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Determination of microplastics in coastal beach sediments along Kattegat Sea, Denmark

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An evaluation of the level of contamination of microplastics in the coastal marine sediment from Kattegat Sea in Denmark has been conducted. The evaluation is based on sediment samples collated from beaches located on coasts partially surrounding the Kattegat Sea: Mou, Bogense, Hasmark, Zealand Odde, and Rorvig. Microplastics were extracted from the sediment samples using a newly developed density and flotation apparatus. Afterwards, the extracted microplastics were categorised under a stereo microscope, and the criteria for visual identification as synthetic polymers were: shape, colour, degradation and surface contours. This study indicate the presence of microplastics in coastal sediments throughout the Kattegat Sea i.e. between two and 55 particles/550 g Dry Weight were found in the sediment across the five locations. The total abundance of microplastics were 210 particles. Among the samples – fragments (46.1%) were the most dominate shape whereas fibres (34.8%) accounted for the second largest fraction. Our results highlight widespread occurrence in coastal sediments from the Kattegat Sea and provide valuable information for further environmental assessments of microplastics in Denmark.

Keywords: microplastic, extraction device, sediment, synthetic polymers.

УДК 504.6

Определение микропластика в прибрежных отложениях моря Каттегат (Дания)

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Была проведена оценка уровня загрязнения микрочастицами пластиков (микропластиками) прибрежных морских отложений моря Каттегат в Дании. Для оценки использовали образцы отложений, собранных на пляжах побережья, окружающего море Каттегат: Моу, Богенсе, Хасмарк, Зеландия Одде и Рорвиг. Микрочастицы были извлечены из образцов отложений с использованием недавно разработанного аппарата для флотации и определения плотности. Извлечённые микропластики были классифицированы под стереомикроскопом. Критериями для визуальной идентификации синтетических полимеров были форма, цвет, деградация и контуры поверхности. Это исследование указывает на присутствие микропластиков в прибрежных отложениях по всему морю Каттегат – в пяти местах было обнаружено от 2 до 55 частиц на 550 г сухой массы отложений. Общее количество микрочастиц пластиков составило 210. Среди образцов доминирующей формой были фрагменты пластика (46,1%), вторую по величине фракцию составляли волокна (34,8%). Полученные результаты показывают широкое распространение микропластиков в прибрежных отложениях моря Каттегат и представляют собой ценную информацию для дальнейших исследований по загрязнению микропластиком окружающей среды в Дании.

Ключевые слова: микропластик, экстракционное оборудование, отложения, синтетические полимеры.

During decades, plastics have become an integrated component of modern society. Global annual production of plastic is now rising to above 335 million tons [1]. Approximately 50% of all plastics today are disposed after a single-use [2] and the quantities recovered as a fraction of total discards shows that recycling rates are relatively low [3]. A consequence of this is that between 4.8 and 12.7 million tons of plastics were estimated in 2010 to enter the marine environment. The cumulative input of mismanaged plastic´s debris to the oceans is predicted to reach as high as 250 million tons in 2025 [4]. Further studies predicts that during the next decades, total production of plastics will increase and approximately 12 billion tons of plastic waste will be in landfills or in the natural environment by 2050 [5].

Carpenter et al. published the first identification of marine microplastics [6]. They found abundant polystyrene spherules in sizes 0.1 to 2 mm (average 0.5 mm) in coastal waters of southern New England, US. Currently, marine plastic pollution receives major attention and has been reported in different marine environments globally including (1) benthic sediments, (2) in the water column, and (3) deep-sea sediments [7].

However, limited focus has been on coastal sediments in the Nordic region. Strand et al. studied microplastics investigate sediments samples from seabed in Danish territorial water northwest of Skagen, and identified microplastics in all collected sediment samples. In that study both fibres and fragments were identified in ranges from 0.6 to 36 particles per 10 g dry weight (DW) of sample [8]. Noren took nineteen samples from the sand sediments at the Swedish west coast sediments: particles found in these samples ranged from 150 and 2400 particles/m^3 [9]. Furthermore, Noren also examined a harbour located in near distance to a polyethylene (PE) production plant, and found 102,000 particles/m^3 with diameter between 0.5–2.0 mm.

This paper presents the first study of microplastic occurrence and types in the coastal sediments in Kattegat Sea in Denmark (Fig. 1, see color inset). Samples has been taken from five beaches along the western and southern boundaries of the Kattegat Sea in order to establish data on potential variations in accumulation and occurrence patterns.

Field campaign

Sampling sites. Five sampling sites (S1–S5) were selected in order to have a good representation of both sites in the inner Danish waters and sites facing the inflow from the Skagerrak Sea and the Baltic Sea. In Figure 1 are the sampling locations plotted, in Table 1 the sampling locations and GPS coordinates are shown, and Table 2 depicts an overview of site-specific features that potentially serves as contributing contamination sources. The following describes briefly the general characteristics of the sample locations:

Mou (S1) is a small town in northeast Himmerland with 1,130 inhabitants [10], located on the southern shores of the eastern mouth of Limfjorden inlet. The town is located in the North Jutland region and belongs to the municipality of Aalborg with a population of 213,558 and an area covering 1,143.99 km² [10]. The sampling area is located where Limfjorden inlet meets the Kattegat Sea.

Bogense (S2) is a small seaport located on the north-western coast of Funen, the population is 3,710 citizens [10], and belongs to the municipality of Nord Funen with 29,030 inhabitants. The municipality covers an area of 451 km . Today, Bogense is a popular tourist-attraction and only little industry is located in the area. This is an interesting site since research has shown that microplastic abundance tends to increase near recreational areas [11].

Hasmark (S3) beach is located on the northeast part of the coast of Funen. The beach areas at Hasmark are a very popular place for recreation activities and many summerhouses are positioned directly in connection to the beach with only a minor dirt and cement barrier between the houses and the sea. This sampling location was selected because it is the most isolated part of beach in the area.

Zealand's Odde (S4) was in recent times an area with great ship traffic due to the ferry routes from Odden harbor on the south coast to Ebeltoft and Aarhus in Jutland. Today fast catamaran ferries with more than 15 daily departures serve these two routes. No heavy industry besides the ferry activities are located in the areas. The city size is small, however due to large areas with holiday homes the actual population differs depending on seasonal activities.

Rørvig (S5) is a cottage and port city in Northwest Zealand with 1,051 inhabitants [10]. Rørvig is located on the northern part of Isefjord at the western shores of the northern mouth of Isefjord inlet. The town is located in Odsherred Municipality and is located in Region Zealand. The area is a frequented recreational area with significant seasonal fluctuations of humans.

Table 1

Sampling locations, ID, and GPS coordinates

Site ID	Location	Longitude	Latitude
S1	Mou	56°58'53.1"N	10°18'25.3"E
S2	Bogense	55°32'29.8"N	10°00'30.8"E
S3	Hasmark	55°33'23.0"N	10°28'49.7"E
S4	Zealand Odde	55°59'03.5"N	11°19'11.2"E
S5	Rørvig	55°57'11.7"N	11°43'00.2"E

Table 2

Five site specific features potentially affecting the abundance of microplastics

Location	Site specific features (located nearby)				
	harbour	ferry berth	recreational area (public beaches, vacation homes etc.)	water stream	inlet
S1 (Mou)	x	x	x	–	x
S2 (Bogense)	x	–	–	–	–
S3 (Hasmark)	–	–	x	–	x
S4 (Zealand Odde)	x	x	–	–	–
S5 (Rørvig)	x	x	x	x	x

Note: x – presence of a factor, – lack of a factor.

The specific locations are chosen due to their spatial coverage of coasts encompassing the Kattegat Sea. Also, the locations are located in or in near-proximity to larger inlets linked to Kattegat Sea. In addition, Kattegat Sea serves as water gateway between the North Sea in northwest and the Baltic Sea in east. Sea currents through the inner Danish waters are mainly northerly direction.

Sampling method. A nested sampling methodology as suggested by [12] was selected for sampling of the coastal sand sediments. Procedures on the sampling locations follows those by [13] and adapted as depicted in Figure 2. The samplings were performed using two transects; the first in the intertidal zone and the second in the berm area of the beach. All sampling were performed on a straight-line perpendicular to the low tide line and approximately 15 meters apart. Samples in the zones were taken 15 meters apart from each other perpendicular to the shoreline, see illustration on Figure 2. At each sampling point a quadrat (30 x 30 cm) was excavated, using a metal spoon. The sediments were stored in pre-rinsed certified glass containers suitable for environmental sample purposes (Fisherbrand, Soda lime glass, 1000 mL, screw cap of PE). Excavation depth was approximately 1.5 centimeters. Larger non-plastic debris, for instance biological material e. g. seaweed, plants and larger stone gravel were removed manually to minimize filling up the sample container.

To collect samples exactly at the low tide beach zone tidal table data from the Danish Meteorological Institute were consulted. The sampling was conducted between September 8th and September 10th 2015. To ensure reproducibility of the study, GPS coordinates were secured using an iPhone 4 device with a GPS tracker app installed. In total, six sand sediment samples were taken from each sampling site.

Laboratory procedures

For extracting microplastic from the collected sediments, a laboratory procedure was developed, in the following the applied laboratory procedures are described in details.

Extraction equipment. Inspired by the extraction instrument developed by [14] we have constructed the extraction instrument (Fig. 3). As can be observed in Figure 3 the instrument consists of two main parts: a separation column (A) where water is filled in and used as carrier of low-density particles, and an inlet chamber (B) where air and water is pushed through. The separation column (A) is equipped with an outlet pipe (E). The inlet chamber consists of an air inlet (C), and water inlet valve (D). In the intersection between the two main parts (A and B) two 5-mm/60-μm bottom mesh filters (F), and a rubber seal (G) is located. To support the weight of a sediments sample, the 5 mm mesh screen is positioned below the 60-μm mesh screen as structural support. Also, the inlet chamber is fitted with two air diffusers (Eheim air diffuser) for aeration upward into

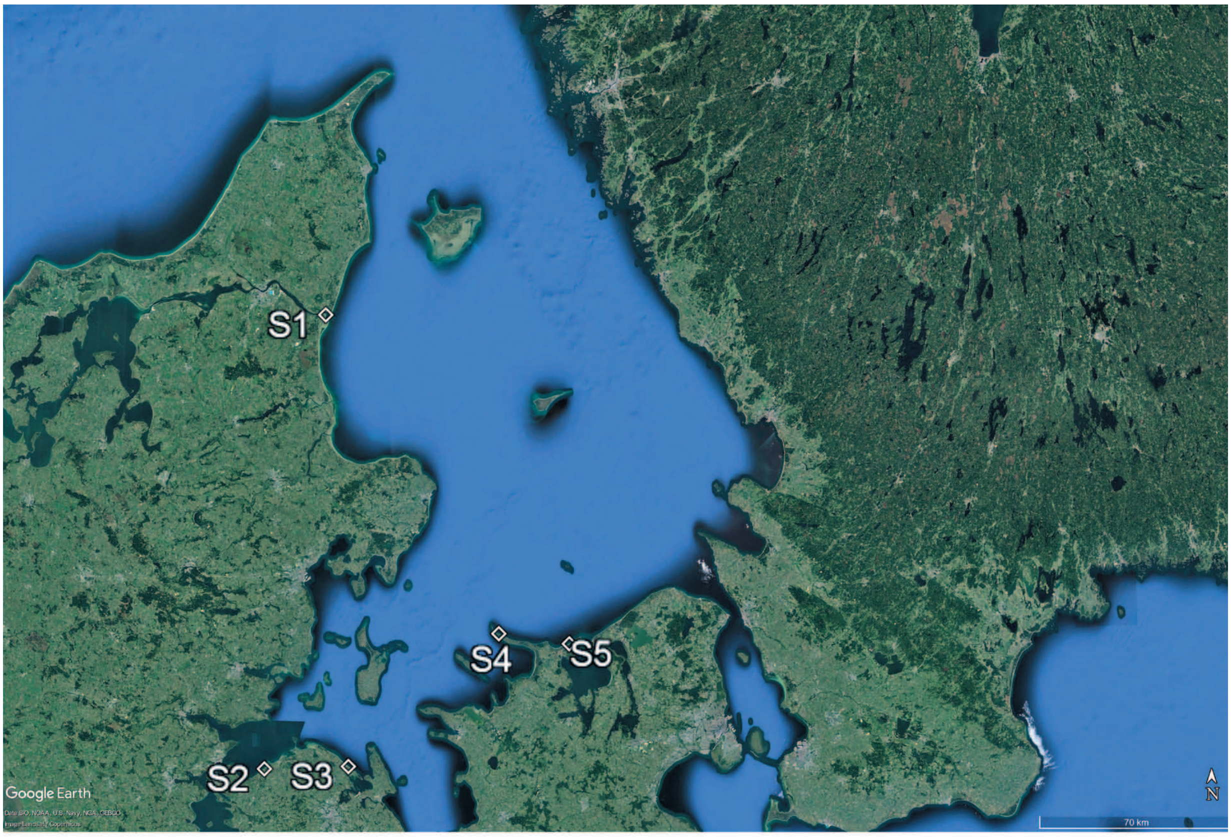

Fig. 1. Sampling locations in Denmark (S1–S5)

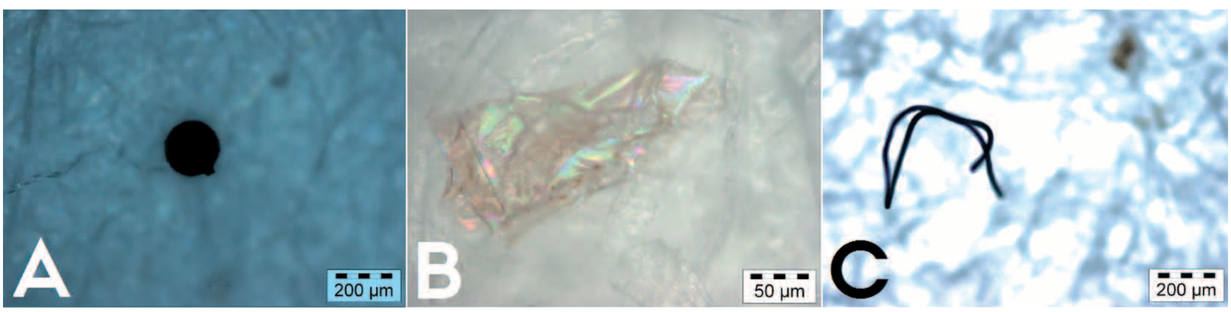

Fig. 5. Selection of particles found: sphere (A), fragment (B), and fibre (C)

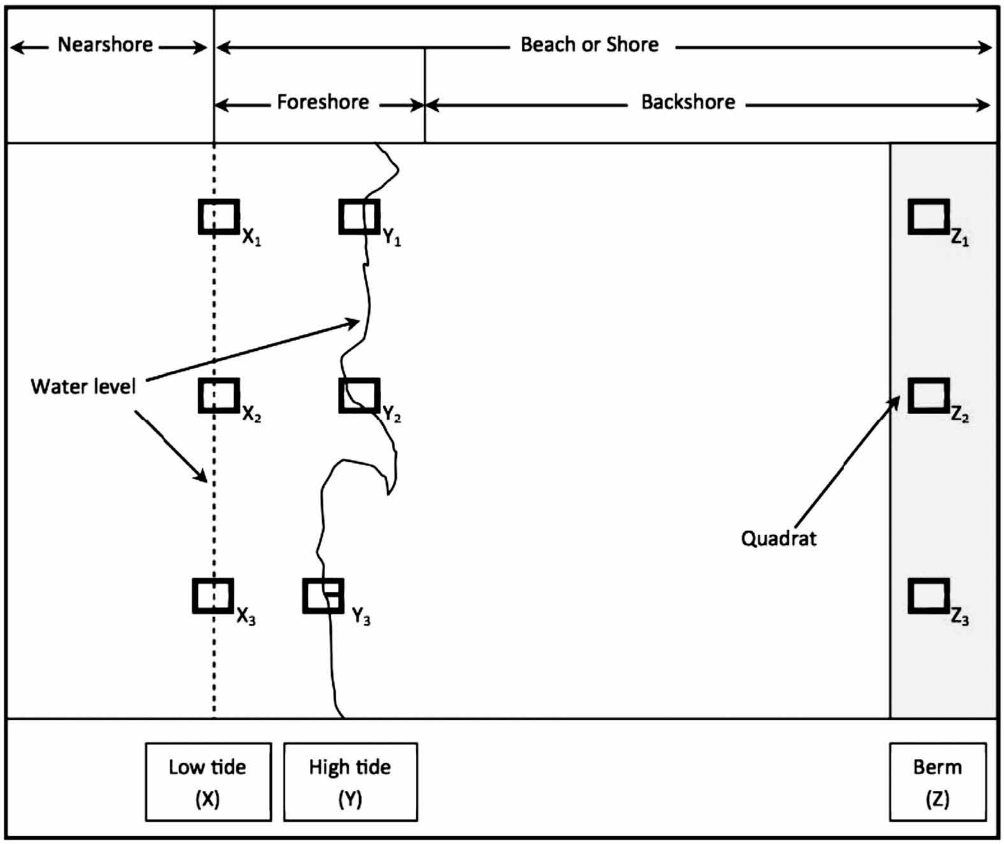

Fig. 2. Nested sampling design (adapted from [13])

the separation column. Even though air diffusers made of lime wood or porous stone exist, diffusers made of PE were selected as these were tested to distributed airflow more homogeneously.

The differences between our extraction instrument and the instrument developed by [14] are:

The material of the instrument was altered from polyvinylchloride (PVC) to food-graded stainless steel, since sediment particles such as quartz might continuously mechanically slide on the inside surface of the column meaning that microplastic particles might be generated and released into the sample.

Using stainless steel components counteracts the risk of the corrosive effect from salt particles in the dried sand sediments coming from the seawater where the samples were taking.

All the steel components were electro-polished to create a smooth surface to allow a smooth transcend of particles from the sediment upwards through the water column to the surface area and output area (Fig. 3 – right part of Figure).

Dimensions of [14] and the developed instrument differs not in height, however 5% in diameter increasing the total water volume assuming this would be beneficial to the release of trapped microplastics during full suspension in water column.

Extraction procedure. First, efficiency test were conducted to evaluation to which degree the instrument could deliver both separation of high- and low-density particles from a sediment matrix. All samples were loaded through the top of the cylinder. The success criteria of the initial experimental process of floatation was that sediment sample should provide full suspension of all sample material in the lower 1/3 section of the flotation cylinder. At an airflow of approximately 1.3 bar was this criteria achieved. The airflow at this level were maintained for 5 minutes to provide time for low-density particles to be released from the sediment and trans-located into the water column from which further separation were possible.

For testing the extraction efficiency of the newly developed instrument used in this study three samples were prepared. Each sample contained 10 fragments of PE (ρ = 0.89–0.93 g/cm^3, size = 300–450 µm), 10 fibres of nylon and polytetrafluoroethylene (PTFE) (ρ = 2.1–2.3 g/cm^3, size = 1000–2000 µm), and a volume of laboratory quartz sand (450–550 g DW, particle size = 0.2–0.4 mm).

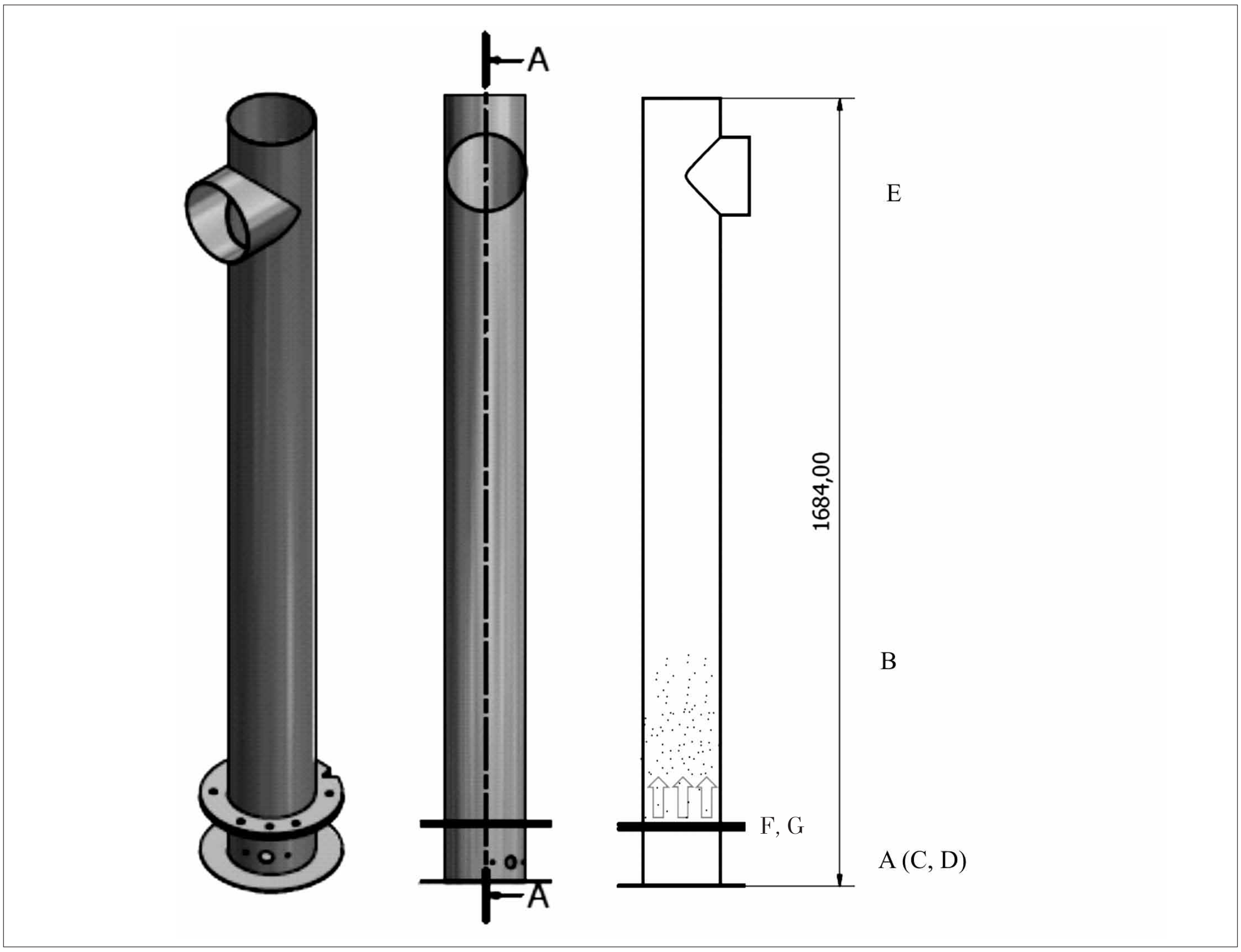

Fig. 3. Schematics of extraction device (proprietary design) (adapted from [14])

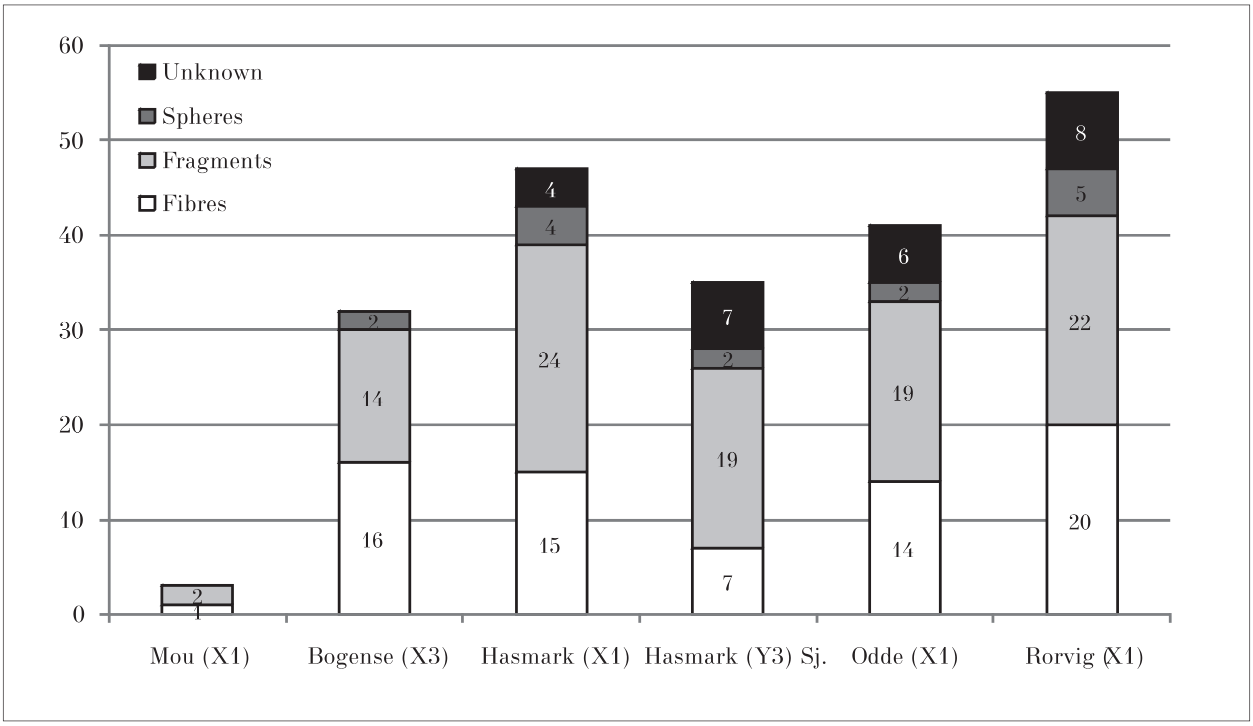

Fig. 4. Types and quantity of found particles at Mou, Bogense, Hasmark, Zealand's Odde and Rørvig in Kattegat Sea

The efficiency tests showed that our instrument performs with an efficiency 98 to 100% with respect to both the PE fragments and the PTFE fibres. In the efficiency tests conducted by [14] their instrument showed an extraction efficiency of 75 (plastic granules) to 98% (fibres). The reason for a higher filtration efficiency is achieved in this study compared to the study by [14] could be the inner surfaces of the separation column. During construction of the column considerable effort were placed on smoothing the surface allowing the microplastic particles not being retained by small irregularities on the inner surfaces of the column allowing the upward particles not to get retained.

Laboratory treatment of sediment samples. Extraction of the microplastics were performed in accordance with the procedure by [14] with minor modifications. Prior to extraction, all samples were placed in aluminium foil and dried at 60 °C yeilding constant weight to minimize the water content in the sample.

Characterisation by visual microscopy. To quantify the number of microplastics the extracted materials were examined by visual microscopy (Motic BA210, CCIS EF-N Plan Achromatic objectives 4X/0.10, 10X/0.25, 40X/0.65). Positive identification was based on (a) particle characteristic of colour and shape proposed by [15] combined with in accordance with criteria formulated by [16] and (b) visually determined according to following criteria:

– no cellular or organic structure are visible in the particles,

– fibers are equal in thickness in their entire length,

– particles show clear and homogenous colours [17].

All the identified microplastic particles were categorised into three types acording to their shape: fibres, fragments and spheres. Each type of identified microplastic were photographed with a microscope equipped with a photo tube option (Motic BA210).

Results

Our study detected microplastics in Danish coastal marine environment with abundance of 2, 32, 44, 35, 41, 55 particles per 500 g DW sediment at Mou, Bogense, Hasmark, Zealand Odde, and Rørvig respectively (Fig. 4). This corresponds to an average concentration of 420 kg/site.

In all samples (apart from Mou) both fibres, fragments and spheres were detected and in different colours ranging from opaque bluish to black. In terms of shape, a higher percentage of fragments were found compared to fibres and spheres. Other findings were black flakes and in some minor instances sphere-shaped particles. Spherical particles, indicating microbeads presence (Fig. 5A, see color inset) this was found only in limited numbers. Fragments (Fig. 5B, see color inset) were the dominant shape observed at all sites (apart from Bogense) with a proportion of 46.1% while fibre (Fig. 5C, see color inset) proportion is 34.8%. Spherical particles and particles of unknown origins were observed occasionally (3.8 and 11.9% respectively).

Discussion

Observed microplastics in sediments. The quantities of microplastics identified in all five samples shows lower level of microplastics in the sediments than the amount of microplastic observed by [8]. Furthermore, the quantities of the found fractions of microplastic particles in the different locations were distributed rather evenly (Fig. 4). It is also observed in Figure 4 the abundance of microplastics at Rørvig were significantly higher than Mou, but only slightly higher than Bogense, Hasmark and Odde. Across all the samples, the quantities varied from 3 to 55 items/550 g DW of sample.

The significant lower concentration of microplastics was identified at Mou compared to the microplastic observed from the other sites. This indicates that Mou represent a kind of outlier among the five sites. This characteristic could be explained by the position of the five sites in relation to the water flow in Kattegat. Bogense, Hasmark, Odde and Rørvig are located directly in the water flow of Kattegat, since the main water streams of Kattegat comes from the North Sea and Baltic Sea. This is opposite to Mou which has a location perpendicular to the dominant current direction through the inner Danish waters. Conversely, Bogense, Hasmark, Odde and Rørvig locations are more exposed to the volume of water flowing through these waters, see also figure 1. Thus, they may function as deposition sites for microplastics.

A Slovenian [18] study found more than 93% of total microplastic were fibrous in the sediments, this is in contradiction with the results from this study where only 34.6% were detected. However, even though fibres not being the largest proportion in this study it is still significant amounts. The high amount of fibres was expected due to research showing that fibres can originated from multitude of sources including the production, washing and natural degradation of textiles [19], e. g. recent research by [20] show that as much as 6 million particles are discharged into the wastewater by washing 5 kg of clothing. Also, research shows that building materials could contribute to microplastics as for instance abrasive beads used in sandblasting, might be discharged or in accidental spills run into the environment [21]. Even wear and tear from tyres contributes to the flow of microplastics into the environment [22].

A recent study by [23] shows that abrasion from paintings and coating might contribute to generation of microplastics, thereby also making shipping industry a potential pathways of microplastic into the environment. Denmark is the major maritime gateway to the Baltic Sea, consequently high levels of maritime freight transportation by larger vessels are sailing close to the sampling sites. According to data from the association of Danish Port, the total annual number of cargo ships and cruise ships between the year 2010 and 2014 varied between 502,405 and 519,613 excluding fishing vessels. All of the five sampling sites in this study are located close to some of the largest regional ship fairways in Kattegat Sea. Furthermore, four of the sites are situated close to a harbour (Mou, Bogense, Zealand Odde and Rorvig) and three of them close to a ferry berth (Mou, Zealand Odde and Rorvig). However, it is not possible from our study to see a similar correlation. For example at Mou was only found three microplastic particles. Hasmark has no harbour in the area but at this site 44 particles were found, i. e. the site with second most pieces of microplastics. This could indicate that the detected microplastic in this study is either from local source or from Skagerrak inlet. It is very difficult to link sources to the particles in the environmental samples and consequently these sources may be speculative in nature.

Conclusion

This paper has presented results on microplastic occurrence in Danish coastal sediments using a newly developed extraction instrument based on the novel design of [14]. In conclusion, a method for extracting microplastics from sediment samples was developed. Samples are subjected to an upward water flow in a column to separate lighter particles from heavier sediment particles. Finding shows that the developed instrument is capable of efficiently extracting small microplastics using tap water exclusively. Though, only testing the efficiency in particle size fractions larger than 1000 μm was conducted, it has been shown that the instrument has almost complete efficiency (29 out of 30 particles were recovered). In these size fractions, it is concluded to be a highly effective instrument for microplastic extractions.

The laboratory sand used in the efficiency study is very homogeneous in terms of particles diameter (0.2–0.4 mm) and may differ from sample of natural sediments were the composition and sizes of grains varies significantly more. In addition, samples of laboratory sand and natural sediment may also differ due to absorbed substances in a natural sample might affect properties such as adhesion. In relation to the methodologies used, it is evident that more research is needed to develop proven methodologies capable of sampling small particles as the areas continue to be exposed to smaller and smaller size fractions.

The results is part of a Danish multi-year field campaign covering five Danish and three Swedish locations headed towards the Kattegat Sea and reveals relatively low concentration of microplastic in sediments of the Inner Danish coastal zone compared to those in similar geographic area. But, the finding still confirms a widespread occurrence in the marine environment.

There is a critical need for further research to understand the impact and contamination level from the “Baltic Sea Port to the Atlantic” towards the Kattegat Sea and the coast areas along Kattegat Sea. The special geographic location of The Kattegat Sea as a water gateway between Baltic Sea and North Sea might also constitute a deposition zone for microplastics. Due to the degraded nature of a microplastic particle, it is difficult to know the specific source of microplastic, microplastics occur in diverse shapes such as spheres, fibres, and fragments in environmental samples.

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