



Microplastics distribution in bottom sediments of the Baltic Sea Proper



Irina Chubarenko ^{a,*}, Elena Esiukova ^a, Mikhail Zobkov ^b, Igor Isachenko ^a

^a Shirshov Institute of Oceanology, Russian Academy of Sciences, 36, Nahimovskiy prospekt, Moscow 117997, Russia

^b Northern Water Problems Institute of the Karelian Research Centre of the Russian Academy of Sciences, 50 A. Nevskogo prospekt, Petrozavodsk, Karelia 185030, Russia

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ABSTRACT

An abundance of microplastics particles (0.2–5 mm, MPs) in bottom sediments is analyzed based on 53 samples (3 to 215 m deep) obtained in 8 cruises of research vessels across the Baltic Sea Proper in March–October 2015–2016. MPs content varied between stations from 103 up to 10,179 items kg⁻¹ d.w., with the bulk mean of 863 ± 1371 items kg⁻¹ d.w., showing a statistically significant increase with water depth. As many as 74.5% of MPs are of fibrous shape, followed by films (19.8%) and fragments (5.7%). The distributions of fibres, fragments, films, and different types of natural bottom sediments are significantly different, highlighting the specific behaviour of each of these kinds of bottom deposits. A statistically significant correlation between water depth and fibres content is found. Based on the analysis of oceanographic factors and sedimentological principles, an erosion/transition/accumulation pattern for fibres in the Baltic Sea Proper is outlined. Fibres can be considered as a specific type of “synthetic sediment”, while principles of distribution of other MPs are not yet certain.

1. Introduction

Microplastics (plastic particles <5 mm, MPs (Arthur et al., 2009; Frias and Roisin, 2019)) are found worldwide in marine environments, including seawater, beach, and seafloor sediments (Ivar do Sul and Costa, 2014; Van Cauwenbergh et al., 2015; Harris, 2020 among many others). Marine sediments are often the most contaminated, having MPs content orders of magnitude larger than the seawater (e.g., Lassen et al., 2015; Booth et al., 2017; Peng et al., 2018). This is the case also for the Baltic Sea: e.g., in the size range of 0.2–5 mm, about 0.01–0.08 MPs items per litre are reported for the water column (Zobkov et al., 2019), while seafloor deposits have on average of about 860 items per kg d.w. (Esiukova et al., 2020), which is about 1200 items per dm³ (litre). A similar contamination level of seafloor deposits is found in other regions, for example, about 900 items per kg d.w. (or about 1200 items per litre; size range 0.063–5 mm) were reported by Alomar et al. (2016) in coastal shallow sediments in the Mediterranean Sea. As many as 44–3463.71 items per litre (42–6595 items kg⁻¹, size range from a few microns to 5 mm) were found by Bergmann et al. (2017) in Arctic sediments. Peng et al. (2018) reported from 200 to 2200 items per litre (270–6200 items kg⁻¹, size range from 20 µm to 5 mm) in hadal sediments of the Mariana Trench. Overall, higher contamination of coastal sediments reflects the contribution of the main sources of marine plastics, while deep-water sediments are suggested to be the ultimate sink of

finer MPs (Woodall et al., 2014).

In both shallow and deeper areas, the authors highlight a very inhomogeneous, patchy picture of MPs distribution (e.g., Claessens et al., 2011; Van Cauwenbergh et al., 2013; Woodall et al., 2014; Lusher, 2015; Bergmann et al., 2017; Pagter et al., 2020). Several studies discuss probable driving mechanisms, relating the observed distributions to either anthropogenic pressure (e.g., the proximity of sources, touristic/fishing activity, etc.: Browne et al., 2011; Lassen et al., 2015; Willis et al., 2017a, 2017b; Peng et al., 2018), or natural factors like weather conditions, hydrodynamics, or sediment characteristics (Claessens et al., 2011; Van Cauwenbergh et al., 2013; Lassen et al., 2015; Duis and Coors, 2016; Bergmann et al., 2017; Zobkov and Esiukova, 2017a).

The relation of MPs abundance to the sediment grain size has not been confirmed up to now (Mathalon and Hill, 2014; Alomar et al., 2016; Dodson et al., 2020; Urban-Malinga et al., 2020; see also Harris, 2020 for the review), except for the tendency of fibres to deposit in organic-rich sediments of deep-sea areas (e.g., Haave et al., 2019; Zobkov et al., 2020). Still, sedimentation of both natural grains and MPs is driven by the same hydrodynamics, and a certain relation between the distributions of sediments and MPs is thus probable. However, it is not clear which parameters of the bed load and MPs correlate, and which parameters of external conditions can be used to predict MPs accumulation patterns. A general theoretical analysis of the MPs particles

* Corresponding author.

E-mail address: irina_chubarenko@mail.ru (I. Chubarenko).

motion under natural drivers is not yet possible (Chubarenko et al., 2018b; Khatmullina and Chubarenko, 2019), thus, further understanding of the logic behind the MPs accumulation pattern can be obtained only through deeper analysis of field data.

In this study, we analyze the contamination by MPs particles (from 0.2 to 5 mm) of the bottom sediments of the Baltic Sea (Proper) – a large semi-enclosed sea of the northern Atlantic Ocean, which receives its

waters from densely populated Europe. MPs abundance, spatial distribution, plastic types, particle shape and size distributions are examined on the base of 53 samples collected from 3 to 215 m deep (see also the related open-access data article (Esiukova et al., 2020)). Results of statistical testing of hypotheses of probable relationships between the abundance of MPs and the sampling depth, the hydromechanical zones, the sediment types and its characteristics (mean grain size, sorting,

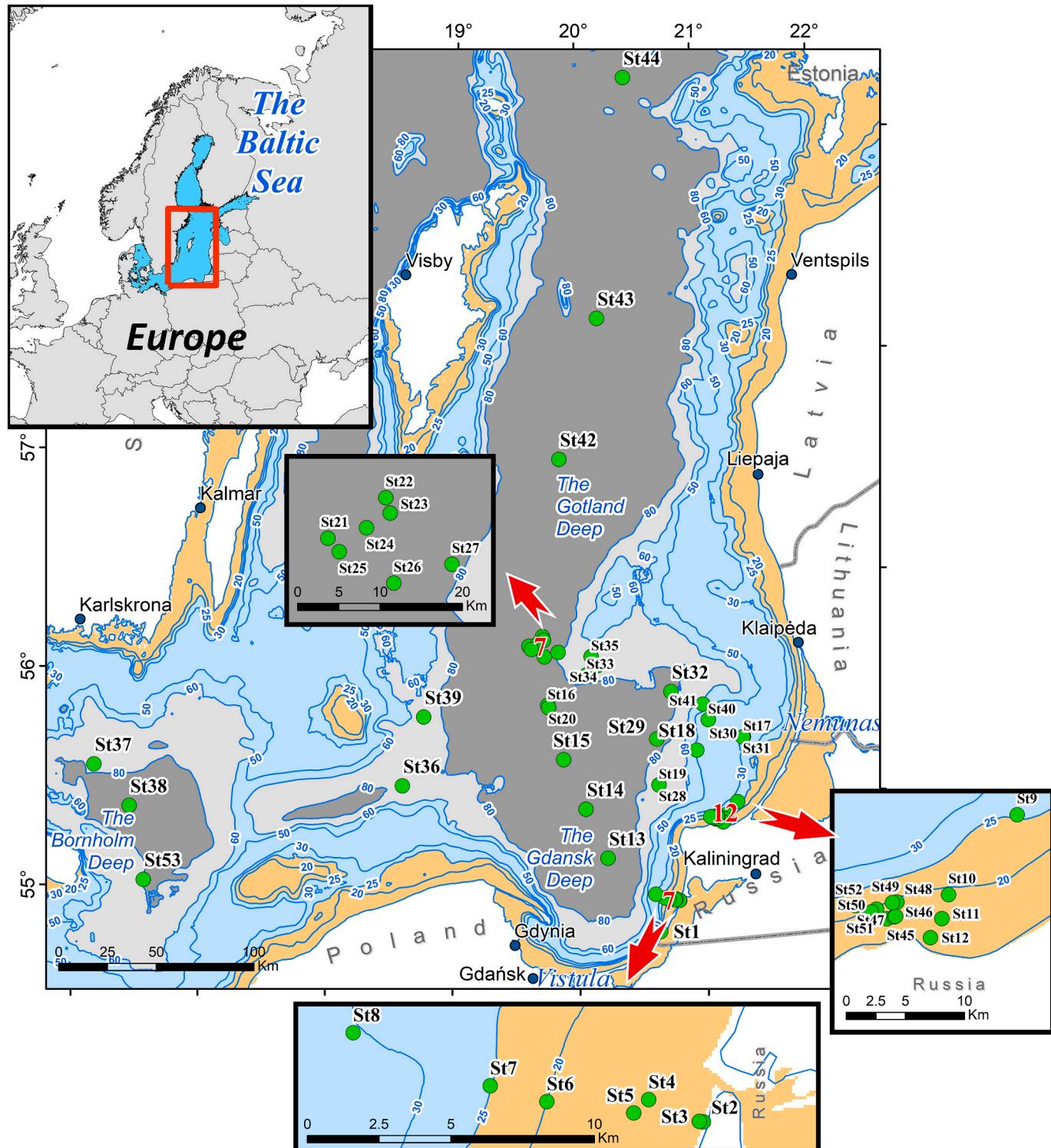


Fig. 1. Bathymetry of the Baltic Sea Proper and the sampling stations. Shading indicates approximate depth ranges of different hydrophysical zones: light brown – shallow coastal area (depth < 20 m), blue – bottom areas in contact with waters of the Cold Intermediate Layer (20–60 m), light pink – the pycnocline depth range (60–80 m), dark grey – deep area (> 80 m). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

particular size fractions) are discussed from the oceanographic viewpoint. Eventually, out of all the variety of MPs particles, synthetic fibres are suggested to compose a specific type of “artificial sediments”, whose distribution differs from those of other MPs particles and natural sediment types.

2. Materials and methods

2.1. Study area

The Baltic Sea is a large tideless semi-enclosed basin (volume more than 21,000 km³, surface area more than 377,000 km²) with very limited water exchange with the Atlantic Ocean, but high anthropogenic pressure from 9 European countries (Leppäranta and Myrberg, 2009). The Danish Straits, connecting the Baltic Sea with the North Sea, are narrow and shallow (mean depths of about 12–15 m) compared to the main basins of the Baltic Proper (Arkona Basin – 53 m, Bornholm Basin – 105 m, Gdańsk Basin - 114 m, Gotland Basin - 249 m (Leppäranta and Myrberg, 2009)), see Fig. 1. This is why the Baltic Sea is supposed to retain and accumulate a significant part of the incoming anthropogenic contamination (e.g., Lassen et al., 2015).

Transport and fate of a particle in sea/ocean waters are defined by oceanographic forcing (such as water currents of different nature, wave-induced motions, turbulence) and physical properties of the particle (density, size, shape, and their variations with time) (van Sebille et al., 2020). For deeper open-sea areas, the settling and transport of particles are of primary significance, while surface gravity waves are fundamentally important for the redistribution of bottom sediments in the coastal zone (Håkanson, 1977; Leppäranta and Myrberg, 2009). Thus, the main oceanographic factors in the Baltic Sea have to be presented for the analysis of MPs distribution.

The general structure of *water currents* in the Baltic Sea is shaped by stratification of the water column: there is a permanent pycno(halo)cline in the Baltic Proper at 60–80 m deep (slightly elevated due to the general weak circulation of estuarine nature), and a seasonal thermocline at the depth of 15–25 m (Leppäranta and Myrberg, 2009; Chubarenko and Stepanova, 2018). Following this density structure, the water circulation is generally three-layered, with the mean current speed in the open sea of an order of 10–50 cm s⁻¹ (maximum 60–70 cm s⁻¹) in the upper layers (0–20 m), 10–20 cm s⁻¹ (up to 40 cm s⁻¹) in intermediate layers (30–60 m), and less than 5–10 cm s⁻¹ below the pycnocline (Terziev et al., 1992). In near-bottom layers, the motions are less intense. However, direct measurements indicate that the maximum current speed near the bottom can reach up to 20–25 cm s⁻¹ even in the deepest depressions, while in coastal areas the maximum is up to 150–200 cm s⁻¹ (Terziev et al., 1992). Mean bottom currents are weaker at the levels of thermo- and haloclines (within the layers of ca. 20–25 m and 60–70 m deep, correspondingly). At the same time, the *turbulent kinetic energy* is higher in the entire upper layer and at the depth of 60–70 m (Lass et al., 2003). This suggests that both vertical and horizontal transport is significant for MPs distribution in sediments. Fingerprints of this sea-scale general hydrophysical structure will be found in the MPs distribution pattern.

The impact of *surface gravity waves* is crucial for the transport of particles in coastal waters. Physically, the “coastal zone” in this case denotes a range of depths where the passage of a surface wave induces “significant” water motion causing the relocation of fine sediments, while deeper seafloor sediments are no longer stirred by the surface wave motion (e.g., Håkanson, 1977; Leppäranta and Myrberg, 2009). The theoretical wave base (which delineates fine-sediment accumulation zones in deeper areas) is shown to be located at about 44 m deep in the Baltic Sea Proper (Håkanson, 1977), while zones of fine sediment erosion (where the bottom is generally hard, covered by sand, consolidated clays and/or rocks) are above 28.5 m. There is a *transition zone* in-between, where fine materials are re-deposited periodically, the spatial distribution pattern of sediments is patchy, and the bottom is covered by mixed

sediments (Håkanson and Bryhn, 2008). Obviously, the values are only general entire-basin estimates, which may vary depending on the particular meteorological conditions. Still, for the considered part of the Baltic Sea, it is generally assumed that, under typical storm conditions, surface waves generate near-bed oscillating motions and currents down to the depth of about 20 m, stronger stormy winds of 18–20 m/s – down to about 30 m (Terziev et al., 1992; Leppäranta and Myrberg, 2009), while under heavy winter storms with the longest wave fetch – down to 50–60 m deep (Chubarenko and Stepanova, 2017).

In accordance with oceanographic driving factors, the following sedimentological zones are proposed (Emelyanov, 2005) in the Baltic Proper: (1) depths down to 20 m on the coastal slope, with intense mechanical sorting of solid particles; (2) depth range 20–80 m, i.e., down to the halocline; (3) deeper than 80 m, the area of accumulation of fine material; and (4) deep areas with hard/rocky bottom. MPs abundance and properties in all the mentioned depth zones (summarized in Table 1), obtained by oceanographers and sedimentologists in the Baltic Sea research, will be analyzed below in order to grasp possible analogies.

2.2. Sampling

The samples of seafloor surface (2–7 cm) sediments were collected from 53 locations (from 3 m to 215 m deep, by drag or grab) in 8 cruises of research vessels in the Gotland, Gdańsk, and Bornholm Basins of the Baltic Sea in 2015–2016 (Fig. 1). The dates, regions, research vessels, types of samplers used are listed in the Table S1 (Supplement), and more detailed information on the sampling sites, depths, quality control procedures, masses of the samples, etc., can be found in the related open-access data article (Esiukova et al., 2020). The collected samples (presumably) represent contemporary sediments: in coastal zone (down to appr. 20–30 m) they are regularly re-located by storms, while in deeper areas of the Baltic Proper for the reported sedimentation rate of up to 0.5 cm/year (Mitchell et al., 2021) the time span may be several tens of years.

2.3. Microplastics extraction and identification

The upper 2–7 cm of sediments were collected from each location and stored in previously cleaned metallic buckets or cans. At all the steps of sample treatment and analysis, precautions were taken to avoid external plastic contamination. Sample preparation, analytical techniques, contamination control and quality analysis, classification methods, and μ -Raman spectroscopy verification for these sediment samples are described in detail in Esiukova et al. (2020).

MPs were extracted from the sediment samples using the method employed by Masura et al. (2015) with modifications by Zobkov and Esiukova (2017a, 2017b). To maximize the extraction rates, sediments with high content of fine fractions were washed through a sieve cascade (0.333 μ m, 174 μ m, 174 μ m) prior to the extraction to remove clayey mud fractions, which hampers the extraction process (Zobkov and Esiukova, 2017b). The extraction procedure included the following main steps (Zobkov and Esiukova, 2017a, 2017b): (1) density separation with $ZnCl_2$ solution (specific density 1.6 g cm⁻³), (2) filtering of supernatant with a filter funnel (mesh size of 174 μ m), (3) wet peroxide oxidation (H_2O_2 (30%) plus Fe(II) catalyst solution) on the water bath (up to 75 °C), (4) calcite fraction digestion with HCl solution (4.5%), (5) filtering with a filter funnel (174 μ m), (6) density separation ($ZnCl_2$) to detach oxidized organic matter, (7) filtering with a filter funnel (174 μ m), (8) MPs detection with a stereomicroscope, and (9) identification of particles materials with a Raman spectrometer. Examples of MPs isolated from the Baltic Sea marine sediments are shown in Fig. S1.

Among 53 samples, as many as 13 were contaminated by paraffin, oil, or fuel, making the extraction process even more laborious. Contamination of sediments by these hydrocarbons makes them sticky, which might have led to losing some MPs during the processing, thus

Table 1

Summary of oceanographic characteristics and sedimentological zones in the Baltic Sea Proper.

Oceanographic characteristics	Depth, m	Type of bottom sediments	Conditions for fine sediment	Near-bottom currents, cm s ⁻¹	Reference
Upper layer (warm season)	0–20				
Thermocline (warm season)	15–25	Coarse	Erosion	10–50 up to 200	Leppäranta and Myrberg, 2009
Erosion/transition border for fine sediments	28.5	Mixed	Transition		Terziev et al., 1992
Theoretical wave base 44 m	44				Håkanson and Bryhn, 2008
Cold intermediate layer	30–50	Fine	Accumulation	10–20 up to 40	Håkanson and Bryhn, 2008
Permanent pycno(halo)cline	60–80				Terziev et al., 1992
Deepwater	>80			5–10 up to 20–25	Leppäranta and Myrberg, 2009
					Terziev et al., 1992

underestimating the MPs content.

A stereomicroscope (Micromed MC2 Zoom Digital) with magnification from $\times 10$ to $\times 40$ was used to identify putative MPs particles according to the recommendations for microscopic determination by Norén (2007). A single operator performed the selection to exclude inter-operator variability. Raman Centaur U (LTD «Nano-ScanTechnology», Russia) spectrometer was applied to verify the results and to obtain the MPs spectra (Araujo et al., 2018; Zobkov et al., 2019). A more detailed description of the extraction and identification procedures is available in the data article by Esiukova et al. (2020).

2.4. Sediment grain size analysis

Sediment grain size distribution was obtained for all 53 samples. For coarse-grain samples, dry sieving on a sieve cascade with a step of 1/2 ϕ was performed. For fine-grained material, the Laser Diffraction Particle Size Analyser Shimadzu SALD 2300 was used. Sediment types are defined according to Folk (1954) and Wentworth (1922).

2.5. Statistical analysis

Several hypotheses were statistically tested to uncover possible links between the MPs distribution (both for total MPs content and separately for fibres, films, and fragments) and environmental parameters. A statistical significance level was set to $p = 0.01$.

(1) The hypothesis that the MPs content is associated with the sampling depth and the distance from the shore was examined using Pearson's correlation test. The depth and the distance increase in

a similar way in our data set (see Fig. 2), however, the dependence of MPs contamination on depth and distance is conditioned by different factors. The variation of contamination with depth would indicate natural transport and accumulation mechanisms, while the variation with the distance from the shore would highlight an influence of the contamination sources.

- (2) The ANOVA test was applied to assess the significance of the difference between the MPs content in different sediment types. The test was run between each pair of sediment type and fibres, films, and fragments content, respectively. The Coarse silt sediment type was not tested because only one such sample was available.
- (3) The parameters of the particular sediment sample (mean grain size (d_{50}), the sediment sorting, and the content of a particular grain-size fraction) were correlated with its contamination by MPs (the abundance of fragments, films, and fibres, separately). The Spearman's rank correlation was used to measure these correlations.
- (4) The ANOVA test was implemented to check the difference in MPs abundance between zones with different hydrophysical regimes. The latter were suggested by Emelyanov (2007) based on the sediment distribution in the Baltic Sea.

The tests were run using the SOFA statistics software (www.sofastatistics.com) and SciPyStats (<https://scipy.org/>).

3. Results

Overall, 6726 MPs items were extracted from 53 sediment samples.

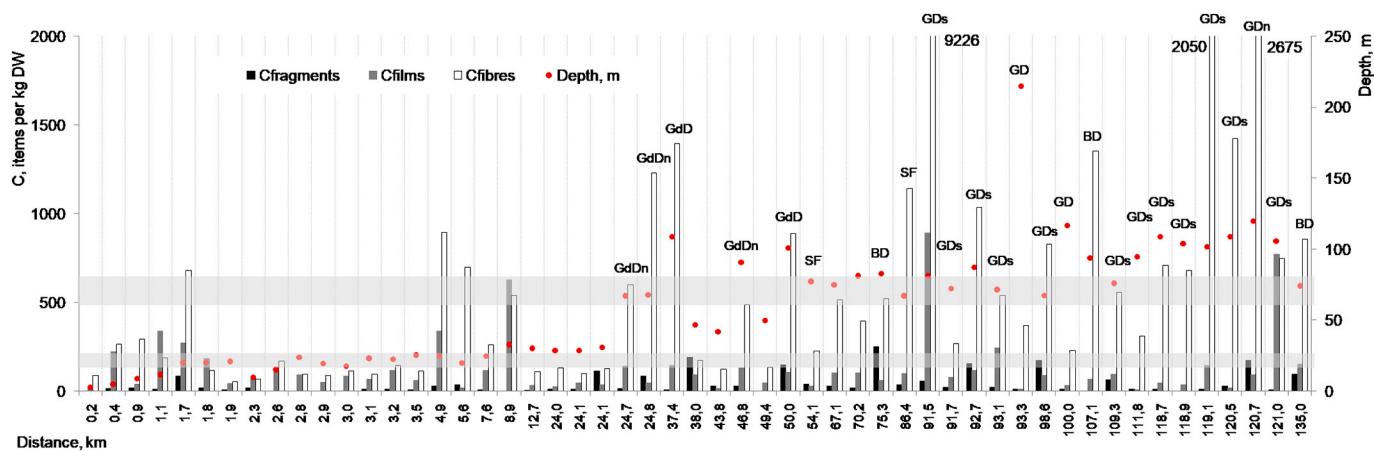


Fig. 2. The MPs (fragments, films, and fibres) content (items kg⁻¹ d.w.) versus the distance from the shore. The dots indicate the corresponding sampling depth. Horizontal grey bands show the ranges of depths of the seasonal thermocline (15–25 m) and the pycnocline (60–80 m). The deep sea areas are labeled: BD - Bornholm Deep; SF - Slupsk "River's foredelta"; GdD – Gdansk Deep; GdDn – Gdansk Deep (northern slope); GD – Gotland Deep; GDn - Gotland Deep (northern slope); GDs - Gotland Deep (southern slope).

The total MPs (0.2–5 mm) abundance significantly varied between the stations - from 103 up to 10,179 items kg^{-1} d.w., with the median of 519 \pm 179 items kg^{-1} d.w. (the 95% confidence interval of the median value without outliers), and the mean of 876 \pm 1427 items kg^{-1} d.w. A detailed presentation of the data set in a tabular form, methods of sampling, extraction, identification, as well as μ -Raman verification results are available from an open-access article (Esiukova et al., 2020).

3.1. Distribution of MPs by shape, size, colour, and polymer type

3.1.1. Distribution by shape

Among all the 6726 MPs identified, the fibres were the most commonly observed shape, accounting for 74.5% of the total number of particles, followed by films (19.8%), and fragments (5.7%), see Fig. S2. In the distribution by size, MPs in the size range of 1–2 mm was the most abundant, making up 36% of the total number of items (Figs. 3, S4). This is mostly due to the prevalence of fibres in this size range (27.5% of the total number of items, or 37% of the total number of fibres). For films, 0.5–1.0 mm fraction was the most abundant, accounting for 9.44% of the total number of items (or 48% of the total number of films) and fractions smaller than 0.5 mm were prevailing among fragments (2.39% of the total number of particles, or 42% of the total number of fragments) (Fig. S4). Items in the range of 0.5–1 mm make up 33.8%, in the size range of 2–5 mm - 22%, and a practically equal number of particles was found in the ranges of <0.5 mm and > 5 mm (4.3% and 4%, respectively).

3.1.2. Distribution by size

The distribution of large particles (> 5 mm; mainly of fibrous shape) was found to be patchy in coastal waters (down to 20 m deep) and decreased in deeper areas (Fig. 3). In the MPs size range (0.2–5 mm), an inhomogeneous increase in the abundance was confirmed down to 20–25 m depth, with patchy distribution farther down (see Section 3.2 for the detailed analysis).

3.1.3. Distribution by colour

Transparent MPs particles were the most abundant, accounting for 64.5% of the total number of particles, followed by blue (12.6%), brown (6.8%), black (4%), white (4%), red (3%), green (2.4%), and yellow particles (2.3%) (see Fig. S2).

Yellowing of MPs with depth (which might indicate the particle's age) was not found for films and fragments. For fibres, only the combined number of the roily, yellowish, and brownish particles increased noticeably in their fraction down to about 70 m deep, with inhomogeneous increase farther down. This may probably indicate that yellowing happens mainly due to exposure to UV radiation (i.e., under sunlight, at the surface), while in the water just general discolouration with time takes place.

3.1.4. Distribution by polymer type

From 130 specimens selected from different samples and different categories, 83% were directly identified as MPs, representing 21 polymer types (see Table 3 in (Esiukova et al., 2020)). Polyethylene (PE/HDPE/LDPE) (11.1%), Polypropylene (PP) (8.3%), Polymer blend (5.6%), Polyethylene terephthalate/Polyester (PET/PES) (4.6%), Polydimethylsiloxane (PDMS) (3.7%), Cellulose/Cellulose acetate (CE/CA) (3.7%), Polyvinyl chloride (PVC) (2.8%), and Synthetic rubber (1.9%) were the most abundant polymer types.

In total, floating polymers of families of PE and PP make up about 20% of all the MPs extracted from bottom sediments, indicating a high contribution of biofouling, aggregation, and other mechanisms capable to increase the integral MPs particle density with time spent in the marine environment (Morét-Ferguson et al., 2010; Chubarenko et al., 2016; Chubarenko et al., 2018b). Deeper investigation of biofouling and aggregation with biological or terrigenous material could not be answered in this study by methodical reasons: sampling by grab and the applied separation/extraction processes destroy any aggregates and remove biological material.

3.2. Distribution of MPs abundance in the Baltic Sea Proper

3.2.1. Dependency on the depth and the distance from the shore

The abundance of MPs (separately for fibres, films, and fragments) versus the distance to the closest shore (varied from 0.2 to 135 km in the data set) and the depth of sampling (from 3 to 215 m) are shown in Fig. 2. The data indicate that, in general, the closer to the shore - the lower is the contamination by MPs. Correspondingly, lower contamination is for smaller depths, while the highest contamination is observed in the deep sea areas, with the maximum value of 10,179 items kg^{-1} d.w. (all the MPs types) in the central part of the sea at the southern slope of the Gotland Deep.

The Pearson's correlation test showed a statistically significant relationship of fibres content with both depth ($p = 4.37 \times 10^{-6} \ll 0.01$; Pearson's $R = 0.584$; $df = 51$) and distance from the shore ($p = 9.76 \times 10^{-7} \ll 0.01$; Pearson's $R = 0.615$; $df = 51$). Thus, fibres content increases with both the depth and the distance from the shore, indicating the overwhelming prevalence of contribution of environmental transport mechanism over the impact of the source locations to the final distribution of the MPs contamination. For the other types of MPs (fragments and films), the correlation was statistically insignificant ($p \gg 0.01$). In particular, this highlights that the transport of fibres in the marine environment is substantially different from that of less mobile fragments and films.

Along with the general tendency of increasing contamination with the depth/distance, the patterns for fibres, films, and fragments are irregular and type-specific. These qualitative features are important for further analysis, so they are described here in more detail.

Across the coastal zone (up to 1.7 km off the coast, depths not

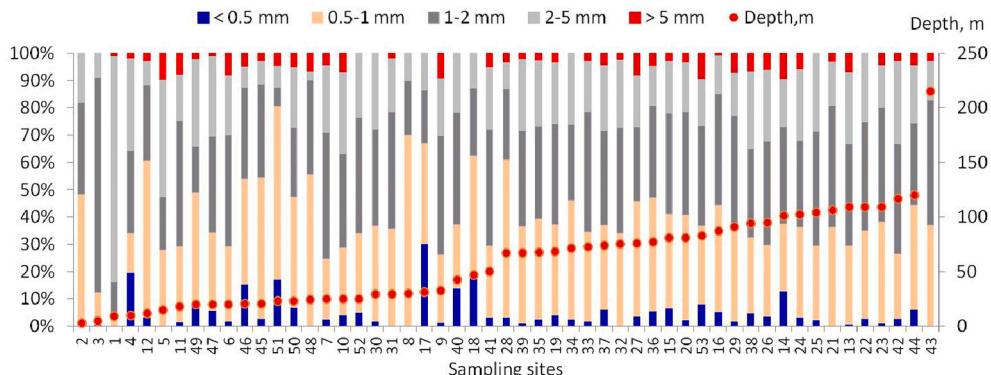


Fig. 3. Distribution of microplastics at all sampling sites by size range and depth.

exceeding 20 m), an order-of-magnitude off-shore increase in abundance of all MPs types is observed: from 0 items kg^{-1} d.w. for fragments/13 items kg^{-1} d.w. for films/91 items kg^{-1} d.w. for fibres at the shallow-most St. 2 (distance 200 m from the shore, depth 3 m) up to 85/273/682 items kg^{-1} d.w., correspondingly (at 1.7 km/20 m deep, St. 47). Farther off-shore, at distances of 1.9–3.5 km/water depths between 10 and 25 m, the presence of films and fibres remains at approximately the same level (43–116/55–172 items kg^{-1} d.w.), while fragments are practically absent (0–20 items kg^{-1} d.w.). At distances of 4.9–8.9 km (20–32 m deep) from the shore, a surge in the abundance of all MPs types is observed: up to 37/627/897 items kg^{-1} d.w. for fragments/films/fibres, correspondingly (St. 6/20 m deep, St. 9/32.6 m, St. 10/25 m).

At the distance of 12.7–24 km/depths of 29–31 m, a 20-fold increase in the number of fragments is found (from 6 to 115 items kg^{-1} d.w.), with the abundance of fibres (99–134 items kg^{-1} d.w.) and films (27–48 items kg^{-1} d.w.) similar to that found at 20–32 m deep but closer to the shore.

Farther off-shore, at distances more than 24 km/depths more than 67 m, a general picture of MPs contamination is patchy, but with obvious remarkable (order of magnitude) increase in the number of fibres, peaking at 2050/2675/9226 items kg^{-1} d.w. These peaks are found, correspondingly, at 102 m deep (St. 24) in the mud of the southern slope of the Gotland Deep, at 120 m deep (St. 44) in the mud of the northern slope of the Gotland Deep, and at 81 m deep (St. 20) in fine silty mud at the southern slope of the Gotland Deep (Fig. 2).

3.2.2. The distribution of MPs in light of general estuarine water circulation

There is no tendency of increase or decrease in MPs contamination towards the exit from the sea for deep-water stations (below pycnocline): bottom sediments of the Bornholm Basin show about the same contamination level as those in the Gdansk or the Gotland Basins (Table 2). This indicates that denser population and higher industrial activity in northern Europe compared to the western/eastern shores of the Baltic Proper do not show their fingerprints in MPs contamination of deep bottom sediments. This speaks in favour of intra-basin exchange and transport by currents as the main drivers of the MPs particles distribution in the bottom sediments of the Baltic Sea.

3.2.3. Coastal litter rim

A certain increase in MPs contamination is observed at distances of 0.4–1.7 km from the shore compared to the data for similar water depths (5–20 m) farther off-shore (Fig. 2). This might be the manifestation of the “litter rim” along the coast, where plastics are permanently re-located by shoaling (typical or stormy) surface waves (Chubarenko and Stepanova, 2017), see further discussion below. As for the identification of the potential MPs contamination sources, the considered data set is hardly representative: even though no specific contamination is indicated near cities, river mouths, or in certain populated coastal regions (Fig. 5), the conclusion on the presence/absence of the particular fingerprints requires more data in sea coastal zones.

3.3. MPs abundance versus sediment characteristics

The results of granulometric analysis of all the 53 samples are provided in a tabular form in Supplement S2. The sediment characteristics

are rather diverse in our data set, covering the range from “Sand, gravel, stones” to “Clayey mud”, with the maximum number of samples classified as sands (16 samples of “Mixed medium or coarse sand”, and 14 samples of “Fine sand”). The set includes almost equal quantities of moderately, poorly, and very poorly sorted sediments (16, 15, and 14 samples, correspondingly), with 7 moderately sorted and only 1 extremely poorly sorted sediment sample.

Correlations were analyzed between the MPs abundance and the mean particle size (d_{50}), sediment sorting, and the content of the particular sediment fractions. One sample was excluded from this analysis: at station St. 20 the abundance of MP fibres (9226 items kg^{-1} d.w.), films (893 items kg^{-1} d.w.), and the total amount of MPs particles (10,179 items kg^{-1} d.w.) exceed the mean values by more than five standard deviations.

A strong correlation in our data set was found between the number of fibres in the sample and the mean value of the sediment particles size expressed in the units of φ (Spearman's $R = 0.729$, $p = 9.25 \times 10^{-10} \ll 0.01$, $n = 52$). A positive R means that fibres are more abundant in fine sediments. There is also a moderate correlation between the fibre content and the standard deviation of the grain size (sorting of sediments) (Spearman's $R = 0.629$, $p = 5.82 \times 10^{-7} < 0.01$, $n = 52$), which means that more fibres concentrate in less sorted sediments.

A correlation was examined between the MPs content and the share of individual size fractions in the sample. A strong correlation was found between fibres content and share of two most fine-grained fractions (4–63 μm (Silt) and $< 4 \mu\text{m}$ (Clay)) in the sample (Spearman's $R = 0.764$, $p = 5.57 \times 10^{-11} \ll 0.01$, $n = 52$ and Spearman's $R = 0.769$, $p = 2.76 \times 10^{-11} \ll 0.01$, $n = 52$, correspondingly). Fig. 4 displays the abundance of MPs of different shapes versus the percentage of all fine sediment fractions ($< 63 \mu\text{m}$, i.e., Silt and Clay together): the Spearman's $R = 0.765$ and $p = 3.96 \times 10^{-11} \ll 0.01$ ($n = 52$) confirm the relation. There is also a statistically significant correlation between the fibres content and the share of sand fractions: Coarse sand - 1000–500 μm (Spearman's $R = -0.69$, $p = 1.49 \times 10^{-8} \ll 0.01$, $n = 52$) and Fine sand - 250–125 μm (Spearman's $R = -0.448$, $p = 8.55 \times 10^{-4} < 0.01$, $n = 52$). This means that fibres tend to accumulate in areas with fine sediments, while they are effectively washed out of sands.

3.4. MPs abundance versus general sediments type pattern

The distributions of both bottom sediments and MPs are known to be patchy. Still, the general distribution of sediment types is rather well known in the Baltic Sea. Thus, a correlation was analyzed between the

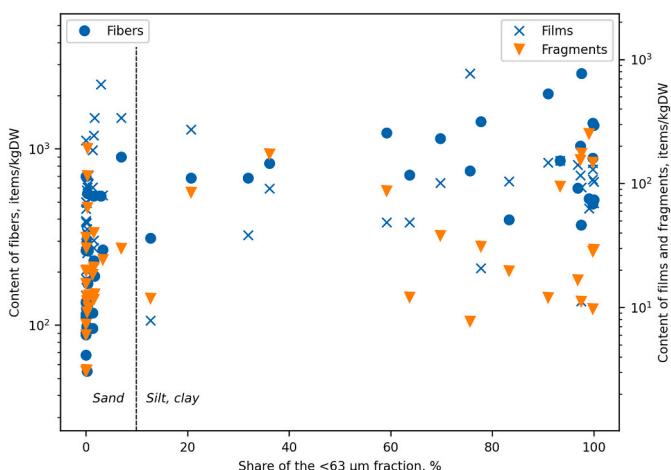


Fig. 4. The abundance of MPs of different shapes versus the percentage of silt and clay fractions ($< 63 \mu\text{m}$). The vertical line separates samples with a $< 63 \mu\text{m}$ -fraction content less than 10%, which in our data set consist mainly of sand of different fractions. Note different scales of axes for fibres and fragments/films.

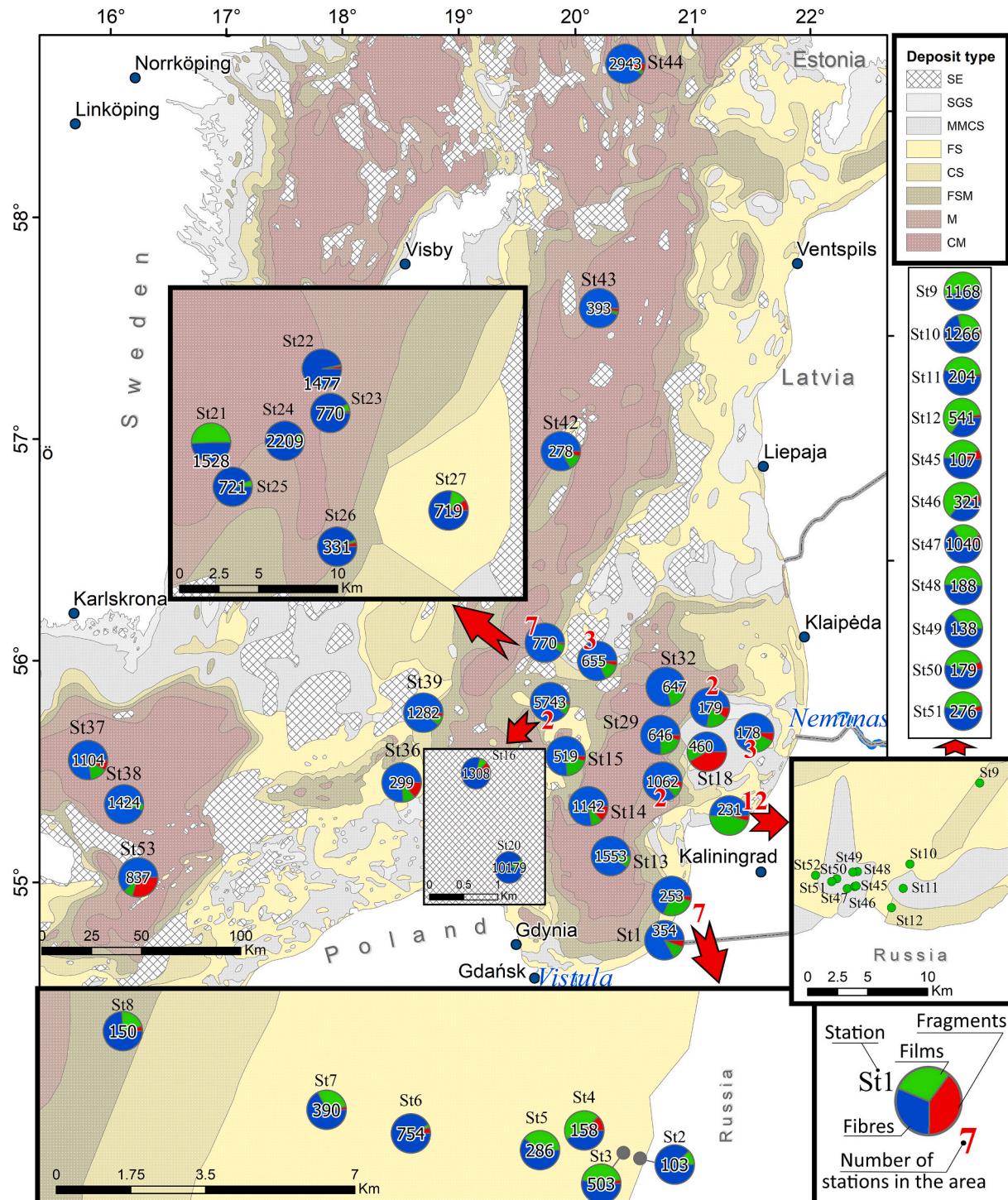
Table 2

MPs content (items kg^{-1} d.w.) in seafloor sediments below the pycnocline in different basins of the Baltic Sea Proper in the format “mean \pm SD (median)”.

	Fragments	Films	Fibres	Total
Gotland basin, >60 m	51 \pm 61 (23)	175 \pm 264 (92)	1425 \pm 2184 (729)	1650 \pm 2386 (947)
Gdansk basin, >60 m	49 \pm 51 (28)	112 \pm 33 (108)	787 \pm 394 (599)	947 \pm 404 (758)
Bornholm basin, > 50 m	116 \pm 127 (95)	95 \pm 50 (69)	911 \pm 419 (857)	1122 \pm 294 (1104)

MPs abundance and types of sediments at the sampling location. To allow for direct visual comparison, the map in Fig. 5 shows the detailed spatial distribution of MPs abundance together with the sediment type distribution compiled by Emelyanov (2007). Generally, sands are found along the shores down to the depths of 15–20 m, with very fine sands farther down to about 40 m. Deeper, at depths of 40–80 m, sedimentary material is practically not deposited and transits, sinking to the bottom of central depressions. At the level of the pycno(halo)cline (60–70–80

m), detrital clay is deposited, while below 80 m, sediments are the fine-dispersed river-borne material. Along with the general tendency of finer sediments to accumulate in deeper areas, the patchiness at smaller scales is an inherent feature of the sediment type distribution. Moreover, our stations 16 and 20 (the last – with the highest MPs abundance in our study) drop onto the area with a hard bottom on the sediment type map, but we were able to take samples there, and the granulometry indicates mud / fine silty mud there. This illustrates high spatial&temporal



variability of the natural sediment distribution pattern, which has to be kept in mind as well.

ANOVA tests show a statistically significant difference between fibres and fragments content in different sediment types (see Supplement, Tables S2-S5). The difference was statistically significant between the coarsest sediment types ("Sand, Gravel, and Stones") and sands ("Mixed Medium and Coarse Sand") in case of fibres ($p = 9.7 \times 10^{-4} < 0.01$; $F = 16.7$; $df = 15$) and fragments ($p = 4.83 \times 10^{-3} < 0.01$; $F = 11.9$; $df = 12$). In case of fibres, the difference was also statistically significant ($p = 3.4 \times 10^{-4} < 0.01$; $F = 16.6$; $df = 28$) between the types "Mixed Medium and Coarse Sand" and "Fine Sand", as well as between "Fine Sand" and "Fine Silty Mud" with ANOVA parameters ($p = 3.8 \times 10^{-3} < 0.01$; $F = 11.0$; $df = 18$). The difference between "Fine Silty Mud", "Mud" and "Clayey Mud" sediments in terms of fibres content was statistically insignificant ($p > 0.01$). The difference was statistically insignificant for other types of MPs and sediment types ($p > 0.01$).

Thus, fibres content is different in sands of different types (coarse, medium, fine); however, within fine sediments (silt, mud), the difference is not observed. This indicates different conditions for fibres burying in sands and finer sediments. For fragments, the MPs content only in the coarse-grained sands was statistically significantly different from that in other sediment types, while the difference between the remaining sediment types was insignificant. This indicates that only rough hydrodynamic conditions can influence the content of hydrodynamically heavy fragments.

Given such complicated relations, analysis of the range of depths, where samples of the particular sediment type were collected (Fig. S5a), might be useful. Sands of different types (coarse, mixed, fine) and Mud are found over the largest depth spans. The variability of the abundance of MPs of a particular shape within the samples of the same sediment type (Fig. S5(b-d), Table S6) was also analyzed. The maximum variability in the total number of MPs (Fig. S5(b-d)) was found for "Fine Sand" and "Mud/Clayey Mud" types of sediment. For fragments and films, an elevated variability was also found for the coarsest sediment type ("Sand, Gravel, Stones"). These features highlight the multifactor character of the MPs deposition in sediments so their distribution cannot be related to the sediment type alone.

3.5. MPs abundance in different hydrophysical zones

The distribution of bottom deposits is closely related to hydrodynamics: sands are found in more energetic zones along the coast, while fine sediments accumulate in calm regions below the pycnocline (e.g., Emelyanov, 2007; Håkanson and Bryhn, 2008). Keeping in mind all the general information about the Baltic Sea waters and sediments (Section 2.1 and Table 1) and in order to disclose just general correlations, an ANOVA test was implemented to check the difference in MPs abundance between zones with different hydrophysical regimes suggested on the base of sediment distribution in the Baltic Sea (Emelyanov, 2007). The stations were divided into three groups: Zone 1 (depths down to 20 m) – with a strong influence of shoaling surface waves and coastal currents; Zone 2 (depths from 20 down to 80 m) – intermediate layers with moderate currents and (generally) small wave impact on bottom sediments; Zone 3 (deeper than 80 m) – below the permanent halocline.

The analysis shows that the difference in abundance of fibres between hydrophysical zones is statistically significant (ANOVA $p = 1.46 \times 10^{-4} \ll 0.01$; $df = 49$; $F = 8.3$), with statistically higher fibres content below the pycnocline (80 m). The difference was mainly due to Zone 3, which has significantly higher fibres content than other zones. When Zone 3 was excluded from the analysis, the difference between other zones became statistically insignificant (ANOVA $p = 0.14 > 0.01$; $df = 34$; $F = 2.1$). The difference in abundance of films (ANOVA $p = 0.95 > 0.01$; $df = 49$; $F = 0.11$) and fragments (ANOVA $p = 0.63 > 0.01$; $df = 42$; $F = 0.59$) between the zones was statistically insignificant.

Thus, the upper layer, the thermocline depth range, an intermediate layer seem to provide too hydrodynamically active conditions to let MPs

accumulate in the sediments of the Baltic Sea. Fibres are obviously accumulated below the pycnocline; however, our data set for films and fragments does not allow for the identification of their accumulation zones.

4. Synthesis and discussion

MPs particles are relatively light and small, thus easily transported across the environment and dispersed all over the planet. In large seas and oceans, MPs contamination is eventually not linked to the particular anthropogenic sources, and their distribution is merely prescribed by the oceanographic factors. The Baltic Sea is known by its severe mid-latitude wind/wave climate, and the reported above observations do not indicate any specific spatial tendency towards the contribution of a particular anthropogenic source(s). Thus, when seeking for the main drivers of the distribution of MPs in marine sediments, natural factors should be considered above all. Fig. 6 shows MPs content in seafloor sediments (down to 120 m deep, averaged over 10-m layers) together with the relevant information on hydrophysical conditions in the Baltic Proper (presented in Section 2.1 and Table 1): general structure of the water column (with density jumps at the thermocline and pycnocline levels), theoretical wave base for fine sediments (the depth, down to which water motions below the surface waves are able to re-locate fine sediments), an order of magnitude of near-bottom currents. The mean and median content of MPs (all forms) in different depth zones is summarized in Table 3.

Synthesis of the information obtained by statistical tests leads to the following key conclusions. Regarding the accumulation in seafloor deposits, MPs particles have such diverse properties that they cannot be related to any type of natural sediments. This is confirmed by other studies, for example, Alomar et al. (2016) did not find a clear trend between the grain size and MPs content in shallow coastal sediments in the Mediterranean. Urban-Malinga et al. (2020) confirmed the absence of this relationship in sediments of 12 beaches of the southern Baltic Sea. No relationship was found by Mathalon and Hill (2014) in intertidal sediments of Halifax Harbor, Nova Scotia. Dodson et al. (2020) concluded "that sediment composition does not control microplastic distribution" on some beaches of the USA and referred to several other studies where MP occurs independently of sediment type (see also (Harris, 2020) for the review). A probable explanation might be that MPs accumulate in sediments in various ways: they may settle freely as they are (Khatmullina and Isachenko, 2017); floating items may undergo heavy biofouling; MPs may stick to sediment grains or be captured in-between grains (e.g., during suspending/settling cycles in wave-induced mixing (Chubarenko et al., 2020)); MPs may be eaten and settle to the bottom together with fecal pellets; they may be suspended and re-located by near-bottom currents. One more reason for the discrepancy might be that MPs settling and deposition are influenced by particle size, shape, and density, which all vary in time (Chubarenko et al., 2016; Khatmullina and Isachenko, 2017; Bagaev et al., 2017a, 2017b).

Only fibres content shows some statistically significant correlations: with the depth and the grain size of the sediment, particularly with the share of the finest sediment fractions (clay and silt). For fragments and films, the correlations are statistically insignificant, while statistically significant is the difference between the fibres and fragments content in different sediment types. Moreover, while the fibres content is statistically different in sands of different fractions (coarse, medium, fine), the difference is not significant within fine sediment fractions (silt, clay). These facts speak in favour of a conclusion that different types of MPs (just like different types of sediments) have their own specific transport-and-accumulation behaviours. In particular, fibres are washed out of sands and settle in the calmest areas on the seafloor (disregarding natural sediments types there, but typically these are exactly fine-grained sediments). Thus, the strong correlation between the fibres content and the share of fine sediment fractions has its roots in their dependence

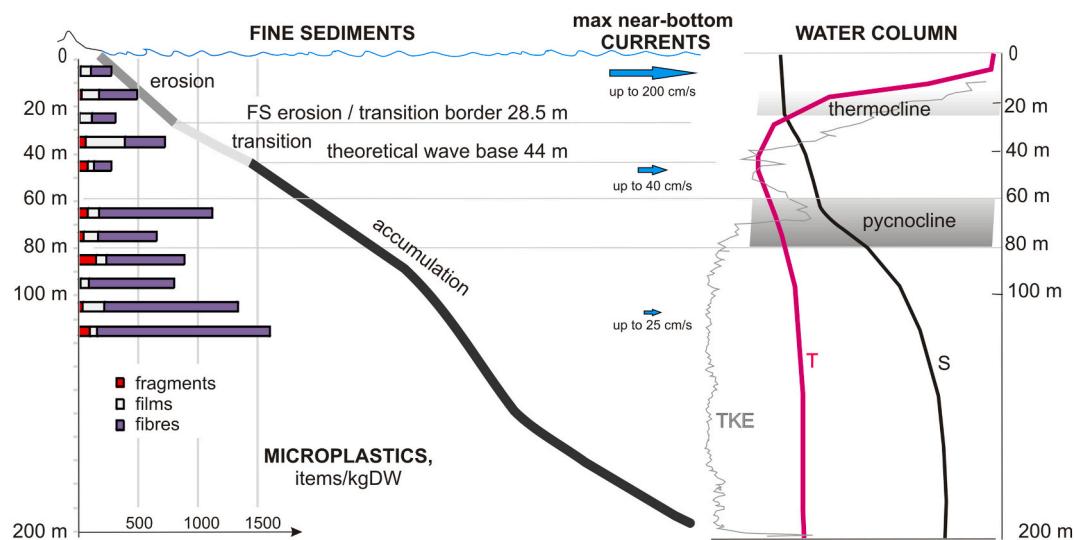


Fig. 6. Microplastics content (in items kg^{-1} d.w.) in bottom sediments of the Baltic Proper (averaged over a 10-m depth) versus the structure of the water column: examples of temperature and salinity profiles after (Leppäranta and Myrberg, 2009), averaged dissipation of turbulent kinetic energy in the Eastern Gotland Basin after (Lass et al., 2003; Reissmann et al., 2009) and near-bottom erosion/accumulation conditions for fine sediments after (Håkanson and Bryhn, 2008).

Table 3
Content of MPs (all forms) in sediments of hydrophysically different depth zones.

MPs, all forms	Mean \pm SD, items kg^{-1} d.w.	Median, items kg^{-1} d.w.
Coastal zone, 0-20 m	286 \pm 167	245
Thermocline depth, 20-25 m	409 \pm 350	276
Below thermocline, 25-30 m	483 \pm 504	186
Cold Intermediate Layer, 30-50 m	273 \pm 162	187
Pycnocline depth, 60-80 m	844 \pm 362	781
Below pycnocline, 80-215 m	1662 \pm 2305	1142

on the depth, i.e., on the intensity of hydrodynamic processes. A similar analysis for bottom sediments in Lake Onego (Zobkov et al., 2020) has also indicated an accumulation of fibres in the fraction of 10–50 μm (medium silt). This suggests that relatively light (material density typically up to 1.4 g/cm^3) and much elongated synthetic fibres of a size range of 0.2–5 mm are transported and accumulated in a marine/lacustrine environment similar in some way to the plate-shaped clay and quasi-spherical silt particles, with their material density of about 2.4 g/cm^3 and size below 0.063 mm. Laboratory experiments on the sinking behaviour of synthetic fibres (Khatmullina and Chubarenko, 2021) have demonstrated that their settling velocity is of the order of a few mm per second, i.e., of the same order as that for silt and clay particles.

It is known that particle-reactive pollutants such as PCBs, PAHs, or trace metals also accumulate in fine sediments, which is due to the increase of available surface area with decreasing sediment grain size. With relatively large MPs (size range 0.2–5 mm is considered here), the mechanism of particle-sediment interaction is different, and mainly driven by surface properties of the very plastic (not sediment) particle. Thus, in the considered case, there is MPs, which defines the distribution of the associated organics/fine sediment particles, but not the pollutant-laden sediment.

This way, the distribution of MPs particles can be understood using not just the direct relation to some sediment characteristics, but via the

general principles behind the sediments distributions developed by sedimentology. Since the abundance of fibres correlates with the share of the finest sediment fractions (and especially with clay ($<4 \mu\text{m}$): Spearman's $R = 0.769$, $p = 2.76 \times 10^{-11} \ll 0.01$, $n = 52$, see Section 3.2), the principles of the distribution of fine bottom sediments are of primary importance. Observations show (e.g., Håkanson and Bryhn, 2008) that for fine materials there is (i) an erosion zone in the most energetic area along the coast, where no apparent deposition of fine sediments is observed and only coarse sediments are found, (ii) the transition zone with mixed sediments, where all the materials are deposited/resuspended periodically under the action of currents and waves, and (iii) the accumulation zone, where fine materials are deposited continuously. For fibres, expecting such zoning and considering the variations in the fibres abundance (Section 3.1, Figs. 2, 6), one may suggest the accumulation zone below 60 m. Since the largest variability and the lowest abundance of fibres are found in “Mixed medium and coarse sands” (see Fig. S5a,b), which are located at depths down to 20–25 m, this gives an approximate location of the border between the erosion and transition zones for fibres.

Quite illuminating is the observation (Chubarenko et al., 2020) that minimum MPs contamination of sediments is observed at the wet shoreface, where wave uprush-and-backwash maintains permanent mixing and the hydrodynamic impact is the most energetic (Efimova

et al., 2018). For the southeastern Baltic Sea, the mean MPs content is (1) in the body and surface sediments of sandy beaches - about 110 items kg^{-1} d.w. (108 items kg^{-1} d.w. (Chubarenko et al., 2018a), 115 items kg^{-1} d.w. (Chubarenko et al., 2020)), (ii) on the wet beach face – about 50 items kg^{-1} d.w. (Chubarenko et al., 2020), and (iii) on the underwater slope down to the depth of about 10 m – 280 items kg^{-1} d.w. (this study), with evident increase farther down, see Fig. 2. A similar observation was reported by De Ruijter et al. (2019): in their study of MPs in beach and shallow marine sediments of the Samos Island (the Mediterranean Sea), the lowest contamination was recorded in the intertidal zone. On the other side, in the review paper by Harris (2020), the highest numbers of MP particles occur in low-energy depositional environments (muddy, low-energy estuaries, fjords, and lagoons are discussed).

Distributions of other types of MPs particles - films and fragments – differ from those of fibres and various sediment types. Our data set does not allow for the identification of the accumulation zones for them, however, from qualitative analyses (Section 3.1) one may suggest that fragments and films accumulate below 30 m. This way, their behaviour is special, differing from both easily movable fibres (which accumulate in the deepest areas) and heavy sands (accumulated along the shores).

5. Conclusions

The MPs (0.2–5 mm) content in 53 samples of the Baltic Sea bottom sediments was analyzed, showing that the MPs abundance increases with depth, with the maximum in the deepest sea part, and is formed by fibres by about 75%. Using a well comparable set of data stretching from the coastal zone to central bottom depressions, it is statistically confirmed that the deep sea is the ultimate end for synthetic fibres.

Total MPs content is not directly related to the type of sediment in the analyzed data set. Instead, the distribution and accumulation of different types of MPs (fibres, films, fragments in this study) are self-specific. This way, it might be profitable for transport and accumulation problems to subdivide “MPs as a whole” to specific classes, exhibiting more or less similar transport properties, distinguishable from those of any sediment or other MPs types.

Fibres are definitely one of such specific types of MPs, and their distribution in bottom deposits can be described in terms of erosion/transition/accumulation zones. This is inherent to fine sediment fractions (silt, clay), however, properties of their particles are somewhat different, so the accumulation pattern for fibres has specific features, and the borders between the zones are shifted deeper. For fragments and films, statistics do not indicate any similarity to either sediment or fibre distribution. Further investigations are required to disclose specific features of these (or other possible) types of “plastic types of sediments”. From this viewpoint one may suggest that larger plastic litter items have the same major drivers as marine sands: they both are abundant in sea coastal zones, sometimes re-suspended and re-located by stormy waves, and exhibit patchy distribution. Hydrodynamical and sedimentological approaches, as well as further laboratory experiments on MPs settling and erosion thresholds, are needed to understand which and how many “plastic sediment types” are required apart from fibres and macroplastics to grasp the main features of the observed contamination pattern.

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CRediT authorship contribution statement

ICh: Conceptualization; Writing- Original draft, Review & Editing, Funding acquisition.

EE: Data curation; Methodology; Writing - Original Draft.

MZ: Investigation, Formal analysis, Writing - Original Draft.

II: Formal analysis, Visualization, Writing - Original Draft.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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