Occurrence and distribution of microplastics in marine sediments along the Belgian coast

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A B S T R A C T

Plastic debris is known to undergo fragmentation at sea, which leads to the formation of microscopic particles of plastic; the so-called 'microplastics'. Due to their buoyant and persistent properties, these microplastics have the potential to become widely dispersed in the marine environment through hydrodynamic processes and ocean currents. In this study, the occurrence and distribution of microplastics was investigated in Belgian marine sediments from different locations (coastal harbours, beaches and sublittoral areas).

Particles were found in large numbers in all samples, showing the wide distribution of microplastics in Belgian coastal waters. The highest concentrations were found in the harbours where total microplastic concentrations of up to 390 particles kg⁻¹ dry sediment were observed, which is 15–50 times higher than reported maximum concentrations of other, similar study areas.

The depth profile of sediment cores suggested that microplastic concentrations on the beaches reflect the global plastic production increase.

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1. Introduction

The global production of plastics was approximately 1.5 million t/y in the 1950’s. Currently it is estimated at almost 250 million t/y and it is still increasing by 10% each year (Plastics Europe, 2008). While reliable estimates of the input of produced plastics in the environment cannot be obtained, substantial amounts end up in the marine environment through industrial discharge, littering and terrestrial runoff (Derraik, 2002). This has lead to increasing levels of plastic litter in oceans worldwide.

The occurrence and distribution of large plastic debris in the marine environment is well documented (Derraik, 2002) and the adverse effects of this type of pollution on marine life have been described extensively (e.g. Baird and Hooker, 2000; Bugoni et al., 2001; Carr, 1987; Laist, 1987; Moser and Lee, 1992). However, these larger items eventually undergo fragmentation which leads to the formation of microscopic particulates of plastic (Barnes et al., 2009). These so-called ‘microplastics’ (plastic particulates ≤1 mm) may become widely distributed in the marine environment through hydrodynamic processes and ocean currents (Ng and Obbard, 2006).

Little is known about the (adverse) effects of microplastics on marine organisms. Recently it has been shown that these particles can be ingested by mussels (Mytilus edulis) and can translocate to the tissue and persist there for at least 48 days (Browne et al., 2008). Other organisms (i.e. polychaete worms, barnacles, amphipods and sea cucumbers) have also been found to ingest microplastics during laboratory trials (Graham and Thompson, 2009; Thompson et al., 2004). No significant adverse effects have yet been observed, but this may be due to the short exposure time used in these studies (Browne et al., 2008). Plastic pellets and fragments have also been shown to (1) absorb and transport hydrophobic chemicals including polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and nonylphenols, and (2) transfer these pollutants to organisms (Derraik, 2002; Endo et al., 2005; Rios et al., 2007; Teuten et al., 2007). Even though it was recently suggested that the severity of both these phenomena is low (Zarfl and Matthies, 2010; Gouin et al., 2011), the lack of other studies confirming these suggestions justify the interest in the occurrence and ubiquity of microplastics in the marine environment. Multiple reports are available on environmental concentrations of plastic fragments larger than 1 mm, including pre-production pellets (e.g. Gregory, 1983; McDermaid and McMullen, 2004). However, it has recently been argued that only fragments of 1 mm and smaller should be classified as microplastics (Costa et al., 2010). Up until now, only three studies on the...
occurrence of plastic fragments have included true microplastics (Ng and Obbard, 2006; Reddy et al., 2006; Thompson et al., 2004) (Table 1). Hence, only limited data on the global abundance of microplastics is available at this time.

The aim of this study was to investigate the occurrence and distribution of microplastics in Belgian marine sediments collected in harbours, on beaches and offshore. At two of the beaches, sediment cores were taken to study time trends of microplastic accumulation. The extracted microplastics were grouped into four categories (i.e. fibres, granules, plastic films and spherules), counted and identified using Fourier transform-infrared (FT-IR) spectroscopy. Results were compared with those of other, similar studies and attempts were made to relate the occurrence of microplastics to local human activities.

2. Materials and methods

2.1. Sampling

An overview of the sampling stations is given in Fig. 1. In each of the three harbours, three to four sampling stations were selected to represent inner to outer harbour conditions (Fig. 1). The three studied harbours differ in their main activities: (1) the harbour of Zeebrugge is the largest coastal harbour used mainly by naval and commercial boats (freight ferries, container vessels, large passenger ships), (2) the harbour of Oostende is used by pleasure crafts, fishing boats and passenger ferries, and (3) the harbour of Nieuwpoort mainly by pleasure boats as it hosts the largest yacht harbour of Northern Europe. In each harbour, one station was located in or near the yachting facilities (stations ZB2, OO2 and NP2 respectively) (Fig. 1).

Three sea sampling stations (S1–S3) were selected near the mouths of the three coastal harbours and three additional stations were chosen further off shore (S4–S6) (Fig. 1). Station S4 was located 11 km off shore in the sedimentation zone of the Scheldt river; stations S5 and S6 were both located 21 km off shore. All subtidal stations (i.e. harbour and sea stations) were sampled using a Van Veen grab (70 kg, 0.1 m² sampling surface).

Sediment was also collected on three Belgian beaches: two known for sand deposition (Koksijde-Bad and Groenendijk-Bad) and one known for erosion (Knokke-Zoute) (Fig. 1). On each beach, sand was collected at the high watermark, in the middle of the intertidal area and in the subtidal zone, except at Knokke-Zoute, where no samples were taken in the latter zone. To study potential trends in time, sediment cores were taken at Koksijde-Bad and Groenendijk-Bad, where the annual local deposition rate, as derived from coastal line maps (Afdeling Waterwegen Kust, 2000a,b), was approximately 7 and 2 cm, respectively. This allowed sediment layers of roughly 4 years (Koksijde-Bad) and 16 years old (Groenendijk) to be studied. Each core was divided in four equal parts, resulting in sediment layers each representing a time span of 1 year for Koksijde-Bad and 4 years for Groenendijk. The cores were taken at the high water mark and in the intertidal zone.

2.2. Analysis

The microplastics in the sediment samples were extracted using the method of Thompson et al. (2004) with some minor modifications. In short, 3 L of a concentrated saline solution was added to 1 kg of wet sediment and stirred for 2 min. The sediment was then allowed to settle for 1 h before the supernatant was poured through a 38 μm mesh sieve. For each sediment sample this extraction was performed twice and the collected particles (sieve) were examined using a binocular microscope. The particle recovery of the extraction procedure was ascertained by spiking known concentrations of microplastics (of similar dimensions as those encountered in the field) into clean sediment and subjecting it to repeated extractions. Recovery of fibres and granules/spheres was assessed separately for both sandy sediments (more than 50% of the sediment particles >63 μm) and sludge (less than 50% of the sediment particles >63 μm). The resulting particle recoveries (ranging from 68.8% to 97.5% for the different sediment and particle types) were used as correction factors for calculating the microplastic concentrations reported in this paper.

Microplastic particles were categorised into four different types: fibres, granules, plastic films and spherules. Particles of each type were identified by Fourier transform-infrared (FT-IR) spectroscopy using an AutolMAGE-microscope attached to a Perkin-Elmer Spectrum GX spectrometer equipped with a nitrogen-cooled Mercury cadmium telluride-detector. The spectra were recorded in reflection mode in the spectral range 400–4000 cm⁻¹ by co-adding 128 scans at a resolution of 4 cm⁻¹. The aperture was set at 100–100 μm using adjustable knife-edges. As in the studies of Thompson et al. (2004) and Ng and Obbard (2006), the particles were identified by comparing FT-IR absorbance spectra of the microplastics to those in a polymer reference library.

Microplastic concentrations were expressed as number of particles kg⁻¹ dry sediment and on a weight basis (mg microplastics kg⁻¹ dry sediment) to allow comparison with other studies.

2.3. Statistical analysis

All statistical comparisons were performed using SPSS software (SPSS, 2007). For multiple comparisons, the nonparametric Kruskal–Wallis test was used. If this test indicated significant differences, the nonparametric Mann–Whitney U test for pairwise comparisons was used to identify the significantly differing groups (significance level: 0.05).

3. Results

The number of microplastic particles observed at the different sampling stations in the Belgian coastal zone is presented in Table 2. All sediment samples collected in the harbours, on the beaches and sublittorally (BCS) contained microplastics (38 μm–1 mm). The four different types of particles were encountered with the majority (based on numbers) being fibres (59%) and granules (25%), which were both present at all sampling locations. Also plastic films (4%) were observed at all sampling stations with a few

<table>
<thead>
<tr>
<th>Country</th>
<th>Location</th>
<th>Maximum concentration</th>
<th>Unit</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>India</td>
<td>Ship-breaking yard</td>
<td>89</td>
<td>mg kg⁻¹</td>
<td>Reddy et al., 2006</td>
</tr>
<tr>
<td>Singapore</td>
<td>Beach</td>
<td>3</td>
<td># kg⁻¹</td>
<td>Ng and Obbard, 2006</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>Beach</td>
<td>8</td>
<td># kg⁻¹</td>
<td>Thompson et al., 2004</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>Estuarine</td>
<td>31</td>
<td># kg⁻¹</td>
<td>Thompson et al., 2004</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>Subtidal</td>
<td>86</td>
<td># kg⁻¹</td>
<td>Thompson et al., 2004</td>
</tr>
</tbody>
</table>

a Only fibre concentrations were reported.
b Original unit (# fibres 50 mL⁻¹ sediment) converted using an average sediment density of 1600 kg m⁻³ (Fettweis et al., 2007) and 1.25 as average wet sediment/dry sediment ratio.
exceptions (OO3, ZB1, ZB3 and S6). The spherules (12%) were found exclusively in harbour sediments and were all identified by FT-IR analysis as polystyrene. The fibres were identified as polypropylene, nylon and polyvinyl alcohol, and all the analyzed plastic films were made of nylon. The granular particles were identified as polypropylene, polyethylene or polystyrene.

Statistical analysis showed that the average microplastic concentration of the harbour sediments (166.7 ± 92.1 particles kg\(^{-1}\) dry sediment) was significantly higher than the concentration found for both the BCS (97.2 ± 18.6 particles kg\(^{-1}\) dry sediment; Mann–Whitney U test, \(p = 0.007\)) and the beach sediments (92.8 ± 37.2 particles kg\(^{-1}\) dry sediment; Mann–Whitney U test, \(p < 0.001\)) (Table 3). The latter two did not differ significantly (Mann–Whitney U test, \(p = 0.407\)).

Fig. 1. The study area; A: map of Europe showing the location of Belgium; B: the Belgian coastal zone, showing the six sampling locations on the Belgian Continental Shelf (S1–S6) and the three sampling locations on beaches (KB: Koksijde-Bad, GR: Groenendijk, KZ: Knokke-Zoute); C: sampling locations in the harbour of Nieuwpoort; D: sampling locations in the harbour of Oostende; E: sampling locations in the harbour of Zeebrugge.

Fibres were the most common type of particles found and the detected concentrations in harbour sediments were comparable to those in sediments collected at the beach and sublittoral stations. Station S6, located at 21 km off shore, exhibited an exceptionally high concentration of fibres at 237.3 ± 22.6 fibres kg\(^{-1}\) dry sediment. Excluding station S6 from the calculations, the average fibre concentrations found at the beach, harbour and sea stations (Table 3) did not differ significantly (Kruskal–Wallis test, \(p = 0.222\)) and were: 81.0 ± 37.2, 65.6 ± 15.3 and 66.3 ± 28.6 fibres kg\(^{-1}\) dry sediment, respectively. Granular particles were the second most common type of particles found with the highest concentration found in harbour sediment (56.4 ± 31.8 granules kg\(^{-1}\) dry sediment) (Table 3). This is significantly higher than the average concentration detected at the sea stations (29.6 ± 5.8
granules kg\(^{-1}\) dry sediment; Mann–Whitney test, \(p = 0.048\)) and on the beaches (5.9 ± 2.8 granules kg\(^{-1}\) dry sediment; Mann–Whitney test, \(p = 0.000\)). The fibre concentration found in the sea station sediments was also significantly higher than those on the beaches (Mann–Whitney test, \(p = 0.002\)). In general, plastic films were present at low concentrations and no significant differences could be detected between the three sampling site types (Kruskal–Wallis test, \(p = 0.412\)) (Table 3). Station NP1 exhibited an exceptionally high concentration (43.4 ± 11.2 plastic films kg\(^{-1}\) dry sediment). At the other stations, plastic films were found at levels ranging from 0 (OO3, S6 and Knokke IT) up to 13.9 ± 3.1 plastic films kg\(^{-1}\) dry sediment (Groenendijk HWMM).

As mentioned above, spherules were only encountered in harbour sediments (Table 2), and all were composed of polystyrene. Generally, the highest concentrations of this category were found in the inner harbours with concentrations up to 94.7 ± 11.2 spherules kg\(^{-1}\) dry sediment at the sluice complex in Nieuwpoort harbour (NP1). In the other harbours, the highest concentrations were also observed at stations in the inner harbour located near yachting facilities and commercial shipping activities (i.e. stations OO2 and ZB2). Outer harbour stations generally exhibited a lower spherule concentration (Table 2).

At the sublittoral sampling sites (BCS) the highest microplastic concentration was found at station S6 (Table 2). As mentioned above, this was due to a very high concentration of fibres. For other BCS stations, total microplastic concentrations varied between 71.5 ± 6.4 (S1) and 115.8 ± 13.3 (S2) particles kg\(^{-1}\) dry sediment (Table 3). Station NP1 exhibited an exceptionally high concentration (43.4 ± 11.2 plastic films kg\(^{-1}\) dry sediment). At the other stations, plastic films were found at levels ranging from 0 (OO3, S6 and Knokke IT) up to 13.9 ± 3.1 plastic films kg\(^{-1}\) dry sediment (Groenendijk HWMM).
Within the harbours, the highest microplastic concentrations were generally found at the inner harbour stations. The concentrations gradually decreased towards the harbour mouths (Table 2). The highest microplastic concentrations were found in the harbour of Nieuwpoort at station NP1 (390.7 ± 32.6 particles kg\(^{-1}\) dry sediment), which is located inside a circular sluice complex where several rivers are connected to the harbour (Fig. 1).

The vertically sectioned sediment cores taken at the beach sections of Groenendijk and Koksijde-Bad enabled us to study the evolution of microplastic concentrations with time (based on the local sediment deposition rate). At the high water mark of the beach in Koksijde-Bad, where each of the four layers of the cores represented roughly one year, the upper and deepest layer contained 95.5 ± 9.0 and 97.3 ± 17.4 particles kg\(^{-1}\) dry sediment, respectively. This indicates that no significant change in microplastic concentration occurred at this site within the last 4 years. At the high water mark of the beach section in Groenendijk, where each layer represented roughly 4 years, a significant increase of microplastic concentrations was found compared to 16 years ago (Fig. 2). For the time periods 1993–1996 and 2005–2008, a microplastic concentration of 54.7 ± 8.7 and 156.2 ± 6.3 particles kg\(^{-1}\) dry sediment was found, respectively. From this it can be concluded that microplastic concentrations in the beach sediments at this site have nearly tripled since 1993.

4. Discussion

Microplastics were present in sediments collected in different parts of the Belgian marine environment and no significant difference in their concentrations was found between offshore and inshore sampling stations. In general, the concentrations we observed were higher than those found in other, similar studies (Tables 1 and 2). Studies in the UK and in Singapore (Ng and Obbiad, 2006; Thompson et al., 2004) noted maximum concentrations that were respectively 15–50 times lower than those observed in our study. Only at a ship-bounding yard in India, a location which is expected to produce a lot of marine debris, reported concentrations were up to twelve times higher (Reddy et al., 2006).

As expected, total microplastic concentrations were highest in the coastal harbours, but it proved difficult to relate the observed concentrations to local human activities. The harbour of Nieuwpoort for example, exhibited the highest microplastic contamination in the inner harbour (station NP1). This harbour hosts three yacht clubs which together have a total capacity of almost 2000 moorings, making it the largest yacht harbour of Northern Europe. These recreational boating activities could be the main contributor to the observed microplastic contamination. This harbour also exhibited the highest concentrations of polystyrene spherules. Particles of this type (0.1–2.0 mm in diameter) were observed in plankton tows made in the coastal waters of southern New England (Colton, 1974). They were identified as ‘suspension beads’, i.e. the bulk material produced by plastics manufacturers and shipped to plastics fabricators. This suggests that the higher microplastic concentrations in the harbour of Nieuwpoort are not necessarily the result of recreational boating activities, but may also have originated from the plastics industry. Given that no shipping of goods occurs in the harbour of Nieuwpoort, it is unlikely that the polystyrene spherules originate from any activity within the harbour itself. Colton (1974) also explained that “the only apparent way in which these clear and opaque polystyrene spherules enter the ocean is via waste-water discharge from a plastic-producing or plastic-processing plant into a river or estuary”. In this context, it is noted that no less than six rivers discharge at the sluice complex in the harbour of Nieuwpoort. The high concentrations of polystyrene spherules at station NP1 may thus be the result of a high input of these particles via rivers. Since polystyrene spherules have a density greater than that of seawater (Colton, 1974), it is possible that they sink to the seafloor before they can flush out of the harbour which would explain their absence in sediment samples collected at open sea.

As partially enclosed harbour areas seem to exhibit the highest abundance of microplastics, the high concentrations within the harbours could also be partially related to the geometry of the harbour compartments irrespective of proximity to any industrial or recreational boating activities. The flushing rate in such compartments can be very low and the narrow entrance can cause the occurrence of tidal eddies (Yin et al., 2000). Microplastics floating into such compartments could get trapped into the vortex, eventually settling on the bottom instead of flushing out of the harbour. This could explain the higher microplastic concentrations at the circular sluice complex in the harbour of Nieuwpoort (station NP1), the yacht harbour in Oostende where the harbour widens (station O02), and the yacht harbour in Zeebrugge which has a very narrow entrance (station Z2B) (Fig. 1).

Polymer type does not allow us to make clear statements about the origin of the particles. The analyzed fibres were identified as nylon, polyvinyl alcohol and polypropylene. Nylon is used in all kinds of fibrous materials like clothes, carpets, ropes and airbags, but also in fishing nets. Polyvinyl alcohol fibres are most likely originating from fishing lines. Polypropylene fibres are used in for example carpets and ropes. The latter are used extensively on ships. The granular particles were identified as polystyrene, polyethylene and polypropylene. Granules can be fractions of hard plastics, and as such these particles could come from numerous land-based and sea-based sources, as all these types of polymers are used extensively for the production of hard plastic used in various applications. Certain types of hand cleaners, cosmetic preparations and some airlast cleaning media could be a source of these granules, as these products can contain granulated polyethylene, polypropylene or polystyrene particles to function as a scrubbing agent (Gregory, 1996; Zitko and Hanlon, 1991). All the analyzed plastic film fragments were identified as nylon. This is surprising, since most packaging material is made of (low density) polyethylene. However, nylon wrapping is also used in for example balloons, packaging of frozen foods, liquids and medical supplies, and in vacuum packaging material. While the occurrence of polyethylene films cannot be excluded, it appears as if nylon wrappings are either more abundant in the marine envi-
The results of this study have revealed relatively high concentrations of microplastics in sediments of the Belgian coastal zone compared to those in similar areas (Ng and Obbard, 2006; Thompson et al., 2004), confirming the widespread occurrence of microplastics in the marine environment. Moreover, spatial variation in microplastic concentrations was observed on a relatively small scale. A clear relationship between local human activities and microplastic concentrations was absent, and elevated concentrations appeared to be primarily linked to the geometry of the respective compartments (i.e. enclosed areas in harbours). The results further suggest that freshwater rivers are a potentially important source of microplastics. Finally, although the used methodology needs further validation, the temporal trends which were observed in this study imply that the concentrations of microplastics in sediments are increasing.

References


SPSS, 2007. SPSS software, version 16.0. SPSS Inc., Chicago, IL.

