Anthropogenic fibres in the Baltic Sea water column: Field data, laboratory and numerical testing of their motion

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HIGHLIGHTS
• A “selective” strategy is proposed: different types of MPs request different models.
• Fibres are the prevailing type of MPs in the Baltic Sea water column.
• Fibres behaviour in the sea: flow with currents, slow sinking, and delayed settling
• Sinking velocity and re-suspension threshold determine distribution of fibres in the sea.

GRAPHICAL ABSTRACT

ABSTRACT

Distribution of microplastics particles (MPs) in the water column is investigated on the base of 95 water samples collected from various depths in the Baltic Sea Proper in 2015–2016. Fibres are the prevalent type of MPs: 7% of the samples contained small films; about 40% had (presumably) paint flake, while 63% contained coloured fibres in concentrations from 0.07 to 2.6 items per litre. Near-surface and near-bottom layers (defined as one tenth of the local depth) have 3–5 times larger fibre concentrations than intermediate layers. Laboratory tests demonstrated that sinking behaviour of a small and flexible fibre can be complicated, with 4-fold difference in sinking velocity for various random fibres’ curvature during its free fall. Numerical tests on transport of fibres in the Baltic Sea Proper were performed using HIROMB reanalysis data (2007) for the horizontal velocity field and laboratory order-of-magnitude estimates for the sinking velocity of fibres. The model takes into account (i) motion of fibres together with currents, (ii) their very slow sinking, and (iii) their low re-suspension threshold. Sensitivity of the final distribution of fibres to variations of those parameters is examined. These experiments are the first step towards modelling of transport of fibres in marine environment and they seem to reproduce the main features of fibres distribution quite well.

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1. Introduction

Presence of plastic pollution is an emerging worldwide threat to the health of marine ecosystems. Small (<5 mm) fragments of artificial polymer objects are commonly termed as “microplastics” (Bergmann...
et al., 2015). Such objects have started to appear in the ocean since 1970th (Bergmann et al., 2015); they were first properly reported in 2000s (Derraik, 2002), and are nowadays found as far as in the Arctic (et al., 2015). Such objects have started to appear in the ocean since 1970th (Bergmann et al., 2015); they were for an important general conclusion: their transport quite a challenging problem. Transport of plastics, different physical mechanisms are of importance, and different time and length scales are of interest: for dense particles, the key parameters are the settling velocity and the near-bottom current velocity magnitude, while, say, for the buoyant particles/pieces - the surface currents direction, windage, and turbulence. Such a “selective” philosophy is quite a common practice at the beginning of investigation of any kind of a complex object, and in modelling as well. Although representing a simplified approach with several limitations, this method could give a valuable insight necessary for development of a more comprehensive model.

Almost every report on MPs research in marine environment shows presence of very small (length ~0.5–5 cm, diameters ~10–50 μm) fibres of anthropogenic origin: in bottom sediments (e.g., Mathalon and Hill, 2014), water body (e.g., Dubaish and Liebezeit, 2013; Lattin et al., 2004; Thompson et al., 2004), at water surface (e.g., Doyle et al., 2011), on beaches and coastlines (e.g., Browne et al., 2011), in the deepest oceanic depressions (e.g., Woodall et al., 2014), and in the Arctic ice (Obbard et al., 2014). Some studies (Browne et al., 2011) showed strong correlation between wastewater disposal areas and textile poly-mer fibres concentration. A recent study (Woodall et al., 2014) suggested that the deep-sea sediments are the main ‘sink’ or accumulation zone of the fibres in the ocean. The review paper (Ivar do Sul and Costa, 2014) reported that in the Atlantic Ocean fibres appeared in much higher concentrations in the sediments than in the water samples. A paper of (Desforges et al., 2014) represented the elevated concentrations of submerged MPs fibres in areas close to land-based sources. In the Baltic Sea, fibres are considered to be a more threatening pollutant than larger particles, especially for invertebrate communities (Setälä et al., 2016), however their sources and concentrations are poorly known. In coastal seawater, (Stolte et al., 2015) reported that off Warnemünde the concentration of fibres varied from 0.1 to 20 fibres per litre (55 μm mesh size); our measurements (this study) show fibre concentrations in coastal and open waters in the range from 0 to 2.7 fibres per litre.

In this paper an attempt is made to simulate with a basin-scale numerical model the transport of fibres, accounting for just their basic transport properties. Our original measurement data on concentrations of fibres in the Baltic Sea serve as a basis for the analysis and modelling.

## 2. Methods

### 2.1. Water sampling in the Baltic Sea proper

With the goal to investigate the presence and distribution of MPs in general and artificial fibres in particular, 95 water samples from various depths, near-surface and near-bottom layers in the water body were collected during six cruises in the Baltic Sea Proper, see Table S1 in Supplementary material for the list and Fig. 1 for the map.

Water samples were collected using standard technique of sampling by means of the 10-litre or 30-litre Niskin bottles. In the cruises of r/v NORD, water samples were taken in the coastal waters from the lowest horizon possible at locations where water depth ranges from 6 to 21 m. The 10-litre Niskin sampling bottle (0.8 m long) was attached to the rope with an anchor and lowered vertically, so that the distance between the anchor and the lower part of the bottle was about 0.7 m. This way, water samples were collected from the depths of 0.7–1.5 m above the bottom. During the ANS (“Akademik Nikolay Strakhov”) cruises the same procedure was carried out with the exception that the distance between the anchor and the lower part of the bottle was around 2 m. Surface samples were collected with the black plastic (poly-ethylene) bucket. During the cruises of “Professor Shitokman” PSH-131 and PSH-132, water samples were collected using Multi Water Sampler SlimLine 12 at different depths. Samples from the r/v NORD were transported to the laboratory and filtered there using 174 μm filters. In the other cruises, water was filtered through the square (7 × 7 cm) filters (174 μm) immediately on-board.

<table>
<thead>
<tr>
<th>Variables</th>
<th>Description</th>
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<tbody>
<tr>
<td>ρ&lt;sub&gt; fibre&lt;/sub&gt;</td>
<td>mean density of the fibre (kg/m&lt;sup&gt;3&lt;/sup&gt;)</td>
</tr>
<tr>
<td>ρ&lt;sub&gt; water&lt;/sub&gt;</td>
<td>mean density of the marine water (kg/m&lt;sup&gt;3&lt;/sup&gt;)</td>
</tr>
<tr>
<td>w&lt;sub&gt;s&lt;/sub&gt;</td>
<td>vertical settling velocity</td>
</tr>
<tr>
<td>ΔT</td>
<td>time step</td>
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<tr>
<td>A&lt;sub&gt; h&lt;/sub&gt;</td>
<td>coefficient of horizontal turbulent diffusivity of the particles</td>
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Conventionally, only upper size limit of 5 mm is set for MPs, while there is no requirement up to now for a “lower bound” in size (Arthur et al., 2009). As a practical matter, the mesh size of the neuston net used for sampling is commonly applied for the lower size limit. Even though fibres (diameter 10–30 μm) can potentially flow through the net, we shall formally declare the limits from 0.174 mm to 5 mm as the MPs size range in this study. For tiny fibres, almost any commonly used net is potentially leaky, and one can only keep in mind that it could result in a certain underestimation of fibres concentration.

The filters and all other equipment were rinsed thoroughly with tap water prior to sampling to minimize contamination, however no ‘blank water samples’ were taken for the contamination control. Each filter was folded twice after the filtering had been over, thoroughly sealed into an individual polyethylene bag with a string lock, and preserved at room temperature until examination in the land-based laboratory. In laboratory, the samples were examined under microscope immediately after recovery from the plastic bags. Torre et al. (2016) reported intense measures taken to avoid contamination in the laboratory conditions, which were necessary due to the long-lasting and multi-stage samples preparation. In our case, such measures were found superfluous, and we only recorded in protocols the colour of the ship deck, colours and material of the staff clothes, etc., and later examined the filtered particles regarding possible listed types of contamination. For fibres and films no contamination cases were registered; as for paint flakes, however, about 50% of them were green, the same colour as the ship deck and shipboards.

In all cases, the filters were visually examined under a microscope with 40× magnification. In comparison with bottom or beach sediment samples, the water-body samples in our case did not contain much filtrate to examine. The only residue on the filters, other than obviously artificially coloured objects, was fading traces of algae/plankton, hence the identification was not problematic. Following (Hidalgo-Ruz et al., 2012; Guide to Microplastic Identification, 2012), the anthropogenic microfibres were identified (i) by colour, (ii) by absence of visible cellular or organic structures, (iii) by comparison with published photographs of polymer fibres (e.g. Folkö, 2015; Desforges et al., 2013; Magnusson and Norén, 2011; Zhao et al., 2015; Torre et al., 2016), and (iv) by means of mechanical manipulations with a thin needle under microscope.

Identification protocol for fibres was similar to those described in (Qiu et al., 2016). Should a fibre be of uniform thickness, distinct colour, elastic material, or not fractionize under the metallic needle, it was considered a polymer fibre. In some cases, the UV-lamp was used to check for possible particle fluorescence, or hydrogen peroxide – to check whether the transparent fibre was inorganic. However, these methods appeared to be excessive for our analysis. Since our research is driven by interest to physical aspects of fibres’ transport in marine waters, we did not define the particular material the fibre was made from. However, in order to keep the possibility of further analysis, any questionable objects (transparent of dark fibres) were reported in Table S2 (in Supplementary material).

2.2. Laboratory testing

Hydrodynamics of sinking of a thin elongated flexible small object is far from being trivial and, to our knowledge, its motion cannot be properly described yet even under still laboratory conditions. In turbulent marine environment and under natural biofouling, the particular sinking rate and the fibre density excess will vary considerably in comparison to laboratory study, making precise laboratory measurements useless. The goal of the performed laboratory testing was (i) to observe the principal character of a fibre sinking and (ii) to estimate the range of
variations of the sinking velocity of the same very fibre under the same laboratory conditions. Sinking behaviour of synthetic fibres, taken from the field samples, was tested in a 50 ml glass vial filled with distilled water. In order to prevent any water motions in the vial due to heat exchange with the surroundings, the air temperature in the laboratory was kept possibly constant and the vial was filled 24 h in advance. Visual observations of settling behaviour were made through the magnifying glass installed on the holder. A particle was placed below the water surface with tweezers. After that, the time the particle took to sink 20 cm was measured with a stopwatch. Material density of fibres was measured by the titration method modified from ISO 1183-1 (2012) in zinc chloride solution (the procedure described in detail in (Khatmullina and Isachenko, 2016)). The density determination was conducted after the sinking experiments in order to avoid contamination of samples by zinc chloride.

2.3. Modelling tools

The velocity fields are of predominant importance for predicting of the pathways of any kind of floating objects in the Baltic Sea. In our study, the trajectories of the particles are calculated from the velocity fields data downloaded from Copernicus Marine environment monitoring service (CMEMS). This reanalysis product was obtained by the Swedish Meteorological and Hydrological Institute (SMHI) (Axell and Liu, 2016) and provided by the Baltic Marine Forecasting Centre (BALMFC). The reanalysis was carried out using HIROMB 2.0 model, which is based on primitive equations and uses Arakawa’s C-grid and z-coordinate (Funquist and Kleine, 2007). The reanalysis covers the Baltic Sea and the Danish Straits. The sea level in the model setup is prescribed at the lateral boundary in the western English Channel and along the Scotland-Norway boundary. Climatological monthly mean values of salinity and temperature are used at the boundary. A buoyancy-extended k-omega model was used for vertical mixing, with parameterizations for internal wave energy and Langmuir circulation (Ulauf et al., 2003). The river runoff is prescribed using daily means from the operational hydrological model of SMHI (about 500 rivers). The model was forced with the output from HIRLAM (High-Resolution Limited Area Model), which has 22-km resolution and spans over the whole reanalysis period. The reanalysis data were available on 5.5 km horizontal resolution grid, with vertical resolution of 4–6 m down to the depth of 125 m, increasing up to 25–30 m below 200 m depth. Reanalysis data spanned over the period of 1989–2014. We used the particular year of 2007 for our tests, with the only intention to model some real rather than a “mean” situation: any kind of ‘averaged’ currents was assumed inappropriate as it would significantly reduce the current velocity magnitude.

A series of numerical experiments was performed using a specifically developed modification of a TracPy model (Thyng, 2014), which, in its turn, is based on a TRACMASS trajectory model (Döös et al., 2013). The TRACMASS includes an algorithm for adjustable space interpolation between the grid cells of a staggered Arakawa C grid. That leads to an increase in precision of a drifter’s position calculation. The code of TRACMASS was originally written about two decades ago and it has since been used in many applications (e.g., Döös and Engqvist, 2007). The efficient FORTRAN code of the TRACMASS model was wrapped up in Python (Thyng, 2014) resulting in TracPy. So the main goal of TracPy is to take advantage of the speed of the TRACMASS algorithm, but to have a possibility for simple setting up batches of simulations. We made a step further: the original code from (Thyng, 2014) was adapted to the numerical grid and velocity fields data from CMEMS. A number of new subroutines were developed, such as re-initialization from the last location of the particles; addition of the extra particles to the experiment; bottom boundary interaction; conditional vertical velocity calculation, and so forth. The resulting model was called MARBLE (MicroPlastics Research in the Baltic marine Environment), after our current project (http://lamp.ocean.ru/).

3. Results and discussion

3.1. Distribution of fibres in water column: field data analysis

There are various kinds of primary and secondary sources of fibres in marine environment: household and industrial waste waters, fishing nets and ship ropes, plastic bags and disposed clothes, etc. (Cole et al., 2011; Kershaw, 2015). In contrast to quite many studies of fibres concentrations in surface waters and bottom/beach sediments, reports on fibres in water column are scarce, the main reason being much lower particle concentrations and lack of convenient methodology of sampling of large water volumes. This way, our measurements are among the very first ones, and definitely the first for the Baltic Sea water body. Microscopic analysis of the samples revealed only three types of micro-particles: fibres, (presumably) paint flakes, and films, see Fig. 2 for some examples. Among all the 95 samples, 37% (35 samples) did not contain visible artificial objects, 7% (7 samples) contained small films (0.2–3 mm, totally 10 pieces), about 40% (38 samples) had flakes, while 63% contained fibres in concentrations from 0.07 to 2.6 items per litre. So, in our samples, microfibres seemed to be the prevailing type of MPs.

Table S2 (in Supplementary materials) presents the information in more detail. It shows fibres concentrations (in fibres per litre), depths of sampling in m and in relative units - dimensionless sigma-layers, obtained by dividing of sampling depth by local depth. The coverage of different sigma-layers by the samples is shown in Fig. 3. During microscopic examination of the filters, we counted only fibres which looked artificial (of uniform thickness and colour, mechanically elastic, etc.). Obviously organic objects (those with cellular structure, or readily decaying under pressure of a thin needle) were discarded. Coloured fibres (having the shades of red, bright green, and blue, see Fig. 2 (c, e, f)) were counted separately from those that looked dark, grey, or transparent (like that on Fig. 2 (d)): if the former must have had artificial origin, the latter might have not. Concentrations of fibres of both groups are shown in Table S2 separately. For 95 samples totally, 1050 l of water were filtered, and about 300 fibres were found, leading to a bulk concentration of coloured/total fibres of about 0.08/0.28 fibres per litre, or, correspondingly, about 1 coloured plus 2 colourless fibres per 12 l of the Baltic Sea water.

Mean concentrations of all the fibres and of the coloured fibres in each sample are shown in the last columns of Table S2. Fig. 4 illustrates the distribution of fibres with depth, taking into consideration three principal layers in the water column: upper (sigma ≤ 0.1), intermediate (0.1 < sigma < 0.9), and near-bottom (sigma > 0.9). It is clear that, on average, near-surface and near-bottom layers in our measurements have considerably higher concentrations of microfibres than intermediate layers. Higher concentrations in surface and bottom boundary layers can be associated with highlighted water turbulence and stronger density stratification there (Thomsen, 2002; Lemckert et al., 2004; Ivey et al., 2000; Richards, 1982; Gordon and Witting, 1977; Holtappels and Lorke, 2011). Turbulent pulsations of stronger and sheared near-surface and near-bottom currents retain light fibres in suspension, slowing down or even preventing them from sinking for some time. In intermediate layers, water currents are weaker, turbulence intensity is much smaller, resulting in just preservation of a free-fall sinking regime of a fibre which had already escaped the surface boundary layer. Higher concentrations of fibres could physically be suggested also right above the pycnoclines (seasonal thermoline and permanent halocline in the Baltic Sea). However, this point cannot yet be examined here, since the present sampling data have insufficient vertical resolution.

Most part of anthropogenic fibres found on the beaches worldwide (Browne et al., 2011) are made of materials with the density slightly higher than that of water - like polyamide (or nylon, 1.02–1.05 g cm−3), acrylic (1.14–1.18 g cm−3), or polyester (1.01–2.3 g cm−3), so they should thus sink in sea water. Positively buoyant fibres - residuals of polypropylene (0.83–0.85 g cm−3) ropes or
polyethylene (0.89–0.97 g cm$^{-3}$) bags – become bio-fouled in the upper water layers and after 6–8 months should also begin sinking (Chubarenko et al., 2016). Thus, most typical behaviour of tiny and flexible fibres in turbulent marine environment seems to be their long-lasting motion together with basin-scale water currents and very slow sinking.

It is physically important to compare available concentrations of fibres in the water column of the open sea and in the coastal waters, as well as in the bottom sediments. Our measurements in the near-bottom water layer of a coastal zone in the Gdansk bay in the cruises NORD-1 and NORD-2 (see Table S2, first 12 lines) show maximum concentrations of 2.6 fibres per litre. This peak concentration was obtained in the vicinity of the damping site for dredged port materials, while the mean concentration for all the measurements in near-bottom layers in NORD cruises (i.e., in the Gdansk bay coastal zone) amounts to 0.71 fibres per litre. This value, characteristic of waters in the coastal zone (7–21 m), is about three times as large as the mean concentration of 0.25 fibres per litre for near-bottom layers and an order-of-magnitude larger than intermediate-layer concentrations in the Baltic Sea, shown in Fig. 4. Thus, coastal waters are more contaminated than open-sea waters, which is quite logical when most contamination comes from the shores.

Along the Baltic Sea coasts, anthropogenic input from the Viikinmäki wastewater treatment plant is reported in (Talvitie et al., 2015); analyses were performed using consequent 200, 100 and 20 μm filters. The study showed concentrations of 2–5 fibres per kg of dried weight sediments and 0.01–0.65 fibres per litre of seawater close to the Helsinki archipelago (see Fig. 1). For concentrations in the beach sediments, study (Stolte et al., 2015) reported tens to hundreds of fibres per kilogram of dry weight (fibres/kg DW) along the German Baltic coast (obtained using zooplankton net with mesh size of 55 μm), while (Esiukova, 2016) found 0.6 ± 0.3 fibres/kg DW of the beach sediment in Kaliningrad area (174 μm mesh size). Bottom sediments in the coastal zone (from 5 to 30 m depth) off the Kaliningrad region are shown to contain from 4 to 45.1 fibres/kg DW (174 μm mesh size, as in Zobkov and Esiukova (2016)). The latter study reports concentrations of fibres in sandy bottom sediments in the Gdansk bay coastal zone (depth range 5–30 m, some 20 km southwards from the area, investigated in NORD cruises). Recalculated to volume units, this makes about 14–43 fibres per litre.

Thus, comparing water and sediments, both the direct measurements and the averaged values (Fig. 4) indicate that the fibres concentration in bottom sediments is one-two orders of magnitude larger than that in the near-bottom water layers. These conclusions are in full agreement with other observations: an order-of-magnitude difference in concentrations between coastal and open sea areas was pointed out in (Van Cauwenberghe et al., 2013; Woodall et al., 2015), while (Woodall et al., 2014) reported that fibres were up to four orders of magnitude larger in coastal sediments than in the intermediate layer of the Baltic Sea.
The sinking behaviour of plastic fibres: laboratory testing

The distribution shown in Fig. 4 and orders-of-magnitude prevalence of fibres in bottom sediments suggest the following scenario of fibres performance in water column. Being small and easily entrained in upper-layer turbulent motions, they, on average, spend some (quite considerable) time in the surface layer, then sink to the bottom, but cannot settle for some time (or are often re-suspended) due to more intense benthic-layer turbulence. Bottom sediments become the ultimate place of their deposition (Van Cauwenbergh et al., 2013). This scenario seems to be generally independent of the fibre material density: relatively heavy fibres ($\rho_{\text{fibre}} > \rho_{\text{water}}$), due to their small size, should be entrained for some time into surface-layer turbulent mixing (Reisser et al., 2015), while relatively light fibres ($\rho_{\text{fibre}} < \rho_{\text{water}}$) get bio-fouled with time (Chubarenko et al., 2016) and eventually sink as well.

What are the sinking velocities of fibres? It is hardly possible to directly observe the behaviour of an elastic, flexible tiny fibre in natural turbulent marine environment. However, laboratory experiments can help to grasp some general features.

An exemplary plastic fibre taken for our laboratory testing had approximate length of 8 mm (see Fig. 5) and measured density of 1.04 g cm$^{-3}$. The diameter of the fibre was in the range of 10 to 30 μm as it was measured by comparing to the known diameter of Neuston net threads. While sinking, the fibre took a curved form (including the loop) during each of the 7 runs. The fibre was sinking with the long axis perpendicular to the direction of the fall (i.e. horizontally) in 4 runs, in 2 runs the long axis of the fibre was maintained vertical and once the spatial orientation of the fibre could not be clearly determined. A horizontally oriented sinking fibre was rotating slowly in a horizontal plane. The measured sinking velocity varied from 0.4 to 1.4 mm/s, with averaged value of 0.9 ± 0.8 mm/s. A vertically oriented fibre was sinking generally faster than that which is perpendicular to the direction of the fall (about 1.4 and 0.8 mm/s, respectively). This is due to the hydrodynamic resistance to the flow, proportional to the cross-flow area of a moving object.

Thus, the sinking velocity significantly depends on spatial orientation of the sinking fibre, which is quite random. It did not change significantly during the tests, however it might vary supposedly under natural turbulent marine conditions. A vertically oriented fibre sinks 3–4 times faster than the one perpendicular to the direction of the fall. The averaged magnitude of sinking velocity – about 1 mm/s – can be taken as an approximate upper bound for free-fall motion in natural sea waters, because (i) natural turbulence should reduce this value, and (ii) plastic-water density difference in salty marine water is somewhat smaller.

In sediment transport problems, the sinking velocity of a particle is shown to be also a measure for threshold velocity of the current, which prevents the particle from settling (Collins and Rigler, 1982; Ballent et al., 2013). For an elongated fibre of a complicated curved shape this threshold might be even smaller than the sinking velocity. The fibre might settle among particles of bottom sediments of various roughness. Having large windage, a fibre might be resuspended by the weakest currents. Thus, fibres per se are able to settle only in areas with very weak currents, and they are indeed found in abundance in bottom depressions with muddy (i.e., very fine) sediments (Van Cauwenbergh et al., 2013; Woodall et al., 2014). On the other hand, enhanced turbulence of the benthic boundary layer re-suspends also particles of sediments, and fibres may be just caught by other particles re-sedimentation. One more possibility to stay at the bottom in a dynamically active area is to get entangled in, e.g., seaweed. In general, settling at the bottom is not so obvious for a fibre, and this conclusion is in full agreement with studies of natural fibres in the marine environment.
agreement with the observed high near-bottom concentrations, shown in Fig. 4.

3.3. Simulation of fibres’ transport: numerical tests

3.3.1. Strategy

Numerical simulation is shown to be an efficient tool for investigating all kinds of problems of environment contamination (Kershaw, 2015). As for pollution of marine waters by synthetic fibres, the performed analysis of field data and laboratory tests allows to suggest proper time and length scales, and those key features of the behaviour of microfibres in marine environment which are to be reproduced by numerical simulation of their transport and fate. This way, presence of fibres everywhere in the water body – in upper/intermediate/lower layers, in coastal and open sea areas – is the consequence of their long residence time in the sea and easy transport by basin-scale currents. Thus, a basin-scale model and possibly close-to-real systems of currents are required. For our tests we chose to use the reanalysis data for the entire Baltic Sea basin spanning a period of one year, and a Lagrangian particle technique for fibres tracking.

An important feature of microfibres to be reproduced by the model is their very slow sinking – with vertical velocity, \( w_v \), from eventually 0 to at most 1 mm/s. It is hardly possible to explicitly prescribe the particular fibre motion within the basin-scale model, with its huge (relative to microfibres size) horizontal numerical resolution of 5.5 km × 5.5 km and vertical layers from 4 m (in the upper 80 m) to 10 m (at about 150 m – the deepest location of the modelled particles). Thus, some way of parameterization is required. We chose step-wise simulations to mimic the motion of a fibre with water currents in a certain layer during the period \( \Delta T \), required for sinking across the layer thickness. After that time this particle is shifted to the layer beneath where it is allowed to follow that layer currents for the next \( \Delta T \). This “sinking time step” \( \Delta T \) was a parameter for calibration.

Another point addressed in our modelling is an attempt to reproduce the fibres settling at the bottom. Since, presumably, a very weak current is required to prevent the fibre from settling, we developed a special numerical procedure which keeps the fibre drifting in the near-bottom water layer, if current speed is above a certain threshold.

3.3.2. Model setup

The main goal of the numerical experiments with the MARBLE model was to test the sensitivity of results to variations of the main parameters, prescribing the fibres motion and distribution – horizontal diffusion coefficient, particle sinking velocity, and the settling threshold, and to find the configuration that reflects the assumed features of the fibres distribution.

We used for our tests the reanalysis data for the particular year 2007 (January–December). The CMEMS data was provided every 6 h, enough for a particle in the upper layer to move out of the initial numerical grid cell (assuming the mean velocity of 50 cm/s). To increase the precision of the calculation, a numerical integration time step was chosen at 1.5 h as a compromise between verbosity and calculation efficiency.

Initial location of the particles was placed on the surface of the Gdansk bay based on its high population density both on the coastline and in the drainage basin. Surface area of the region initially filled with the fibres was set to be approximately 5000 km². As much as 10,000 particles were uniformly distributed over the surface on January 2007. Such a number of particles was defined in a series of preliminary test runs for a combination of sufficient consistency of the final particles distributions and computational efficiency. Additional 10,000 particles were distributed to the same locations on the 1st day of each month from February till May.

The coefficient of ‘numerical diffusion’ \( A_h \) had to be introduced in the numerical simulation. Despite the fact that some of the recent papers on the plastics transport modelling (Liubartseva et al., 2016 and Lebreton et al., 2012) mentioned the problem of turbulent diffusion for Lagrangian particles, there was no direct information on the values of that coefficient or any other ‘random walk’ properties that were used during the experiments. The only available estimation of 10 m²/s in (Critchell and Lambrechts, 2016) was made for shallow areas; hence for the open sea we tested that coefficient as 20 m²/s (Case 1) and 100 m²/s (Case 2). As a result the value of \( A_h = 50 \) m²/s was selected. The simplest estimation showed that around 30% of the Baltic proper in such a case would be covered by the particles cloud in 1 year only due to numerical diffusion. Generally speaking, the value should decrease with depth, due to weakening of the velocity magnitude, but there are different approaches to that problem and we have no data to speculate on how the deep layer turbulence influences the MPs behaviour.

3.3.2.1. Vertical sinking velocity

In order to test the sensitivity of final distribution to the magnitude of sinking velocity of a particle, \( w_v \), the simulations were carried out with two velocity magnitudes: movement of the particles from one level to the following one (downwards) took place once in 3 (Case 3) or 10 (Case 4, Case 1, and Case 2) days. This yielded sinking velocity in upper 4-m thick layers of about 0.02 mm/s (Case 3) and 0.006 mm/s (Case 1, Case 2, Case 4), correspondingly. Below 80 m depth, where the thickness of the model layer increased, the sinking velocity increased as well, which was quite right since it mimicked smaller turbulence below pycnocline. In our simulations, the sinking velocity increased with depth by 3.5 times maximum.

Particles’ settling to the bottom was assumed as one-way only. When the particle hit the bottom, it was removed from the numerical run and its location was saved. The critical shear velocity (or ‘settling velocity’), which prevents fibres from settling, was estimated as follows. Taking 1 mm/s (e.g. \( u^* = 0.001 \) m/s) as a scale of magnitude, we obtained the critical shear stress to be \( \tau_c = u^* \rho_w = 10^{-6} \cdot 10^{-3} = 0.001 \) N/m². This value was small - one order of magnitude smaller than 0.01 N/m², which had been used by (Ballent et al., 2013) for industrial pellets, and looked physically correct for tiny and light fibres. However, as it was mentioned before, fibres might get entangled in bottom material or entrained by other settling sediment particles. From the results of the test cases Case 6 (\( u^* = 0.001 \) m/s) and Case 7 (\( u^* = 0.01 \) m/s), we decided on the critical shear stress of 0.01 N/m² for modelling of fibres. Besides, due to discretization of processes and various parameterisations used in numerical modelling, natural intimate link between sinking and settling velocities could hardly remain in numerical simulations. Thus, both velocities mentioned above should be considered as calibration parameters for the particular model.
Since reanalysis data were too coarse to reproduce complicated coastal processes, we restricted our analysis to areas deeper than 4 m, and no coastline boundary interactions were parameterized, with perfect-slip conditions at the bounding grid-cells.

In general, physical behaviour of a particular fibre in the model was as follows. It was placed at the surface in the centre of one of the grid cells within the Gdansk Bay (a red rectangle in Fig. 6) and was allowed to drift together with surface currents for a few (3 or 10) days. Then, being already at another location, it was moved one layer downwards, to drift there for the next few days; etc. When, after the next step down, the particle appeared to reach the near-bottom layer, the flow velocity in this cell was checked. If the flow velocity was smaller than the critical shear velocity - the particle was assumed to have settle; if the flow was stronger – the particle was kept moving for the next (3 or 10) days. The final settling point was saved and the final distribution of particles was analysed.

3.3.3. Results of modelling

3.3.3.1. Sensitivity to the variation of the turbulent diffusivity coefficient. Case 1 and Case 2 aimed to test the sensitivity to the variation of the turbulent diffusivity coefficient. The initial location of the fibres in the model coincided with the centre of the grid cell. Horizontal grid cell size was 5.5 km, thus, if there were no turbulent diffusion added, the "stacked" particle patch would follow the velocity field during the numerical run. In order to simulate sub-grid diffusion of the particles, the coefficient \( A_h \) was introduced, the so called "random walk" (Lynch et al., 2015). In the paper (Critchell and Lambrechts, 2016) the authors showed that turbulent diffusion parameterisation has a determinant impact on the modelling result in shallow coastal areas.

The initial source location of 10,000 particles was placed inside the Gdansk Bay in the surface water layer. Simulations started on 1 January 2007 and ended on 30 December 2007.

For Case 1, the horizontal turbulent diffusion coefficient was set to 20 \( \text{m}^2/\text{s} \), and for Case 2 - 100 \( \text{m}^2/\text{s} \). In both cases, vertical settling velocity in the upper layers, \( w_s \), was set to 4 m per 10 days, which is -0.006 \( \text{mm/s} \).

For 10 June, locations of fibres suspended in the water column are shown in Fig. 6. And for 31 December, a 2D-histogram of the final fibres position at the bottom of the Baltic Proper is given at Fig. S3. At the end of the simulations in Case 1/Case 2, i.e., after 1 year of drift, as many as 2756/2634 particles (or about 1/4 out of initial 10,000 ones) still remained suspended, mostly below the 100-m depth isobath.

Areas covered by the particle cloud on 10 June (Fig. 6) were not much different for turbulent diffusion coefficients of 20 \( \text{m}^2/\text{s} \) and 100 \( \text{m}^2/\text{s} \) (note that the settled particles are not shown in the figure). However, the distribution of the particles within the area was much more uniform for higher diffusion, which is natural. Along with that, the comparison of the panels in Fig. 6 shows that the influence of diffusion is much more pronounced in the water column, while final settling maps look quite alike (Fig. S3). The main differences were observed in the shallow areas, only supporting the idea of (Critchell and Lambrechts, 2016). Besides, this manifests that the settling threshold may be of greater importance in comparison with the intensity of spreading in the water column: the particle settles where and when it is able. In the present simulation, we set the fibres’ critical velocity ten times as high as that in the calm laboratory tank – and still it seems to be too small and in fact controls the final distribution of fibres at the bottom. This poses once again the question of proper use of the laboratory threshold velocity in the particular numerical model, or, oppositely, proper model calibration regarding this value. The latter is not yet possible, since real distribution of fibres at the bottom is not yet known.

On the other hand, obviously higher concentrations of settled particles in the coastal zone (Fig. S3) highlights another problem: too low velocities in boundary-cell currents in the re-analysis data. This effect is caused by the averaging over a large numerical cell, while naturally the coastal currents are much stronger than those in the open sea. This way, the limitation should be imposed on the use of large-scale models and re-analysis data for problems of distribution of micro-particles in coastal areas.

Thus, the area of coverage by this sort of pollution and the final distribution of the fibres over the bottom do not seem to be sensitive to the value of the turbulent diffusion coefficient. At the same time, the diffusion intensity influences the form of the particles cloud and complexity.
of their trajectories. Henceforth, an intermediate value of 50 m$^2$/s for turbulent diffusion coefficient was chosen for further experiments.

3.3.3.2. Sensitivity to the magnitude of the sinking velocity. As it was discussed above, the sinking velocity, $w_s$, of the fibres in the turbulent surroundings (i) was obviously smaller than the velocity measured in a still laboratory water tank, and (ii) definitely influenced the travel distance of the microfibers from the initial location. Cases 3 and 4 were aimed to test the sensitivity of the modelling results to the variation of the vertical sinking velocity magnitude. It was set to one layer per 3 days (Case 3) and one layer per 10 days (Case 4), making up about 0.02 mm/s and 0.006 mm/s, correspondingly, in the upper 80 m. The turbulent diffusivity coefficient remained intact at 50 m$^2$/s.

As it is shown in Fig. 7 (a, b), intermediate locations of the fibres on 10 June 2007 are significantly different for “fast sinking” (a) and “slowly sinking” (b) particles. The rapidly sinking ones (a) contaminate the area about three times smaller, which stretches from the initial location in the direction of prevailing currents. Relatively slowly sinking particles (b) are distributed much wider, their areal expands in all directions, and quite a large proportion of fibres is transported down to the deep layers (the darker the colour in Fig. 7 (a, b), the deeper the fibre is). Towards the end of the simulations (31 December, panels (c, d) in Fig. 7), as much as 3629 particles (36%) remained in the suspended state (mostly between 30 to 50 m depth) in Case 3, and 2649 (26%, mostly below the 100-m depth isobath) in Case 4. In other words, finally, slowly sinking particles are transported by currents farther, reach deeper layers, and this eventually increases the probability of their settling.

In Case 3, about 25% of the particles settled within the restricted area in the Gdansk bay (dark spot in Fig. 7 (c)), while in Case 4 the particles are spread from the centre of the Bornholm basin to the northern end of

![Fig. 7. Distribution of fibres with different sinking velocities: (a, c) - 0.02 mm/s, Case 3; (b, d) - 0.006 mm/s, Case 4. (a, b): maps of suspended fibres locations in water column on 10 June for “fast” (Case 3) and “slow” (Case 4) particles. The darker the colour – the deeper the fibre is. Initial position of 10,000 particles on the sea surface in the Gdansk bay is shown by a red rectangle. Note that settled particles are not shown. (c, d): 2D-histograms of final fibres locations at the bottom of the Baltic Proper at the end of 365 days in Case 3 (c) and in Case 4 (d). The more intense the colour – the more fibres settled in the cell. Isobaths are drawn for 5, 10 m, and from 20 to 300 m every 30 m. Dotted line on the left depicts the western bound of the numerical domain. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image-url)
the Gotland island. Thus, only 3-fold difference in sinking velocity is able to drastically change the final particle distribution. Having in mind that fibres are found in abundance in the deepest ocean depressions (Woodall et al., 2014), the sinking velocity magnitude of 0.006 mm/s looks more realistic.

Once again, higher concentrations are observed in shallow coastal areas, especially for slowly sinking particles (Fig. 7 (d)) along the Curonian spit. We suggest that the same numerical reason is responsible as in previous cases: underestimated near-bottom and near-shore velocities that lock the particles movement.

Sensitivity of the final fibre distribution to the magnitude of the settling threshold was tested. In Case 5 and Case 6 the critical velocity of the bottom current which stops the particle from settling was set to 0.001 m/s and 0.01 m/s, correspondingly. The vertical sinking velocity was set to 0.006 mm/s. The turbulent diffusivity coefficient remained intact at 50 m²/s.

The main differences between these two cases are shown in Fig. 8 (a, b). Variation of the critical velocity affected the settled particles density in the shallow water areas. In Case 6, on 31 December, only ~3000 particles remained in the water column, while in Case 5 up to 9000 of initial 10,000 particles remained in the water column, distributed across different layers but suspended. As it is shown in Fig. S4 (a), when the fibres have low critical settling threshold (i.e., even low current velocity leads to particle re-suspension), they concentrate in the near-bottom areas, often locked there by the currents, slowly drifting away from the initial position but unable to settle down to the bottom even after a year of simulation.

Only those particles that had travelled far enough, to the centres of the deepest depressions, were able to settle due to the weakness of the currents velocities. In Case 6 (see Fig. S4 (b)), on the contrary, the fibres started to settle pretty fast as they reached the regions away from the strongest currents, hence accumulating both along the slopes and at the deepest parts of the sea. The main drawback of the model is the strength of the currents, hence accumulating both along the slopes and at the deepest parts of the sea. The main drawback of the model is

Taking into account the numbers of settled particles in Case 5 (~1000) and Case 6 (~7000), we suggest using critical settling velocity for the fibres as 0.01 m/s in order to reproduce the fibres’ removal from the water column. Our suggestion is supported by our observations (see paragraph 3.1) and the hypotheses by Van Cauwenberghe et al. (2013) and Woodall et al. (2014).

The performed analysis of field data and laboratory tests indicated that modelling of transport and distribution of fibres in the sea requests a basin-scale model, with model runs lasting for several months. For the Baltic Sea, we chose to use the open HIROMB reanalysis data, spanning over a period of one year, and a Lagrangian particle technique for fibres tracking. Analysis of sensitivity of the final modelled distribution of fibres in the Baltic Sea Proper to changes in the horizontal diffusion coefficient, particle sinking velocity, and the settling threshold was performed. It was shown that the area of coverage by this sort of pollution and the final distribution of the fibres over the bottom seemed not to be too sensitive to the value of the turbulent diffusion coefficient. However, the magnitudes of the sinking velocity and the re-suspension threshold significantly influenced final fibres distribution. Slowly sinking particles were transported farther by currents and finally reached deeper layers. Smaller threshold re-suspension velocity led to the analogous effect. Overall, the use of HIROMB re-analysis data allowed us to simulate the transport and distribution of fibres quite adequately.

Since large-scale models have rather coarse numerical grids, resulting in quite weak (in comparison with natural) currents and much simplified coastline, their use for MPs transport in shallow coastal zones is limited. This is highly important for fibres distribution along the coast, because they are suggested to migrate together with bottom sediments under the influence of surface waves (Chubarenko and Bagaeva, 2016). This way, for simulation of the behaviour of MPs particles from relatively heavy plastics (like industrial pellets, plastic wreckage, etc.), a sediment transport model seems to be required.


Our field sampling at various depths in water body showed that fibres are the prevalent type of anthropogenic micro-particles pollution in the Baltic Sea (Proper). Surface and bottom layers are more contaminated in comparison with intermediate layers, and open-sea waters are cleaner in this regard compared to the coastal ones. Objective difficulty

![Fig. 8. Distribution of fibres with different critical settling velocities: (a) ~0.001 m/s, Case 5; (b) ~0.01 m/s, Case 6. (a, b): 2D-histograms of final fibres locations at the bottom of the Baltic Proper at the end of 365 days in Case 5 (a) and in Case 6 (b). The more intense the colour – the more fibres settled in the cell. Isobaths are drawn for 5, 10 m, and from 20 to 300 m every 30 m. Dotted line on the left depicts the western bound of the numerical domain.](image-url)
of reporting the microplastics fibres concentrations is the applied sampling methodologies: water surface samples are typically collected by nets with different mesh sizes, particles from bottom sediments are extracted by flotation with filtering of supernatant (Zobkov and Esiukova, 2016). Fibres, with their typical diameters of 10–30 μm, are caught in samplings together with other anthropogenic particles and MPs only when they get entangled in the filter/net or its content. Thus, with present-day methods, one cannot yet fully rely on quantitative data on fibres concentrations. Having this severe limitation in mind, we still were able to make use of the fact that fibres are extremely widespread in marine environment and learn some principal features of their behaviour in the sea water body. Taking into account many factors (like beginning of water contamination from the sea surface, different initial buoyancy of different particles, etc.), we concluded that principal behaviour of fibres in the water column is as follows. First, they spend some time in the surface layer: buoyant fibres just float, while heavy ones tend to sink, although for some time the latter are captured by upper-layer turbulent motions. With time, all of them should begin sinking, due to bio-fouling or catching the suspended matter at least. A long-lasting sinking process favours settling in those very calm environments.

References


