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Microplastic concentrations in beach sediments along the German Baltic coast

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ABSTRACT

The contamination with microplastic particles and fibres was evaluated on beaches along the German Baltic coast. Sediments were sampled near the Warnow and Oder/Peene estuaries, on Rügen island and along the Rostock coast to derive possible entry pathways. Seasonal variations were monitored along the Rostock coast from March to July 2014. After density separation in saline solution, floating particles were found to be dominated by sand grains. Water surface tension is shown to be sufficient to explain floatation of grains with sizes less than 1.5 mm. Selecting intensely coloured particles and fibres, we find lower limits of the microplastic concentrations of 0–7 particles/kg and 2–11 fibres/kg dry sediment. The largest microplastic contaminations are measured at the Peene outlet into the Baltic Sea and in the North Sea Jade Bay. City discharges, industrial production sites, fishing activity and tourism are the most likely sources for the highest microplastic concentrations.

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1. Introduction

1.1. Definition and risks of marine microplastics

Over the past decade, microplastic detections have become a growing concern in the scientific community, with a wide range of concentrations between one and thousands of potential plastic particles per kg of dry sediment or per litre of seawater reported (see Tables 4.1 and 4.3 in Leslie et al., 2011, for an overview). Seawater sampling with varying mesh sizes indicates that number concentrations of microplastics are increasing with decreasing particle sizes (Norén, 2008). Size ranges of microplastics reported in sediment, seawater, and biota samples range from several micrometres (μm) to a few millimetres and depend heavily on the employed mesh width of zooplankton net and the sieve or filter pore size (see e.g. the discussion in Leslie et al., 2011, Chpt. 6). In the Venice lagoon, a systematic spectroscopic analysis of 20 sediment samples found more than 93% of microplastics with sizes <500 μm, of which 55% were <100 μm for a lower size limit of 32 μm (Vianello et al., 2013). Particles with sizes 5–25 μm are found in mussels grown for human consumption (Van Cauwenberghe and Janssen, 2014), while a size peak at 1.25–2.5 mm is observed in North Atlantic and Celtic seawater samples for a lower size limit of 250 μm (Lusher et al., 2015). These size scales found in sediment, seawater, and biota samples cover the size range from micro- to mesoplankton and therefore bare the risk to infiltrate the marine food web from the lowest trophic levels.

These numbers raise the concern that contamination levels might inadvertently affect the marine food chain from the smallest planktivores to the largest fish and marine mammal species. Today, the chemical fingerprints of microplastics are detected in the muscle and blubber tissue of large filter feeders such as basking sharks and fin whales (Fossi et al., 2012, 2014). Although health adverse effects are demonstrated on marine organisms in experimental settings Barnes et al. (2009) and von Moos et al. (2012), whether health and natural development are compromised at environmental particle and additive concentrations is presently not clear. As microplastics cannot easily be removed from the marine environment and plastics are highly persistent materials, the risk of health adverse effects to marine organisms on all scales increases with increasing microplastic concentrations. This risk might return to us as the top predator since microplastics are already shown to infiltrate the human food web (Van Cauwenberghe and Janssen, 2014). With increasing levels of microplastic concentrations (Morét-Ferguson et al., 2010), health-adverse effects to humans have to be anticipated with the long-term presence and exposure to microplastics. The European Union (EU) has acknowledged microplastics as a pollutant with potential health risks to marine

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species as well as humans, and requests the characterisation of *"trends in the amount, distribution and, where possible, composition of microparticles (in particular microplastics)"* (Criterion 10.1.3), including microplastics ingested by marine animals (Criterion 10.2.1), in Annex III of the Marine Strategy Framework Directive (MSFD). The fact that all layers of the marine food web are exposed to microplastics implies that understanding the amount and origin of microplastic pollutants is necessary to identify counteracting measures.

In the executive summary of the *International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris* (IRW), microplastics are defined as particles with sizes less than 5 mm (Arthur et al., 2009). No lower boundary is determined, although seawater samples are frequently limited to 333 μm by the mesh size of neuston nets. The minimum boundary of sediment samples is frequently lower when 50–100 μm sieves or 1–5 μm filters are used to collect particles (see also Dubaish and Liebezeit, 2013). Two kinds of marine microplastics are distinguished throughout the literature on the basis of their origin, and were also defined by the IRW (Arthur et al., 2009). *Primary microplastics* originate from spillage during plastic production or recycling, from sandblasting in shipyards and other abrasives, and from microcleansing particles in personal care products. All of these primary microplastics share the common property that they are *designed to be small* during their production process. *Secondary microplastics* comprise broken fragments of larger plastic pieces, including, but not limited to, marine litter, derelict fishing gear from industrial and recreational fishing, litter from landfills, painting flakes from ship hulls, synthetic fibres from laundry discharge, and foil fragments from packaging, industrial or agricultural sources. Given the large diversity of sources, the entry pathways of secondary microplastics and possible mitigation measures are particularly difficult to define.

Biodegradation of macroplastic litter entering the environment from land- or sea-based sources is extremely slow (Thompson et al., 2004, see also Andrady, 2011, for a review). As all rivers flow to the sea, the oceans provide the largest sink for undegraded synthetic polymers down to molecular sizes. With UV radiation, oxidation, mechanical or bacterial degradation times of several hundred years (Thompson et al., 2004), the current rate of increasing plastic production and the expected enrichment of the environment and oceans with both macro- and microplastics imply that contamination of the food chain will proceed, even if particle input could be stopped instantaneously. The contribution of fishing line fibres and the degradation timescale of synthetic net material are presently unknown. Synthetic clothing likely comprises a major fibre source especially in coastal waters. A single polyester fibre shirt released 1900 fibres in a single washing (Browne et al., 2011). In a comparative study between micro- and macroplastics, Van Cauwenberghe et al. (2013) estimate that microplastics contribute between 8% and 40% of the plastic weight in beach sediments at the Belgian coast. Both fibres and particles are found to significantly contribute to the microplastic concentrations, and the large variation in the spatial distribution and local weight concentration between micro- and macroplastic fractions implies that microplastics have to be monitored separately (Browne et al., 2010; Van Cauwenberghe et al., 2013; Dekiff et al., 2014).

1.2. Microplastic concentrations in seawater and sediment

Marine microplastics have evolved into one of the major marine pollution research fields over the past decade. Although more than one hundred studies are presently available on marine microplastic pollution (see Ivar do Sul and Costa, 2014 for a comprehensive review), spanning beaches, continental shelf areas, central ocean gyres, and harbours worldwide, the spatial coverage is limited by the few research centres where microplastics are investigated (see Fig. 2 in Ivar do Sul and Costa, 2014). The largest concentrations of microplastic particles and fibres are observed in coastal and harbour areas and near industrial production sites (e.g. Norén, 2008; Claessens et al., 2011; Desforges et al., 2014). Microplastics have been found in several estuarine environments, indicating rivers as one of the entry pathways of microplastics into the marine environment. Microplastic concentrations are reported for the UK Tamar estuary in Europe (Browne et al., 2010) and seasonal and spatial variations correlated with rain floods were derived for the Goiana estuary in Brazil (Lima et al., 2014), where the ingestion of nylon threads in estuarine fishes was found at all life stages (Possatto et al., 2011; Ramos et al., 2012; Dantas et al., 2012). River basins and mangrove systems are affected by microplastic contamination along the Singapore coastline (Nor and Obbard, 2014). Particularly large concentrations of microplastic particles are reported in enclosed bays with industrial activity, such as the Jade bay (Dubaish and Liebezeit, 2013). On a larger scale, the Baltic Sea provides only limited exchange with the North Sea during storm events and has to be considered a sink for microplastics. With discharges from the Oder, Neva, Vistula, Western Dvina, and Neman rivers and major industrial coastal cities (Szczecin, Copenhagen, Malmö, Stockholm, Helsinki, Gdansk, Saint Petersburg, Rostock, Lübeck, Kiel), the Baltic Sea represents one of the largest enclosed marine bay areas worldwide.

Despite this predestined location, only one microplastic study was conducted in the Baltic so far. In Baltic Sea coastal waters, Magnusson and Norén (2011) found average concentrations of 4 fibres/litre (F/l) and 32 anthropogenic debris particles/litre (P/l), as quoted in the WP3 GES-REG report (Ojaveer et al., 2013, p. 3, original study not in English). Several groups have addressed microplastic contamination in the North Sea. In seawater samples obtained in a Skagerak transect at the outlet of the Baltic into the North Sea, Norén and Naustvoll (2011) found blue particles with sizes 10–100 μm in 15 of 17 samples, and 102 microplastic spheres per litre of seawater are found in Stenungsund industrial harbour near a polyethylene production plant (Norén, 2008). The characteristic size range of 0.5–2 mm of these spheres is large for marine microplastics and covers the size range of prey for juvenile fish. Increasing evidence indicates that the vicinity of urban areas increases the concentration of microplastics in surface waters and in beach sediments. In excess of 1200 particles/l, by far the highest microplastic concentrations reported in the North Sea environment, are detected in seawater samples in the densely populated Jade Bay serving as a discharge site for industry and the Wilhelmshaven sewage treatment plant (Dubaish and Liebezeit, 2013).

In deep, soft sediments in the UK, microplastic particles and fibres are found in 23 out of 30 samples (Thompson et al., 2004), indicating that microplastics were efficiently transported from the water column to sediments over the past decades and are omnipresent in benthal environments today. As in seawater samples, a wide variety of concentrations of potential microplastic particles is reported in sediments. In remote locations, microplastic contaminations between 1 and 2 particles/kg dry sediment are found at the island of Norderney (Dekiff et al., 2014), while a maximum of 50,000 particles/kg is reported for the island of Kachelotplate (Liebezeit and Dubaish, 2012). However, Lorenz (2014) recently found between 34 and 74 particles/kg dry sediment in three off-shore locations on the wider Helgoland shelf and two beach sediment samples on the island of Sylt, and showed that a significant fraction of particles after extraction in dense saline solution are natural minerals when scrutinised with FTIR spectroscopy, rendering previous high number counts uncertain. Along the Belgian coast, microplastic concentrations in harbour sediment

exceeded with 167 ± 92 pieces/kg dry weight the concentrations observed in beach sediment (93 ± 37 pieces/kg) by almost a factor of two (Claessens et al., 2011). The same study found the average microplastic concentration in beach samples to be identical to the concentration in offshore locations on the Belgian Continental Shelf (97 ± 19 pieces/kg), suggesting that three beaches along the Belgian coast accumulated microplastics at the same rate as is deposited on the shelf. These numbers include particles, fibres, microspheres and films, and spectroscopic verification was used to confirm microplastics.

Taken together, the seawater and sediment analyses suggest that harbours and industrial production plants are predominant discharge sites for microplastics. Yet, the entry pathways of microplastics into the Baltic Sea are completely unknown. Microplastic contributions might be expected from city discharge and sewage treatment plants, overseas harbours, industrial production sites and shipyards, as well as touristic activities. With the aim to provide a first identification of microplastic entry pathways into the Baltic beach environment, we have investigated beach sediments near the Warnow and Oder/Peene river outlets, and at heavily frequented beaches on the island of Rügen and in the Rostock area. One location for comparison with earlier studies was included near a paper recycling plant in the North Sea Jade Bay (Dubaish and Liebezeit, 2013).

The reported microplastic concentrations severely depend on the employed extraction and visual inspection methods. A frequently used extraction method from sediments is air-venting in high-density saline solutions, such as zinc-chloride solutions with densities in excess of 1.6 g/cm^3, the maximum density of pristine polymer material (PVC). This procedure is prone to two biases. While a large number of refilling and handling steps causes losses on the container walls even if smooth glass or aluminium containers are used (Imhof et al., 2012), the floatation of mineral sediment on saline solutions contaminates the potential microplastic samples, as discussed below (see also Lorenz, 2014). The optimal way to avoid these biases is the minimisation of handling steps and the spectral confirmation of the extracted particles (Hidalgo-Ruz et al., 2012; Lorenz, 2014). As discussed extensively in Lorenz (2014), material identification requires microimaging spectroscopy as a time-consuming process with costly FTIR equipment. For large surveys as required by the EU Marine Strategy Framework Directive (MSFD), a complete spectral classification will not be feasible. Here, we made use of standard laboratory equipment with the aim to investigate the efficiency of microplastic extraction and the potential biases. With this study, we hope to contribute to the quantification of microplastic particles and fibres in the Baltic Sea environment, as well as to the definition of standardised methods for the extraction, observation, and quantification of microplastic contents in sediments.

2. Materials and methods

2.1. Materials

The materials used in the laboratory were restricted to glass wherever possible. Only glass flasks were used, including in particular 2 l Erlenmeyer flasks employed for air-venting. The surface solution was extracted with a 30 ml graded glass pipette after air-venting. Nevertheless, the use of synthetic materials was unavoidable at several stages. The suction bulb attached to the glass pipette was made of red rubber, and the lint-free cleaning cloth consisted of light-blue polyamide. Glass fibre pre-filters and 5 μm polyacetat membrane filters were used to clean the saline solution after every experiment. Even though no clean room was available to analyse the samples, all clothes worn by the author in the lab were made of cotton. In order to test the fibre and particle contamination to be expected from air-born synthetic fragments in the laboratory, we used blind reference samples where the pure, filtered saline solutions without sediment were processed through all filtering and air-venting devices. Blind samples treated in the same way and over the same durations as sediment samples indicated a laboratory background contamination of up to 2 coloured fibres. No coloured or transparent particles were found in blind samples.

Materials in direct contact with samples:

- Stainless steel sieves with pore sizes 0.063, 0.5, 1.0, and 2.0 mm
- 2 l Erlenmeyer flask
- 25 cm glass tube for air-venting (approximate opening diameter 2 mm)
- 30 ml glass pipette
- 250 ml filtration glass flask
- 55 μm mesh size zooplankton net, cut to 7 cm filter size
- plastic & wire-mesh filter holder

2.2. Methods

2.2.1. Density separation and microplastic extraction

With the general steps of density separation, filtration, and visual inspection we follow the suggestions of Hidalgo-Ruz et al. (2012) for microplastic extraction from sediments and sea surface water samples.

Air-venting in high-density calcium-chloride solution with densities of 1.30–1.35 g/ml was employed to extract lower-density particles from mineral sediments with characteristic densities of 2.45–2.65 g cm^{-3} (Nuelle et al., 2014). Sediments were filled into a 2 l Erlenmeyer flask and covered with 900–1100 ml of $CaCl_2$ solution. The sediment was air-vented through a 25 cm glass tube with opening diameter 2 mm for 4 h while regularly rotating the Erlenmeyer flask, and was allowed to settle overnight after turning off the air flow. The cleared solution above the settled sediment was extracted in two stages. (1) Pipetting 200–400 ml off the surface onto zooplankton net filters was used to extract very light particles and fibres from the surface solution. This part of the solution is classically analysed in air-venting experiments. (2) The remaining $CaCl_2$ solution above the settled sediment was decanted to maximise extraction of higher-density particles and fibres, including particles affected by biofouling. The pipetted and decanted solutions were sucked through zooplankton net filters with a mesh width of 55 μm, which provides a strict lower limit to all detected potential microplastic particles and fibres. Zooplankton net samples were treated with hydrogen peroxide (H_2O_2) to digest residual organic matter and rinsed with deionised water after 24 h.

All filters were inspected under a dissecting stereo microscope (Olympus SZ51 or similar) with 3–4× magnification. Suspicious particles and fibres were analysed under the Olympus SZX16 stereo microscope equipped with the DP21 digital camera with a magnification of up to 11 to facilitate the distinction between microplastics and natural sediment or organic matter, as well as between synthetic/anthropogenic fibres and organic fibres. Because spectroscopy was not available, no distinction is made between synthetic and non-synthetic anthropogenic fibres in the remainder of this article. Microplastics and natural materials were distinguished on the basis of colour, surface structure, and morphology (shape). As transparent particles are most susceptible to misclassification by visual inspection, transparent particles are only included as potential microplastics if their surface structure was clearly distinct from natural sediment. All particles and fibres investigated by visual inspection alone are considered *potential* microplastics (e.g. Dekiff et al., 2014). As material proof via

spectroscopic identification was not available, we implicitly assume all pieces to be *potential* microplastics when the terms microplastic particles and fibres are used. Particles and fibres with colours different from natural sediment, such as intense blue, green, pink, and violet, are visually identified as the most certain microplastic contaminants.

This procedure is designed to maximise the extraction rates of potential microplastic particles and fibres while minimising the exposure of samples to laboratory air and minimising the number of handling steps to reduce the risk of contamination with fibres. Following the recommendations in Imhof et al. (2012), the number of refilling stages is kept to a minimum to avoid the sticking of microplastic particles and fibres to flask walls and the corresponding biases.

2.2.2. Counting procedure

All number counts were carried out under a dissecting microscope with 3–4× magnification. The lower limit for particle counts is given by the zooplankton filter mesh width of 55 µm. Fibres longer than 200 µm are included in the total number counts, and very thin fibres of a likely natural origin are excluded to maximise the synthetic contribution. Because dry counts on zooplankton filters were frequently hampered by large amounts of residual sediment, zooplankton net filters were rinsed with deionised water into petri dishes, and particles and fibres settled to the bottom of the petri dish were counted separately from fragments floating on the surface of the aqueous solution (*ground* and *float* number counts). Although particles and fibres floating on the surface are expected to have a higher likelihood to be composed of synthetic polymers, intensely coloured particles and fibres were routinely discovered in the ground fraction as well (as expected for nylon or polyamide with a higher specific density than deionised water). The *ground* and *float* fractions from both the pipetted and the decanted solutions of the same sediment sample were added to obtain the final particle and fibre number counts and concentrations for each sample.[1]

2.2.3. Artificial samples

Two artificially enriched samples were created by adding 200 polyethylene particles (PE) with a density of 0.9 g/cm^3 to ~500 ml of sediment with grain sizes <0.5 mm, corresponding to a sediment dry weight of 802.6 g and 743.3 g, respectively. Before enriching sediment with PE particles, microplastic particles had been extracted from Nienhagen beach March and April sediment samples as described in Section 2.2.1. The PE particles were cleaned, post-processed recycling fragments covering the approximate size range 100 µm–1 mm. Artificially enriched samples were processed and visually inspected in the same way as all other samples.

2.2.4. Beach sediment sampling

Samples of wet sand were obtained predominantly at the drift line above sea water level. All samples were obtained during calm conditions with low wave activity. Fine sediment was sampled as a larger number of microplastics were expected to be bound in the fine-grained layer than among coarse grains regularly rinsed with sea water. The majority of samples was collected at the drift line where small shell fragments were found to concentrate, under the assumption that microplastics would also accumulate here. Shallow-water samples were retrieved below the characteristic ridge of coarse gravel found a few metres below the drift line. Beach sediments at the Baltic Coast cover a wide variety of grain sizes from fine sediment <0.5 mm to large rocks. As a consequence, layers with grain sizes larger than 2 mm (coarse gravel) are found at varying height levels less than 1 cm below the sand surface inside and outside the water near the surf zone. These conditions prohibit single-height sediment cores to be extracted. With the aim to avoid the coarse gravel zone, samples were scraped off the surface layer with a stainless steel table spoon either at the drift line or were carefully spooned off the surface of sand ripples under water with the same flat table spoon. Samples were limited to the top 1–2 cm at most and frequently did not exceed 1 cm depth. Studying the stratification of sediment cores to a depth of 25 cm, Carson et al. (2011) found that 50% of mircoplastic fragments were contained in the topmost 5 cm of each core, and that the top 15 cm hosted 95% of all detected plastic particles. Similar results are found at the high-water line at a Belgian beach, where the top 16 cm layer of 32 cm cores contained 65% of all microplastics, with 40% detected in the top 8 cm layer alone (Claessens et al., 2011). We therefore expect to capture the largest concentrations of microplastics when sampling the sediment surface. Samples were collected 500 ml each into screw cap glasses. Dry weights ranged from 450 to 960 g, with 600–800 g for most samples (Table A.1).

2.2.5. Water sampling

In addition to sediment samples, three water samples were obtained for comparison with the sediment content. Seawater was sampled at Warnemünde beach, the location used for all methodical tests. In addition, one North Sea water sample was drawn at Dangast at the Jade Bay and one freshwater sample was drawn in the *Nordender Leke* in Varel near a paper recycling plant for comparison with earlier microplastic measurements extracted from water samples near these locations. For all water samples, surface water at the top layer at a depth of 2–4 cm was allowed to flow freely into 5–10 l canisters previously rinsed several times with the ambient water. These samples were filtered over 55 µm zooplankton net and treated with 30% H_2O_2 solution for 24 h to dissolve organic matter. In the case of the *Nordender Leke* freshwater sample, organic content was so high that zooplankton mesh filters were treated for a second 24 h period after rinsing with deionised water.

2.2.6. Sampling locations

Three sampling locations were chosen to represent potential entry pathways of microplastics into the Baltic coastal zone. The four beaches along the west-east current in the greater Rostock area were focused on low anthropogenic influence sites (Nienhagen/Börgerende, Wilhelmshöhe), extensive touristic activity (Warnemünde), and the Warnow outlet with city and harbour discharge (Markgrafenheide). These locations were sampled four times from March to July 2014 to probe seasonal variability. Four locations on the island of Rügen were chosen with varying levels of touristic activity without nearby city or harbour effluent. Two locations along the Oder/Peene estuary represented the inner bay of the Oder discharge (Kamminke) and the Peene outlet into the Baltic Sea near the Freest fishing harbour. In addition to these Baltic sampling sites, two locations in the Jade Bay were sampled because some of the highest levels of microplastic concentrations were reported here (Dubaish and Liebezeit, 2013). For direct comparison with this earlier study, water samples were obtained in addition to two sediment samples. Freshwater was sampled in the rivulet *Nordender Leke* at a viewing distance of ~50 m to Varel's paper recycling plant, and both seawater and sediment were sampled at Dangast beach 5 km North of Varel along the Jade coast. The latter location was chosen to probe the dispersal of microplastic flakes and fibres from paper recycling effluent emitted into the central Jade Bay.

[1] A detailed analysis of individual pipetted and decanted *ground* and *float* number counts can be found in Stolte (2015).

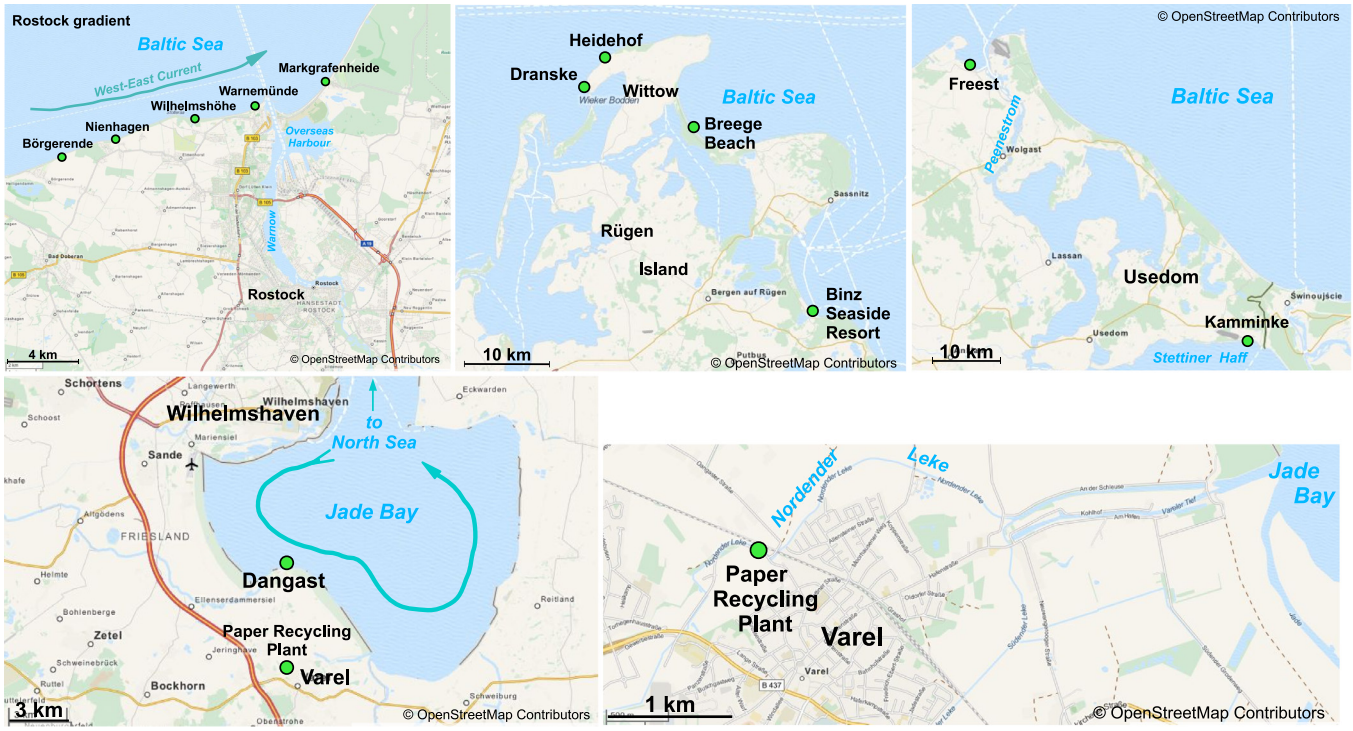

Fig. 1. Sampling locations along the German Baltic coast and in the Jade Bay. Locations cover a gradient along the west-east current of the Rostock Bay area (*top left*), the island of Rügen (*top middle*), the Oder/Peene flow (*top right*), and Dangast beach (*lower left*) as well as the Varel freshwater sampling site (*lower right*) in the Jade Bay. The flow direction of North Sea water through the Jade Bay is also shown.

All sampling locations are illustrated in Fig. 1 with dates, coordinates and results provided in Table A.1.

3. Results

3.1. Artificially enriched samples

The recovery rates of the samples where 200 PE recycling particles were added to pre-extracted sediment are shown in Fig. 2. Values represent the average recovery rate of the two samples, and error bars indicate the variation between sample 1 and 2. Recovery rates are highest for coloured particles with blue, violet, or green hues, where fractions between 60% and 100% of polyethylene pieces are recovered. Lower fractions between 0% and 40% are found for yellow, orange, and pink colours. Despite their similarity to mineral sediment, between 45% and 63% of transparent polyethylene particles are recovered on the basis of their surface structure and shape alone. Total recovery rates of coloured and transparent particles are 49% in sample 1 and 62% in sample 2. We therefore expect at least half of the microplastic particles with sizes 100 μm–1 mm to be extracted with the adopted density separation procedure, with higher extraction fractions for particles with blue or green colouring.

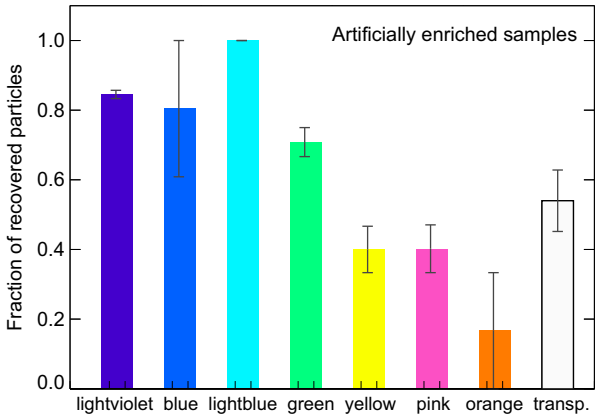

Fig. 2. Recovery fractions of artificially added polyethylene particles in sediment samples. Recovery rates are sorted by colour, and the recovery rate of transparent particles is shown as the rightmost bar. Bar heights represent the mean of both samples, and error bars indicate the variation in the recovery rate between sample 1 and 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.2. General results from sediment samples

All sediment samples contained large numbers of particles in both the pipetted and decanted solutions. In the pipetted solutions, particle numbers ranged from 10 particles to several 100 particles per sample, with typical values between 20 and 60 particles. In the decanted solutions, several 100 to several 1000 particles were extracted onto the zooplankton net filters. The vast majority of these particles were visually indistinct from natural sediment grains. Rinsing the net filters with deionised water caused about half of the extracted particles to sink immediately, while the remainder floated on the surface initially, but sank upon contact with the lancet. The majority of extracted particles are therefore indistinguishable in density and in appearance from natural sediment.

As expected from the artificially enriched samples, coloured particles and fibres stand out most prominently in the sediment samples. Both particles and fibres are dominated by intense blue, violet, green, or red colours, providing strong evidence for their anthropogenic origin. Without spectroscopic confirmation, natural anthropogenic coloured fibres cannot be distinguished from synthetic fibres here. However, long persistence times are expected

for synthetic materials, while natural fibres such as cotton or wool can be expected to dissolve under marine conditions more rapidly. Coloured particles and fibres will therefore be used as the dominant tracers for microplastic contaminations in the following discussion (Section 4).

3.3. Rostock gradient

Four seasonal samples were obtained in March, April, May, and July 2014 along four locations of the wider Rostock coast (Fig. 1). Over this time period, almost no systematic seasonal and spatial variations are observed in the fibre concentrations (Fig. 3). Total fibre counts ranged from a few to 80 fibres per sample in the pipetted solutions, and from 10 to 220 in the decanted solutions for both the fine-grained (<0.5 mm) and coarse (0.5–1 mm) size fractions. Total fibre concentrations, as derived by adding the fine-grained and coarse size fractions as well as the pipetted and decanted solutions, are shown in Fig. 3. For beaches along the Rostock coastal area, fibre concentrations ranged from 42 F/kg dry weight (DW) sediment to ~200 F/kg for most samples, with only two exceptions. In the Warnemünde samples, the July fibre counts exceeded all other months in both pipetted and decanted solutions in both size fractions. In excess of 400 fibres are counted in the combined sediment sample, leading to a fibre concentration of 532 F/kg dry sediment, which exceeds characteristic fibre concentrations in the wider Rostock area by at least a factor of two. The second highest fibre concentration is measured to be 307 F/kg in the Nienhagen April sample. The trend is not reflected in the number of coloured fibres or particles. The number of coloured fibres in the Warnemünde July sample is with 4 fibres (5 F/kg) moderate, and 3–4 coloured particles (4–5 P/kg) are observed in all Warnemünde samples (Fig. 4).

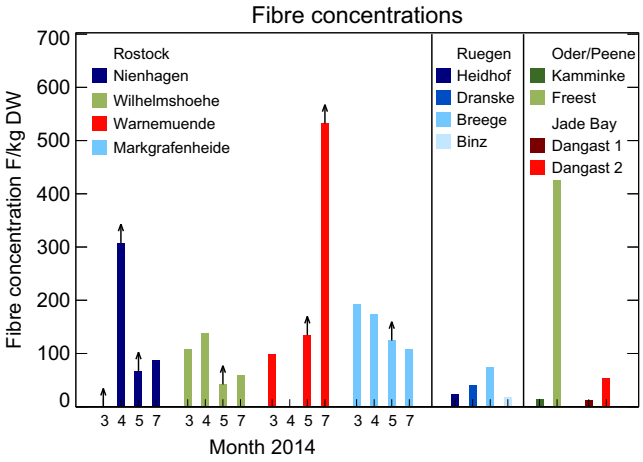

Fig. 3. Fibre concentrations in all sediment sampling locations. In the first 16 positions, the seasonal variation in the fibre concentration along the wider Rostock coast is shown with respect to the sampling month (3-March, 4-April, 5-May, 7-July). Rügen, Oder/Peene, and Jade Bay sediments were not sampled for seasonal variations. All fibres with lengths >200 µm, including transparent and coloured fibres, are included, and no distinction is made between potentially synthetic and potentially organic transparent fibres. Fibre numbers were not counted in the March Nienhagen sample, and no April sample was obtained in Warnemünde. Arrows indicate lower limits where large amounts of contaminating sediment prohibited accurate fibre counts. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.4. Fibre concentrations in all other sampling locations

Large spatial variations are observed in the fibre concentrations among the four sampling regions (Fig. 3). In contrast to the Rostock sediment samples, Rügen samples contain a maximum concentration of 73 fibres/kg in the Breege sample, with all other locations displaying lower fibre loads. The lowest overall fibre load of 14 F/kg is observed in the Kamminke sample at the inward Bay of the *Stettiner Haff*, while the second highest fibre content (425 F/kg) is found at the Oder/Peene outlet near the Freest fishing harbour. In the seawater sample drawn in August near Warnemünde beach a total fibre concentration of 3.3 F/l was observed, which appears low in comparison to the fibre concentration of 532 F/kg extracted from beach sediment at the same location in July. The Varel freshwater sample contained with 19 F/l a moderate fibre concentration, while both sediments (13–53 F/kg) and seawater (3.1 F/l) at Dangast beach did not show enhanced fibre numbers as compared to the Rostock samples.

3.5. Coloured particles and fibres

Almost all samples contained coloured fibres and 13 out of 22 sediment samples contained intensely coloured particles. In the Rostock area, only Warnemünde beach contained coloured particles in all three seasonal samples, while only the April sample contained coloured particles in the three other locations along the Rostock coastline (see Fig. 4).

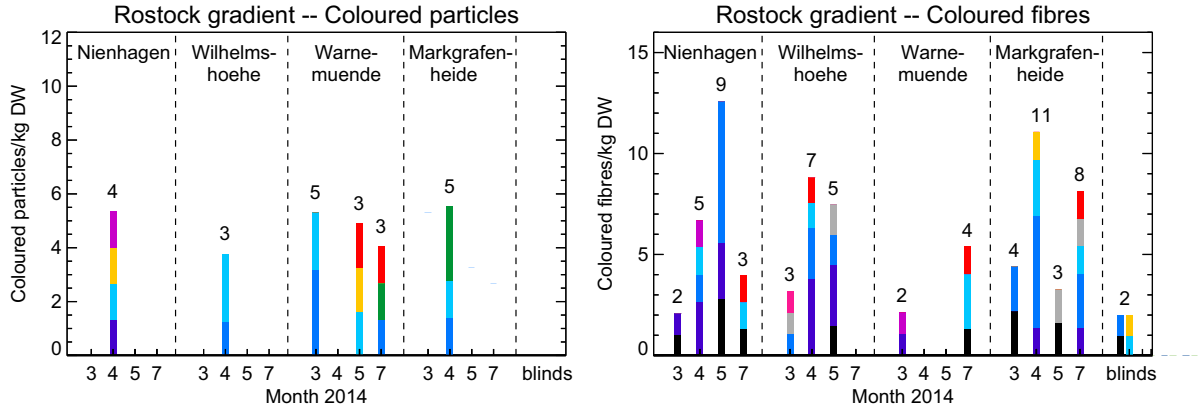

Fig. 4. Concentrations of intensely coloured particles (*left panel*) and fibres (*right panel*) observed from March (3) to July (7) at each Rostock sampling location. Each bar is labelled with the absolute number count to illustrate the number of fragments detected in each sample. Colours are approximately coded to represent the real detected colour range and contribution in each sample. No bar indicates no coloured pieces were found, except for the April (4) Warnemünde position, where no sample was obtained. Coloured particle and fibre number counts of laboratory blind samples are shown in the last bins to the right in each panel. No coloured particles were observed in the reference samples. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

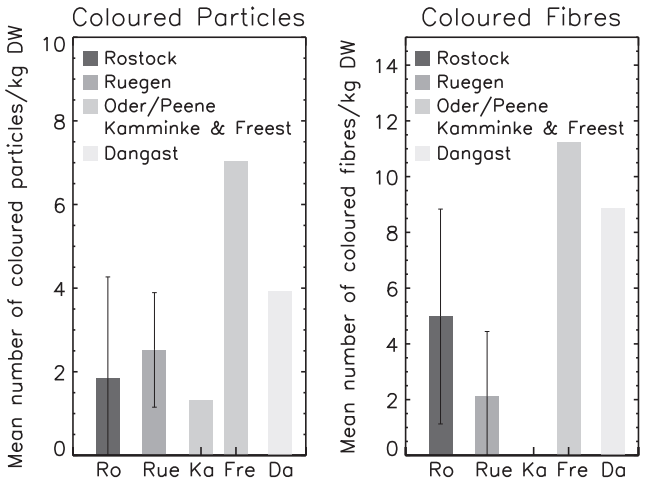

Fig. 5. Mean concentrations of coloured particles and fibres observed in each sampling area with standard deviations where more than one measurement was available. No coloured fibres were observed in the Oder estuary sample collected at Kamminke beach.

Coloured particle concentrations in all samples range from 0 to 7 P/kg DW sediment, and coloured fibre concentrations range from 0 to 11 F/kg DW (Fig. 5). Particularly low coloured particle and fibre concentrations are observed with 1 P/kg and 0 F/kg in Kamminke along the inland side of the Oder flow through the *Stettiner Haff*, while the highest concentrations are detected at the Oder/Peene outlet into the Baltic Sea near Freest. A large coloured fibre concentration of 9 F/kg DW is also observed at Dangast beach to the North of the Jade outlet near Varel. All four Rügen locations contained 2.5 ± 1.5 P/kg, while fibre counts were consistent with the laboratory background level of two coloured fibres per sample. Coloured particle and fibre concentrations are with 2 ± 2 P/kg and 5 ± 4 F/kg moderate in the Rostock area, and similar to the particle concentrations found at Rügen locations.

In addition to coloured microplastics, green glass pieces are regularly detected among sediment samples. The fragments feature smoothed surfaces and characteristically round shapes. While only a few glass pieces were observed in all Rostock and Rügen samples, the Freest sediment sample contained at least 206 glass fragments (Fig. 6), or a concentration of 290 glass pieces per kg dry sediment.

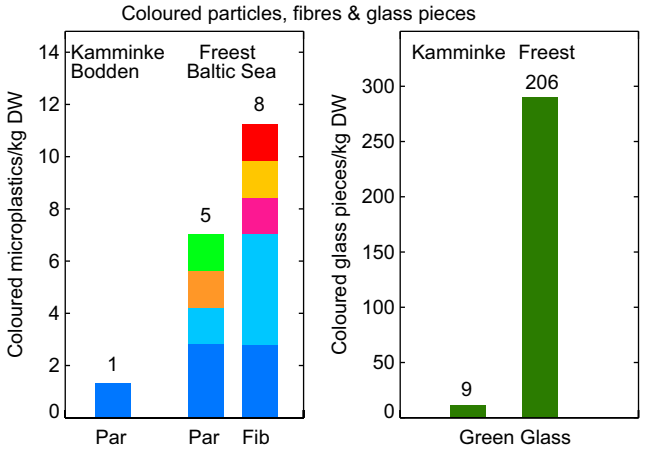

Fig. 6. Concentrations of coloured particles, fibres (*left*), and glass fragments (*right*) detected in the Oder/Kamminke (*left bars*) and Peene/Freest (*right bars*) samples, respectively. Bars are labelled with absolute numbers of detected pieces, and bar colours indicate the colours of discovered particles and fibres. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

This concentration is contrasted by only 12 glass pieces/kg in the Kamminke sample.

3.6. Seawater and freshwater samples

Coloured particle and fibre concentrations extracted from sea- and freshwater samples are shown in Fig. 7. The concentration of coloured fibres is with 5 F/l particularly high in the freshwater sample extracted from Varel's *Nordender Leke* near the paper recycling plant. The seawater sample drawn at the surface at Dangast beach contains only 0.8 coloured fibres per litre, and even lower concentrations of 0.4 coloured F/l are detected in Warnemünde surface water. In contrast to the coloured fibre concentrations, between zero and one coloured particles are found in the water samples, and the concentrations are correspondingly low (Fig. 7).

4. Discussion

4.1. Fibre concentrations

Transparent fibres are particularly numerous in the July Warnemünde sample, while coloured fibres are only moderately enhanced, suggesting rapid bleaching under the influence of UV-intense solar radiation. The origin of the exceedingly high increase in the fibre load is likely seasonal touristic activity. Warnemünde is the most frequented beach in the Rostock area, and a major tourist destination as well as a major place for weekend recreation for the local communities. Visitor numbers in Warnemünde are on the order of one million beach guests in the vacation season, with July and August being the peak months (Statistisches Amt der Stadt Rostock, 2013), consistent with the exceptionally high fibre concentration observed in the July sample. Despite similar visitor numbers in the seaside resort of Binz, comparably high fibre concentrations are not found at beaches on Rügen island. A reason for this discrepancy might be the water current around the island carrying off microplastic influx more rapidly than in the shallow, semi-enclosed Warnemünde Bay. Establishing a correlation between water currents and fibre concentrations was beyond the scope of this study, but would be a valuable investigation in the future.

An unexpectedly high fibre concentration of more than 300 F/kg was observed at Nienhagen beach in the April sample (Fig. 3). This

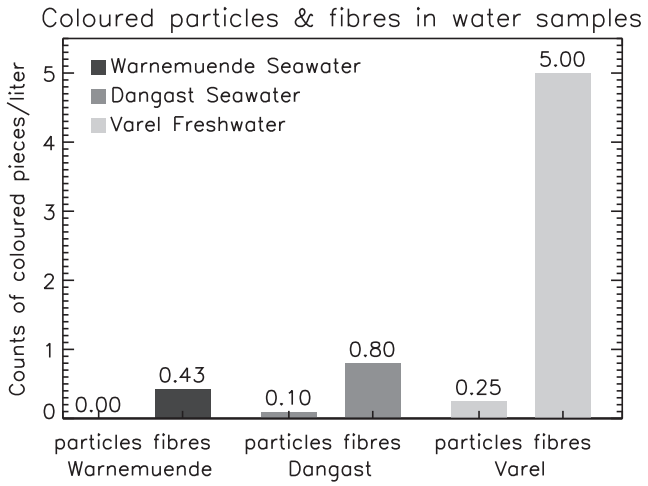

Fig. 7. Numbers of coloured particles and fibres per litre in seawater and freshwater samples. The freshwater sample in Varel was drawn from the canal *Nordender Leke* near a paper recycling plant, while the Dangast seawater sample represents a beach location northward of Varel in the Jade Bay (see Fig. 1).

location was chosen specifically as a low anthropogenic influx site. A possible origin of fibres is the artificial reef located 1.5 km into the Baltic Sea from the Nienhagen coast. The reef is built from a combination of concrete structures and net material (http://www.riff-nienhagen.de), and the mesh netting could be a continuous source of disintegrating fibres, which might lead to particularly enhanced fibre loads after the stormy spring season. As chitin-based fibres and synthetic fibres both persist treatment with H_2O_2, increased natural activity in zooplankton/crustaceans and insect species in the spring season cannot be excluded as the origin for the large fibre contents in the April samples at the present time.

A surprising result is that the fibre concentrations are with 100–200 F/kg in Markgrafenheide only moderately enhanced. The effluent of the Warnow with its sewage treatment plant discharge as well as all discharges from the overseas harbour passes on the easterly current from the Warnow outlet directly in front of Markgrafenheide (see Fig. 1). Further analysis of Warnow water samples and direct samples of the sewage treatment plant effluent would be valuable to compare these fibre concentrations with the discharge contamination.

The second highest fibre content is measured in the Peene estuary sediment sample with 425 F/kg obtained near Freest fishing harbour. With 5 coloured particles and 8 coloured fibres, the unambiguous microplastic contamination is also high in Freest sediments. Substantial fibre loads, coloured particles, and the large number of green glass fragments (see below) render the Freest sample the most evidently contaminated sample with anthropogenic particle and fibre influx.

4.2. Coloured particles and fibres

Almost all sediment samples contained coloured particle or fibre loads (Fig. 5). Coloured microplastic particle contamination is found to be low in 500 ml samples, ranging from 1 to 5 particles, with most samples featuring 1–3 coloured synthetic particles. Coloured particle concentrations range from 3.8 to 5.5 P/kg DW at all Rostock locations, and the maximum particle concentration is observed at the Peene outlet near Freest with 7 P/kg DW. Although number statistics are small, the fact that all Warnemünde samples contain coloured microplastics strengthens the strong anthropogenic influx of synthetic materials at this most touristic Rostock beach. While the transparent fibre load is found to be exclusively large in the July seasonal sample, coloured fibre concentrations are with 2–5 F/kg not enhanced compared to the other three Rostock locations, where concentrations up to 12.5 F/kg are observed (Fig. 4), similar to the large coloured fibre concentration near the Freest fishing harbour (Fig. 6). Coloured particles are persistently present in all seasons at Warnemünde beach, while they are only detected in the April samples at the less frequented Rostock beaches. This suggests that the influx of fragments/particles might be particularly large at Warnemünde beach, and possibly that the transport mechanisms for fibres are different than for particles, which needs to be investigated further. Beach activity and waste, frequently seen at the Warnemünde drift line, might contribute to a higher influx of particles. Whether the large concentration of transparent fibres is of anthropogenic synthetic origin or organic material needs to be investigated with spectroscopic material identification.

The Freest beach at the Peene outlet with 7.0 P/kg DW and 11.3 F/kg DW stands out in both coloured particle and fibre concentrations. The large concentration of 290 glass pieces/kg in Freest sediments yields additional evidence for severe influx of anthropogenic materials into the Baltic at the Peene outlet. A potential origin for the high glass concentration is tentatively suggested to be the Freest fishing harbour. The harbour hosts 54 registered small-scale fishing vessels (Fischereigenossenschaft Peene/Freest 2010), and glass bulbs were common for net floatation in the 1960s and possibly later in small family-run enterprises. The remnants of shattered glass spheres could contribute to the large load of fragments in nearby beach sediments. Likewise, the fishing industry might contribute to the comparatively large concentration of coloured and transparent fibres from netting material. With no coloured fibres and only one coloured particle, the lowest microplastic contamination is observed at Kamminke beach along the Oder flow inside the *Stettiner Haff*. This finding is difficult to reconcile with the large anthropogenic contamination observed near the Peene/Oder outlet at Freest, and supports the hypothesis that the fishing harbour contributes to the particle and fibre load. Further studies are required to distinguish the fishing industry contribution from potential synthetic fibre contamination transported in the Oder/Peene runoff with Szczecin city discharges into the Baltic, or to unambiguously conclude on other major sources of anthropogenic contamination in the Oder/Peene estuary.

4.3. Comparison between Baltic beaches and the Jade Bay

With 20 coloured fibres (5 F/l, Fig. 7), the freshwater sample obtained in Varel's *Nordender Leke* near a paper recycling plant contains the largest coloured fibre load of all samples. As the plant does not discharge into the rivulet at a viewing distance of 50 m to the paper stacking court, the origin of the large influx is most likely wind carriage. Fibres, including synthetic fibres from acrylic colour layers, are a natural byproduct of paper recycling due to the shredding process. Hydrogen peroxide is a potent bleaching agent that acts destructive to cellulose. The 24 h treatment with hydrogen peroxide in all our samples is expected to either dissolve or bleach cellulose paper fibres. The observed coloured fibres are therefore likely of a synthetic composition. In contrast to the freshwater sample, the seawater and sediment samples obtained at Dangast beach ~5 km North of Varel contained coloured fibre and particle loads only moderately enhanced as compared to Rostock and Rügen samples (Figs. 5 and 7). Transparent and coloured fibres and coloured particles were lower in the Jade Bay samples than in Freest sediments at the Oder/Peene outlet. As touristic activity is limited at Dangast beach, and as the North Sea current circles from North to South along the West side of the Jade Bay (Fig. 1), Wilhelmshaven city and harbour discharges are a likely source of the observed microplastic contamination.

4.4. Comparison to previous studies

Comparison studies are primarily selected on the basis that extraction and counting methods are comparable to the methods presented here. For scientific merit, only studies in the North Sea and Baltic areas are selected, as conditions in terms of ship traffic, touristic activity, waste treatment, and fisheries are assumed to be more comparable between the North Sea and the Baltic Sea than between the Baltic and the large oceans. In addition, both seas are too small for the built-up of gyres capturing plastics, although smaller eddies might contain plastic waste for the limited time of their existence (Omstedt et al., 2014). The microplastic concentrations reported in the text below are summarised in Fig. 8.

This is one of the first systematic studies of microplastic content in beach sediments at the Baltic Coast. The closest region probed for microplastics previously was sampled by boat between the Danish and the Norwegian coast. Norén and Naustvoll (2011) counted coloured or structured particles with sizes 10–500 μm in seawater samples across a Skagerak transect. Their counting procedure using visual inspection and unnatural properties of particles was similar to the procedures established in this work. They discovered blue particles in 15 of their 17 seawater tows. FTIR

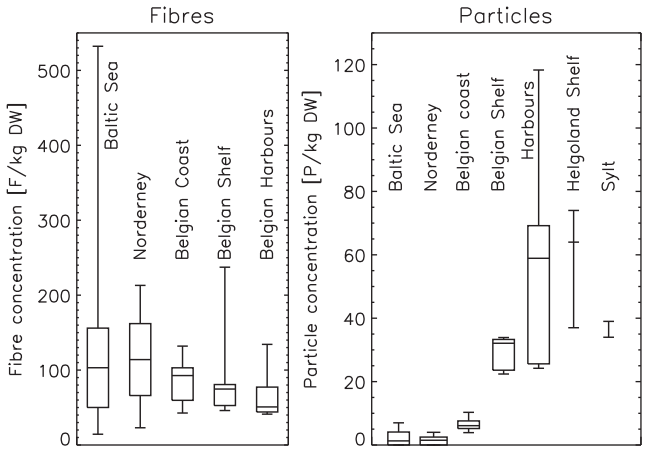

Fig. 8. Comparison of potential microplastic concentrations in sediments measured in the North Sea and at beaches along the German Baltic Sea coast. Total fibre concentrations including both coloured and uncoloured fibres are shown in the *left panel*. The *right panel* summarises the concentrations of particles confirmed to be microplastics either spectroscopically or on the basis of their colour. Boxes denote the median (central line) and the 25% and 75% quartiles, while whiskers indicate the minimum and maximum value reported for each location. For Baltic Sea values (this work), the median and quartiles are derived from all ten Baltic coast locations. Beach sediments were sampled at Norderney (Dekiff et al., 2014), Sylt (Lorenz, 2014), and the Belgian coast (Claessens et al., 2011). Studies on the Belgian Continental Shelf, Belgian Harbours (Claessens et al., 2011), and the Helgoland Shelf (Lorenz, 2014) report concentrations in sublitoral sediment. For the Norderney fibre concentration, only mean and standard deviation (box and central line), minimum and maximum concentrations (whiskers) of colourless fibres are provided (Dekiff et al., 2014). The Helgoland shelf and Sylt contain only three and two values, respectively, such that no quartiles could be derived.

analysis indicated that the particles were epoxy-based paint flakes as used for ship hull sealing. With sizes of 30–70 µm, the particles depicted in their Fig. 5 are similar in shape and appearance to 7 tiny blue fragments detected in our sediment and seawater samples. We therefore tentatively conclude that the tiny blue fragments might also be paint flakes with ship paintings being a likely origin.

A second study using air-venting in saline solution counted microplastics in beach sediments at the North Sea island of Norderney (Dekiff et al., 2014). In their study, the means of visual selection of microplastics on the basis of colour and structure/morphology were particularly similar to the selection choices employed here. Dekiff et al. (2014) distinguish particles and fibres as colourless and intensely coloured, and found between 23 and 213 colourless fibres/kg dry sediment with a mean of 114 ± 48 F/kg, which covers the same range and large variation as observed in Baltic sediment samples (Fig. 8). For coloured fibres, Dekiff et al. (2014) found 4–25 F/kg with a mean of 16 ± 4 F/kg, which in their study was indistinguishable from laboratory blank fibre counts. In our Baltic Sea beach samples, between 2 and 11 fibres/kg dry sediment are found, with a substantial number of samples well above the level of our blanks, where a maximum of 2 fibres per blank is observed. In three different beach locations on Norderney, Dekiff and colleagues found a total of 59 potential microplastic particles in 26 of their 36 samples, implying that 72% of the beach samples contained microplastic particles identified on the basis of colour and structure. We observe coloured potential microplastic particles in 12 out of 23 samples, or slightly more than 50% of our samples. Numbers range from 1 to 7 coloured particles/kg dry sediment in our samples (Fig. 5). Dekiff et al. (2014) report 1–2 particles/kg with a maximum of 4 particles/kg detected, covering a similar range of particle concentrations as compared to the Baltic beach sediments. In their study, the authors were able to confirm 15 particles with gas chromatography as polymers with PP, PE, and PET being the dominant contributors, giving high confidence to the adopted selection procedures. The similarity of North Sea island and Baltic beach sediments is surprising in view of the fact that numbers as high as hundreds to thousands of microplastic particles were claimed in the comparable island location of Kachelotplate at the North Sea coast (Liebezeit and Dubaish, 2012). While the island of Norderney might be particularly pristine in view of the west-east current and the moderate touristic activity, it is also one of the most comparable locations to the more remote Rostock and Rügen beaches investigated here.

Using colour as the major distinction criterion for microplastics, the resulting concentrations are lower limits of the true microplastic contamination in each sample. Claessens et al. (2011) analysed a large number of beach and subtidal harbour sediment samples along the Belgian coast with a lower size limit of 38 µm. Detected microplastic particles and fibres were confirmed spectroscopically. Consistent with the observations presented above, they found that the majority of all synthetic pieces were fibres (59% fibres as compared to 25% granules). Polystyrene microspheres constituted as much as 12% of their micropieces and were exclusively found in harbour sediment. Harbour microplastic concentrations exceeded with a mean of 167 ± 92 pieces/kg dry weight the concentrations observed in beach sediment (93 ± 37 pieces/kg) and in offshore locations on the Belgian Continental Shelf (97 ± 19 pieces/kg), by almost a factor of two. These numbers included particles, fibres, microspheres and films, and spectroscopic verification allowed the inclusion of transparent pieces as well. In beach sediments, Claessens et al. (2011) found on average 82 ± 33 fibres/kg and 6.3 ± 2.5 plastic fragments/kg with sizes >38 µm. A large variation of 43–132 fibres/kg is also observed in the fibre content, while concentrations for fragments range from 4 to 10 P/kg. The amount and variation observed in total fibre and plastic fragment concentrations in Belgian beach sediments are comparable to the total fibre and coloured particle concentrations in the Baltic and North Sea Jade Bay sediments analysed here.

Claessens et al. (2011) compare the increase in microplastic concentrations derived from beach sediment cores on a timebase of 15 years (1993–2008) to the increase in annual global plastic production (see their Fig. 2), and conclude tentatively that a correlation between the global plastic growth rate and the deposited microplastics might be present. Microplastic concentrations are shown to have tripled in beach sediments from 55 to 156 pieces/kg (including fibres, granules, foil, and spheres) over just 15 years. The highest concentrations of microplastic particles are observed in three harbour locations with recreational or industrial activity. Here, particle numbers are with 24–118 P/kg one order of magnitude higher than in beach sediments (4–10 P/kg). Claessens et al. (2011) suggest that microplastics might be trapped in the harbours due to the enclosed geometry. On a larger scale, such trapping might affect the Jade Bay samples investigated here, leading to increased microplastic loads in the bay area, as discussed above. As in most of the Baltic coast samples, no clear correlation is found between human activities and microplastic content when beach sediments and offshore sediments along the Belgian coast and continental shelf are compared. This suggests that microplastics are more uniformly distributed by natural forces with time than macroplastics, where accumulation on beaches near urban and recreational areas is more often observed, stressing the necessity to sample the spatial and temporal distribution of microplastics individually. Individual monitoring of microplastics is emphasised by a comparative study of the water surface, beach sediments, and the seafloor, where the accumulation of microplastics in seawater and on the seafloor is found to exceed the weight of macroplastics by a factor of 100 and 400, respectively (Van Cauwenberghe et al.,

2013). The same authors estimate that a concentration of just 13 particles/kg dry sand correspond to a total microplastic load of 3.3×10^6 to 7.7×10^7 particles on a beach extent of just 100 m. With particle concentrations only marginally lower in the Baltic coast samples presented here, beach sediments must be considered an important entry path of microplastics into the marine and coastal environment and food chain.

The size range of particles targeted in our study was 55 μm–1 mm, where the lower limit was fixed by the mesh size of the zooplankton net used for filtration and the upper limit was set by the majority of sediment grains by weight. As the fine-grained sediment surface layer at the drift line or in shallow water was collected, the 1–2 mm sieve contained at most a few grammes of coarser grains, which did not contain visible plastic particles and were not considered in the analysis. The largest fraction by weight was composed of grains <500 μm in all samples with the exception of the Markgrafenheide beach sample, where the grain size range 500 μm–1 mm was most common, such that the maximum grain size included in the analysis was 1 mm. In seawater samples drawn with plankton net tows, mesh sizes frequently range from 333 to 450 μm, such that most of the seawater findings are not directly comparable to sediment studies. Studying the amount of small plastic particles in seawater in 16 locations along the Swedish west coast, Norén (2008) found microplastic numbers to be steeply increasing with decreasing sampling size. A 1000–100,000 times higher concentration of particles plus fibres was found with 80 μm mesh samples as compared to a 450 μm mesh width.

With 80 μm mesh, concentrations of 0.15–2.4 plastic P/l of seawater are detected, comparable to the concentrations of coloured particles and fibres found in seawater samples at both Warnemünde and Dangast/Jade Bay (Fig. 7). Three harbour sites were sampled by Norén (2008) for sediment, one industrial harbour and one commercial harbour near Stenungsund, and one small harbour near Tjuvkils. Stenungsund industrial harbour was sampled near a plastics production plant and yielded by far the highest microplastic concentrations in water and sediment. Two to five milkwhite or transparent plastic particles per 100 ml sediment with sizes 1–7 mm are found in the small harbour of Tjuvkils, while 332 microspheres with sizes 0.5–1 mm were counted in Stenungsund industrial harbour per 100 ml sediment. Concentrations hence range from 20 particles/kg (conversion factor 1.6 g sediment/1 ml volume from our own wet sample experiments) to more than 2000 particles/kg sediment. Similarly high concentrations were reported in the studies of Jade Bay sediments and seawater for particles down to a few micrometre in size near the paper recycling plant by Dubaish and Liebezeit (2013). Despite the inherent uncertainties with visual identification, the alarmingly high concentrations of microplastic particles, spheres, and fibres found near industrial discharge sites call not only for more consistent scientific monitoring of a representable range of locations in Europe, but also require the rethinking of production discharge practices in view of the EU Marine Strategy Framework Directive to reduce microplastic particle entry into the marine environment in the future.

4.5. Discussion of problems and biases

4.5.1. Grain floatation by surface tension

The fact that hundreds of the particles extracted with the adopted density separation method float on the surface of deionised water after rinsing off the filters suggests that sediment grains are supported by surface tension. Both the Warnemünde and the Dangast seawater samples contained substantial amounts of 69 and 450 particles as well. Both samples were drawn from the surface in shallow water near the beach. Finding sediment fragments on the water surface strengthens our conclusion that microplastics and sediment particles are not unambiguously separated on the basis of their floatation properties. If these particles are kept afloat by surface tension, the force exerted by the water surface has to balance the gravitational pull causing high-density particles to sink. The gravitational force of a single particle, assumed to be spherical for simplicity, is given by

$$F_g = g\,V\rho = g\,\frac{4}{3}\pi r^3\rho \tag{1}$$

where g is the gravitational acceleration (9.81 m/s), ρ the particle's density, and V and r the particle volume and radius, respectively. Gravity is counterbalanced by the force exerted by surface tension, which acts to minimise the energy E and hence the area: $\Delta E = \sigma \Delta A$, where σ is the surface tension and ΔA is the change in area (GerthsenPhysik, pp. 100–101). For a spherical particle:

$$\Delta E = \sigma\, 2\pi r\, \Delta h \tag{2}$$

where r is the radius of the sphere touching the surface, and Δh is the height of the sphere segment with surface area $\Delta A = 2\pi r \Delta h$ causing the indentation. Here, the surface area of the sphere segment is the area supporting the particle, and hence is used to approximate ΔA in the energy balance (see Appendix A). The surface tension force, $F_s = \Delta E/\Delta h$, acts as the force parallel to the surface of contact between the particle and the water. The vertical component of F_s counterbalances the gravitational force and increases with the angle of indentation:

$$F_s = \sigma\, 2\pi r \sin\theta \tag{3}$$

where θ is the indentation angle between the object and the horizontal water surface, and σ, the surface tension of the liquid, is 72.7×10^{-3} N/m for water at 25 °C. For natural sediment, a characteristic density of 2.5 g/cm^3 (Nuelle et al., 2014) and a particle radius of 100 μm are assumed, consistent with observed grain sizes. With these values, the gravitational force of a single sediment particle is estimated to be $F_g = 1.0 \times 10^{-7}$ N. Assuming an indentation angle between the water surface and the surface of the sphere of 30°, the surface tension exerts a force of $F_s = 2.3 \times 10^{-5}$ N on the particle. Thus, the suspending force due to water surface tension is two orders of magnitude larger than the gravitational pull of particles with a radius of 100 μm and a density of 2.5 g/cm^3. Note that the suspending force depends linearly on the size of the particle, while the gravitational force scales as r^3 (Fig. 9). With the same simplified assumptions, the gravitational force exceeds the surface suspension force for a particle with a radius of 1.5 mm ($r_{lim} = \sqrt{3\sigma \sin\theta/(2g\rho)}$), causing larger particles to sink. Given these considerations, the assumption that particles floating on the surface are of a lower density than water and hence have to be plastics needs to be revised. A very low surface tension appears to be sufficient to keep mineral grains floating for an extended period of time after being stirred into the water column from the bottom, e.g. by wave motion, touristic or boat activity. This conclusion implies that more sophisticated techniques are required to unambiguously distinguish microplastics from natural sediment even in seawater (or freshwater) samples. The major conclusion from this estimate is that natural sediment grains with diameters $\lesssim$1 mm are also expected to float on the surface of the aqueous solution after rinsing of the zooplankton net filters. The fact that smaller particles are more easily supported by surface tension is consistent with increasing numbers of particles extracted from the finer-grained sediment samples in Dangast at the Jade Bay. Caution must therefore be taken to avoid counting natural sediment as microplastics especially in fine-grained sand.

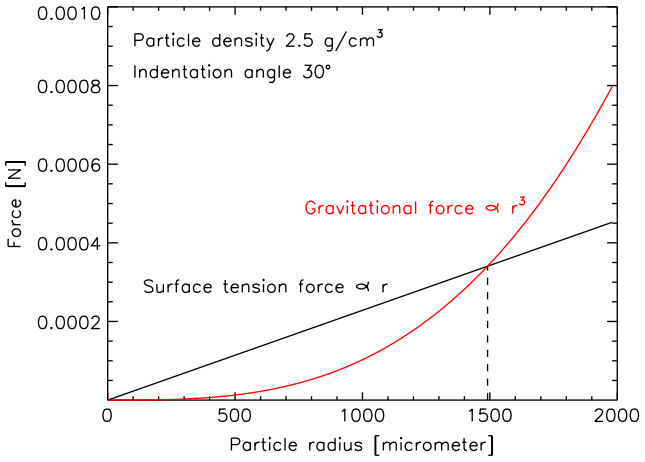

Fig. 9. Gravitational force and force exerted on the water by surface tension for particle sizes from 100 μm to 2 mm. The particle density is assumed to be 2.5 g/cm^3 and the indentation angle of the spherical particle with the horizontal water surface is assumed to be 30°.

Because a distinction between floating natural sediment grains and microplastic particles could not be made with the given setup of the experiment, total particle numbers were not discussed above.

4.5.2. Biases by fine-grained sediment

Previous studies by Dubaish and Liebezeit (2013) of seawater in the *Vareler Tief* at the outlet of the discharge pipeline of Varel's paper recycling plant found in excess of 1200 potential plastic particles per litre of seawater. Fibre counts reported near Varel and Dangast ranged approximately from 300 to 900 F/l (Dubaish and Liebezeit, 2013, see their Fig. 5). These very high fibre and particle loads are not confirmed in our samples. One reason for this discrepancy might be the fact that we counted particles down to sizes of 55 μm and fibres above a length of ~200 μm, while fibres as small as a few micron could be counted with up to 80× magnification by Dubaish and Liebezeit (2013). Sampling at offshore locations with higher wave activity and strong mechanical stirring by the pipeline discharges, as well as the time of the day and year, tidal effects, and weather patterns might influence the concentrations. When included in the particle count, sediment-like particle concentrations are also high in our samples. In 10 l of surface seawater, we detect 451 particles or 45 P/l. Most of these particles have the visual appearance of the ambient natural sediment, suggesting that the small silicate grains were suspended on the water surface by surface tension. The fine-grained, clay-like sediment in the Jade Bay might be particularly prone to floatation. As discussed in Section 4.5.1 and shown in Fig. 9, a correlation of floatation and size scale is expected if particles are suspended by surface tension. Furthermore, the surface tension in North Sea water at a measured salinity of 30.7‰ might be sufficient to suspend fine-grained silt for extended periods of time, which would explain the large particle numbers found in seawater samples at the pipeline discharge site. This problem has to be addressed further before final conclusions on the potential contamination with microplastics in the Jade Bay can be made.

4.5.3. Colour selection bias

The artificially enriched samples have shown that particles with blue, green, and intense red hues are particularly easy to detect among residual sediment because of their strong colour contrast. This colour detection bias is a likely cause for the large fraction of these particles and fibres found in all samples. Blue particles and fibres contribute by far the largest fraction of coloured pieces (see Figs. 4 and 6). Counting all coloured particles in all sediment samples (including the Rügen samples with few colour pieces present), a total of 36 coloured particles are detected, of which 33 (92%) are composed of blue (24), green (4), and intense red (5) materials. Recovery rates above 70% for green particles and of 80–100% for blue particles (see Fig. 2) suggest that these colour ranges are almost completely detected. As expected, lightly coloured particles are much more difficult to discern in residual natural sediment. Low recovery rates of between 16% and 40% are observed for particles with yellow-orange hues and for transparent pieces. Only 3 particles (8%) show yellow-ochre hues in all sediment samples. With recovery rates of at most 40% for yellow particles, it has to be assumed that at least 60% of lightly coloured particles evaded visual detection. Such a correction factor would imply that at least 5 particles were missed, leading to a total of 7.5 lightly coloured particles. While this bias appears low, the true correction factor is likely larger, as a wider range of naturally coloured plastic materials might be found, including brown, ochre, beige and black hues not present in the PE recycling fragments used to derive extraction efficiencies.

A similar bias is found for the coloured fibre detections. Among a total of 56 coloured fibres in all Baltic sediment samples, 46 are blue, none is green, and 10 are intensely red, pink or violet. Only 2 yellow and 5 grey fibres are found, in addition to 8 black fibres. The fraction of lightly coloured fibres is therefore 12.5%, and if the same 60% correction factor could be applied as for particles, 17 lightly coloured fibres instead of 7 would be expected. From the large number of transparent fibres observed in all Rostock samples and in the Freest sample, the bias in the number of lightly coloured fibres is likely larger than the bias derived for particles. Especially bleached colours are harder to discern during visual inspection given the filamentary structure of fibres.

The true bias in the coloured particle and fibre detection rate can only be quantified with a substantial suite of laboratory experiments using artificially enriched samples with microplastics of different shapes, colourings, and materials. As a consequence, the concentrations reported above have to be considered lower limits to the true microplastic contamination in Baltic beach sediments, as already indicated by the results of the artificial samples (Section 3.1).

4.5.4. Advanced detection methods

The observed difficulties in the distinction of microplastics from natural sediments and organic fibres have likely affected studies that were based on the visual inspection of samples under the microscope. At the same time, photo-induced bleaching and the plastic production process cause substantially larger numbers of transparent and lightly coloured microplastics to be expected in the marine environment. A complete microplastic extraction with more accurate contamination rates requires micro-imaging spectroscopy of transparent floating particles, as conducted in Lorenz (2014). In beach sediments from the island of Sylt and sublitoral sediments from the wider Helgoland shelf, Lorenz (2014) found 34–74 particles/kg dry sediment (Fig. 8). In her study, imaging microscopic spectroscopy (μFTIR) was applied to identify the materials of all particles extracted after zinc-chloride floatation. These values are about one order of magnitude larger than particle concentrations detected in the Baltic sediment samples by visual inspection, suggesting that only 10% of the particles might be detected with colour selection alone. However, this technology is costly and analysis is time-consuming, such that more practical solutions for plastic-sediment separation have to be developed. Preliminary experiments suggest that adding a centrifugation step after density separation in saline solutions improves the plastic-sediment separation efficiency (Stolte, 2015).

Centrifugation provides a simple means to separate real low-density particles from suspended higher-density sediments with standard laboratory equipment. Centrifugation was already suggested by Claessens et al. (2013) as a subsequent step after floatation to improve the extraction efficiency of microplastic fibres and granules from sediments. Claessens et al. (2013) combined a first elutriation/floatation step by fluidising sediment in a water flow of 300 l/h during air-venting with a second centrifugation step to extract microplastics in sodium-iodine solution (density 1.6 g cm^{-3}) from the small amounts of residual sediment after elutriation. Using artificially enriched samples similar to the procedures employed in Section 3.1, they find extraction efficiencies of 68–97% after two to three consecutive centrifugations. This procedure is similar to the combination of air-venting extraction and centrifugation suggested from preliminary testing here (see Stolte, 2015 for details), and hence is expected to lead to high extraction rates of microplastic particles and fibres (Claessens et al., 2013). To avoid additional biases, special care has to be taken that extracted surface fragments are not lost during refilling into centrifugation tubes as a consequence of adhesive forces at the container walls (see discussion in Imhof et al., 2012). For the distinction of organic and synthetic fibres, however, this method would not be sufficient. Here, adding a chitinase digestion step would allow a cleaner sample of transparent synthetic fibres to persist (Lorenz, 2014). With these concerns in mind, we focused the microplastics analysis on coloured particles and fibres. The reported concentrations per kg of dry weight sediment or per litre of seawater are therefore strict lower limits of the true microplastics contamination in Baltic Sea beach sediments and surface waters.

5. Summary and conclusions

In beach sediments along the German Baltic coast, microplastic concentrations of 0–7 particles/kg dry sediment and 2–11 fibres/kg are found. These values are strict lower limits, as selections were based on intensely coloured pieces. Both coloured particle and fibre concentrations were largest in the Peene outlet into the Baltic Sea and in the Jade Bay. Combined with similar findings in previous studies, nearby industrial discharge sites (paper recycling plant, sewage treatment plants) and harbour areas can be identified as major entry pathways for microplastics. Coloured particle and fibre sediment concentrations were comparable to measurements obtained on the North Sea island of Norderney and at Belgian beaches, and were one order of magnitude lower than comparable observations on the island of Sylt and in the area of the Helgoland shelf and Belgian harbour and shelf locations. While this renders the Baltic coast one of the less contaminated areas for microplastics, it has to be kept in mind that the Baltic proper serves as a sink for pollutants. At the same time, the fact that almost all beach samples contained several unambiguously identified microplastic particles or fibres implies that microplastic contamination is omnipresent along the German Baltic coast, and must be considered as a contaminant of the marine food web. The comparably high concentrations of particles in the size range of zoo- and phytoplankton measured in estuarine environments are particularly critical as these locations serve as breeding and development grounds of juvenile fish and other species.

The total fibre concentration including both transparent and coloured fibres showed a dramatic increase during the summer season in the month of July at Warnemünde beach, suggesting that touristic activity has to be considered a major entry path for potentially synthetic fibres as well. Further studies are required to derive unambiguous seasonal and spatial trends in the microplastic contamination along the German Baltic coast. In particular, spectroscopic characterisation of selected sediment samples after extraction would be beneficial to include transparent particles and fibres and hence identify a more complete microplastic census as well as the chemical composition of microplastics for further clues on their origins and the major entry pathways into the Baltic Sea environment.

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Appendix A. Sampling locations, dates, fibre and particle concentrations

All sampling locations, sampling methods (sediment, sea- or freshwater), sampling dates, sample sediment dry weight or water volume are provided in Table A.1. All samples without explicit mentioning of the sampling method were sediment samples. The last three columns contain the concentrations of transparent fibres, coloured fibres and coloured particles after air-venting extraction in $CaCl_2$ solution and visual inspection under the dissecting microscope.

Appendix B. Calculation of the area change of a sphere indenting a water surface

For simplicity, we have made the assumption that the supporting water surface area represents the change in area contributing to the change in potential energy ΔE that leads to the suspension force. This simplifying assumption allowed the immersing depth Δh of the sphere to cancel in the equation of the suspension force, rendering the result independent of the indentation depth.

More precisely, the change in area would only be given by the difference between the sphere mantel area and the base of the mantel piece:

$$\Delta A = 2\pi r \Delta h - \pi a^2 = \pi(a^2 + \Delta h^2) - \pi a^2 = \pi \Delta h^2$$

where a is the radius of the sphere segment with height Δh causing the indentation of the surface. This treatment leads to

$$F_s = \Delta E/\Delta h = \sigma \Delta A/\Delta h = \sigma \pi \Delta h$$

where Δh remains in the force equation, and the surface tension cannot be estimated from the radius of the sphere alone. If the additional assumption is made that the sphere sinks approximately half into the water surface, or $\Delta h \approx r$, the surface tension force results in $F_s = \sigma \pi r \sin\theta$, which in comparison to Eq. (3) is exactly half the force estimated above. Hence, the approximated surface tension

Table A.1
Concentrations of fibres, coloured fibres and coloured particles per kg dry weight in all sediment samples, and per litre in sea- and freshwater samples.

Location	Date	GPS coordinates degrees	Sampling location	Amount gDW or litre	All fibres N_F/kg	Coloured fibres N_F/kg	Coloured particles N_P/kg
Rostock sediment samples							
Nienhagen	31-03-2014	54.166184 11.962618	Drift line	957	–	2.09	0.00
	23-04-2014		Drift line	747	307.69	6.69	5.35
	25-05-2014		Drift line	715	67.13	12.59	0.00
	26-07-2014		Drift line	759	86.96	3.95	0.00
Wilhelmshöhe	31-03-2014	54.176462 12.026697	Drift line	937	108.86	3.20	0.00
	23-04-2014		Drift line	793	137.47	8.83	3.78
	03-06-2014		Drift line	670	41.79	7.46	0.00
	22-07-2014		Drift line	737	58.35	0.00	0.00
Warnemuende	31-03-2014	54.183315 12.083082	Shallow water	939	99.04	2.13	5.32
	22-05-2014		Drift line	610	134.43	0.00	4.92
	19-07-2014		Drift line	744	532.19	5.38	4.03
Markgrafenheide	31-03-2014	54.193767 12.139279	Shallow water	455	193.41	4.40	0.00
	23-04-2014		Shallow water	722	174.52	11.08	5.54
	22-05-2015		Drift line	611	124.39	3.27	0.00
	19-07-2014		Drift line	737	107.12	8.14	0.00
Rostock Seawater sample							
Warnemünde	06-08-2014	54.183315 12.083082	–	7 l	3.3	0.43	0.00
Rügen Sediment samples							
Binz	22-06-2014	54.402833 13.614562	Drift line	756	17.2	0.0	1.3
Breege	22-06-2014	54.602267 13.389711	Drift line	737	73.3	5.4	4.1
Dranske	28-06-2014	54.633245 13.220834	Drift line	610	41.0	1.6	3.3
Heidehof	28-06-2014	54.666044 13.267386	Drift line	704	24.1	1.4	1.4
Oder/Peene estuary August 2014							
Kamminke	23-08-2014	53.866751 14.212367	Drift line	766	14.4	0.00	1.3
Freest[a]	23-08-2014	54.140276 13.728513	Mud flat	711	424.7	11.2	7.0
Jade Bay September 2014							
Varel/Nordender Leke Freshwater[b]	15-09-2014	53.405249 8.132402	–	4 l	19.2	5.00	0.25
Dangast Seawater[c]	15-09-2014	53.451859 8.123518	–	10 l	3.1	0.80	0.10
Dangast Sediment[d]	15-09-2014		Mud flat	765	13.1	0.00	3.9
Dangast Sediment[e]	15-09-2014		Mud flat	677	53.2	8.9	0.00

Notes: The location and date are given in the first two columns with the GPS coordinates of each sampling location provided in column 3. Details on the exact sampling situation are given in column 4 and the dry weight of each sediment sample in column 5. For water samples, the volume is given in column 5 in litres. The concentration of all fibres with lengths in excess of ~200 μm is provided in column 6 without distinction of transparent natural and synthetic fibres. The concentrations of coloured fibres and particles are provided in columns 7 and 8, respectively. All concentrations are on the basis of sediment dry weight or litre of sea-/freshwater as given in column 5. Large amounts of residual sediment prohibited the total fibre count in the Nienhagen March sample.

[a] Shallow sand islands between silt and mud.
[b] Freshwater sample near paper recycling plant.
[c] Seawater sample near beach, few metres into mud shallows.
[d] Fine-grained sediment islands between silt/clay/mud.
[e] Second 500 ml fine-grained sediment sample taken few metres apart.

derived with the full area of the immersed sphere segment as the supporting area instead of the surface area difference might overestimate the surface tension force by at most a factor of two. In this conclusion, the only remaining assumption is that a particle denser than water is not expected to sink further than half its size while still being supported on the water surface. The surface suspension force would still allow particles with a radius of up to 1.05 mm to be suspended on the water surface, changing the conclusions drawn above only marginally.

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