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PROGRESS TOWARDS MONITORING OF MICRO- LITTER IN SCANDINAVIAN MARINE ENVIRONMENTS

State of knowledge and
challenges

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Martin Hassellöv, Therese Karlsson, Karin Mattsson, Kerstin Magnusson, Jakob Strand, Robin Lenz, Bert van Bavel and David Pettersson Eidsvoll

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Summary

Four different case studies were carried out to determine dominating microlitter types from urban environments to the regional Scandinavian seas (eastern North Sea). The sampling was both from sediment near sources (urban runoff and road dust sediment), and further out from coastal sediments. The sea surface layer and subsurface samples was taken in two different gradients, in the Oslo and Roskilde fjords, where also blue mussels were sampled. Best available technologies for sampling each compartment was used and evaluated, and while the water samples was analysed as collected, the sediment and biota samples needed some pretreatment of chemical digestion and/or heavy density liquid floatation or elutriation. In order to develop visual identification as objective as possible, a visual and physical observation scheme was proposed. The visual identification scheme should be complemented with spectroscopic identification to different degrees depending on the size fractions.

Spectroscopic identification is still often a quite time-consuming process, meaning that for monitoring purposes it is not currently advisable to aim to identify all particles during monitoring studies. Until fully or partly automated spectroscopic methods are available they are still important tools for verification of representative types of particles in samples above 100 μm .

The amount of particles that should be identified to provide adequate compositional information would be dependent on the aim of the study as well as the type and composition of the samples. However, in order to do monitoring and include sample composition in the results a minimum of 100 of the fewest particles should be counted in a sample to achieve 10% standard deviation in terms of counting statistical uncertainty.

The field is however rapidly evolving, and automated procedures are already being published. For research purposes and more detailed monitoring and screening studies spectroscopic methods can aside from providing particle identification also give clues on additives and level of degradation.

The most common types of microlitter found varied between studies but common trends could be identified between the road tunnel sediment and the urban creek sediment that these contained black particles resembling tire rubber from both visual

and tactile tests, and also asphalt, charcoal, oil/tar particles and road marker particles. In the coastal water samples the surface layer was dominated by polystyrene foam particles and polyethylene fragments and films. In the subsurface water samples fibres, films and fragments of plastic was most common. In both the Gothenburg urban creek sediment and Oslo fjord surface water samples particles that could be related to artificial sports turf (polyethylene green grass and clear cut, tire granulate) was observed. The microlitter in mussels was dominated by fibres. The approach of using gradient studies, and include both near source sampling as well as recipient gradient sampling, was concluded to be very suitable to determine sources and fate.

1. Introduction

Marine litter, including microlitter, is highlighted as one of the most prioritized stressors for the oceans and coastlines today, for example in the recent UN First Global Integrated Marine Assessment (Nations, 2016), and as preparation for the UN Oceans conference 2017. Microlitter is defined as anthropogenic particulate pollutants below 5 mm in length and comprise a wide range of material with different densities, shapes and chemical makeup. Microlitter, especially but not exclusively microplastics, have been frequently reported from accumulation zones in the subtropical ocean gyres during the last decade (Enders *et al.*, 2015). Knowledge about abundance, size distributions, morphologies, polymer type and other chemical information, from these areas have consequently been growing. From coastal seas, there are actually less amount of published data available. Although a proportion of the marine macro litter is subject to long range transport, there are also fate processes such as weathering, fragmentation, fouling and sedimentation (Andrady, 2011) that are sorting out some fractions closer to the sources, and the types of litter that are abundant in the oceanic accumulation zones have such properties that is favouring buoyancy (low density, thick material, resistant to weathering and fragmentation, low adhesion of fouling etc.) (Enders *et al.*, 2015). Furthermore, from beach litter inventories, it is also known that although many common types of litter objects are found all over the world, the litter composition can also reflect the local or regional litter input with sanitation articles or packaging or fishing related litter showing a higher or lower relative abundance. The compositional differences of litter in marine environments are crucial to understand sources, transportation and fate of marine litter but also to be able to assess risk and exposure.

The Marine macrolitter in Nordic coastal waters have been inventoried relatively well (Strand *et al.*, 2015, OSPAR Commission, 2014, Svärd, 2013, HELCOM, 2015). In the eastern North Sea and Norwegian Sea coasts, the composition of beach litter is showing traces from both regional litter situation and from long range transport around the North Sea. The items are both rich in consumer goods such as packaging, fast food articles, certain especially floating articles, and much more fishing and

shipping related litter goods than in the Baltic Sea, that is more consumer litter dominated (Marlin, 2013).

Other than the regional input of litter the composition of litter in beach surveys and surface water samples may also depend on long range transportation with ocean currents. In Nordic waters there are a few large coastal currents that are acting as conveyor belts for marine macrolitter. Especially important are some branches of the Gulf Stream that transport litter from the eastern UK and up the English Channel to join in the Jutland Current along the Danish coast. The Jutland Current consequently transports litter of the drainage from a large part of Western Europe. The Baltic surface waters is mostly flowing out of the sounds between Denmark and Sweden and can thus transport both litter from those coasts and from the greater Baltic Sea. There is less known about transport and accumulation of marine litter within the greater Baltic Sea area. The Jutland Current have higher salinity than the Baltic current and the denser Jutland waters are being submerged in convergence zones in the Skagerrak and transient accumulation of floating debris can often be seen. This floating macrolitter is though effectively washed ashore on the Swedish or Norwegian Skagerrak coasts when there is a westerly or southerly wind. The situation of transport and accumulation of microlitter along the Nordic coastal waters is however, virtually unknown. So is the contributing effect of the regional input on more long-range transport. Model simulations show that litter that is emitted to the North Sea, or transported from the Nordic coastal zones to the North Sea, has potential to be transported with surface water currents to the Arctic ocean (Lebreton *et al.*, 2012), where a rapid increase in marine litter on the seafloor have been observed since 2010 (Tekman *et al.*, 2017). In fact, a majority of the litter that enter the marine environment is expected to sink to the ocean floor due to vertical transportation processes such as biofouling and the formation of marine snow (Holmström, 1975). Therefore it is important, in order to understand the exposure risk and fate of microlitter, to include compositional studies of microlitter in sediment.

The overall aim of the project was to investigate common microlitter types in Nordic coastal waters. The first specific objective was to evaluate and transfer knowledge between the partners of different methods to sample, prepare and analyse microlitter samples from surface water, pelagic and sediments. Discussion about selecting suitable particle size classes will be included. The second specific objective was to compare, learn from each other and report about the common microlitter types found in selected case studies of the partners. The scopes of the case

studies have been to sample gradients from potential sources, as water or sediments. That has been done in order to differentiate between local sources and long range transport. The third specific objective was to initiate a discussion with stakeholder authorities that have already or may show interest and responsibility to monitor microlitter in the Sweden, Denmark and Norway. The report is thus concluding with a section on the prerequisites and potential for a marine microlitter monitoring programme in the Scandinavian countries.

2. Materials and Methods

2.1 Case studies overview

To investigate important local sources of different microlitter types in the Nordic waters three gradient studies was performed; that include transects reaching from areas with a high density of anthropogenic activity, towards the open sea, and in an urban creek from more clean to heavy impact to the sea, as well as some sampling of potential hot spot sources (Figure 1). Since some microlitter composition and size fractions is expected to be more prone to sedimentation while others more buoyant, then both surface water and sediment has been sampled. The case study areas, environmental compartments and the applied methods are listed in Table 1.

Figure 1: Overview map including the four sampling sites in Denmark, Sweden and Norway

Table 1: Sampling and analysis overview from case studies

Environmental compartment	Case study area	Sampling method	Mesh/Pores	Treatment	Analytical method
Road runoff sediment	Karmøy-tunnellen	Manual scooping	38 µm	Elutriation	Visual
Sediment	Roskilde Fjord	Van veen grab	1,000, 300, 100, 38, 20 µm	Chemical digestion, density separation and decantation	Visual
Sediment	Kville-bäcken creek, Gothenburg	Box corer	300, 100 µm	Density separation	Visual+ SEM
Water	Oslo fjord	Trawl	300 µm	None	Visual+ FTIR
Water	Oslo fjord	Ruttner	10, 1 µm	None	Not yet analysed
Water	Roskilde Fjord	Pump	1000, 300, 100, 38 µm	None	Visual
Biota	Roskilde Fjord	Pooled 10x3 blue mussels		Chemical digestion	Visual

2.2 Method description: Karmøy tunnel study (NIVA)

Sludge from a sedimentation pool and dust from a noise reduction chamber in Karmøy-tunnellen just south of Haugesund, Norway (Figure 2), were removed with a spoon and transferred to plastic boxes. The samples were taken by Karmøy-tunnellen sent to NIVA where they were stored in dark at room temperature.

Figure 2: Map of location of the Karmøy tunnel in Haugesund, Norway

The technique used to extract microlitter from sediments from Karmøytunnelen was an elutriation device (ED). Elutriation is a separation technique that entails an upward stream of gas and/or liquid to isolate lighter particles from heavier. Particles are pushed upwards with the water and gas/air through a cylinder column and pours out of the nozzle at the top. Any particles that follows this flow can be captured on a filter or mesh under the nozzle at the top. At NIVA we used an ED (Figure 3) made to the same specifications as Claessens *et al.* (Claessens *et al.*, 2013), only slightly modified to fit the use in our lab. The full length of the ED is a 167.8 cm consisting of a 35 cm base, a 105 cm PVC cylinder column and the 27 cm “nozzle” at the top. The base of the device fastened by bolts circling the flanges measures and can be opened and closed. In between the PVC cylinder and the base, a 1 mm mesh screen was placed on top of a 38 μm mesh screen to allow water to flow through, but no particles $>38 \mu\text{m}$ to escape nor enter the water column. The whole system was aerated by a Hiblow HP-80 air pump with a capacity of 80 L/min⁻¹. Three standard aquarium air stones

measuring 50 x 20 x 20 mm was placed in a “Y” pattern to minimize the formation of dead zones inside the column. A 9 mm one-way valve controlled the amount of air flowing from the pump and could manipulate the amount of air flowing into the system (approximately between 40 and 20 L/min⁻¹). The ED was connected to a tap in the lab by a garden hose. The fresh water flowed in at the base of the ED pushing itself up the column. At approximately 10 cm below the nozzle mouth, the water was turned off and the aquarium stones lowered into the cylinder from the top down to the bottom. Once in place and the air flow sufficiently strong, a sediment sample was added from the top. The sediment and the aerated water was allowed to mix (approximately 30 seconds) before the water was turned on again. The water then overflowed out the nozzle and through a 100 µm mesh. The overflowing water and any particles passing through the mesh were directed to the drain through another hose. After approximately 20–40 min, the dark and sediment-mixed water column became clearer until all smaller particles had been washed out of the sample (the time varied from sample to sample). What remained in the 100 µm mesh at the top nozzle and from the bottom 38 µm mesh were collected for further analysis.

Figure 3: Road runoff sediment elutriation device

M. Claessens et al. / Marine Pollution Bulletin 70 (2013) 227–233

Note: A) The left picture is a sketch of the original ED for microplastics extraction (Claessens *et al.*, 2013).
B) The picture on the right is the ED used at NIVA (Photo: D.Eidsvoll).

2.3 Method Description: The Oslo fjord study (GU)

A series of 13 stations in the regular monitoring program carried out by the Oslo University research vessel Tryggve Braarud, was visited during December 2015, from the innermost harbour and city of Oslo out through the fjord to less populated coastal areas (Figure 4).

Figure 4: Oslofjord case study maps

Note: A) An overview map of the sampling stations in the Oslo fjord area, with the innermost stations highlighted in B).

Trawl samples were collected with the Avani high speed trawl, developed by 5 Gyres institute, LA, USA (Figure 5). To avoid contamination and microlitter loss a 300 μm mesh was attached to polypropylene tubes with hose clamps. Prior to sampling the tubes were prepared in the lab, back-flushed, wrapped in aluminium foil and attached to the cod end of the trawl, using double hose clamps. Individual tubes were used for each station. Between stations the trawl was rinsed in the water to minimize carryover. On deck the trawl was kept clean from dust at the entrance.

Figure 5: Sea surface microplastic trawl

Note: A) Avani high speed trawl with 300 μm mesh size.
B) The “cod end” of the net with preassembled 300 μm mesh and protected from contamination with aluminium foil.

Source: M. Hassellöv.

For smaller fractions surface water samples were collected with a Ruttner sampler and filtered through stainless steel filter holders housing polycarbonate filters with pore sizes of 10 μm and 1 μm (Figure 6). The filter holders were prepared in a clean air hood and wrapped in aluminium foil before and after filtration to minimize air exposure. The filter samples will be analysed with light microscopy, electron microscopy and Raman microscopy, once new instrumentation has been made available (June 2017).

Figure 6: Sampling pelagic microparticles with on line membrane filtration

Note: A) Collecting a “clean” subsurface water sample with a Ruttner sampler.
B) Preassembled membrane filter holders that connect directly to the Ruttner sampler for clean vacuum filtration.

Source: T. Karlsson.

2.3.1 *Analysis with visual identification and FTIR spectroscopy*

All 300 μm samples were identified, classified and counted under a stereo microscope. From each sample 20 random particles was selected for FTIR analysis. These particles were first photographed under the stereo microscope before transferred to FTIR microscope. If reflectance mode did not find reliable spectra with high signal to noise ration then ATR mode was employed.

2.4 Method Description: Roskilde fjord study (AU)

2.4.1 *Sampling*

Samples of water filtrates, sediment and blue mussels were collected from each of the four sampling site in Roskilde fjord (Figure 7).

Water filtrates were collected by pumping one m³ of sea water collected about 10 cm below the surface of the water through sequential filter system with mesh sizes of 1,000, 300 and 100 µm consisting of zooplankton net. 100–300 litre of water could generally only be sampled before for the smallest mesh size before water flow was blocked by phytoplankton.

Sediment samples were collected with a hand held box core at depths of 3–4 meters. For each samples, the top 2 cm sediment layer from three subsamples were pooled into one sample at about 500 g.

Blue mussels living on top of the sediment were collected with a small dredge.

2.4.2 *Sample treatment*

For water filtrates, whole filters in each size fraction were examined for microplastic. Sediment samples were first homogenized carefully by shaking and a subsample of ~100g (corresponding to ~50ml) was then transferred to for further treatment consisting of 2 hours chemical digestion for reduction of content natural organic matter using a 100 ml solution of 5% NaOCl, 3% KOH and 1% STPP. This was followed by density separation using 200 ml saturated saline water and the total water fractions were decanted into a new glass container. This step was repeated 3 times and the decanted fractions were pooled into one sample container that afterwards was size fractionated with metal test sieves of 1,000, 300, 100, 38 and 20 µm using wet filtration with filtered water.

Blue mussels were first open and soft bodies of 10 individuals (corresponding to about 20g wet weight) were pooled into one sample that following were chemically digested for 2 hours using a 100 ml solution of 5% NaOCl, 3% KOH and 1% STPP. The sample was then filtered using 20 µm filter. Three replicate pooled samples were analysed from each site.

All applied glassware have been baked and reagents and solutions were filtered before use to reduce the risk for internal contamination. Daily control of assessment of potential risk for airborne contamination risk was performed using three water filled petri dishes placed at different locations in the laboratory.

2.4.3 *Visual identification and counting*

Each sample was examined using a stereo microscope. Identified microplastic particles were classified according to shape and colour and counted. Only fibres longer than 100 µm were targeted in the sample fractions below 100 µm.

Figure 7: Map of the sampling stations in Roskilde fjord

2.5 Method Description: Kvillebäcken study (IVL and GU)

Sediment samples were collected with a sediment grab along the creek Kvillebäcken that passes through an area with numerous urban activities, and eventually is emptying into the river Göta Älv just before it opens up into the sea (Figure 8). Sampling was done at three sites in the creek (KB1–KB3) and two in the river, one where the creek opens up into the river (KB4) and one at some distance downstream (KB5). Samples were kept cool but not frozen until analyses.

Figure 8: Map of Gothenburg with the sampling sites in Kvillebäcken urban creek and sites in the river towards the sea

The sediment was thoroughly homogenized and a subsample of around 500–700 g sediment (wet weight) was taken out from each sampling location. To separate microplastics and other microlitter particles from sediment minerals a density separation was performed using saturated NaCl solution ($1.2 \text{ g}\cdot\text{cm}^{-3}$). The separation

was carried out in a separation tower of stainless steel, slightly modified from a construction by Imhof *et al.* (2012). In short the sediment and around 4 L of saturated NaCl solution was poured into the tower from the top while a rotor (14 rpm) was turning in the bottom which kept the sediment suspended. The sediment was left like this overnight and thereafter allowed to settle to the bottom of the tower. The entire water fraction down to a few centimetres above the sediment surface was filtered in sequence over a 300 and a 100 µm filter.

Analyses was done by visual inspection using a stereo microscope, 60x, and also through manipulating particles with tweezers and melting tests. A selection of the black anthropogenic particles were analysed with scanning electron microscopy (SEM), Zeiss Sigma VP with secondary electron detector (surface sensitive, topography), backscattered electron detector (atomic number contrast), and energy dispersive X-ray spectroscopy detector (elemental maps). The particles were mainly imaged in variable pressure mode up to 150Pa N₂.

3. Results and discussion

3.1 Case study results overview

An overall aim of the project was to identify the most commonly found microlitter types in Nordic waters. We have realized that the dominating types differ to a large extent depending on the sampled environmental compartment, but that there are also some common elements. In Table 2, can be seen that in surface waters the low density buoyant polymers microplastics (polyethylene, polystyrene and polypropylene) of fragments, foil are together with textile fibres dominating while in urban and road sediments black particles appearing to be wear from tires, asphalt, soot and charcoal and tar-like particles are common. Some additional semi-solid classes typically not considered as microlitter was found to be fat and oil/tar like particles.

Table 2: Identification of most common microlitter types in each case study

Type	Case study area	3 most common litter types
Water	Oslo fjord	FTIR identification: Polystyrene (PS), Polyethylene and Polypropylene Visual: White foamed particles, Fragments and Fibers/Filaments Also: Fat particles common
Sediment	Roskilde Fjord	Visual: Fibres, films, granules, fragments
Sediment	The creek Kvillebäcken, Gothenburg	SEM identification: carbon rich Visual and tactile: Elastomer (rubber like particles), Charcoal-like (black porous, brittle) particles, Plastic Fragments Also: Semisolid oil tar-like particles and fat common
Road sediment	Karmøy tunnel	Visual: Tire rubber-like particles, road markings, asphalt
Water	Roskilde Fjord	Fibres, plastic films, granules/fragments
Biota	Roskilde Fjord	Fibres

3.2 Case study results from Karmøy tunnel runoff sediments (NIVA)

The results from the nine sediment samples, eight from a sedimentation pool and one from a noise reductions chamber, are summarized in the Table 3.

Table 3: Sampling scheme for NIVA tunnel study

Sample	Sample location	Microlitter	For future	Sample
9	Karmøytunnelen	Yes	FT-IR	Sludge separator
8	Karmøytunnelen	Yes	FT-IR	Sludge separator
7	Karmøytunnelen	Yes	FT-IR	Sludge separator
6	Karmøytunnelen	Yes	FT-IR	Sludge separator
5	Karmøytunnelen	Yes	FT-IR	Sludge separator
4	Karmøytunnelen	Yes	FT-IR	Sludge separator
3	Karmøytunnelen	Yes	FT-IR	Sludge separator
2	Karmøytunnelen	Yes	FT-IR	Sludge separator
1	Karmøytunnelen	No	-	Sound trap

From the visual identification, potential microlitter particles were identified in all samples collected from the sludge separators (minimum mesh size of 38 µm). One sample analysed from the noise reduction chamber did not contain any potential microlitter particles retained by our mesh size. The visual identification of microlitter in sample no. 4, 6, 7 and 9 are described in the pictures below and represent examples of the type of microlitter found in all the samples.

Figure 9: Road runoff grain showing complex composition

Note: This picture from sample no. 4 (see the table above) is from a part of the surface layer of the sediment sample (this does not imply it necessary was part of the surface of the sediment in the sludge separator). The picture reveals several pieces of potential microlitter in the size range of 50–100 µm.

Figure 10: Black road runoff particles

Note: A) These two pictures are of the same particle (different exposure in the two pictures) from sample no. 6 (see table above).
B) This black “mixture” particle is potentially a composite mixture of car tires, asphalt, sediment and other particles captured in the tunnel, mixed together. Similar particles could be identified in all the other samples from the sludge separator.

Figure 11: Yellow road marking particles

Note: These three pictures identify different particles from sample no. 7 (see table above).
A) The particles in the upper picture are most probably wear of the road paint from the middle part of the road (Norway has often yellow road paint).
B, C) In the two lower pictures similar yellow particles can be identified on top of a cigarette filter also found in the sample.

Figure 12: Complex mixture of road runoff wear particles and microlitter

Note: These three pictures identify different particles from sample no. 9 (see table above).

A) The particles in the upper left picture are an example of the dried particles from the sample. Again we can identify yellow road paint particles this time amongst the black "mixture" particles with an orange and a round clear particle.

B) The particles in the upper right picture are a representative of the wet sample just after separation in the ED.

C) The lower picture is a concentrated picture of the sample identifying yellow particles, blue and white flakes, the black "mixture" particles and round clear particles identified as possible microlitter.

3.3 Case study results from the Oslofjord (GU)

The results from the trawl samples in the Oslofjord show a 30 times higher microlitter concentration per m^3 in the innermost samples compared to the outermost (Figure 13).

Figure 13: Particle concentration per m^3 as calculated from trawls samples with a mesh size of $300\ \mu\text{m}$

A closer examination of the composition showed that more than 50% of the microlitter particles as identified by visual identification consisted of white foamed particles. FTIR analysis ($n=100$) identified them as polystyrene.

Spectroscopic analyses of 60 random particles confirm that the majority consist of polystyrene (PS) but that there is also some polyethylene (PE) and polypropylene (PP) as well as one polycarbonate (PC) (Figure 14).

Figure 14: Polymer identification made with FTIR spectroscopy (Thermoscientific NicoletIN10) on 60 random particles

The composition differed between the 7 samples taken in the innermost part (Figure 15) of the fjord and the 4 samples taken in the outer parts (Figure 16). In the inner samples 62% of the microlitter was polystyrene compared to 5% in the samples taken outside. Instead there was a 5% increase in fibres and filaments, a 3% increase in other particles and a 44% increase in fragments.

Figure 15: Microlitter composition in trawl samples taken from the inner parts of the fjord, as identified by visual identifications

Figure 16: Microlitter composition in trawl samples taken from the outer parts of the fjord, as identified by visual identifications

3.4 Case study results from Roskilde fjord water, sediment and biota (AU)

The case study is based on parallel sampling of water filtrates, sediment and blue mussels from four stations in Roskilde fjord and of water filtrates from different sources which are thought to be important for inputs of microplastic to Roskilde fjord, i.e. WWTP effluent, harbour, freshwater stream and storm water.

The results of the water filtrates showed that the sources with 28–58 particles per m^3 characterised as microplastic in size fractions $>100\text{ }\mu\text{m}$ generally contained more particles in water filtrates from the fjord, where 2–11 microplastic-like particles per m^3 were found (Figure 17). The size fraction $100\text{--}300\text{ }\mu\text{m}$ contributed with 88–100% in all samples. High uncertainty was introduced on the presence of articles $>300\text{ }\mu\text{m}$ due to only finding single particles (0–7 particles) when filtration of 1 m^3 water. Fibres were in all samples the dominating shape for the microplastic-like particles, followed by films and fragments. Dominating colours were blue followed by black, transparent whitish and red particles.

Figure 17: Levels of microplastic-like particles in size fraction 100–300 μm and >300 μm in water filtrates from Roskilde fjord and at potential sources

Figure 18: Classification of microplastics categories from Roskilde fjord

Note: Composition of the microplastic-like particles according to their shapes found in the samples collected at the B) sources and C) in the fjord.

The results of the sediment samples from the fjord showed higher levels in the inner part of the fjord (Roskilde Vig) inclusive the harbour compared with the stations located in the more outer parts, i.e. in Kattinge Vig and Roskilde Bredning especially for particles sizes >100 μm . If data were normalised to content of 5% total

organic carbon (TOC) a clear trend appeared in the dataset from the harbour and outwards (Figure 18). This supports that adjusting to total organic content in sediments can have an important effect when comparing microplastic data. Somewhat unusual, particles with sizes in the range 20–100 µm seem to be underrepresented in the samples, and further studies inclusive reanalyses are needed to understand this trend as it differ from results from sediments analysed from other areas in Denmark. Regarding shapes, fibres were again dominating followed by film and granules/fragments, Blue was also here the dominating colour, followed by black, transparent and red.

Figure 19: Microlitter sediment abundance, distribution and classification

Note: Levels of microplastic-like particles in sediment samples from Roskilde fjord reported on:
A) Dry weight basis.
B) Basis of total organic carbon (TOC) normalized to 5% TOC content.
C) Shape composition of the microplastic-like particles found in the sediment samples from the fjord.

Microplastic-like particles were also found in all samples of blue mussels collected in the fjord, but with no larger difference in levels between the different sites (Figure 19). A similar pattern was also reflected in the water filtrate samples. Fibres were again the dominating shape category followed by films and fragments.

Figure 20: Microlitter biota abundance, and classification

Note: A) Levels of microplastic-like particles in blue mussels from Roskilde fjord and at potential sources.
B) Composition of the microplastic-like particles according to their shapes found in the samples.

3.5 Case study results Kvillebäcken creek sediments (IVL and GU)

The character of the sediments from the creek (KB1–KB3) differed significantly from the sediments collected from the mouth of the river Göta älv (KB4–KB5). The creek sediments were dark and contained dead leaves and seeds from trees. Sediments from KB2 and KB3 had a distinct smell of oil. The Göta Älv sediments were oxygenated and at station KB5 there were remains of marine macroalgae and empty shells of marine mussels. The creek sediments KB2 and KB3 were expected to be closest to obvious microlitter sources, e.g. fragmented litter particles from industrial and shopping areas and various kinds of black particles from parking lots and roads. KB1 was selected as a site upstream intense urban areas and KB4 and KB5, although situated in a busy maritime environment, were supposed to be less affected by land-based activities. However, analyses showed that all three sites from the creek, KB1–KB3, had dramatically elevated microlitter concentrations compared to the river sediments KB4 and KB5, and they were therefore grouped together when presenting the results.

Black particles dominated the anthropogenic microlitter found in the sediment of the creek Kvillebäck, whereas the microplastics dominated in the sediments of Göta älv. Concentrations of both black anthropogenic particles and microplastics were considerably higher in creek than in river sediments, with 15,490–16,798 black particles and 312–868 microplastics $\geq 100 \mu\text{m}$ at stations KB1–KB3 and 121–436- black particles and 24–48 microplastics $\geq 100 \mu\text{m}$ at stations KB4 and KB5 (Figure 20 and 21). The majority of the black particles at the stations KB1–KB3 were in the size range $<300 \mu\text{m}$ – $\geq 100 \mu\text{m}$ (i.e. particles collected on the $100 \mu\text{m}$ filter), whereas the microplastics were more equally divided between the fractions collected on the 300 and $100 \mu\text{m}$ filters respectively (Figures. 22 and 23).

Figure 21: Black anthropogenic particles $\geq 300 \mu\text{m}$ in sediment (dry weight) from three locations in the creek Kvillebäck (KB1–KB3) and two stations where the creek empties into the Göta älv estuary (KB4–KB5)

Note: Grey part of the bars indicate particles collected on $300 \mu\text{m}$ filter and the black bar represent particles collected on $100 \mu\text{m}$ filter.

Figure 22: Microplastic particles $\geq 300 \mu\text{m}$ in sediment (dry weight) from three locations in the creek Kvillebäck (KB1–KB3) and two stations where the creek empties into the Göta älv estuary (KB4–KB5)

Note: Blue part of the bars indicate particles collected on $300 \mu\text{m}$ filter and the red part particles collected on $100 \mu\text{m}$ filter.

Three categories of black anthropogenic particles were detected, sticky, oily, tar-like particles that were compressed when squeezed with a pair of tweezers, elastomeric particles that retook their original shape after compression and charcoal particles that fell to pieces when squeezed. Two kinds of elastomeric particles were detected, one with clear cut edges and one more crumbled. Both kinds were however categorized only as elastomers (Figure 23). The oil particles were the most numerous of the three categories at all stations (on average 65% at KB1–KB3 and 53% at KB4 and KB5) (Figure 24). In the creek sediments the second largest group were the rubberlike elastomers (32%) and least abundant were the charcoal particles (3%). In the river sediments (KB4 & KB5), where the total number

of black particles were two orders of magnitudes lower than in the creek, charcoal particles were more abundant than elastomers (~37% compared to 10%). Microplastics in KB1–KB3 were dominated by fragments (on average 94%), followed by fibers (6%) and very few plastic flakes (~0%) (Figure 25). Fragments were also the dominant group of microplastics in KB4 and KB5 (on average 73%), although plastic fibers and flakes were more frequent (~19% and 8% respectively). When separating microplastics into colours, semitransparent particles of different shapes were the most commonly occurring in all sediments. In the sediments at site KB1 green plastic filaments, probably deriving from an artificial turf close to the creek, were also particularly common (Figure 26).

Another kind of particulate matter origin from human activities that was found at relatively high abundances, particularly at stations KB2 and KB3, were irregular lumps of fat (Figure 27).

Figure 23: Black microlitter particles from urban creek sediments

Note: A) Two categories of black anthropogenic particles, a rubberlike elastomer with clear cut edges and a sticky oil particle.
B). Elastomer particle with crumbled appearance.

Figure 24: Relative abundance of three different categories of black anthropogenic particles, oil particles, rubberlike elastomers and charcoal

Note: The sample sites were divided into those close to sources like industrialised areas, parking lots etc., which were sediments from the creek (KB1–KB3) and those sites away from obvious point sources which were sediments from the river (KB4–KB5). Data is based on number of particles per sediment dry weight.

Figure 25: Relative abundance of three different categories of microplastics, fragments, flakes and fibres

Note: The sample sites were divided into those close to sources like industrialised areas, parking lots etc., which were the sediments from the creek (KB1–KB3), and those sites away from obvious point sources which were the sediments from the river (KB4–KB5). Data is based on number of particles per sediment dry weight.

Figure 26: Green plastic fragments from sediments at KB1

Figure 27: Lumps of fat were frequently occurring in the sediments at sites KB2 and KB3

The different types of black particles was also analysed in SEM with energy dispersive X-ray spectroscopy (SEM-EDS). The secondary electron detection (VPSE) provide high surface topography imaging, while the backscatter electron detection (BSD) respond to the atomic number composition of the material. The substrate is aluminium so anything carbon based will be darker and anything made out of minerals/metals can be lighter in contrast. The fine structure can be shown to confirm the visual and physical testing as well as the elemental maps showing that the material is carbon rich and contain some other elements depending on the particle. The rubber like particles contained C, Ca, S, and to some extent Zn.

Figure 28: Black clear-cut elastomer particle in Scanning electron microscopy

Note: A) Above, overview backscattered detection image indicating a particle lighter than aluminium background, with clear-cut edges.
B) Lower left showing the secondary electron surface topography.
C) right the elemental map of carbon.

Figure 29: Black charcoal-like particle in Scanning electron microscopy

Note: A charcoal like particle (black, porous, brittle to tweezer pinch):

- A) Overview in secondary detection mode.
- B) High magnification, in backscattered detection mode.
- C) Elemental map for carbon also showing porous structure.

Figure 30: Black crumbled elastomer particle in Scanning electron microscopy c

Note: A–D) Secondary electron and elemental EDS maps indicated composition including carbon, calcium and sulfur and to some extent zinc.
E) Lower panel show detailed structure in high magnification.

Figure 31: Black elastomer particle in Scanning electron microscopy

Note: A) Secondary electron image of black particle with worn like appearance, rubber like elasticity and sticky texture. Site KB₃.
B) Same particle with elemental map for carbon.
C) With titanium and iron together indicating a carbon based particle with mineral grains on the surface.

Figure 32: Images showing a reference tire granulate particle from an artificial football turf collected from the field for comparison to above elastic black particles

3.6 Discussion on methods

Microplastics and microlitter analysis is a relatively recent scientific discipline which is rapidly developing but still relatively immature. The field of microplastics abundance measurements was sprung out of zooplankton methods of sampling and identification with mainly neuston nets, and stereomicroscope identifications. The scope have been expanded to include smaller sizes, sediments and biota, and complemented with chemical identification methods to improve the quality control, since analysis based on solely visual identification can lead to both positive negative as well as negative positive identifications (Hildago-Ruz *et al.*, 2012). Here is briefly discussed the whole analytical chain from design of monitoring programmes, sampling, sample processing and analysis and quantification including quality assurance aspects.

3.6.1 Considerations for design of monitoring programmes

The microlitter field is just starting to be established so it is very early to have conclusive opinions on design of complete monitoring programmes, but some suggestions at least applicably to screening studies will be given. To support environmental management to take measures to mitigate the pollution of microlitter or macrolitter that can fragment into microlitter, or to understand long range transport or accumulation of microlitter the following should need to be covered.

Point sources

Potential point sources from the land to sea should be sampled and include storm water effluents (as near different types of activities as possible), sewage water treatment facilities, major rivers representing mixed sources, industrial effluents especially plastic industry, marinas and commercial ports. Specific activities that have been pointed out as potentially most important (Magnusson *et al.*, 2016) include tire and road wear, styrofoam from construction and marine operations, industrial plastics, and artificial sport arenas.

Diffuse sources

Both long range transport, marine sources (e.g. fishing, shipping, aquaculture and offshore industry), mixed sources, and macro litter fragmentation can contribute to what can be considered diffuse sources, and the design of near shore and offshore sampling points should be selected to as far as possible be able to differentiate those contributions.

Environmental compartments

Different microlitter compositions and sizes will inevitably transport and accumulate differently so both sea surface layer, the pelagic and sediments need to be monitored. Sampling the sea surface layer with trawling devices is directly relevant to the exposure of surface foraging sea birds and therefore relevant and relates to the monitoring of stomach content of Northern Fulmar birds. However, the variation of sea surface concentrations is believed to be most variable due to wind and currents, and need to be frequently sampled and correlated to those variables. Accumulation bottom sediments is already included in many monitoring programmes and should appropriately be monitored for microlitter both for exposure of sediment organism and for assess loadings and distribution in the environment. Since most marine sediments are effectively mixed by the benthos, assessing loading trends can be difficult so therefore filter feeding organisms such as bivalves may be valuable complement. Which species that is suitable for biomonitoring is however not yet established.

3.6.2 Overview QA/QC

In any analytical measurement there is critical to assess and report the complete measurement uncertainty. For microlitter analysis the field is presently not mature enough for that but certain elements are becoming established.

Blanks

It has been recognized that certain types of microlitter is very easily contaminated from the surroundings (the ship, the analyst, the laboratory environment where the samples is handled). There are many early studies where adequate analytical procedures were not reported and the results therefore uncertain, but other fields of trace environmental analysis can give guidance. The materials in sampling equipment should be free from

plastics and thoroughly washed. Blanks should be measured and preferably follow the entire sample chain from field, extraction, analysis.

The blanks could be subtracted from the particle count but it may often be considered beneficial to report them separately as in “maximum blanks counts corresponds to 5% of total MP count in samples” or as counted numbers.

It is perceived that the risk increases with decreased particle size, but also commonly observed that certain types of microlitter is more common in the blanks, such as textile fibres but this may vary with each study.

Recovery/reproducibility

There are many different types of methods used for microlitter studies. In order to increase comparability it is important that the method of choice is tested for recovery and reproducibility. In order to make quantitative measurements it is crucial to include a QA. Similar to the blank measurements the aim should be to integrate these steps on all stages; sampling, extraction and analysis.

Until a certified reference material is adapted an internal reference material to assess recovery is recommended. To repeatedly re-analyse a laboratory control sample is another way of internal QC. Such QC sample should ideally include the most common polymers in different shapes and appropriate size-fractions adapted for the different methods. An internal standard material should include both a distinctly different (e.g. coloured) particles but otherwise with relevant properties to assess recovery of sampling and sample treatment.

Sampling

Particle analysis based on microscopic identification is a counting exercise and as such obeys counting statistics, so called Poisson statistics. Poisson statistics describe the relation between number of counted particles as a function of sampled volume with a certain concentration on a filter and the associated measurement uncertainty. The standard deviation (%) relates to 100% divided by the square root of number of counted particles. As can be seen in Figure 33 for the total number of particles the standard deviation descend 10% after 100 counted particles, but when one need to differentiate also the “red” particles at 10% uncertainty then 1000 total amount of particles need to be counted, or at least minimum 100 particles of each class that should be differentiated.

Figure 33: Hypothetical example describing relation between # of particles counted and uncertainty

Note: Standard deviation calculation based on Poisson statistics for a hypothetical example of counting of particles on a filter with the distribution between three different types as black: blue :red as 7:2:1.

Therefore one need to choose the sampled volume of water or sediment or biota tissue, so one obtains a particle count that exceed the number of counted particles for the least abundant category one need to determine with a targeted maximum uncertainty.

Generally, there are fewer particles the larger they are, since particles are generated by wear and fragmentation processes, and therefor sampling methods designed for sampling larger sized particles also need to process larger volumes. The selected sample volume also need to be adjusted for the abundance of a given sample, so off shore samples with lower concentrations than a near point source also need to be processed larger sampled volume. Typically, >300 μm trawl samples, requires 50–100 m^3 of volume near the sources to be sieved since typical abundances are 1–10 particles/ m^3 , while offshore with a concentration 0.5–2 particles/ m^3 requires 100–200 m^3 . For >10 μm membrane filtration on the other hand requires only 1–4 L of sample, but is not accurately sampling the largest particles. Similar considerations need to be taken for pump sampling and sediment sample volume. An outstanding question

that relates to selection of pristine reference locations; how much do we need to sample to say there are no particles?

The counting statistics uncertainty in relation to other sample to sample variation factors still need to be empirically demonstrated.

Pumps vs trawls

Although different types of trawl nets, sometimes called neuston nets or manta trawls) are most commonly used for sampling large volumes of larger size fractions, there are also examples of pump filtrations. There are few side-by-side comparisons and here will only be given some general pros and cons for the two methods.

Pump sampling is practical for point sources, that trawls have more difficult to achieve (vertical nets could be used where appropriate). Pump filtration can be equipped with mesh size that is smaller than what a trawl can be equipped with before a hydrodynamic bow wave is build up in front of the trawl that reduce the sampling efficiency. Pump filtration however, generally has problem with sampling the sea surface layer that the trawl does include. Contamination issues may be important if the pump or hoses are placed in front of the filter. The trawl has advantage to be place outside the influence by the ship, and integrate over larger areas of the sea, it captures the sea surface. A weakness is that the sampled depth is less accurately determined and conversion to sampled volume therefore less accurate than for a pump.

3.6.3 Sample treatment

Chemical digestion

In order to reduce the amount of natural particulate matter, and extract the microlitter from natural matter to facilitate subsequent separation and/or analysis, a chemical procedure can be employed. There are currently a number of digestion protocols for different types of samples available in the literature (Kühn *et al.*, 2017, Mintenig, 2014). Although it is not clear which methods is overall superior, some discussion towards the approaches used by the partners will be given here.

Some methods including strong oxidizing acids like nitric and perchloric acid, as suggested in for example ICES (ICES special request advice published 3 June 2015), but have shown to affect the physical and/or chemical properties of the plastic (Enders *et al.*, 2017a). Although there is no current consensus on which method to use four main tracks can be identified amongst the Nordic groups (Table 4).

Table 4: Four main tracks of chemical digestion protocols with strengths and weaknesses

Chemical agents	Matrix removal efficiency	Plastic matter integrity	Comments
KOH	Applicable to tissue	Potential impact on some type of plastics	
KOH 3%+ NaOCl 5%+ Na ₄ P ₂ O ₇ 1%	Applicable to all matrixes	Potential impact on some type of plastics	
Enzymatic methods	Different enzymes effective to different matter. Need adaptation	Not affecting plastics	Some enzymes expensive
H ₂ O ₂	Effective on all matrices than can be oxidized	May influence some plastics	Create lots of foam

Choice of digestion method might depend on the needed level of removal for identification method, if density separation is going to be applied after, cost of chemicals, if identification is only purpose or also degree of weathering of the plastics (then oxidation influence on plastics is critical).

Density separation and elutriation

Since sediments contain very high proportion of natural particulate matter it is critical to obtain an effective separation of the microlitter from the natural solids. The sediment matrix consist of mineral grains and an organic matrix that often act as a glue between the microlitter and the minerals. Therefore it is advantageous to first treat the samples with a digestion protocol to break up this structure as a first step.

The density separation include creating a dilute slurry in a heavy density liquid, and in its most simple form repeat a slurry step and decanting procedure, and with a closed system approach use a separating tower that collects the buoyant fraction through a filter on top (Claessens *et al.*, 2013, Imhof *et al.*, 2012).

There is also a method to separate lighter particles from heavier that is based on elutriation with water and air bubbles according to (Claessens *et al.*, 2013) and have been used by NIVA in this study.

The choice of density liquid depend on which type of plastics or microlitter compositions that is targeted to be separated and other considerations such as cost and green chemistry aspects. Quinn (Quinn *et al.*, 2017), investigated the recovery of different sizes of different polymers in a number of density liquids and found that

saturated NaCl (density 1.2 g/cm³) gave with some exceptions a recovery above 80% for sizes 200–400 µm, and above 70% for 800–1,000 µm, while 25% of saturated ZnBr₂ (density 1.7 g/cm³) gave a recovery between 95–100 %.

3.6.4 Analytical

At presently there are many opinions regarding a visual versus spectroscopic identification methods, but the consensus from this project was similar as the recommendation from the JRC guideline of (MSFD Technical Subgroup on Marine Litter, 2013). Generally our experience is that a visual identification is more accurate the larger the size classes, because the objects is closer in size to their origin, the optical performance is better and tactile (tweezer pinch) or otherwise physical tests (melting or solvent) can complement the visual appearance. Still the visual only schemes is highly subjective until a strict guide and particle identification scheme have been developed, and ideally complemented with automated image analysis routines. For smaller sizes the uncertainty in determination increases that include higher number of false positives and missed true positives as well as erroneously identification of class.

Visual analysis (identification, counting and measuring)

For any kind of identification protocol it is important that ample training is provided and diligent protocol followed. For someone with experience the visual investigation is a strong tool that can provide a lot of insight into the sample composition, (Helm, 2017), but the results only comparable to other studies once a objective protocol and criteria can be transferred. A common reference library would help but until there is an agreed upon protocol for plastic identification it is important to include a detailed description of what the operator count.

Table 5: Suggested structure for physical identification scheme

Visual			Tactile			Size Class
Shape	Colour	Texture	Solid/semisolid	Brittleness	Elasticity	(µm)
Sphere	Blue	Transparent	Semisolid	Fragile	Rubbery	100–150
Film	Red	Glossy			Smearing	150–200
Fragment	White	Porous	...	...	Hard	300–500
Fibre	...	...			...	10x150
...	...	...			...	...

In visual analysis it is often useful to include melting, needle tests, and touching (only practical for bigger particles than $\sim 100\ \mu\text{m}$). For quantitative comparisons an initial tentative recommendation would be that the visual identification for monitoring is restricted to particle sizes $>100\ \mu\text{m}$, as JRC recommended in their report. As the field develops further quality assurance may allow for monitoring of smaller particles.

Preferably description of samples should be based on visual characteristics rather than hypothesis about the sources. (e.g. black elastomer, rubber-like particle rather than tire particle). Any particle definition that is related to a source description should include comparisons with reference materials – for example pictures of tire particles in comparison with the sample particles. This should be coupled with detailed analytical studies of the common classes.

We welcome development of a visual identification scheme that can be supplemented with spectroscopic data, and build a knowledge base of microlitter types, that has a direct link to sources. That should ideally have a structure that includes colour and optical appearance, morphology descriptors (e.g. fragments, flakes, fibres, film, spherules, foam, filaments), texture descriptors as well as physical tests), as well as different size measures (e.g. maximum ferret diameter, and exposed area) that is provided by common image analysis software (Table 5).

Spectroscopic methods

Fourier transform infrared spectroscopy (FTIR) in different modes have been applied to complement and validate visual identification in many microplastic studies (Hildago-Ruz *et al.*, 2012) and more and more comes other vibration spectroscopy methods in use. These include near infrared (NIR) absorption spectroscopy as well as Raman scattering spectroscopy (Kappler *et al.*, 2016). The trend is going from validating a small subset of particles (Mecozi *et al.*, 2016) towards more automated workflows that allow measuring many or even the majority of the microplastic particles on a sample (Shim *et al.*, 2017), (Primpke *et al.*, 2017), (Ivleva *et al.*, 2017). Some researchers are strongly advocating the requirement to identify all particles by spectroscopy, while others argue against the use of very expensive and labour intensive instrumentation. The project team has a consensus that it is important to develop methods towards objective, automatized, evidence based workflows that use as many sources of information on the particles as possible, but still maintains a smooth feasible labour time per sample.

FTIR is available in many labs in academia, and governmental institutes, and represent a medium cost investment. FTIR can be applied in transmission mode where

the infrared light is shined through the particle and the reduction of light on each wavelength is measured. Similarly thin semi-transparent samples can be applied on a mirror and the light absorbed both going down and up through the sample. Then there is a third form of FTIR called attenuated total reflection ATR-FTIR where a hard crystal is pushed into the material and in the interface the light is reflected and bounces back and forth between the probe and the sample, that generates an standing electromagnetic wave called evanescent wave, that excites the bonds in the sample and some of the energy of the evanescent wave is absorbed by the first few micrometer of the sample and the rest is reflected back towards the detector.

ATR has the advantage of being able to probe dark, light absorbing polymers that is otherwise not possible, and ATR often has good signal to noise ratio but it is a surface sensitive technique rather than a bulk material technique. FTIR is suitable for material identification in microlitter research, but limited by some factors such as: presence of moisture, biofilms, degraded polymers hindering spectral matches, and also weak signal to noise for small particle size (in reality typically less than 20 μm).

Raman scattering micro-spectroscopy have become increasingly studied for microplastic investigations due to that for many polymers Raman spectral peaks have a higher signal to noise ratio and due to the confocal microscope design able to probe particles below 1 μm in size (Lvleva *et al.*, 2017). Raman is also not influence by presence of water, but on the other hand sometimes limited by fluorescence of biofilm coatings and from Raman interference from the dye pigments in the plastic particles. Due to the confocality feature focusing on the particle is a critical issue in Raman but less in FTIR.

Until now most FTIR and Raman microlitter investigations is based on a search, point and shoot approach, while a few examples from automated mapping is under development (Primpke *et al.*, 2017).

JRC report recommend that above 100 μm visual inspection complemented with spectroscopic analysis of one tenth of the particles would be sufficient, but for smaller than 100 μm all particles should be subjected to spectroscopic identification to provide evidence based determinations (MSFD Technical Subgroup on Marine Litter, 2013).

Scanning electron microscopy (SEM) is the third microspectroscopic method used. For studies of small microlitter particles (<100 μm) the resolution of optical microscopes on around 1 μm is limiting the amount of fine structure information that can be generated (Hassellöv and Kaegi, 2009). That is one reason why one may lean towards SEM for higher magnification characterizations. Additional reasons are that SEM can be equipped with several types of detectors that provide different chemical informations, like atomic number

contrast or even elemental concentrations in points or maps over the particles. However, SEM is probed by electrons and does consequently not provide any visual colour information. In order to combine the two sources of information one need to work with correlative microscopy tools that University of Gothenburg are currently developing.

3.7 National monitoring authority perspectives

3.7.1 Sweden

In Sweden the marine environmental monitoring is generally managed by the Swedish Agency for Marine and Water Management (SWAM), although the Swedish Metrology and Hydrology Institute (SMHI) has responsibility for biogeochemistry and plankton sampling, Naturvårdsverket (EPA) has overall responsibility for hazardous substances, and the Swedish Geological Survey (SGU) for sediments. Furthermore, the regional EPA, and especially the EPAs with Water authority offices, has responsibilities for coastal water monitoring, and management to assure environmental quality standards.

The project team has been in contact with SWAM (Agnes Ytreberg), SGU (Anna Apler), SMHI (Pia Andersson) and county administration board(CAB), west Sweden (Anna Dimming). The responses confirm that there is on one hand not so clear differentiation of responsibility for monitoring programmes of microlitter between SWAM and EPA but this is something they are aware of and realize it need to be solved. All authorities realize they have big knowledge gaps in relation to marine microlitter but have great interest to build up knowledge and to launch some pilot screening studies, and Anna Dimming at CAB has already commissioned several such surveys.

SGU is interested in taking on sampling for microlitter in their regular sediment sampling, SMHI is interested in the offshore water sampling and CAB in the inshore sampling. SMHI is considering microlitter when outfitting their new vessel. Analysis capacity is something the authorities are thinking about. Anna Dimming at CAB says that according to the Marine Directive implementation in Swedish monitoring, the surveillance of Descriptor 10, should be conducted regionally. But so far no assignment or funding from SWAM has come yet. Currently, they are also working on how to inform the coastal municipalities about artificial football turf management. The Swedish EPA have had a government assignment to investigate the major sources of microplastics from land to sea and that report may lead to further studies or legal measures.

3.7.2 Norway

In Norway environmental monitoring is generally managed by the Norwegian Environment Agency (Miljødirektoratet) and has initiated several literature studies on marine litter and micro plastics (Klima- og Forurensningsdirektoratet 2010, Miljødirektoratet 2014a). Followed by reports on distribution occurrence and effects (Miljødirektoratet 2014b) and sources (Miljødirektoratet 2015a). Until recently monitoring activities in Norway have mainly been focussed on Norway's reporting to OSPAR and limited to beach surveys, surface observations or plastics in fulmar stomach coordinated by the Norwegian Environment Agency (Miljødirektoratet 2015a). The Norwegian Environment Agency has also recently initiated a monitoring study of micro plastics from sewage treatment plants to the marine environment. Micro plastic have also been added to the Contaminants in Coastal Waters of Norway program to start in 2017 and initiated a small monitoring study of micro plastics in mussels and sediments in the Oslo fjord. The Ministry of Climate and Environment have given the Environment Agency the task to find measures to reduce marine litter and the emission of micro-plastic to the marine environment. Microplastics are also specifically mentioned by the Norwegian Foreign Office in the strategy document "Sea Foreign and Development Politics" (Foreign Office 2017).

3.7.3 Denmark

In Denmark the marine environmental monitoring is at a national level managed by the Danish Environmental Protection Agency (EPA) under the Ministry for Environment and Food of Denmark. The MSFD monitoring of marine litter indicators was initiated in 2015 by the Danish Agency for Water and Nature Management (SVANA), which since February 2017 have been merged with the Danish EPA as part of organisational restructuring of the environmental agencies in Denmark.

The monitoring activities on marine litter indicators have also included surveys on microplastic in Denmark as part of the MSFD oriented monitoring program. This includes also surveys on microplastic particles in sediments (Strand and Tairova, 2016), (Strand and Lundsteen, 2017), in press) as well as in fish (Enders *et al.*, 2017b) from the Danish parts of the North Sea, Inner Danish waters and the Baltic Sea and plastic particles in fulmars from the North Sea. The sampling of the sediments has been coordinated with the national monitoring activities targeting hazardous

substances. These microplastic surveys are scheduled to be repeated with comparable surveys in the period 2017–2021 under the marine part of the Nationwide Monitoring and Assessment Programme for the Aquatic and Terrestrial Environments, called NOVANA.

In addition, the Danish EPA has launched several projects to gain more knowledge on sources, occurrence and fate of microplastic in the Danish environment, where the first projects have been reported in e.g. (Lassen *et al.*, 2015) and (Vollertsen and Hansen, 2017). The Danish EPA have also funded a project that includes microplastic studies in Greenland 2016–2017 (Strand, pers. comm). The Danish EPA has also identified a need for establishment of more standardised and quality assured methods for analysing microplastic at sources and in the environment, although they are also aware of the current challenges in doing so.

3.8 Concluding guidelines and future perspective

Very little is known on the settling rates of microlitter in the marine environment. Particles with a density higher than water may sink because of this, but they may also be entangled in matter floating on the surface and therefore remaining here for prolonged periods of time. Particles with a lower density than water may float on the surface but could also be entangled in marine snow, or embedded in zooplankton fecal pellets and sink to the bottom with these matrices. We therefore recommend that both water, sediment and biota should be monitored for microlitter in selected stations.

In conclusion, the state of knowledge of methods for sampling is relatively well established with protocols outlined, but comparability between approaches is still not tested. Protocols for sample treatment, especially sediments and biota, that include chemical digestions, need to be further evaluated but some good candidates that are efficient and low influence of the plastic polymers have been found and used. Different setups for sediment extraction and use of different heavy density liquids is also an area of intense research but the two options of saturated sodium chloride (low cost, environmentally friendly, but not so high recovery) and zinc chloride (expensive, toxic, with quantitative recover) are the two most commonly used and for different purposes both may be motivated.

Analysis approaches is likely to go more and more towards objective, stringent protocols where visual inspection methods are complemented by spectroscopic

analysis for the larger microlitter, and for the smaller microlitter automated micro-spectroscopic methods that is now under development will be the methods of choice, since the small microlitter particles (<100 µm) is so diverse and visual information very limited at these sizes (Shim *et al.*, 2017).

In the conducted case studies it was identified in the urban sediments that black particles of characteristics resembling tire wear (crumbling, elastic, rubbery), and charcoal (porous, brittle), and oil/tar or asphalt particles and road markers was common and supplemented with some fragments of microplastics. In the surface seawater samples polystyrene foam particles and polyethylene films and fragments was dominating, and in the submerged water samples also fibres, films fragments of plastics was common types. The microlitter in mussels was dominated by fibres.

From these observations we recommend that monitoring programmes will be designed to capture both point sources (sampling near the source to define the particle characteristics and abundance) and to capture load from diffuse pollution and long range sources on the regional seas.

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Progress towards monitoring of microlitter in Scandinavian marine environments

Microlitter consists of minute particles of anthropogenic or processed natural material. The project brings together research groups to conduct specific case studies in gradients from near urban sources such as the traffic environment and cities to the coastal water and sediments in order to study the relative occurrence of specific sources and their environmental dispersion and distribution. The conclusion were first that in sediments from the road environment (tunnel runoff water), tire particles, asphalt and road markings could be identified, and in the urban creek sediments many black particles including elastomers, charcoal-like and oil and soot where in high abundance and decreased rapidly out in the recipient. The results emphasize the role of the cities as hotspot source functions for microlitter in the coastal environment and also where mitigating measures could be directed.

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