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Distribution Patterns of Floating Microplastics in Open and Coastal Waters of the Eastern Mediterranean Sea (Ionian, Aegean, and Levantine Seas)

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Microplastic pollution is a pervasive anthropogenic phenomenon at the ocean surface. Numerous studies have been performed worldwide; nevertheless, the distribution patterns, morphological properties, and sources of origin in the Eastern Mediterranean Sea are still poorly explored. The purpose of this study is to investigate the distribution patterns of surface floating microplastics (MPs) in the Ionian, Aegean, and Levantine Seas in relation to their sources and sea surface circulation. In total, eighty-four samples were collected using manta nets from 2014 to 2020, covering open waters, coastal waters, and enclosed gulfs (Corfu and Saronikos). MPs concentration measurements revealed high variability ranging from 0.012 to 1.62 items m^{-2} and did not present maximum concentrations close to MPs hotspot areas. The presence of sea surface slicks, as recorded visually during our samplings, seems to play a key role on the distribution pattern of MPs, and highest concentrations were recorded in samples affected by these formations. The dominant MPs shape type identified were fragments (50–60%), whilst filaments (1–23%), films (3–26%), and foams (0–34%) varied among the studied areas. The majority of MPs in open waters had sizes $\leq$2 mm peaking between 0.6 and 1.4 mm. Spectroscopic analysis of MPs revealed the presence of 11 polymer types in both open sea and gulfs; the most abundant type was polyethylene (PE), followed by polypropylene (PP), and polystyrene (PS). The relative abundance of polymer types was more diverse in Saronikos Gulf, compared to the open sea due to the proximity to major urban and industrial sources. Our findings suggest that the vicinity to coastal population centers determined the properties, size and polymer types of MPs and highlight that MPs concentrations are affected significantly by local oceanographic conditions, such as surface slicks.

Keywords: plastics, marine litter, seasurface, windrows, surface slicks, sources, OpenSpecy, FT-IR

INTRODUCTION

During the last decades, dispersion of plastics in the oceans has become a global pollution problem that poses a great threat to the marine ecosystem. The extensive production of short use-cycle plastic products leads to considerable waste generation, and subsequent leakages to the environment. It is estimated that over 269,000 tons of plastic debris float on the surface of the oceans (Eriksen et al., 2014). Despite being durable and long-lasting materials, plastics deposited in the environment are subject to weathering processes, such as UV-photooxidation and hydrolysis, causing degradation, and eventually fragmentation to smaller particles, called microplastics (MPs) (Thompson et al., 2004; Ioakeimidis et al., 2016).

Microplastics are defined as plastic pieces smaller than 5 mm, classified as primary and secondary. Primary MPs are small plastic particles consisting of raw industrial plastic pellets and processed particles added intentionally in health-care and cleaning products (cosmetics, detergents, etc.). Secondary MPs are particles derived either from larger plastics' fragmentation or from materials' wearing off during use (GESAMP, 2016; Veiga et al., 2016). MPs formation takes place at beaches and land, and reach the sea through rivers, road run off, city storm water, and wastewater treatment plants (Moore et al., 2011; Cheung et al., 2016; Kalogerakis et al., 2017). MPs could be found both in populated and remote places, and they are ubiquitous in the marine environment, from the ocean surface to the deep sea sediments (Van Cauwenberghe et al., 2013; Bergmann et al., 2015; Lusher et al., 2015; Courtene-Jones et al., 2017; Morgana et al., 2018). At sea, MPs are redistributed under the influence of physical factors and despite the well-known accumulation in the oceanic gyres, concentrations of MPs along the surface ocean exhibit increased variation in space and time. In addition, environmental fate, vertical transport, and biological effects, may be affected by microbial colonization on MPs and biofilm formation (Kaiser et al., 2017; Kooi et al., 2017). Biofilm may lead to an increase in the density of MPs and a decrease in their buoyancy with a strong impact on both sedimentation potential and upward transport (Rummel et al., 2017; Nguyen et al., 2020). Furthermore, despite that the understanding of MPs trophic transfer in marine ecosystems is increasing, several aspects remain unknown, primarily including the role of the microbial biofilm living on MPs surface (the so-called "plastisphere;" Zettler et al., 2013) on trophic transfer and its effects on marine organisms' health. MPs are known to absorb harmful contaminants (Karapanagioti et al., 2011; Koelmans et al., 2016; Torres et al., 2021) whilst records on the ingestion of MPs by marine biota are increasing exponentially (Galloway, 2015; Wright et al., 2017; Digka et al., 2018). More recently, the role of MPs has also been investigated in relation to the carbon cycle (Galgani et al., 2018, 2020; Romera-Castillo et al., 2018; Taipale et al., 2019). Our understanding of the factors affecting the MPs variation at sea and their harmful effects in organisms, is still limited. Consequently, there is insufficient capacity for designing and implementing successful mitigation and regulatory policies (Thompson, 2015; Galgani et al., 2021).

In the Mediterranean Sea, the estimated average plastic concentration from both model and field data is comparable to the oceanic gyres (Lebreton et al., 2012; Eriksen et al., 2014; Cózar et al., 2015). This observation potentially results from several factors including the high population concentration (∼150 million), the increased tourism activity in the Mediterranean area (1/3 of the world's tourism), the expanded shipping activity (15% of the global shipping; UNEP/MAP, 2017), in combination with the enclosed character of the basin. The distribution of floating MPs in the Mediterranean Sea, has been mainly investigated in the northwestern and central part of the basin (Collignon et al., 2012, 2014; Fossi et al., 2012, 2016; De Lucia et al., 2014; Cózar et al., 2015; Pedrotti et al., 2016; Suaria et al., 2016; Zeri et al., 2018). Even though there are some studies in the open and coastal waters in the eastern Mediterranean Sea (Cózar et al., 2015; Gündoğdu and Çevik, 2017; Güven et al., 2017; van der Hal al., 2017), there is still lack of information for the floating MPs distribution. In the present work, we aim to fill in the gap on the magnitude of MP pollution in the Eastern Mediterranean Sea, by providing data on concentrations and properties of floating MPs at the sea surface of the Ionian, Aegean, and Levantine Seas in addition to two enclosed gulfs, Corfu and Saronikos. We further discuss the factors affecting the observed distribution patterns and properties considering the vicinity to MPs sources and oceanographic variables.

MATERIALS AND METHODS

Study Area

The investigated study area covered parts of the Eastern Mediterranean Sea: the Aegean Sea, the Eastern part of the Ionian Sea and parts in the Levantine Sea within the Greek territory. In addition, two gulfs were studied: Corfu Gulf in the Ionian Sea and Saronikos Gulf in the Aegean Sea (**Figure 1**). Surface waters originating from the Levantine and Cilician basins enter the eastern Aegean Sea from the eastern Cretan Straits and Rhodes passage and travel northward along the eastern coasts, until they meet lighter waters of Black Sea origin in the North Aegean. These relatively light surface waters are progressively mixed with saline ones following a cyclonic circulation along the eastern coasts of the Greek peninsula. Sea surface circulation in the Ionian Sea shows considerable seasonal and interannual variability with reversals of surface circulation from cyclonic to anticyclonic and vice versa. Within this pattern exist several cyclonic and anticyclonic mesoscale and/or sub-mesoscale features (Poulain et al., 2012).

Corfu Gulf is situated in the North Ionian Sea and is a secluded elongated bay formed between the coast of Corfu and the Greek mainland. The gulf is burdened by dense and frequent movement of vessels and ships, including recreational and fishing boats; the town of Corfu (28,185 inhabitants) which is an international touristic destination and the port of Igoumenitsa (25,814 inhabitants) on the mainland. Kalamas River (length 115 km; water flux 74 m^{-3} s^{-1}) drains mountainous and agricultural land and outflows in the gulf by the port of Igoumenitsa. The Saronikos Gulf is situated in the central Aegean

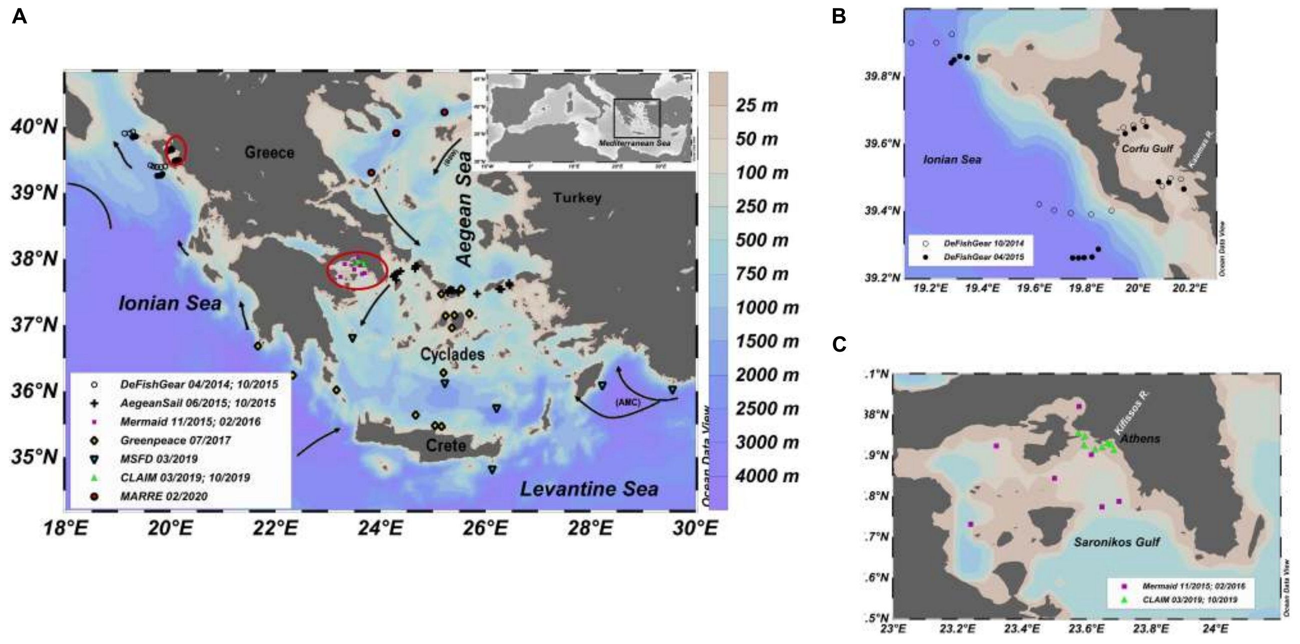

FIGURE 1 | Map of sampling positions in the Ionian, Aegean, and Levantine Seas for each of the seven campaigns from April 2014 to February 2020. In red circles the two enclosed gulfs sampled **(A)**. Sampling positions in Corfu Gulf **(B)** and Saronikos Gulf **(C)**. Black arrows denote surface currents.

Sea and is the marine gateway of the Athens greater metropolitan area (∼3.8 million inhabitants), including the port of Piraeus and increased navigation activities. Several point and non-point pollution sources are concentrated in the inner part of the gulf. A small urban river, Kifissos River (length 25 km), ends up close to the port of Piraeus; the river does not have a stable flow and is subject to flooding events, depending mainly on the annual precipitation. Approximately 70% of its catchment is currently a built-up urban area characterized by mixed land uses, such as operating and abandoned factories, small and medium enterprises, warehouses, illegal areas for fly tipping of solid waste etc. Another important pollution source is the outflow of the treated sewage of Athens/Piraeus (∼800,000 km^{-3} per day). Several other point sources are spread along the coasts and include marinas, touristic facilities, fish farms, and the treated effluents of smaller towns and settlements.

Sampling of Microplastics

A total of eighty-four samples of sea surface water were collected for MPs investigation during 11 sampling campaigns from 2014 to 2020 (**Figure 1**). In most cases sampling sites were visited once. Only for two cases, two samplings were conducted in autumn and spring at same locations. In particular, the North Ionian Sea and Corfu Gulf were visited in October 2014 and in April 2015 and the inner Saronikos Gulf in March 2019 and October 2019. Five of the samplings were conducted with research vessels (R/V Filia and R/V Aegeao); two with sailboats; one with a fishing boat and one of the samplings in the inner Saronikos Gulf with an inflatable boat. The start and end sampling positions were obtained either from the vessel's or from a portable Global Positioning System (GPS). Details on samples position and wind conditions are given in **Supplementary Table 1**. Sea surface MPs were collected using manta nets. Manta net dimensions were: W60 × H24 cm rectangular frame opening; net 3 m length and mesh size of 330 μm with the exception of the Greenpeace campaign which were: W84 × H15 cm rectangular frame opening; net 4 m length mesh size 335 μm. For samplings conducted after 2017 the manta net was equipped with a flow meter (HydroBios). In all cases the manta net was towed from the side of the vessel and beyond the vessel's wake. The duration of the manta net tows varied from 15 to 30 min, depending on the size of the net used, assuring collection of a representative sample. The vessel speed was always kept <2 knots. At the end of each tow, the net was washed in order to gather all particles in the cod end. In order to avoid sample contamination on board, immediately after sampling, the cod net was transferred into a glass jar and kept frozen until analysis in the laboratory.

Wind Correction

Sampling of MPs with manta net trawls is subject to limitations related to the sea state during the time of sampling. Wave-induced turbulent mixing causes the downward flux of plastic particles deeper than the height of the manta net frame. For this reason, it is recommended that net tows are carried out under light wind conditions <10 knots (<3 B) (GESAMP., 2019). Wind speed was recorded by a portable anemometer or by ship's instruments and was <10 knots during most of our samplings, except nine tows conducted with sailing boats under wind force up to 14 knots (**Supplementary Table 1**). For these cases, we have applied a correction factor on the MPs field data following the model described by Kukulka et al. (2012) and Reisser et al. (2015). Wind stress (τ) and water friction velocities (u_w^*) were calculated from measured wind velocity onboard, while the significant wave height (H_s) was based on typical wave heights experienced at corresponding Beaufort numbers (for 3 B: H_s 0.6–1 m; for 4 B: H_s 1–2 m) and further refined using photographs of the sea state during our samplings. The use of photographs was considered helpful as sometimes wave propagation may lag wind or vice versa.

Microplastics Separation and Counting

Analysis was conducted in line with the GESAMP. (2019) guidelines. After thawing, the samples were sieved through a stack of metallic sieves (1 mm and 300 μm), to facilitate the separation of organic material, especially in organic rich samples. In case of presence of large natural organic items (seaweed, branches, and leaves) these were thoroughly washed with deionized water above the stack of sieves to collect all MPs adhered onto them. Then the two fractions were dried at 40°C. MPs were visually identified under a stereomicroscope. For samples holding a significant amount of natural organic matter (gels), a step of H_2O_2 digestion at 60°C was performed, until the digestion was complete, and no natural organic material was visible. The digests were filtered through Glass Microfibre Filters GF/C filters and then examined under a stereomicroscope for MPs presence. The counting of MPs, identification of shape type (fragment, filament, film, foam, and pellet) and size was performed using the OLYMPUS SZX10, SZX stereomicroscopes, equipped with a digital camera (Luminera; Nikon, and DSL3) and the INFINITY ANALYZE software. Image analysis was used in order to measure the longest dimension (mm) of MPs. To avoid lab contamination the filtering equipment was placed in a plastic hood (Sigma-Aldrich Pyramid) while for the 2019–2020 samples all processing was conducted under a laminar flow bench (HN14). Furthermore, fiber free (tyvek) lab coats were used and airborne contamination was estimated by using blank filters at all stages of the analyses. Particles present in samples with features similar to those collected on the blanks were excluded from the MPs analysis. MPs concentration was expressed per m^2 and per m^3 when a flowmeter was used.

FT-IR Analysis

For the characterization of the MPs polymer type, ATR-FTIR spectroscopy was used [Agilent Cary 630; Perkin Elmer Spectrum Two with (UATR) accessory with a 9-bounce diamond top-plate]. Spectral range was 4,000–650 cm^{-1} with a resolution of 4 cm^{-1} and 32 scans s^{-1}. The threshold for spectra similarity was set to 80%. Polymer identification was based on a combination of instruments' built-in and in-house libraries. ATR-FTIR analysis was done in 12% of the total particles counted. Cross-reference of spectra retrieved from the ATR-FTIR instrument was performed with the open source database Open Specy (www.openspecy.org; Cowger et al., 2021). The Open Specy tool includes 636 spectra of 276 materials from three libraries of pure polymers, and materials relevant to microparticles and fibers found in the environment (Primpke et al., 2018; Chabuka and Kalivas, 2020; Suja Sukumaran, Thermo Fisher Scientific).

Statistical Analysis

Differences in MPs concentrations were examined by the Mann–Whitney *U* test and the non-parametric Kruskal–Wallis test *post hoc* pairwise, since data did not meet the assumptions of normality as shown by the Shapiro–Wilk test and homogeneity of variance as shown by the Levene's test. Level of significance was set to <0.05. All statistical analyses were performed using IBM SPSS statistics 25.

RESULTS

A total of 23,800 microparticles were counted. Mesoplastics (>5 mm) caught in the manta net corresponded to ~5% of total particles and were not included in the present analysis. In the subset of 2,971 items, subjected to FTIR analysis 2.7% were identified as natural materials, and 0.7% were suspect to laboratory contamination (e.g., filter material). These percentages are considered low, and so all microparticles counted were considered as MPs.

Distribution of MPs

Results on MPs concentrations per sampling area are presented in **Figure 2A** and **Supplementary Table 1**. Concentrations showed increased variability ranging from 0.012 to 1.62 items m^{-2} (average ± SD: 0.26 ± 0.36; 1.18 ± 1.27 items m^{-3}; median: 0.12; 0.72 items m^{-3}), while most of the data (80%) fall below 0.3 items m^{-2}. The statistical analysis showed a significant variation between the sampling areas (overall $P = 0.007$). Data from the Levantine Sea were very limited, so this area was not included in this analysis. Furthermore, the *post hoc* pairwise analysis revealed that the difference occurs only for the Corfu Gulf and the Saronikos Gulf ($P = 0.010$) data sets. It is understood that MPs distribution follows overall similar patterns, with some differentiations in the two enclosed gulfs (**Figure 2A**). In order to map the spatial distribution of MPs in the studied areas, we grouped the MPs concentrations in five classes as depicted in **Figure 2B**. Areas visited more than once (N. Ionian Sea, Corfu Gulf, Saronikos Gulf), show increased spatiotemporal variability of MPs, while the highest concentrations were recorded in the N. Ionian Sea and the Corfu Gulf. Few elevated concentrations were recorded in between the islands of the Aegean Sea and in the Levantine Sea. The lowest concentrations of MPs in our dataset appeared systematically along the west–east transect in the central Aegean Sea. To investigate any potential relationship between MPs concentrations and surface currents velocities, we retrieved surface current velocity data from Copernicus - Marine environment monitoring service, during the same dates of our samplings and at the closest position (Lat, Lon). The comparison of MPs concentrations in our samples with corresponding current velocities did not show any systematic pattern and no relationship could be established (**Supplementary Figure 1**). This can be attributed to several reasons such as (i) the fact that velocity data correspond to average daily values integrated over the grid and not to exact conditions at the time and place of the samplings, and (ii) other factors, besides surface currents, affecting the transport of plastics on

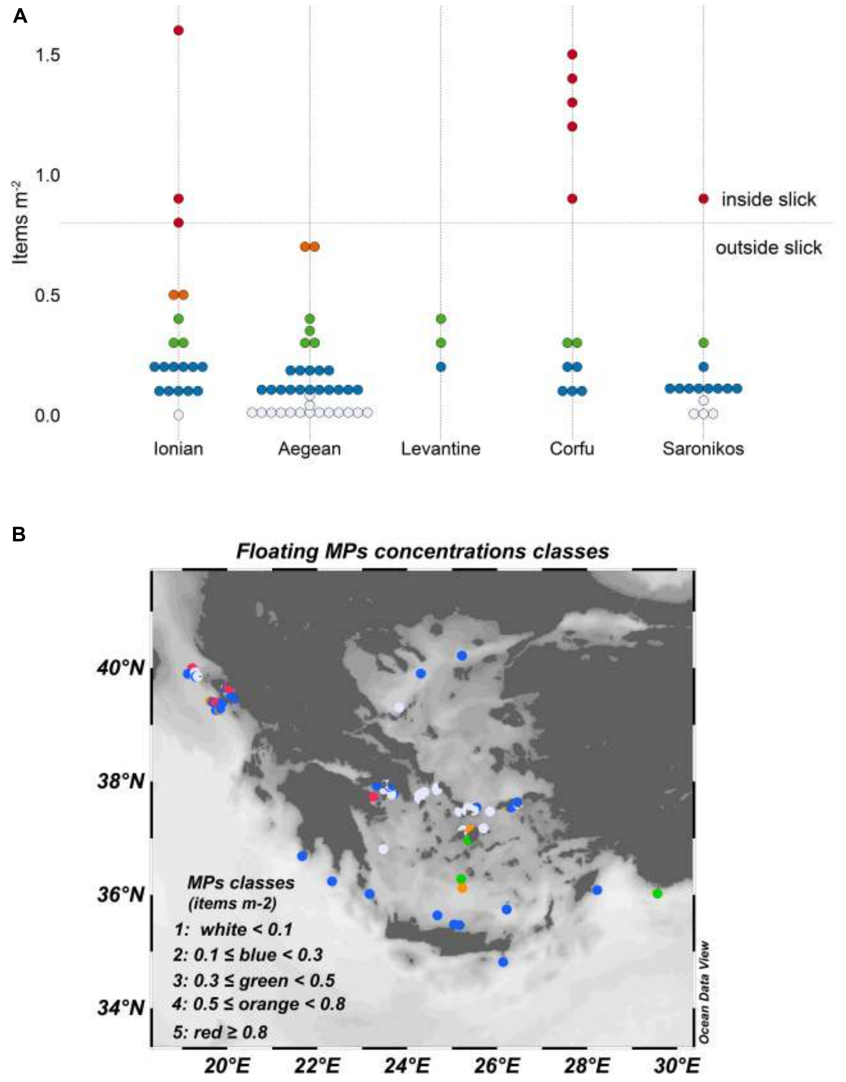

FIGURE 2 | Dot-plot diagram of microplastics (MPs) concentrations (items m^{-2}) in the five sampled areas. Concentrations of samples collected inside and outside the slicks are separated by the horizontal line at 0.8 items m^{-2} **(A)**. Map showing floating MPs concentration classes for all sampling positions **(B)**. Colors in figures correspond to 5 MPs concentration classes (items m^{-2}) as follows: 1: white < 0.1; 2: 0.1 ≤ blue < 0.3; 3: 0.3 ≤ green < 0.5; 4: 0.5 ≤ orange < 0.8; and 5: red ≥ 0.8 items m^{-2}.

the sea surface, i.e., wave action, beaching, and the shape of the particles.

At three of the investigated areas, two repetitive samplings were conducted: in the N. Ionian Sea and Corfu Gulf in October 2014 and April 2015 and in the inner Saronikos Gulf in March 2019 and October 2019. In all cases, MPs concentrations where found higher during the first sampling occasions (October 2014, March 2019), which coincided with rain events either during the same day or 10 days before samplings took place[1] (**Table 1**). These differences are statistically significant for the N. Ionian Sea and Corfu Gulf data (Mann–Whitney *U* test, $P = 0.009$), while the test was not possible for the inner Saronikos Gulf data due to the limited number of samples. In the two gulfs (Saronikos Gulf and Corfu Gulf) where the sampling transects were conducted in very short distances from land and from the mouth of the rivers (<2 km), the differences observed could be an indication of direct MPs inputs from land during rain events. For the sampling transects, however, in the N. Ionian Sea with distances from land farther offshore (>2.5 km), land runoff of MPs might not be the only factor which affects the seasonal variation observed. In the N. Ionian Sea as well as in the Corfu Gulf, during October 2014, formations of long stripes with mirror-like appearance of the sea surface water (slicks) were visible and occasionally accumulation of flotsam was apparent (windrows; **Figures 3A,B**). In many cases, the manta net trawls coincided and/or crossed the slicks and this information was recorded. Data obtained inside and outside the slicks are indicated (**Figure 2A**). Elevated concentrations of MPs were consistently found inside the slicks

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[1] http://meteosearch.meteo.gr/

TABLE 1 | Microplastics (MPs) concentrations (items m^{-2}) in the areas with repetitive samplings and rain (mm) during 10 days before samplings.

	Rain* (mm)	Area	MPs (items m^{-2})
October 14 *n* = 13	179.2	N. Ionian Sea	0.56 ± 0.52
		Corfu Gulf	0.98 ± 0.58
April 15 *n* = 13	0.4	N. Ionian Sea	0.22 ± 0.16
		Corfu Gulf	0.28 ± 0.32
March 19 *n* = 5	29.6	Inner Saronikos Gulf	0.11 ± 0.06
October 19 *n* = 3	6.4		0.06 ± 0.05

**10 days before sampling.*

(>0.8 items m^{-2}), almost one order of magnitude higher than outside the slicks ($P = 0.005$); all samples were collected under calm conditions. The appearance of slicks on the sea surface is a clear indication of the action of specific sub-mesoscale physical structures such as Langmuir circulation and river fronts related to wind-wave forcing or alongshore currents (van Sebille et al., 2020). Although, we do not know the exact mechanism causing the slicks formation and whether it is related to the storm event of the preceding days and/or the river outflow for the case of Corfu Gulf, our data show that slicks have a strong influence on MPs concentrations. Additionally, MPs concentrations > 0.8 items m^{-2} were recorded in two sampling locations, one in Corfu Gulf close to the Kalamas river mouth in April 2015 and associated with windrow formation in (**Figure 3C**) and a second sample collected in the Saronikos Gulf in November 2015 (**Figure 2A**). The latter was collected at the western part of the gulf away from pollution point sources and coincided with the presence of an oil slick. Although our samplings were not designed in relation to the slicks, these results highlight that information on local oceanographic conditions during samplings is needed for the interpretation of MPs distribution on the sea surface.

Microplastics Properties

All counted MPs were classified in five different shape types (fragments, filaments, films, foams, and pellets) as shown in **Figure 4**. Fragments had the highest percentage contribution exceeding 60% in all regions except Saronikos Gulf where they contributed by 50%. Filaments contribution was highest in the Aegean Sea (23%) while in the other areas fluctuated between 1 and 7%. Films had the highest share (26%) in the Levantine Sea followed by 7% in the Saronikos Gulf and 3.4% in the Aegean Sea. About one third (34%) of the MPs found in the Saronikos Gulf were foams, while in other areas foams were almost not recorded. Pellets were found only in the Saronikos Gulf by 1.3% (**Figure 4**). Data shows that fragments were the most common shape type caught by manta nets while filaments and films varied largely. In the enclosed Saronikos Gulf the variety in the shape types of MPs was highest.

Measurements of the longest dimension of MPs by image analysis were conducted in a subset of particles (4,125 corresponding to the 17.3% of the total). Average length of MPs was 1.71 ± 1.07 mm. The frequency diagrams of **Figures 5A,B** show the size distribution of the MPs collected in the three areas (Ionian, Aegean, and Levantine; denoted as "open waters") separately from those collected in the inner Saronikos Gulf, close to input sources. About 70% of MPs from open waters have lengths ≤ 2 mm, peaking between 0.8 and 1.4 mm. In the Saronikos Gulf the contribution of ≤ 2 mm MPs, is only 56%, and the histogram shape suggests that MPs are more or less evenly distributed within the measured size range. It should be noted that these data are operationally defined by the manta net mesh size 330 μm, and in particular for small sized particles <1 mm which may pass the net opening based on their shortest dimension.

Spectroscopic analysis with ATR-FTIR revealed that the majority of particles were made of synthetic polymers. Nevertheless, it was not possible to identify (ND) about 288 particles, either because their material was not included in our databases or because of their small size, as particles within the size range 300–500 μm fall within the limits of detection of ATR-FTIR instruments. In all these cases however, it was possible to obtain a spectrum. These ND spectra were further analyzed against the Open Specy database, in order to check whether comparison with vast types of materials could possibly refine our results. Out of the 288 ND spectra re-analyzed, 68 had spectral similarity (>76%) to natural materials, and 220 to synthetic polymers (142 with spectral similarity >80% and 78 in between 76 and 80%). The natural materials identified had the following characterizations in the Open Specy databases: leaf plant, algae fucus serratus, cellulose, chitin, animal fur, fiber linen, and broodcomb. Apart from elucidating the nature of the ND particles, the OpenSpecy databases were used also for cross-referencing of our polymer spectra. We chose to check all spectra characterized as general polymer types (e.g., thermoplastic polymers-TPE) and materials (e.g., adhesive tape, bag), as well as a subset of well defined polymers [e.g., High Density PolyEthylene (HDPE), PolyVinyl chloride (PVC)]; in total 61 spectra. Results from this exercise are given in **Table 2** (**Supplementary Table 2** in detail). Overall, there was a 100% agreement between our instruments' and OpenSpecy libraries for well defined polymers HDPE, LDPE, Ethylene-Vinyl-Acetate (EVA), PP, PS while a more specific polymer characterization was achieved for the spectra of general polymer types and materials. Polyvinyl-alcohol (PVA) was the only polymer that presented dissimilarities between the two ways of characterization. In almost all cases spectra characterized as PVA by the instrument's library matched with natural materials when compared to the OpenSpecy libraries (**Supplementary Table 2**). Final polymer characterization of the MPs was conducted by refining our results based on the Open Specy re-analysis (excluding re-analysis results on PVA, which were considered as such). The percentage contribution of the different polymer types is presented for the open waters and the enclosed gulfs separately (**Figure 6**). In total, eleven different polymer types were identified, seven of them were common for all the areas (open waters, Corfu Gulf, and Saronikos Gulf) namely; Polyethylene (PE; including high and low density), Polypropylene (PP), Nylon-Polyamide (Nylon-PA), Polystyrene (PS, including expanded polystyrene EPS), Polyurethane (PU), Polyvinyl-chloride (PVC), and PVA. For the latter, the high

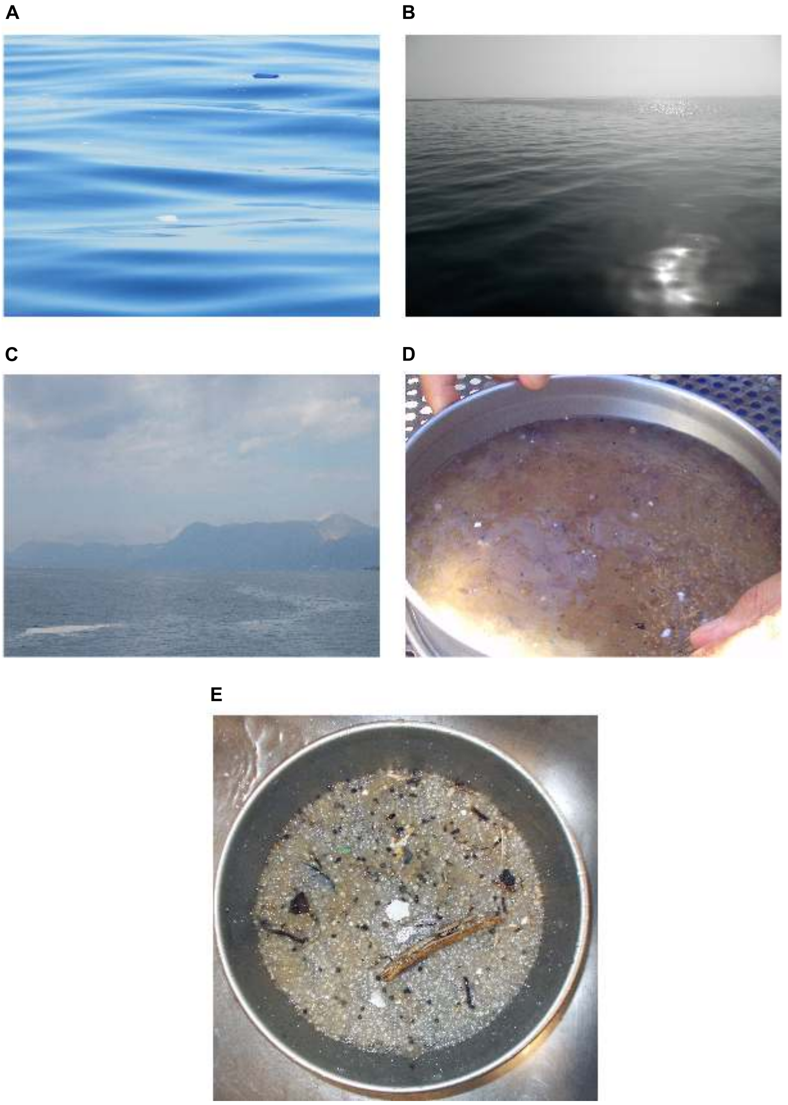

FIGURE 3 | Slick appearance of the sea surface on 9/10/2014 in the N. Ionian Sea (sample 10-1M) with apparent floating macroplastics **(A)**; on 14/10 2014 in Corfu Gulf (sample 10-13M) **(B)**; on 25/4 2015 in Corfu Gulf with apparent formation of windrow (sample 4-15M) **(C)**; gelatinous zooplankton in sample 10-1M **(D)**; fish eggs in sample 10-7M **(E)**.

degree of uncertainty, based on spectra re-analysis, should be noted. The other non-common polymers found are Antifouling paints (AF-paints), Ethylene Propylenediene monomer (EPDM), EVA, Polyester-tetraphthalate (PET/Polyester), wax materials. The relative contribution of the various polymer types in open waters and Corfu Gulf appear similar to each other in contrast to the Saronikos Gulf. There is a clear dominance of PE and PP particles in open waters (79%) and Corfu Gulf (91%). In Saronikos Gulf, PE and PP hold a lower share (47%) and an elevated contribution of PS (18%) and AF-paints (20%) is observed (**Figure 6**). Saronikos Gulf is the only area were AF-paints as well as EPDM (2%), a polymer with industrial and building insulation uses, were found.

DISCUSSION

The Mediterranean Sea is characterized as a hot spot area for plastic pollution and floating plastic concentrations are found comparable to those of the oceanic gyres (Eriksen et al., 2014; Cózar et al., 2015). Despite this statement, increased variability in MPs has been recorded with concentrations differing 2–3 orders of magnitude from place to place and at different time instances (Suaria et al., 2016; Fossi et al., 2017; van der Hal et al., 2017; Baini et al., 2018). This can be attributed to the permanent, quasi-permanent mesoscale circulation features, seasonal structures, and significant temporal changes in the surface currents that characterize this basin (Poulain et al., 2012).

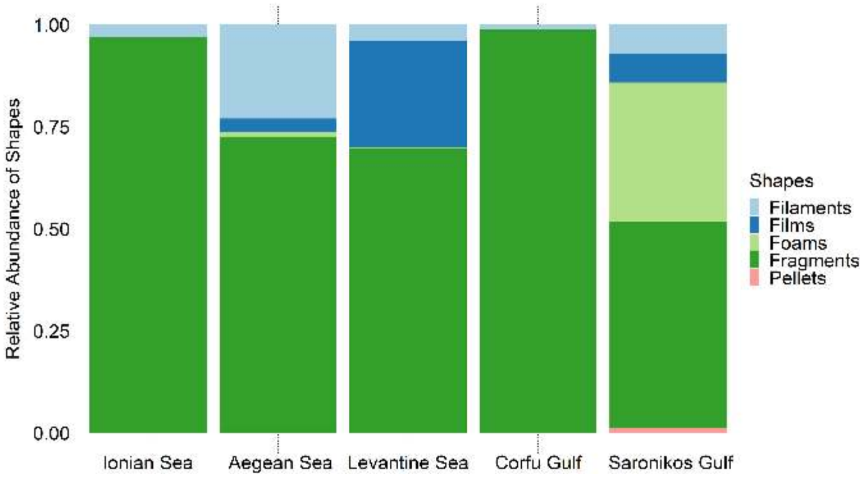

FIGURE 4 | Relative abundance of MPs shape types at the studied areas.

Concentrations of MPs reported in the present study (0.012–1.62 items m^{-2}) capture this variability and are in line with other case studies reported at the Mediterranean Sea (**Table 3**). Models on plastics distribution in the oceans relate inputs from large rivers, navigation routes, and coastal population centers to surface currents circulation (Lebreton et al., 2012; Eriksen et al., 2014; van Sebille et al., 2015). Relevant studies for the Mediterranean Sea provide justification for the increased spatiotemporal variability observed and define that elevated plastic concentrations are expected close to input sources (river mouths, large cities) and along a 50 km strip parallel to land (Zambianchi et al., 2017; Liubartseva et al., 2018; Mansui et al., 2020). Liubartseva et al. (2018), in particular consider the stokes drift, as well as the beaching and sedimentation effects, to conclude that any long-term accumulation of plastics at the sea surface would be unlikely in the Mediterranean, mainly due to the beaching effect on long and complex coastlines. This is particularly true for the Aegean Sea, which is characterized as a "least polluted" area, especially the central Aegean Sea where simulated plastic concentrations appear less than 2 g km^{-2} (Liubartseva et al., 2018). Seasonal simulations show a patchy distribution during winter (December–April) with relatively elevated concentrations in the N. Aegean and S. Aegean Seas and a cleansing action during summer (Politikos et al., 2017; Mansui et al., 2020). For the Levantine Sea, models agree on the formation of a coastal accumulation strip along the southern coasts of Turkey throughout the year, coinciding with the Asian Minor Current (AMC) and least polluted waters south of Crete island (Liubartseva et al., 2018; Mansui et al., 2020). For the eastern Ionian Sea, model simulations of floating litter transport conclude that moderate amounts of litter are consistently found around Corfu island, transported by the northward coastal current of the eastern Ionian Sea (Politikos et al., 2020). In accordance to model simulations, our results show the lowest concentrations of MPs in the Aegean Sea (0.15 ± 0.17 items m^{-2}), particularly in the Central Aegean transect (**Figure 2B**), while higher concentrations were recorded (0.27 ± 0.08 items m^{-2}) in the Levantine waters especially at the edge of the AMC which transports MPs from the Cilician basin as described above (**Figure 2B**). Our data set presents the highest MPs concentration in the areas of Corfu Gulf (0.63 ± 0.60 items m^{-2}) and in the N. Ionian Sea (0.38 ± 0.40 items m^{-2}) while, contrary to what one would have expected not in the highly populated Saronikos Gulf. The elevated MPs concentrations observed in these two areas may be related to the presence of sea surface slicks during October 2014. In fact, van Sebille et al. (2020) in their extensive review on the physical processes controlling the transport of plastic particles in the ocean, explain how the appearance of slicks denotes the presence of sub-mesoscale (<10 km) convergence zones due to specific physical structures (i.e., sub-mesoscale eddies, Langmuir circulation). Along these convergence zones floating objects (flotsam) i.e., seaweed, wood, oil, and plastics, show strong concentration factors (D'Asaro et al., 2018) sometimes forming visible windrows. Apart from visible objects, these formations favor also the accumulation of MPs, organic matter, planktonic organisms, fish eggs, and small fish. Pictures of the samples collected inside the slicks provide evidence of the proliferation of larvae and eggs (**Figures 3D,E**). Gove et al. (2019) investigating MPs occurrence and ingestion by larval fish inside and outside surface slicks around Hawaii islands showed that MPs concentrations were 126-fold higher inside than outside the slicks, while ingestion by larval fish was found 2.3-fold higher inside the slicks. The authors found surface slicks to be a conducive nursery habitat for small fish. In our study, MPs concentrations were found to be significantly higher (1.08 ± 0.46 items m^{-2}) when crossing or inside the slicks than those in the waters outside the slicks (0.21 ± 0.14 items m^{-2}), this regardless the studied areas. Surprisingly, surface slicks concentrations resulted to be equal to those reported recently for the N. Pacific gyre (average 1.08 items m^{-2} for size range 0.5–5 mm; Lebreton et al., 2018). The high accumulation of MPs in such sensitive habitats becomes a matter of ecological relevance due to the increased possibility for interactions (and

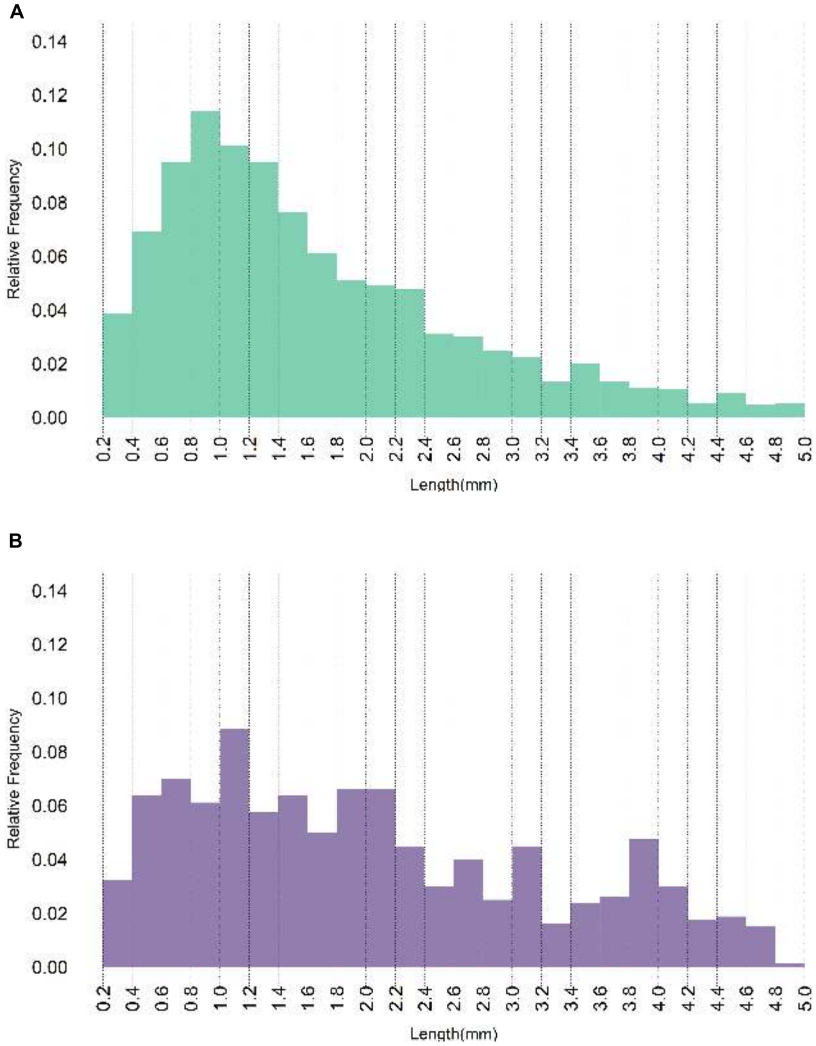

FIGURE 5 | Histograms of the relative frequency of the MPs length (mm) for MPs collected in the Ionian, Aegean and Levantine Sea (Open Sea) **(A)** and in the inner Saronikos Gulf **(B)**.

ingestion) between organisms and MPs. It is important to specify that the sampling design of this study was not arranged according to the presence of slicks, i.e., concentrations may have been even higher if trawls were conducted 100% inside the slicks, suggesting that this finding is worthy for further investigation. The importance of windrows in understanding marine litter and MPs pollution and the need for targeted windrow research has been acknowledged very recently by Cózar et al. (2021), in their relative perspective article.

It is documented that MPs show different buoyancy characteristics depending on their density, but also on their shape type, size, and biofouling degree (Kooi et al., 2016, 2017; Kaiser et al., 2017). In fact, small sized and elongated particles tend to float deeper in the water column due to increased water friction forces (Reisser et al., 2015; Kooi et al., 2016). This pattern can be further enhanced under the action of convergence and downwelling cells such as those described previously. Relatively large particles tend to stay at the surface and accumulate, while very small and elongated ones are more effectively transported below the surface. In addition, the predominance of fragments on the sea surface compared to small sized and elongated particles may be explained by the interactions between MP and biofilm and its effect on their buoyancy. In fact, biofouling should remove small fragments faster than large ones as a result of their higher surface area to volume ratios (Cózar et al., 2014); because the buoyancy of an item is a function of its volume, while its susceptibility to fouling is dependent largely on its surface area (Ryan, 2015). Given that the manta net is a neustonic sampling device, the buoyancy patterns described above, may explain the high contribution of fragments (more than 50%) in all the collected samples, and in most other places worldwide. Moreover, the minimal contribution of filaments in the N. Ionian Sea and the Corfu Gulf could be related to the sorting action of the convergence and downwelling structures that occurred

TABLE 2 | Cross-referencing of spectra identified by our instruments' and in-house libraries against the OpenSpecy libraries.

Polymer characterization		Number of spectra
ATR-FTIR libraries	**OpenSpecy libraries**	
EVA	EVA	1
HDPE	HDPE	3
LDPE	LDPE	4
Nylon	Nylon 6.6	1
PP	PP	1
PS	PS	2
PVC	HDPE	2
PVA	PVA	1
PVA	Polyacrylamide	1
PVA	Natural	26
SPE	HDPE	6
TPE	PE	2
TPE	Styrene ethylene butylene	1
TPE	EPDM	1
TPE	PP	2
Adhesive tape	PU	1
Bag	Polyester fiber	3
Carnauba wax	Honeycomb, PVA	3

there which further enhanced the downwelling of less buoyant shapes such as filaments. MPs floating in the Ionian, Aegean and Levantine Seas occur mostly in sizes smaller than 2 mm peaking at the size range 0.8–1.4 mm, that is in accordance with other surveys using manta nets (330 μm; Cózar et al., 2014, 2015; Isobe et al., 2015; Maes et al., 2017). The similarities in the size distribution of MPs caught in the manta nets in various marine regions reflect that the MPs assortment has undergone substantial mixing during long residence time at sea.

Global Plastics production in 2019 reached 368 Mt, out of which 58 Mt correspond to European production.[2] About 40% of all polymers produced are used in packaging, with polyolefin polymers PE and PP having the highest demand, followed by PVC, PET, PS and EPS, and PA (European market demand: 9, 7, 5, and 2%, respectively). In the oceans' surface waters, this may be modified according to the buoyancy characteristics of each of the polymers. In fact, the relative contributions of light polymers such as PE and PP is further enhanced due to their low densities (0.86–0.96 g cm^{-3}), lower than the surrounding seawater (1.28 g cm^{-3}), while denser materials tend to escape from the surface layer. In line with the above, the chemical composition of sea surface MPs in the Ionian, Aegean and Levantine Seas is dominated by PE and PP polymers by 80%. Other polymers present are PA-Nylon, PET and PVA. Polyamides such as Nylon are used in fiber manufacturing while PET is used for packaging and for textile manufacturing in the form of fibers (polyester type fibers). Although other studies have reported the presence of PVA in environmental samples (Ng and Obbard, 2006; Claessens et al., 2011; Chae et al., 2015; Suaria et al., 2016; Zeri et al., 2018), its considerable proportion is quite surprising as it is not one of the commonly used materials. PVA is a hydrolizable, glue-like adhesive polymer, used also as a coating for other materials such as cloth and paper and more recently for 3D printing. The cross-referencing of our spectra with a larger data base revealed that PVA is one of the most questionable materials, as 26 out of 28 spectra presented high similarities to various natural materials, glue like natural resins (wax, brood comb), chitin, algae, linen, wool (animal fur), rather than the polymer spectra. A recent study on fibers distribution in the oceans (Suaria et al., 2020) highlighted that natural fibers are dominant in surface oceanic waters (0–5 m) (cellulosic 79.5% and of animal origin 12.3%) in comparison with those of synthetic origin (8.6%). Here we should mention that at least one of the OpenSpecy libraries (Primpke et al., 2018) used for the cross-referencing of the spectra is common to that used by Suaria et al. (2020). The spectral similarity by more than 70% with either PVA or natural materials, depending on the library used, indicates the presence of common functional groups among all these materials. Our findings suggest that checking against a greater data base of spectra for environmental particles is considered useful in refining the results obtained only from polymers data bases (Cowger et al., 2021). At the same time, the question is raised whether the obtained spectra indicate deviations from the pure PVA spectrum, related to its fast degradation/hydrolysis in the environment (Min et al., 2020) or are indicative of natural materials with functional groups similar to PVA. Furthermore, the use of PVA in paper and textile coatings further complicates the issue. Experimental work with known materials and combination of libraries including degraded polymers could be useful in clarifying the questions raised above.

Many studies have shown a direct relationship between MPs elevated concentrations and input sources such as large urban centers or river mouths (Frias et al., 2014; Zhao et al., 2014, 2015; Gewert et al., 2017; Sun et al., 2019) and have highlighted the importance of flood events for the washout and transport of MPs in coastal areas (Moore et al., 2002; Veerasingam et al., 2016; Gündoğdu and Çevik, 2017). In our study, for the case of Corfu Gulf, we cannot ascertain if the elevated MPs concentrations recorded reflect direct inputs of MPs from the Kalamas river, or rather the subsequent concentration effect caused by the formation of sea surface slicks. On the other hand, although Saronikos Gulf has been identified by models as a hot spot area, due to the high plastic inputs from Athens metropolitan area (Liubartseva et al., 2018), our field data, do not capture constant elevated MPs concentrations there (0.16 ± 0.22 items m^{-2}), even in the inner part of the Gulf and after the heavy rain event (**Table 1**). It is possible, that local conditions may favor fast dispersion or beaching of plastic particles. While some authors have reported that large sized MPs, meso- and macro- plastics become less abundant close to the shores than in offshore waters (Pedrotti et al., 2016; Zeri et al., 2018), others have shown the opposite. In particular, close to river mouths and urban centers the relative abundance of large sized MPs has been linked to shorter residence time in the marine environment (Morét-Ferguson et al., 2010; Schmidt et al., 2017). Isobe et al. (2014) combined modeled and field data and demonstrated that meso-plastics are selectively drifted close to the shores,

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[2] www.plasticseurope.org

TABLE 3 | Sea surface MPs concentrations (items m^{-2}) reported for the Mediterranean Sea, using surface manta nets.

Area	Net mesh (μm)	MPs (items m^{-2})	Sources
Mediterranean Sea	200	0.24	Cózar et al., 2015
Mediterranean Sea	330	0.14 ± 0.025	Ruiz-Orejón et al., 2016
W. Mediterranean Sea	333	0.12 ± 0.13	Pedrotti et al., 2016
W. Mediterranean Sea	330	0.10 ± 0.09	de Haan et al., 2019
W. Mediterranean Sea	780/330	0.11	Schmidt et al., 2017
W. Mediterranean Sea	330	0.082 ± 0.079	Fossi et al., 2017
W. Mediterranean and Adriatic Seas	200	0.4 ± 0.7	Suaria et al., 2016
Adriatic Sea	330	0.3 ± 0.5	Zeri et al., 2018
E. Mediterranean Sea	333	0.14 ± 0.12	Güven et al., 2017
E. Mediterranean Sea	333	0.38	Gündoğdu and Çevik, 2017
E. Mediterranean Sea	333	1.5	van der Hal et al., 2017
E. Mediterranean Sea	330	0.26 ± 0.36 (0.012–1.62)	Present study

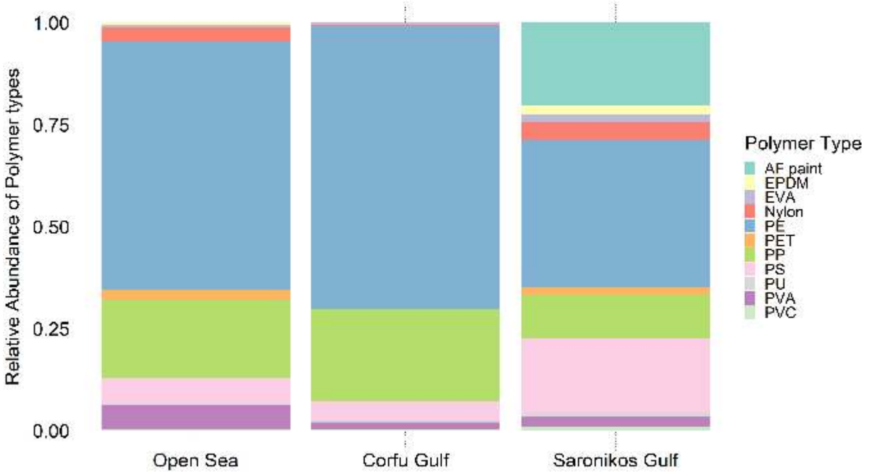

FIGURE 6 | Relative abundance of polymer types of MPs collected in the Ionian, Aegean and Levantine Sea (Open Sea), Corfu Gulf and inner Saronikos Gulf.

independently from the presence river mouths. Based on these outcomes, it is understood that the size distribution of plastic particles close to the coastline is controlled by both the vicinity to sources (short residence time) and the prevailing dispersion-concentration modes (Doyle et al., 2011; Frere et al., 2017). Recent data on MPs in Kifissos river water further upstream confirm the high level of large MPs and mesoplastics, mostly PE films (Zeri et al., 2021). In addition, the small beach by the river mouth has been characterized as a hot-spot area for beach litter (>10,000 items/100 m) with ~50% of items corresponding to small sized plastic and polystyrene fragments (Greek Marine Strategy Framework Directive monitoring program, HCMR unpublished data). These local conditions may explain the observed MPs distribution and properties in the inner Saronikos Gulf. In fact, as already described, in this area, several known input sources of MPs coexist (Kifissos River mouth, WWTP, ports and marinas, anchorage points). Apart from the typical contribution of PE and PP, a considerable presence of foam particles in the inner Saronikos Gulf was confirmed by their chemical signature, PS/EPS being 18%. In Greece, the market demand for plastics in 2018–2019 was ~1 Mt per year; while recycling and energy recovery rates in the country are of the lowest in Europe (70% of waste is landfilled) (see text footnote 2). The EU Directives on the banning plastic bags (EU/720/2015) and single use items (EU/904/2019) came into force only very recently (in 2019 and 2020 respectively). This situation has favored plastic leakage in the environment and is reflected in the MPs chemical composition in the inner Saronikos Gulf where a higher diversity of polymer types has been recorded. In fact, different types of polymers indicative of specific economic sectors such as industry, construction, maritime, were found in this area (EVA, PU, EPDM, and AF-paints). AF-paint particles are usually painted metal chips not expected to stay afloat for long periods, and their presence in the inner Saronikos samples is a strong indication of the direct inputs of MPs from the anchorage points, ports operations and increased marine traffic.[3] Lastly, the presence of WWTP outflow in the analyzed samples were not evidenced, this is probably related to the fact that sewage enters the gulf via a diffusive pipe situated at 65 m depth. In addition, MPs mostly found in sewage correspond to small sized

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[3] www.marinetraffic.com

filaments (Talvitie et al., 2015), which are unlikely to be caught by surface manta nets.

The results presented in this work highlight that local oceanographic conditions, such as slicks, significantly affect MPs concentrations, posing risks in these sensitive habitats. Our study suggests that MPs sources from coastal population centers were not detectable based on MPs concentrations, but rather on properties such as size and polymer type. Elevated MPs concentrations were not found close to the major MPs sources at the Athens metropolitan area. The assortment of MPs there, consisted of large sized MPs and more variable polymer types, holding rather "local" features and differentiated from the well mixed MPs assortment found in open waters and less impacted areas. To our knowledge, this is the first study presenting field data of floating MPs concentrations in relation to the presence of sea surface slicks in the Mediterranean Sea, and provides evidence that slicks act as strong MPs concentration factors. It is anticipated that the outcomes of the present study will provide insights toward a better interpretation of floating MPs data from systematic monitoring activities.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author/s.

AUTHOR CONTRIBUTIONS

AA processed the samples, analyzed the data, and wrote and reviewed the manuscript. CZ conceived the study, collected the samples, and wrote and reviewed the manuscript. FG processed the samples and reviewed the manuscript. CG collected and processed the samples and reviewed the manuscript. CI collected the samples and reviewed the manuscript. EP processed spectroscopic data and reviewed the manuscript. All authors have read and agreed to the published version of the manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fmars.2021.699000/full#supplementary-material

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