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RESEARCH PAPER

Microlitter Pollution in Sea Water: A Preliminary Study from Sinop Sarikum Coast of the Southern Black Sea

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Abstract

In this study, microplastic pollution for the first time is reported in the sea surface and water column in Sinop Sarıkum Lagoon coast of the southern Black Sea. Distribution, density and type of microplastics were investigated in the study area where is one of the significant wetlands of the Black Sea. Study was carried out between 2015 and 2016 as seasonally and two types of net for sea surface and water column were used for three different depths. Results showed that microparticle density was 2.667±2.325 pieces m^{-3} for sea surface and 24.475±26.153 pieces m^{-3} for water column. The most common material type is ship paints for sampling areas (Sea surface: 55.45%; Water column: 54.21%) followed by fibres, hard plastic pieces and nylons. Our results show that microplastic is found a large amount in the Black Sea and this pollution type is a growing problem in our day. The presence of this pollution type is an important issue and requires further examination about transportation, origins, types and effects on biota.

Keywords: Microplastic, marine litter, Black Sea, pollution, Sarıkum Lagoon.

Introduction

Marine litter may be defined as: "*any persistent, manufactured or processed solid material disposed of or abandoned in the marine and coastal environment*" (UNEP, 2005). Unexpected or planned human behaviours and activities are the sources of marine litter (UNEP, 2009).

Plastics are commonly integrated into today's way of life and make a major contribution to virtually almost all product areas. The typical characteristics that render them so useful relate primarily to the fact that they are flexible and long-lasting (Hammer, Kraak, & Parsons, 2012). Over the last 60 years, plastic production has increased enormously from 1.7 to 288 million tons (Plastics Europe, 2013).

The endurance and surge usage of plastics make a big waste management problem (Thompson, Swan, Moore, & vom Saal, 2009). Huge amounts of plastic accumulate in natural environments and garbage dump site (Thompson, Moore, vom Saal, & Swan, 2009). In these days, plastics dominate marine litter and plastic litter constitutes 60-80% of all marine litter (Gregory & Ryan, 1997; Derraik, 2002). One of the most important changes that have taken place in recent times is the accumulation and fragmentation of plastics (Barnes, Galgani, Thopson, & Barlaz, 2009).

After entering the environment most polymeric materials are subject to degradation that impact by factors such as thermal oxidation, photo-oxidative degradation, biodegradation and hydrolysis (Hammer *et al*., 2012). Microplastics have been defined as synthetic polymer particles, being less than 5 mm (Arthur, Baker, & Bamford, 2009). Microplastics enter the marine environment as preproduction pellets (primary microplastics) or arise from breakdown of larger items already present as marine litter in the marine environment (secondary microplastics) (Andrady, 2011; Cole, Lindeque, Halsband, & Galloway, 2011; CIESM, 2014).

In recent studies, the presence of microplastic was reported beaches, sea floor and sea water and even freshwater (Mathalon & Hill, 2014; Cauwenberge, Vanreusel, Mees, & Janssen, 2013; Collignon *et al*., 2012; Eriksen *et al*., 2013) and also it was reported that microplastics was found many marine species from zooplankton to invertebrates, fish, sea birds and marine mammals (Boerger, Lattin, Moore, & Moore, 2010; Lusher, McHugh, & Thompson, 2013; Codina-Garcia, Militão, Moreno, & González-Solís, 2013; DiBeneditto & Ramos, 2014). Microplastics are consumed by filter-feeding at the base of the food web (Cole *et al*., 2013) and transfer of the trophic level (Setälä, Fleming-Lehtinen, &

Lehtiniemi, 2014; Farrell & Nelson, 2013). Toxicity concerns are raised by ingestion of degraded plastic pellets and fragments, because plastics are known to adsorb hydrophobic pollutants. The potential bioavailability of added to plastics during manufacturing (such as Bisphenol-A), besides those adsorbed from the environment are complex issues that deserve more widespread investigation (Moore, 2008).

The Black Sea is about 2000 m deep basin with zonal and located roughly between 28° and 42°E longitudes, 41° and 46°N latitudes. It has only a narrow opening to the shallow (less than 75 m deep) Bosphorus Strait restricting exchange with the Mediterranean Sea. Bathymetry of the Black Sea is characterized by a flat abyssal plain with a maximal depth of 2200 m in the central area and a continental shelf. Unlike the Mediterranean Sea, the Black Sea is an estuarine type basin due to the large river discharge, especially on its north-western shelf area. The low salinity surface waters reach the Turkish Black Sea coast in modified form due to mixing. Throughout the Black Sea, the low salinity surface waters of riverine origin overlay salty deep waters of the Mediterranean Sea origin. Inflow of the salt water through the Bosphorus Strait determines the density stratification of the Black Sea (Oguz, La Violette, & Unluata, 1992; Oguz *et al*., 1993; Oguz, Malanotte-Rizzoli, & Aubrey, 1995; Ozsoy, Unluata, Top, 1993; Ozsoy & Unluata, 1997). In addition to all these factors, the Black Sea has a very dynamic current system allowing cross-border transportation of waste materials (Topcu & Ozturk, 2010), which in turn makes this enclosed sea very vulnerable to marine litter (Topcu, Tonay, Dede, Ozturk, & Ozturk, 2013).

During the last decade, some governmental and private institutions in Bulgaria, Romania, Russia, Turkey and Ukraine conducted marine litter research using different approaches and methods (UNEP, 2009). Scientific studies on this issue have gained speed in recent years but it is still quite inadequate and especially the researches about microplastics are quite little. Most recently, Aytan *et al*. (2016) reported that first evaluation of neustonic microplastics in the Black Sea waters. They reported that the relatively high microplastic concentrations suggest that the Black Sea is a hotspot for microplastic pollution and there is an urgency to understand their origins, transportation and effects on marine life.

Marine Strategy Framework Directive (2008/56/EC) (MSFD) published by European Union (EU) in 2008, includes necessary measures to provide or maintain "Good Environmental Status" (GES) of the Member States up to 2020. In Annex I to Directive 2008/56/EC is determined Good Environmental Status according to 11 qualitative 'descriptors' for marine regions and sub-regions and Descriptor 10 is related to marine litter. Marine litter (Descriptor 10) has been defined as "*Properties and quantities of marine litter do not cause harm to the coastal and marine environment*" (2010/477/EU) and in this descriptor microlitter take in consideration "*10.1.3. Trends in the amount, distribution and, where possible, composition of microparticles (in particular microplastics)*". Turkey as an associated member of the EU is seeking to harmonize national legislation with EU MSFD (Aydın, Guven, Salihoglu, & Kıdeys, 2016).

Sarıkum Lagoon is one of the significant wetlands of the Black Sea. Lagoon and its surroundings due to the geographical location are exposed to a significant accumulation of solid waste both with sea currents and influence of the prevailing winds in Sinop peninsula. To evaluate the level of the microlitter pollution on sea surface and in water column conducted investigations in region shores during 2015-2016 and used proposed methods of MSFD (MSFD GES Technical Subgroup on Marine Litter, 2013).

Materials and Methods

Study Area

Sarıkum Lagoon is located on the western coast of Sinop Peninsula in Central Black Sea Region (Figure 1). Sinop is located 21 km west of the city centre, between 42°00'00"-42°02'42" N and 34°54'46"-34°58'22" E latitudes (Yılmaz, 2005). Sarıkum Lagoon which is one of the significant wetlands of the Black Sea has been announced as Nature Reserved Area in 1987 and then lagoon surroundings have been declared as Natural Protected Area in 1991.

Sample Collection

In 2010, following the Commission Decision on criteria and methodological standards on GES of marine waters, the Marine Directors requested the Directorate-General for the Environment (DG ENV) to establish a Technical Subgroup on Marine Litter (TSG-ML) under the Working Group on GES (WG GES), to address these gaps and further develop Descriptor 10. Proposed methods by Guidance on Monitoring of Marine Litter in European Seas' published by European Marine Strategy Framework Directive Technical Subgroup on Marine Litter section of microlitter adopted the region (MSFD GES Technical Subgroup on Marine Litter, 2013).

Study was carried out in May 2015- February 2016 as seasonal. Samples were taken from sea surface and water column at 3 different depths (5 m, 15 m and 30 m). Two types of net were used for sampling. Neuston net which was 60x20 cm rectangular mouth opening and 250 cm long and 300 µm mesh size was used to get neuston samples (NS) from sea surface. Cylindro-conical plankton net which was 45 cm mouth opening was used to get column samples (CS).

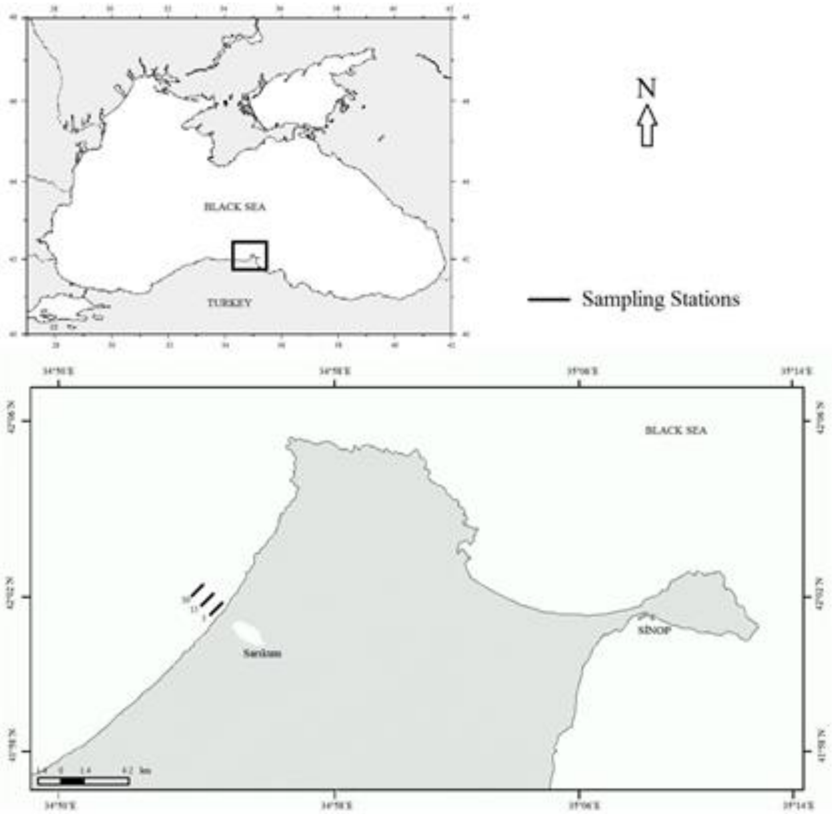

Figure 1. Sampling area.

Collected samples retained from plankton nets passed through 1 mm sieve and then filtered through 4.7 µm retention glass microfiber filter paper (GF/C; 1.2 µm; 47 mm Ø; Whatman) using vacuum filtration system. Filter papers were examined under the stereo microscope. The abundance of any pieces of unnatural appearance was recorded. The amount of microparticles was calculated in m^3. They were categorized according to types (ship paint, plastic fragments, fibres, polystyrenes, other etc.), colors (white, transparent, blue, black, brown, green, yellow, grey, red etc.).

Results

Litter Abundance

A total of 2314 pieces of microparticles were found all seasons and all stations in NS. Study results show that microparticle density was found as 2.67±2.33 pieces m^{-3} for sea surface. The most intense season was found in spring and lowest season was found in winter in terms of microparticle density. When the study results are evaluated according to depths, in terms of the amount of microparticle the farthest station from the beach at depth of 30 m was the highest value, the station at depth of 15 m was the second highest station and the station at depth of 5 m was the station with the lowest value. In this context, it has been observed that the microparticle density increased on the sea surface as it moves away from the shore. There was no statistically significant differences between stations and seasons in NS ($P>0.05$).

In CS 687 pieces of microparticles were found total of all seasons and all stations. Study results show that microparticle density was found as 24.48±26.15 pieces m^{-3} for CS (Figure 2). The most intense season was found spring and lowest season was found autumn in terms of microparticle density. When the study results are evaluated according to depths, in terms of the amount of microparticle most polluted station was found 5 m and least polluted station was found 30 m. In this context, it has been observed that the microparticle density increased on the CS as it closer to the shore (Figure 2). There were no statistically significant differences between seasons ($p>0.05$) but there were statistically significant differences between 5 m and 30 m stations (Tukey test $F=5.698$, $p=0.025$).

Litter Composition

Results of NS and CS were categorized according to type of material. Samples were separated to 6 groups which are plastic fibres, hard plastic, nylon pieces, polystyrene pieces, ship paint and others (other group contains glass pieces, bitumen residues

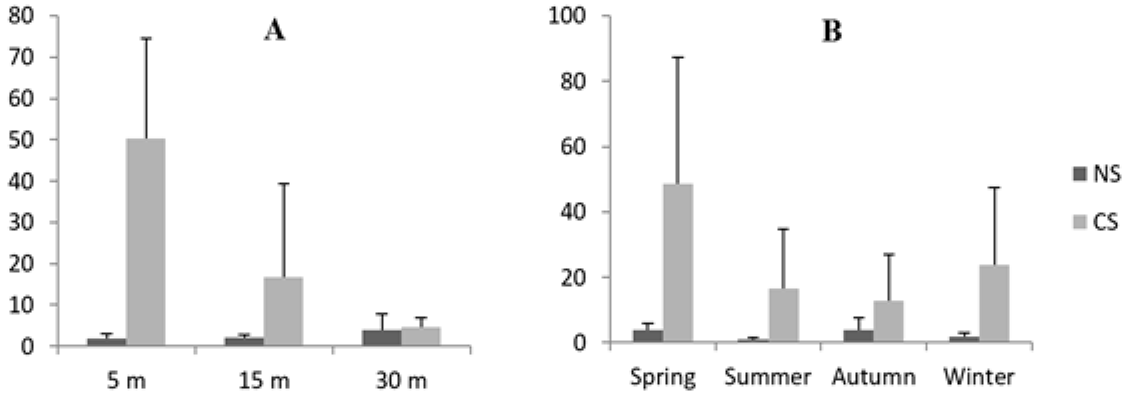

Figure 2. Distribution of microlitter in sampling area among different depths (A) and seasons (B).

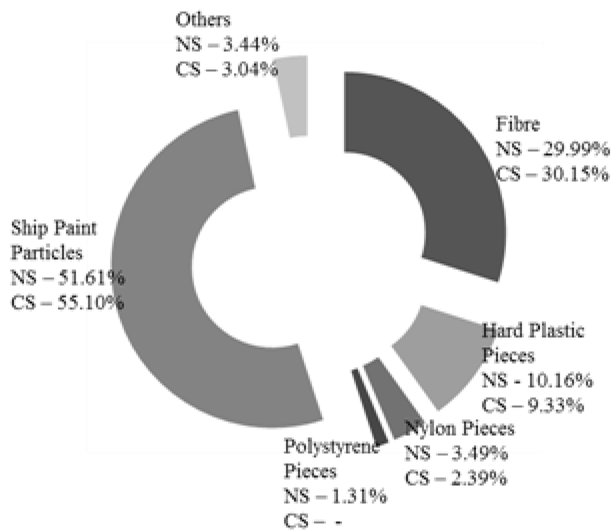

Figure 3. Percentage distribution of microlitter according to their type.

and unidentified items). In samples taken from NS, the highest amount of microlitter category was found ship paint followed by plastic fibres, hard plastics, nylon pieces, other and polystyrene particles, respectively (Figure 3). In CS samples the most dominant microparticle type was observed in ship paint followed by plastic fibres, hard plastics, others and nylon pieces and no polystyrene pieces were found in CS (Figure 3).

Microlitter in sampling stations were categorized 10 diffent groups acording to their colors and themost common color was found white both NS and CS (Table 1). The intense presence of this color group is due to the fact that the most common material group which is ship paints are white in general.

Discussion

The presence of microparticles on the sea surface and column was recorded in the study area as the other studies in different seas of the world (Table 2). It was observed to be present in a large amount compared to other studies on NS (Eriksen *et al*., 2013; Gago, Henry, & Galgani, 2015; Collignon *et al*., 2012). However, it is compared with the study which is conducted in the SE Black Sea (Aytan *et al*., 2016), microlitter pollution is quite little both NS and CS in our study.

Compared to the density of the microlitter on the sea surface and in the water column, it was observed that more intense microlitter was found in the water column. Rivers dilute the Black Sea water - its surface layer salinity is 17‰, two times less than those in the ocean (average 35‰). Thus microlitter may have a tendency to sink due to density variation. It can be explained that the microlitter density in the CS is higher than the NS. Furthermore, the data obtained in the part of the sea bottom survey of project which the results of this part is supporting this hypothesis. Sinking experiments were conducted with diverse polymer particles using fluids with different salinity (Kowalski, Reichardt, & Waniek, 2016). It is reported

Table 1. Percentage color ratio in NS and CS

Sampling area	Color (Percent-ratio %)									
	Black	Blue	Brown	Green	Grey	Purple	Red	Trasparent	White	Yellow
NS	10.70	14.35	-	6.52	8.52	0.35	12.74	3.91	40.04	2.87
CS	1.94	15.98	0.43	5.62	20.09	-	5.83	11.45	37.80	0.86

Table 2. Neustonic microplastic amount comparison with other studies

Region	Pieces/m^2	Pieces/m^3	Pieces/km^2	References
Black Sea Turkey	0.67± 0.55	2.67±2.33	656,000	This Study
Black Sea Turkey	-	$1.2x10^3 \pm 1.1x10^3$ $0.6x10^3 \pm 0.55x10^3$	-	Aytan *et al.*, 2016
Mediterranean Spain	0.034±0.032 0.176±0.278	-	-	Gago *et al.*, 2015
Mediterranean	0.116	-	-	Collignon *et al.*, 2012
South Pacific	-	-	26,898	Eriksen *et al.*, 2013
East Asian Japan	-	-	1,720,000	Isobe *et al.*, 2013
SE Korea	-	1.92±1.84 5.51±11.24	-	Kang *et al.*, 2015
Korea	-	0.19 ± 0.14	-	Chae *et al.*, 2015

that the sinking experiments with six different polymer types distinctly show the relevance of particle size, density and shape as well as salinity of the ambient fluid for the sinking velocity. They clarified that sinking velocity is decrease with increasing salinity (Kowalski *et al*., 2016).

Thornton and Jackson (1998) and Carlos Astudillo, Bravo, Dumont, and Thiel (2009) have proposed that physical factors such as the wind and wave-action have a role in influencing spatial patterns of accumulation. In the Mediterranean Sea microplastic particle concentrations measured before the strong wind event were 5 times higher than those measured after the strong wind in sea surface. Thus, it is suggested that significant difference can be explained by the extreme differences in wind conditions. The mixing and vertical redistribution of the plastic particles in the upper layers of the water column raised the result of wind force (Collignon *et al*., 2012). Especially in the winter when the weather conditions were negative, our study results showed a significant decrease in NS but this situation is exactly the opposite in CS, namely the micro litter density showed an increasing effect in CS. On the other hand, it is observed that microlitter density decreases in the CS as it moves away from the shore, which is the opposite situation on the NS (Figure 4). Microlitter density at the NS has reached the maximum level at the farthest station from the beach. This may be related to the fact that water is more mixed in shallow regions due to wave action.

The Black Sea is characterized by a predominantly cyclonic and strongly time-dependent basin wide circulation that follows approximately the continental slope around the basin. Sinop eddy may form repeatedly once or twice a year for about a season in region and the presence of this eddy mostly depends up on propagation characteristics of the meanders superimposed on the Rim Current system (Korotaev, Oguz, Nikiforov, & Koblinsky, 2003). Sampling area is under the influence of regional winds, wave and currents. When considering the special condition of the region, such factors are important for interpreting the distribution of microlitter and when the Black Sea current plots are examined (Figure 5), the region attracts attention as one of the important solid waste accumulation areas.

There are many factors that affect the horizontal circulation of water and internal water exchange in the Black Sea. The most important ones can be emphasized that they are the stable quasi two layered density stratification of the water, unsteady and spatially irregular wind forcing, and the characteristic features of topography of the continental slope. One of the important characteristic features of the horizontal water circulation in the Black Sea is the general cyclonic gyre in the upper layer, i.e., the contour current, which flows counter clockwise along the coast. Since eddies are one of the main mechanisms of the interaction of the near-shore zone with the deep sea (Zatsepin *et al*., 2003). The wind effect seems to be the most important factor that determines both the structure and the intensity of the horizontal water circulation. Our results have shown that the density of microplastic was found less in summer, this may be explained that the weakening of the wind stress leads to a decrease in intensity of the mean currents in summer (Stanev, 1990) (Figure 6).

Korotaev, Saenko, & Koblinsky (2001) pointed out that the Black Sea circulation has a strong seasonal variability, with attenuation in summer– fall and intensification in winter–spring. Thus the seasonal variability of the Black Sea upper layer circulation might not be so simple. However, The

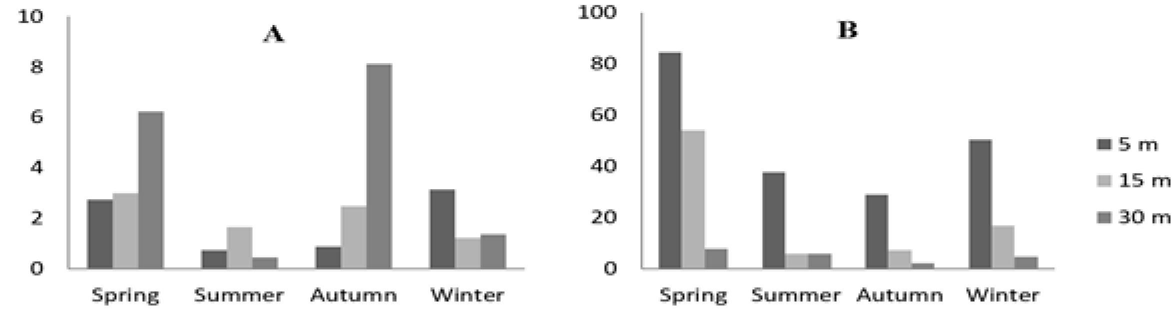

Figure 4. Amount of microlitter depend on seasons and stations (A-Sea Surface, B-Water coloumn).

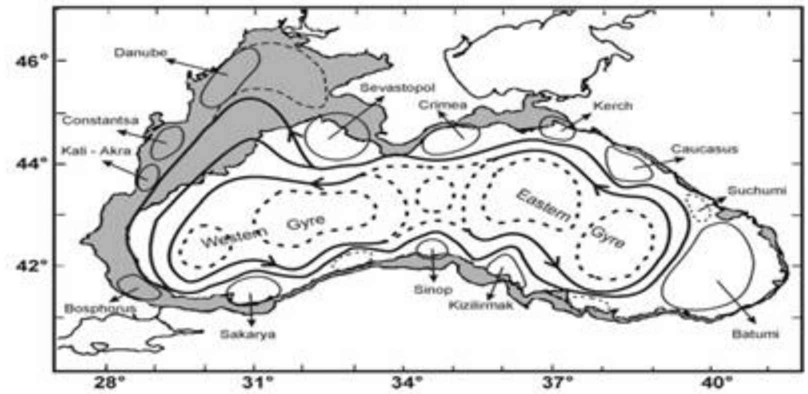

Figure 5. The schematic diagram for the main features of the upper layer circulation of the Black Sea (taken from Korotaev *et al.*, 2003).

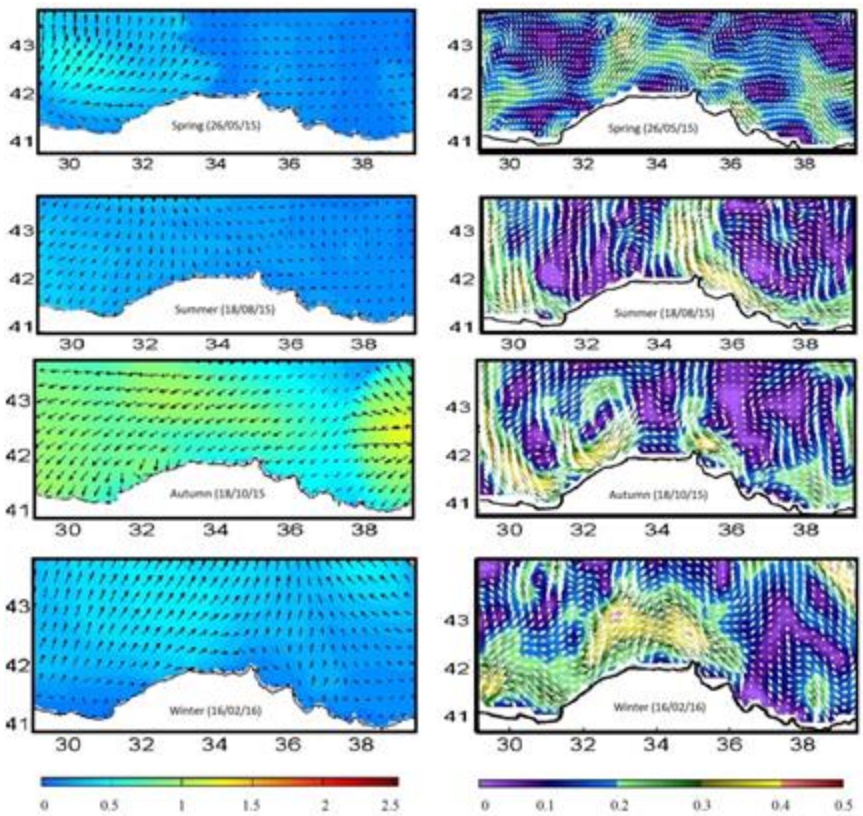

Figure 6. Black Sea current (taken from Marine Hydrophysical Institute of RAS) and wind directions and speed (taken from Poseidon System Hellenic Centre for Marine Research) during sampling days.

Rim Current transports water around the perimeter of the Black Sea in geostrophic balance and is the most intense in winter-spring seasons (Korotaev *et al*., 2011). Stanev, Le Traon, & Peneva (2000) explained that the general cyclonic circulation intensifies in winter and spring corresponding to pronounced positive sea level anomalies in the coastal areas. They also indicated that the wind is the important force driving the seasonal intensification of the circulation. Since there are no marine litter sources neither land nor river based sources in the study area, the microlitter come through only wind and currents. It is suggested that increasing trend in the microlitter amount especially in the spring and winter samples, may be due to regional current flow and wind stress.

Formal identification of particles uses FT-IR (Fourier Transform Infra-Red Spectroscopy) or Raman Spectroscopy and these analyses is suggested by Guidance especially for fragments <100 µm (MSFD GES Technical Subgroup on Marine Litter, 2013). In this study microlitter samples consist of relatively large particles (>300 µm) and our study don't include this analyse. However, interpretations can be made on the possible sources of microplastics found during the study although the polymer types have not been determined. Paint particles were observed in this study area a high frequency especially in fishing season. This area is affected by shipyard-based pollution, like many other studies (Song *et al*., 2014; Chae, Kim, Kim, Song, & Shim, 2015). Various paints with anticorrosive and antifouling properties were used by local fishermen. Anticorrosive polymer resin paints used on ships contain various inorganic and/or organic chemicals (Hamid, Blustein, Deya, Amo, & Romagnoli, 2011; Gade, Heiaas, Lillicarp, & Hylland, 2012; Chae *et al*., 2015) and also antifouling paints (Zhou, Guo, & Hao, 2007; Lin *et al*., 2008; Chae *et al*., 2015). The existence of paint related pollutants was verified with our study but the presence of the toxic chemicals in the ship paints was beyond the scope of this study. However, Song *et al*. (2014) ratified the existence of various elements at considerable levels in the alkyd particles.

The second most common material group was found plastic fibres. Synthetic plastic fibres have many uses in the field of from clothing to ropes and for this reason it can be mixed in intense amounts through various activities in the marine environment. From municipal wastewater to maritime activities, many sources may have led to enter of these materials (Hidalgo-Ruz, Gutow, Thompson, & Thiel, 2012). A major source of plastic fibres reported as disposal of municipal wastewater contaminated with fibres from washing clothes and experiments sampling wastewater from domestic washing machines demonstrated that a single garment can produce >1900 fibres per wash (Browne *et al*., 2011). Sinop is not populated area but the study area is affected by prevailing winds of BS and pollutants are transported from neighbouring cities and even countries. Therefore, the municipal wastewater came from these regions can be major sources of the fibres in the region and at the same time study area is an important fishing area in Sinop and fishing ropes and fishing gear can be an important source of fibre founded in samples. Recently published paper which was made in the SE Black Sea, plastic fibre concentration was found the highest percentage (~49.4%). However in same region no mention about ship paints (Aytan *et al*., 2016). Hard plastic pieces, which are third in terms of their presence in the marine environment in our study area, are generally found in the marine environment due to the breakdown of larger plastics (Hidalgo-Ruz *et al*., 2012).

The study area is one of the fishing points of the Sinop region and, there are high amounts of fishing boats in the region during fishing season. Especially in the autumn, fishery activities as a source of microlitter are attracting the attention. One of these is polystyrene boxes residues and most of the polystyrene fragments in NS were found during the autumn season. This is thought to be due to the fact that the polystyrene-based foam boxes used in fishing were left unconsciously in the environment. However, the polystyrene fragments found on the NS are not found in CS, and it is thought that the low density of the polystyrenes move directly from the water surface to the shore sediments. The other pollutant which is come from fisheries activities is fuel contamination. Bitumen residues were encountered during autumn sampling. In this season fishing activities reaches maximum level in sampling area and this is the result of the mixing of the fuel wastes of fishing boats. Possible sources of the microlitter encountered in the results of the study shows that marine environment is involved in intensive amounts of pollutants as a result of maritime activities.

There is an increasing concern about consumption of microplastics by marine organisms and found of this pollutant in fish stomach reported from all over the world (Boerger *et al*., 2010; Lusher *et al*., 2013; Codina-Garcia *et al*., 2013; Anastasopoulou *et al*., 2013; DiBeneditto & Ramos, 2014). One of the first reports on this issue that in New England examined 8 species out of 14 species consumed selectively white, opaque spherules (Carpenter, Anderson, Harvey, Miklas, & Peck, 1972). When microlitter samples were examined according to colour category both NS and CS samples in the Sarıkum Lagoon coast, white was found most common colour.

Microplastics are consumed by filter-feeding at the base of the food web (Cole *et al*., 2013). The dominant planktivorous fish in the Black Sea is European anchovy (*Engraulis encrasicolus*) which is main commercial fish stock of Turkey (Bat, Kaya, & Öztekin, 2014) and the possibility of this species encounter with microlitter is high (Aytan *et al*., 2016). Resulting from ingestion of plastic litter, concerns have also been raised about physical problems such as block feeding appendages or hinder the passage of food (Lusher *et al*., 2013) and more transferring toxic pollutants into marine food chains. The potential danger of compounds added to plastics during production as well as the compounds that plastics absorb from nature pose a significant risk, and this also poses the danger of digestive toxicity of plastic particles. Moreover, the transfer of the microplastics in trophic level has been reported in experimental studies (Setala *et al*., 2014; Farrell & Nelson, 2013) and the problem of transport of toxic pollutants from plastics in the food web is emerging.

Conclusion

The Black Sea poor ecological conditions are a result of its limited water exchange with the open basins, weak vertical mixing (due to the strong density stratification and weak tides), and enhanced pollutants including macro and microplastics by river discharges, domestic and touristic wastes, fisheries activities and other discharges from shipping. Because most of the pollutants come from the shore and near-shore regions of the marine environment, the processes of horizontal mixing and shelf sea water exchange are of the great importance. The presence of this pollution type is an important issue and requires further examination about transportation, origins, types and effects on biota.

The results show that Sinop Sarıkum Lagoon shores exposed to a high level of microlitter pollution and the region is particularly contaminated with sea-based pollutants (ship paints, polystyrene box pieces etc.). Unfortunately, there is no protection against this pollution type in the study area. Therefore, it is vulnerable to monitor marine litter pollution.

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