibers and fragments, while carbonate and sand are factors that well explain the distribution of MP film and spherule in sediment. Recent studies have indicated the interaction of OM and clay fraction with MPs by attaching to MP particles and influencing their fate and transport in aquatic environments (Li et al., 2021). The stability and migration capability of MPs is influenced by humic and fulvic acid attached to MPs (Dong et al., 2019; Hou et al., 2020). Maes et al. (2017) found a positive correlation between concentration of MPs and total organic carbon (TOC) in sediment of the Southern North Sea. Green and Johnson (2020) observed that finer sediments with high TOC, trap more MPs than sediments with low TOC. The transport of MPs can be affected by clay minerals such as kaolinite that adsorb onto MP surfaces (Li et al., 2020). Enders et al. (2019) found a significant correlation between fine sediment fraction (<63 μm) of sediment and MPs in the Warnow estuary in Germany. Elevated MP particles separated from fine-grained sediments are probably the result of higher surface tension of fine-grained particles (Stolte et al., 2015). In this study, a large number of MP particles, fibers and fragments, indicated good correlations with colloidal particles, clay and OM, reflecting that the colloidal particles in aquatic ecosystem of Arvand River may have played a crucial role in transport and fate of MPs.

Conclusion

This study examines the occurrence and characteristics of MPs in urban, industrial, agricultural and natural areas of the Arvand River and it leads to new understanding of pathways and fate of MPs in a large river system. The MPs aboundance in the sediment were superior than in water ($p < 0.05$). Larger MPs were more prevalent in sediment and it was identified as a major sink for MP debris. There was positive correlation between MP concentrations and colloidal particles including clay and OM. This point towards a potential role of aqueous OM hydrophilicity and particle size of natural matter on the transport and ultimate fate of MPs. The decreasing order of the average aboundance of MPs in water and sediment was urban > industrial > agricultural > and natural areas ($p > 0.05$). Natural areas of the Arvand River are still relatively pristine in term of MP pollution. In contrast, no statistically significant differences of MPs distribution were found among urban, industrial and agricultural sources and this indicate that they are all potential sources or pathways of MPs pollution. The Arvand River is highly affected by human activity with regards to plastic pollution and thus, corrective actions should be taken by scientific community, the industry, policy-makers and civil societies to minimize the continuous flux of plastics into the environment. The results of this research will support future MP pollution monitoring in water and sediment in the Arvand River and other grand rivers.

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Table 1. Distribution of MPs in water (items L^{-1}) and sediment (items kg^{-1} dw) of different land uses in the Arvand River. Sum corresponds to total number of MP particles in each land uses and Total comes from the addition of sum values in four land uses of the sediment and water samples. n= number of samples, SD= standard deviation.

Environment		Urban area (n=7)	Industrial area (n=4)	Agricultural area (n=9)	Natural area (n=4)
Water	Mean	71.4	20.2	16.9	1.6
	SD	103.8	7.6	14.9	0.6
	Median	13.7	20.8	9.7	1.5
	Min	3.0	10.3	4.7	1.0
	Max	290.7	28.7	46.3	2.3
	Sum	500.0	80.7	152.3	6.3
Total	739.3				
Sediment	Mean	3073.6	726.3	681.1	111.3
	SD	5621.2	471.9	579.3	41.3
	Median	590.0	527.5	485.0	105.0
	Min	125.0	420.0	220.0	70.0
	Max	15620.0	1430.0	1850.0	165.0
	Sum	21515.0	2905.0	6130.0	445.0
Total	30995.0				

Table 2. p-values obtained from the independent t-test and one-way ANOVA with normalized MP concentrations in water and sediment in different land uses.

Statistical test	Constant Variable	P-value	
		Water	Sediment
Independent t-test	Water & sediment	0	0
Levene Statistic test		0.657	0.657
One-way ANOVA test			
Levene Statistic test		0.003	0.019
Welch test		0	0.001
Brown-Forsythe		0.003	0.032
ANOVA Between Groups	Land-use	0.002	0.036
Dunnett T3 test			
Urban	Agricultural	0.835	0.961
	Industrial	0.994	0.997
	Natural	0.014	0.078
Agricultural	Urban	0.835	0.961
	Industrial	0.791	0.991
	Natural	0	0.002
Industrial	Urban	0.994	0.997
	Agricultural	0.791	0.991
	Natural	0.001	0.011
Natural	Urban	0.014	0.078
	Agricultural	0	0.002
	Industrial	0.001	0.011

Figure 1 Click here to access/download;Figure;Fig. 1.docx

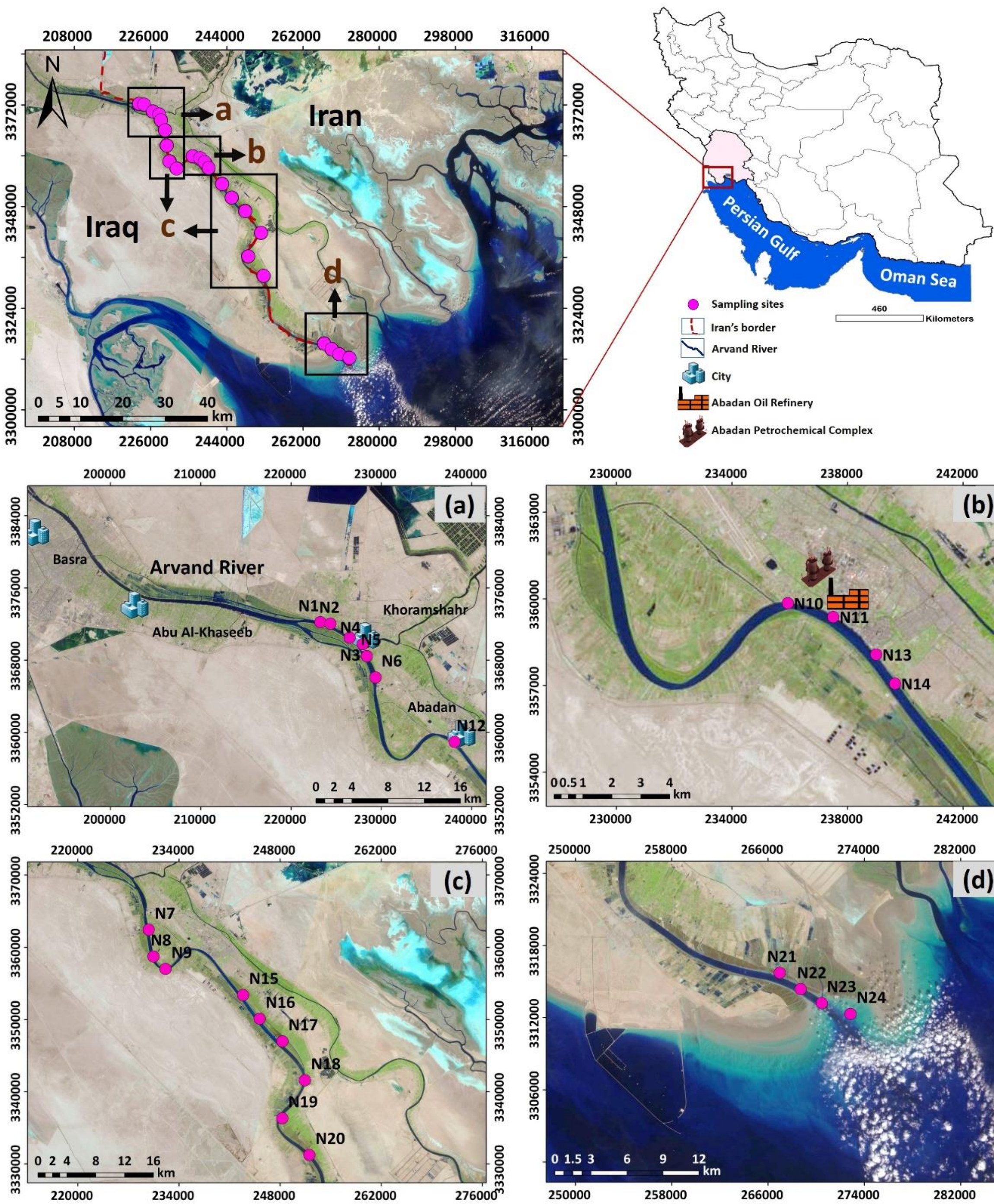

Fig. 1. Sampling locations of surface water and sediment in different land uses: (a) urban; (b) industrial; (c) agricultural; and (d) natural areas of the Arvand River, Persian Gulf. Background image (Landsat 8 OLI/TIRS C1 Level-2) is obtained from earthexplorer.usgs.gov.

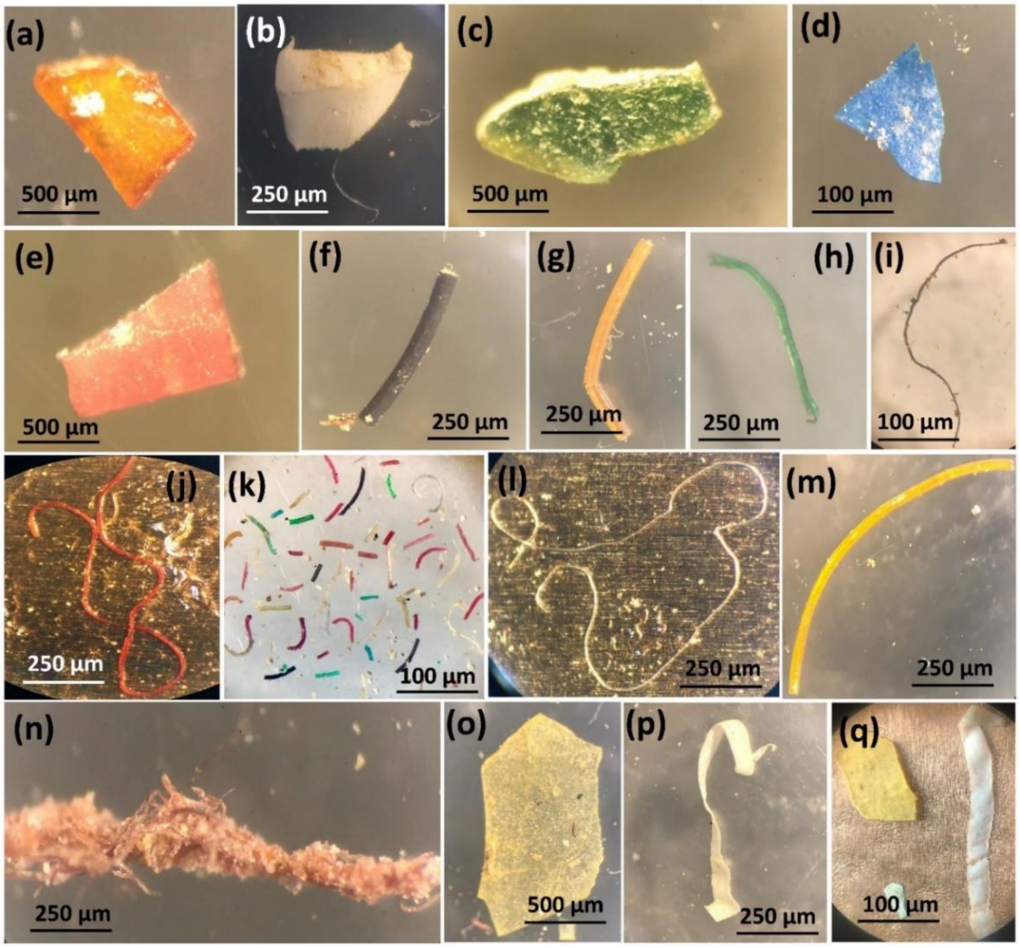

Fig. 2. MPs extracted from sediment and water from the Arvand River. The images were taken with a binocular optical microscope (Carl-Zeiss, Oberkochen/West Germany). a–e: Fragments, f–m: fibers, n: tangled fibers o–q: films.

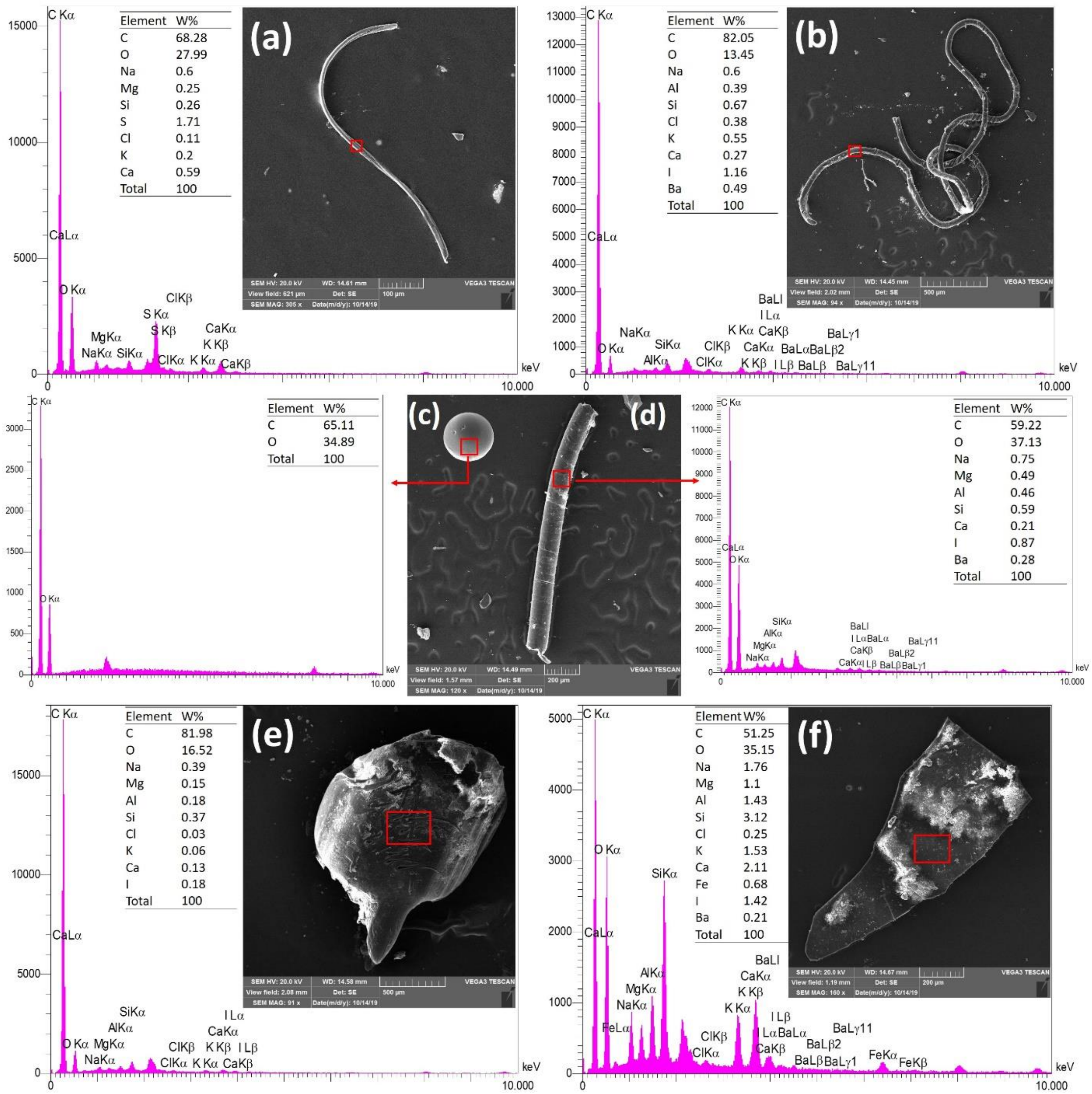

Fig. 3. SEM images and EDX of representative MPs found in the Arvand river including: (a) fiber particle separated from water sample; (b) long fiber MPs extracted from sediment sample in urban area; (c) spherule found in sediment sample of urban area; (d) line/fiber MPs in sediment sample of urban area; (e) fragment MPs extracted from sediment sample of agricultural sector; and (f) film particle in water sample of urban area.

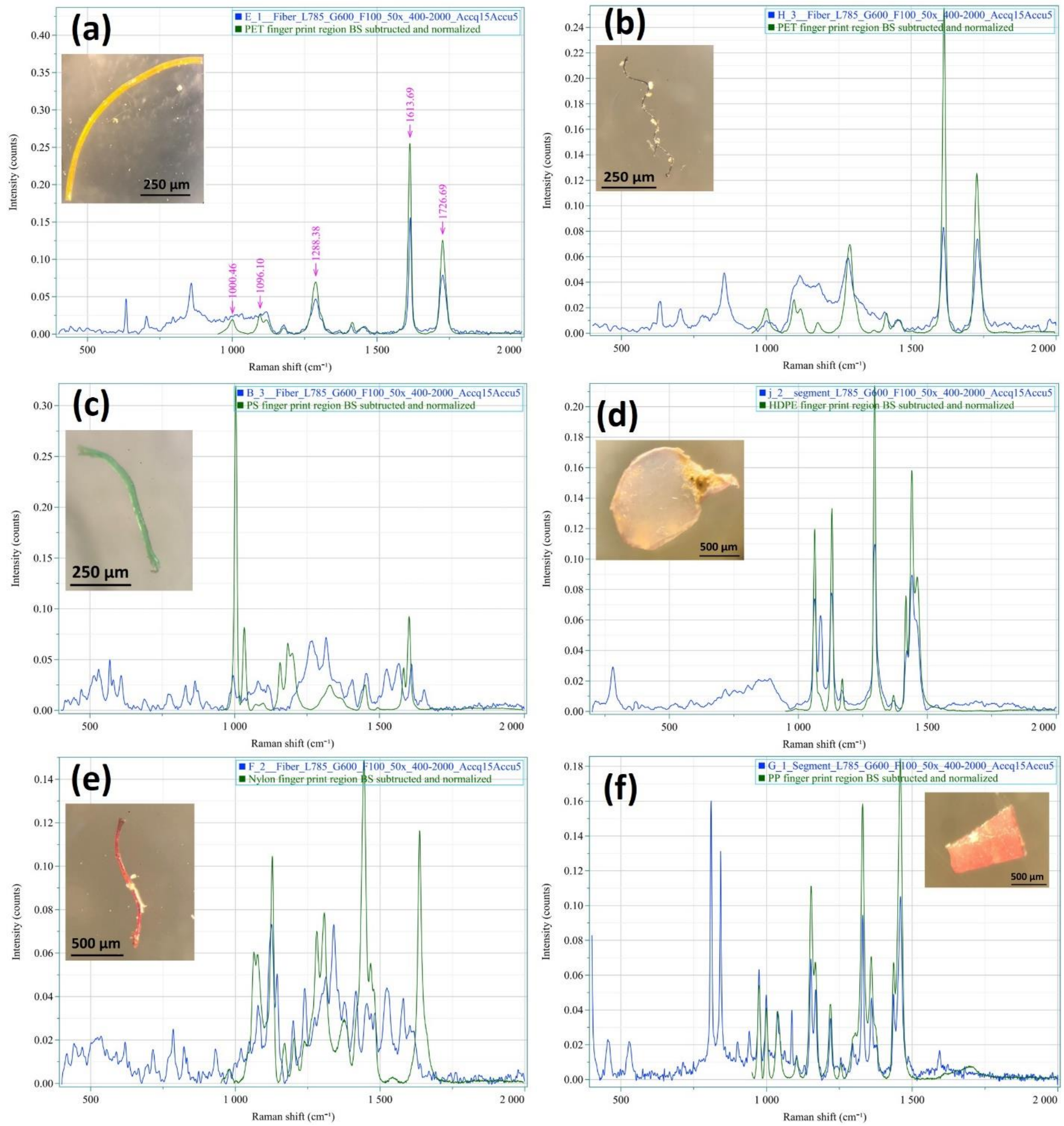

Fig. 4. Raman spectra of some MPs found in surface water and sediment: (a) yellow fiber, polyethylene terephthalates (PET); (b) black fiber, PET, (c) green fiber, polystyrene (PS); (d) transparent fragment, high density polyethylene (HDPE); (e) red fiber, nylon; and (f) red fragment, polypropylene (PP).

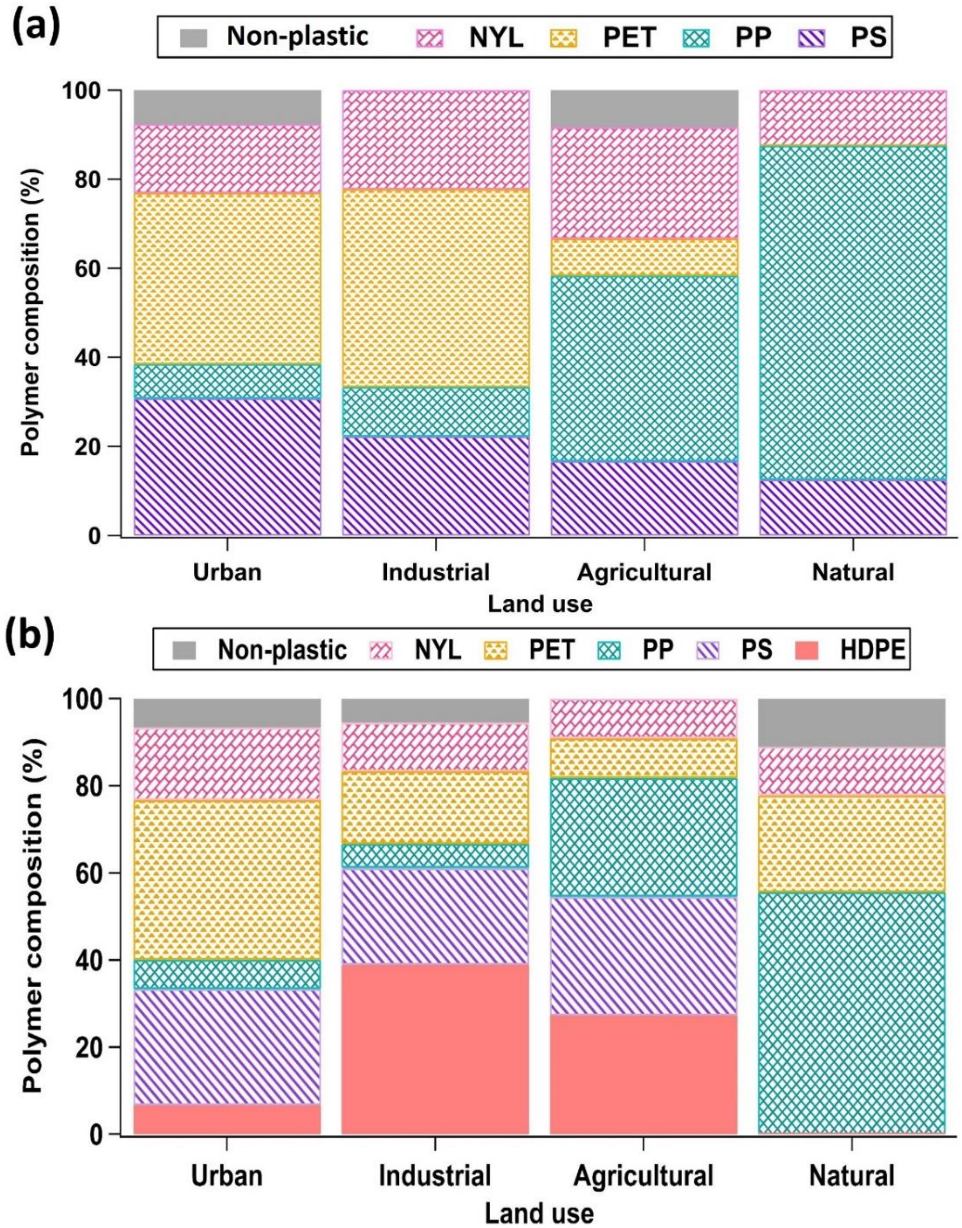

Fig 5. Polymer compositions in (a) surface water and (b) surface sediment from the different land uses across the Arvand River.

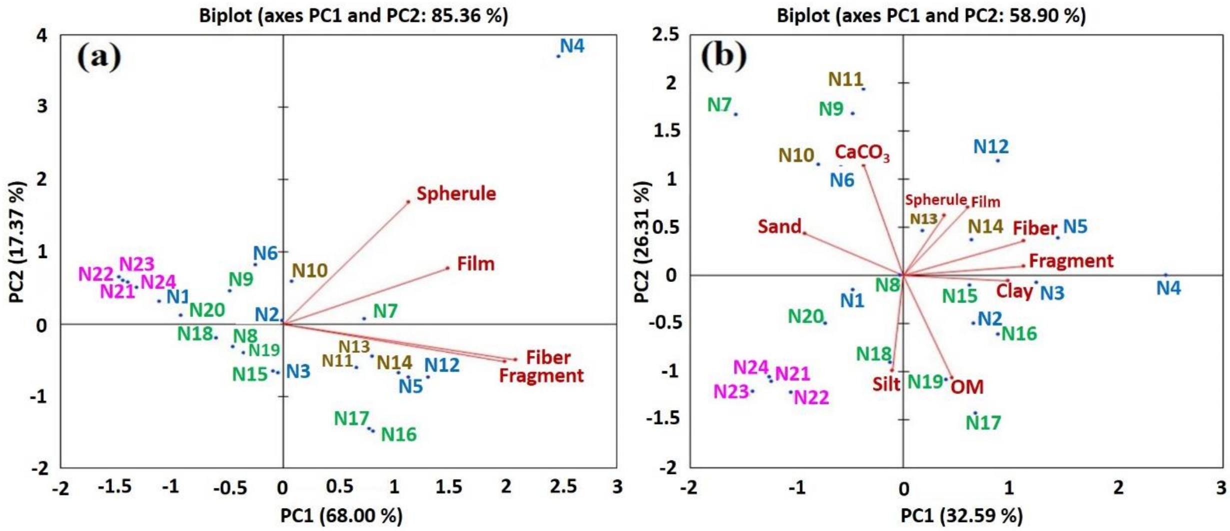

Fig 6. PCA biplot showing interrelations of different types of MPs in (a) water and (b) sediment sampling sites. Samples collected from the urban, agricultural, industrial and natural land uses are distinguished by blue, green, brown, and pink colors, respectively.

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

CRediT authorship contribution statement

Naghmeh Soltani: Conceptualization, Investigation, Writing - original draft. **Behnam Keshavarzi:** Supervision, Validation, Resources. **Farid Moore:** Supervision, Review & editing, Resources. **Rosa Busquets:** Review & editing. **Mohammad Javad Nematollahi:** Writing, Formal analysis. **Sylvie Gobert:** Review & editing. **Reza Javid:** Investigation, Logistical supporting. All authors discussed the results and contributed to the final manuscript.