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Differences in the Fate of Surface and Subsurface Microplastics: A Case Study in the Central Atlantic

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Abstract: Plastic is recognized as a threat to marine ecosystems, and estimating the level of plastic and microplastics (MPs) pollution of the World Ocean is, nowadays, the goal of many studies. However, the use of different methods for the sampling and analysis of MPs leads to the problem of comparing the results obtained. Studies on surface MPs pollution of the surface sea water are based on the application of the manta or neuston nets that collect water from the upper 10–20 cm layer (the "surface" MPs) or submersible pumping systems that collect water from the water layer 3 to 5 m below the sea surface (the "subsurface" MPs). These two techniques allow the collection of particles of different size fractions, i.e., >300 μ m for the surface and subsurface layers differ not only in the size of the items found, but also in morphology, types of polymers, abundance, weight concentration and their spatial distribution. Different hydrodynamic processes affect the fate of the plastic found exactly at the sea surface and several meters deeper. The aim of this work was to study the distribution of surface and subsurface MPs and to reveal an influence of oceanographic conditions on their spatial distribution, using as an example the open ocean waters of the Central Atlantic.

Keywords: microplastic pollution; different types of microplastics; FT-IR; visual analysis

1. Introduction

Microplastics pollution is a modern challenge for scientists around the world. There are a number of reasons for this. First, there is an annual growth in the production of polymeric materials. The beginning of the active production of polymers and plastic products can be considered the middle of the 20th century [1]. Since then, the growth of production volumes has been a global trend. Estimations from 2020 amounted to about 367 million tons and are likely to continue to grow [2]. Apparently, the trend will continue in the near future, as at the same time prices for hydrocarbon raw materials for polymer synthesis are decreasing [3]. If these trends continue, by 2050 there will be about 12,000 million tons of plastic waste in the environment [4]. Therefore, it is not at all surprising that microplastics particles are found in almost all environments: in the sea water [5] and fresh water [6], in bottom sediments and soils [7], and in snow [8]. After all, plastic waste is one of the main sources of secondary microplastics [9]. It is formed as a result of the fragmentation of debris into smaller particles under the influence of ultraviolet radiation, mechanical stress and atmospheric oxygen [10].

Fragments of plastic waste are usually divided into size categories: Macro (25–1000 mm), Meso (5–25 mm), Micro (<5 mm), and Nano (<1 μ m) [9]. As a rule, studies focus on one size category. Historically, the first works were devoted to the study of larger size groups [11]. Currently, the focus is shifting towards smaller sizes.

To date, it is known that different particle size groups have similar properties, despite the general heterogeneity due to the morphology and type of polymer. At the same time,



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). the difference between different size groups becomes just as obvious. It is known from field studies and modeling that larger plastics with positive buoyancy will accumulate in the surface layer, while small particles' distribution is more dispersed [12]. The difference is not limited to the distribution of particles in vertical and horizontal directions; apparently, particles of different sizes and morphologies cause damage to wildlife in different ways. At high concentrations, the growth of some algae is impaired, and the effect is enhanced with a decrease in particle size [13]. Particle uptake by marine organisms is also affected by both size and morphology. Larger particles of secondary microplastics are eaten less actively, all other things being equal [14]. Therefore, in our opinion, it is important to correctly divide the studied particles into certain groups.

Despite all of the above, most of the work focuses on a certain size range and works with one sampling method. Here, by "surface" microplastics, we define the particles from the upper 2 to 3 dm layer of the sea water, including those floating directly at the sea surface, that are collected with manta or neuston nets with the mesh size $300-330 \mu$ m, and by "subsurface" microplastics we define particles collected from 3 to 5 m depth below the sea surface with a pump with $100 \mu m$ filters. One of the first studies using parallel sampling of surface and subsurface microplastics was a study of Arctic waters [10]. This study used parallel sampling with a manta net and sampling from a flow-through pumping system. Firstly, a difference in abundance for surface and subsurface plastic was found. Secondly, no other differences, such as size or polymer composition, were shown. The joint use of parallel selection by two different methods has been further developed in Setälä et al. [15]. In this paper, the differences between these methods are described in detail. The paper also describes the difference in the data obtained, and that they can complement each other. The first study of the South China Sea was also carried out using these two approaches [16]. The concentrations were determined; the contribution of the coastal runoff was estimated. The presence of more buoyant polymer particles in the open waters of the sea is also shown. Our scientific group continued similar studies and also applied parallel sampling in the waters of the seas of the Siberian Arctic [17]. The difference in concentrations of surface and subsurface microplastics was also shown. Larger particles were more common in surface microplastics samples than in subsurface microplastics samples.

This approach makes it possible to obtain more detailed data on the qualitative and quantitative distribution of particles of different morphology and sizes in the horizontal and vertical directions. More detailed data can further help simplify the search for sources of plastic pollution and help build models of the distribution of plastic particles. In this case, great attention should be paid to the identification of potential plastic particles obtained. At the moment, there are several main approaches to particle identification: visual analysis, identification of only 10% [10] or 50% [18] of a sample of all potential particles using spectroscopy, and complete identification of particles using spectroscopy methods. Visual analysis compares favorably with its speed of operation and the absence of the need for special equipment; while the probability of false determination is high, there is no information about the polymer composition, and in general there is an overestimation of concentrations. The latest AMAP guidelines [19] talk about the requirement for identification of all found particles (for particles > 300 μ m), as this provides more complete data on the nature of the polymers and more accurate concentrations. Fourier transform infrared (FT-IR) spectroscopy is a classic method for identifying polymers by a special region of the spectrum called "fingerprints". This methodology is considered the most reliable by the latest recommendations for analysis and guidelines [9].

In this work, we simultaneously investigated particles from the surface and subsurface (3–5 m) layers, and also compared the visual initial assessment of particles with the further identification of all potential plastic particles using FT-IR spectroscopy. The goal of this work was to compare the features of the MPs inhabiting on the sea surface and several meters deeper, as well as to study an influence of oceanographic conditions on the distribution of surface MPs on the example of the open ocean waters of the Central Atlantic.

2. Materials and Methods

2.1. Microplastics Sampling

Floating plastic debris were sampled during the 79th research cruise of the R/V Akademik Mstislav Keldysh in December 2019 in the central part of the Atlantic Ocean (Figure 1). Samples of floating particles were collected from surface water using a neuston net and from subsurface water using a ship-board underway pump-through system. Subsurface samples were collected underway, and the points on the map show the middle positions between start and end of the water pumping. A total of 38 subsurface water samples and 7 surface samples were collected (Figure 1). During net towing, the relevant vessel characteristics, including speed and GPS coordinates, were recorded. Physical characteristics of surface water (temperature, salinity) and air (temperature, wind speed, atmospheric pressure) were continuously measured along the ship track.



Figure 1. Station map for surface (middle) and subsurface (right) samples.

Surface water samples were collected using a neuston net with mesh size of 330 μ m and a mouth opening of 40 cm \times 60 cm. The net was towed along a straight line for 30 min per sample at an average speed of 2–3 knots. Only half of the mouth of the net was immersed in water due to the floats. Despite the fixed duration of the towing time intervals, towing distances varied by approximately 5% due to occasional instabilities in the vessel speed and impact of sea surface currents. The amount of filtered water was equal to $220 \pm 11 \text{ m}^3$ according to the GPS track of the vessel. After towing, the net was rinsed outside the vessel with a deck hose and returned to the ship deck. The cod-end was removed and taken to the vessel laboratory, where it was rinsed, and the volume reduced into a metal sieve with a mesh size of 1 mm. The collected particles (1–5 mm) were visually estimated and stored in hermetically closed centrifuge tubes until the analysis in the onshore laboratory.

Subsurface seawater was collected by a ship-board underway pump-through system with an intake located at a depth of 3 m on the right side of the vessel. In order to perform microplastics sampling, flowing subsurface seawater was passed under pressure through two stainless steel meshes (1.5 mm and 100 μ m pore size) within the filtration system, which consisted of two sequentially established first step water appliance protective systems and food grade PVC pipes. Two flow meters in the system provided accurate registration of water volume for each sample, which varied from 1.2 to 7.8 m³ per sample. After every sampling period, collected material was rinsed from the filtration system and filtered through stainless-steel fabric mesh filters (\emptyset 25 mm, pore size 50µm) using a filter holder attached directly to the sampling system. This was carried out to avoid air pollution. After that, the metal fabric-mesh filter was placed in plastic centrifuge tubes pre-washed with Milli-Q water. A total of 20 samples were visually analyzed on board [20], while 18 tubes were stored until the analysis in the onshore clean laboratory. In this study, surface and subsurface samples were collected in parallel, but if the surface samples were collected with a net during 30 min, the subsurface samples were collected continually, with approximately 12 h pumping minimal sufficient volume of water for one sample (>1 m^3). In the laboratory, samples were processed to remove organics using an optimized protocol with 10% KOH in the same tubes where the metal filters were stored. The treated samples were filtered from a metal filter onto 47 mm GF/A paper with a pore size of 1.6 μ m. The paper filter with the material was immediately transferred to a Petri dish and covered for drying and further analysis.

2.2. Contamination Control

Field and processing blank controls were performed to check possible samples contamination (according to [21] for surface samples and [22] for subsurface samples). Two cellulose fibers were found in the blanks that did not affect the results, as only synthetic fibers were included in the MPs assessment. In our study, all plastic objects that came into contact with the samples were controlled for potential contamination. This was carried out to subtract particles that could carry contamination from the sampling or analysis system itself. In this particular case, no PVC particles of the same color and similar morphology were found in our samples and blanks.

2.3. Microplastics Identification

All surface samples and 18 subsurface samples (as well as procedural and field blanks) were analyzed using a combination of visual inspection and chemical identification of polymeric composition via spectroscopy methods. Subsurface samples on GF/A filter papers were visually examined under a dissecting microscope Nikon SMZ745T fitted with an Infinity 1-3C camera and the associated Infinity Analyze software. Photographs of all potential microplastics were recorded, size (mm) and surface area (mm²) were measured using Image software. Identification of chemical composition of the items was made using a Fourier Transform Infrared spectroscopy (FT-IR) analysis on PerkinElmer Spotlight 400 FTIR, Frontier ATR for surface samples, and transmission micro-FTIR with DCC for subsurface samples. Due to the difference in particle sizes, different configurations of FT-IR were used: ATR or μ FTIR, with the same software and database of polymer spectra. μ FTIR cannot be used for big particles and with ATR it is possible to work with particles which can be transferred to the instrument without microscope. All spectra were manually inspected to ensure that the library matches were acceptable. All particles from surface samples were weighed (mg). The weight of the subsurface microplastic was estimated based on the density of the polymer and the volume of each particle. It was assumed that the fibers were cylinders of visible diameter. The thickness of the fragments was indirectly estimated by comparison with the dimensions of neighboring fibers (according to the procedure described in [17]).

3. Results

3.1. Surface Samples

In total, 71 potential plastic items were found under visual analysis (0–26 items per station, Figure 2). Only 56 of 71 potential plastic items (79% from visual analysis, Figure 2) were confirmed as plastic, where 43 items were microplastics (1–5 mm) and 13-mesoplastic (5–25 mm). Non-plastic particles were identified as cellulose and wood. The average size of MPs items was 2.8 mm \times 2.1 mm (Table 1). The polymer composition of items found at the surface included only polymers with density lower than seawater density and was represented mainly by PE (62%) and PP (29%) (Figure 3).

Abundance of MPs at the seven studied stations varied from 0 to 0.07 items/m³ (0–18 MPs in one transect). Three stations, 6561, 6562, and 6563 in the equatorial Atlantic (10° N– 10° S), were free of MPs; a low abundance of MPs was found at station 6551, around 23° N, 0.005 items/m³; and the maximum MPs abundance was observed south of 20° S, stations 6563, 6566 and 6573, and amounted to 0.04–0.07 items/m³ (Figure 4). Weight concentration of MPs varied between 0 and 0.5 mg/m³.



Figure 2. Number of items found by visual identification and identification using FT-IR for surface (**left**) and subsurface (**right**).

Table 1. Microplastics characteristics for different sampling methods and size fractions.

Sample Type, Size, mm	Abundance, Average (Min–Max), Items/m ³	Weight Concentration, Average (Min–Max), µm/m ³	N of Stations, MPs Found/Total	Average Item Size, mm	N of Polymer Types
Surf, 1–5	0.026 (0-0.073)	154.3 (0–503)	4/7	2.8 imes2.1	2
Subsurf, 1–5	0.17 (0-0.88)	4.3 (0–17.5)	9/17	2.8 imes 0.3	7
Subsurf, 0.3–1	0.44 (0-0.88)	1.9 (0-8.8)	12/17	0.55 imes 0.2	9
Subsurf, 0.1–0.3	0.12 (0-0.45)	0.06 (0-0.67)	4/17	0.25 imes 0.1	6
Subsurf, 0.1–5	0.78 (0-2.4)	7.0 (0–26)	15/17	1.1 imes 0.2	12



Figure 3. Surface square (**left**) and polymer composition (**right**) of found microplastics ("square" refers to the surface area of a particle).



Figure 4. Microplastics concentrations at different stations.

Mesoplastic items were found at three southern stations only: one, four and seven particles at stations 6563, 6566 and 6573, respectively. The maximum total plastic pollution was observed on the southernmost studied station and accounted for the total abundance of plastic—0.1 items/m³ and mass concentration—0.96 mg/m³.

3.2. Subsurface Samples

In total, 92 potential plastic items were found under visual analysis (0–12 per station, Figure 2) and 42 (46%) were confirmed as plastic, where 40 items were microplastics (0.1–5 mm) and 2 mesoplastic (5–25 mm). The average size of MPs was 1.1 mm \times 0.2 mm (Table 1). The polymer composition of the subsurface microplastics particles is more diverse than for surface samples and includes polymer particles with a density higher than that of sea water (Figure 3). Fragments amounted to 87.5% of found items (Figure 4) that are not typical for subsurface microplastics [10,17,22].

The abundance of MPs at 17 studied stations varied from 0 to 2.4 items/m³ (0–5 MPs in one transect). The weight concentration of MPs varied between 0 and 25 μ g/m³. The

highest MPs abundance and weight concentration in the subsurface layer in the Central Atlantic were found at stations 7908, 7910, 7913, 7916, 7917, from 1° S to 18° N (Figure 4).

The detailed data for different size fractions according to AMAP recommendations [19] are given in Table 1.

4. Discussion

4.1. Visual vs. FT-IR Analysis

The obtained data of visual analysis and analysis using FT-IR confirm the assumption of the overestimation of concentrations in visual analysis (Figure 2). After identification using spectroscopy, false positive (2 stations) to 15–250% overestimation (3 stations) was found for surface samples. Only at one station did results of visual and FTIR analyses coincide. Visual identification gave the wrong result in 83% of surface samples. For subsurface samples, one false positive and 20–300% overestimation at 14 stations (117% average) were found. Only at one station did results of visual and FTIR analyses coincide. Subsurface MPs concentrations obtained on board by visual analysis [20] were on average two times higher than for samples with FTIR analysis. Visual identification gave significantly less accurate result for 94% of subsurface samples. In this work, the found particles were mainly represented by fragments that are easier to correctly identify. When identifying the fibers, the error in the determination was maximum. Based on this, we do not recommend using the visual analysis method for fibers. Where possible, full identification of all potential plastic particles using IR spectroscopy should be applied. In the absence of the possibility of such an analysis, we recommend that the concentrations of fragments and fibers be given separately, since the accuracy of determining fragments is higher than that of fibers.

4.2. Characteristics of MPs Particles

MPs sampled at the sea surface and at 3–5 m depth are often considered as a single group—MPs in the surface water—with a difference only in the size range according to the method used. Differences in abundance, weight concentrations and particle morphology were noticed for surface and subsurface MPs in [10,22], with a clear tendency that subsurface microplastic abundance appears to be higher than that of the surface microplastics. Here, we found, in addition, other differences between surface microplastics and subsurface ones. First, size differences present even within the same size ranges, 1–5 mm (Table 1). Items are 3D and if the longest dimension has the same average for both surface and subsurface particles, the second dimension is seven times smaller for subsurface particles; that holds true for the third one, too. This results in differences in particle surface area up to two orders of magnitude (Figure 3). According to the data, larger particles of a certain density tend to remain on the sea surface, while smaller ones would be suspended in the water column. MPs below a certain size no longer have their own vertical velocity and become passive tracers in the water column, which can be transported by ocean currents until their properties are altered by biota [22]. The identification of all found particles revealed the second important difference, which is a polymer composition. In the surface layer, only polymers with positive buoyancy were observed, while a more diverse composition was associated with the small-sized subsurface MPs. Biofouling and aggregation with suspended organic matter contributed to the buoyancy of microplastics inversely proportional to their size, making their initial density irrelevant. Based on these findings, it can be hypothesized that surface and subsurface microplastics are, in fact, two different groups of microplastic particles that are worth studying individually, as they do not behave similarly in the open ocean and could have different sources.

4.3. Role of Hydrophysics in the Spatial Distribution of MPs

The highest concentration of MPs in the surface and subsurface layers was found in the different parts of the studied area: at 23–33° S and 18° N–1° S for the surface and subsurface MPs, respectively (Figure 4). It is well known that surface MPs accumulation zones occur in subtropical ocean gyres [12,23]. Stations with the maximum abundance of MPs in the surface layer in this study (St. 6563, 6566, 6573) were close to the South Atlantic subtropical gyre (Figure 4). Floating larger plastic particles (5–30 mm) were found only at the same three southern stations. This coincides with modern concepts about the nature of the micro- and macroplastics distribution on the ocean surface. For these items, ocean currents and wind are the main driving factors. All found particles had positive buoyancy. The garbage island in the South Atlantic subtropical gyre could be the source of these floating MPs. As the distance from the coast for most stations was more than 200 km, it is unlikely that coastal sources can be the main source of MPs in the Central Atlantic.

The distribution of subsurface MPs was more uniform, without such pronounced zones of absence and maximum abundance of MPs as for the surface layer (Figure 4). No high MPs abundance in subsurface water at stations near the South Atlantic subtropical gyre was observed. Higher concentrations were found between Cabo Verde islands and Africa and on the equator, which are, according to hydrophysical data, the upwelling areas, where the equatorial divergence zone and Canary upwelling are located [24]. Upwelling zones seem to be important factors for increasing the concentration of subsurface MPs, but not for long-term accumulation there due to the strong seasonal variability of their dynamics. The importance of mesoscale convective flows for subsurface MPs' distribution was earlier shown in [25].

Summarizing the results of this work and our previous studies [17,22], we would like to highlight the difference in the fate of surface (0–20 cm depth) and subsurface (3–5 m) MPs in ocean water.

Surface MPs:

- distribution is sporadic (found at 25–57% of stations);
- items with positive buoyancy, mainly (usually three types: PE, PP, EPS);
- lower abundance but higher weight concentration (in comparison with the same size fraction for subsurface).

Subsurface MPs:

- more uniform distribution (up to 90% of stations);
- items with both negative and positive buoyancy (up to 18 different types);
- smaller and lighter particles, even in the same size range.

5. Conclusions

MPs inhabiting the subsurface waters (about 3–5 m depth) have buoyancy close to neutral and appear to be suspended in the surface mixed layer and are readily transported from the sources to the distant regions by ocean currents. Subsurface turbulence is probably the main process that maintains the MPs in the near-surface part of the water column. In contrast, the surface MPs (which are captured by sampling with surface nets from the upper 10–20 cm layer) have positive buoyancy and their spatial distribution is significantly influenced by water dynamics, wind and waves, which lead to a more scattered distribution on the ocean surface. It was shown that surface and subsurface microplastics differ significantly in a number of properties and, apparently, they should be considered as two independent groups that may have different sources, and their distribution is driven by different hydrophysical processes. Thus, MPs data collected using both methods simultaneously could provide additional information about MPs fate in the ocean. The identification of all potential plastic particles using spectroscopy is essential for the correct determination of MPs pollution level.

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