

A Nordic perspective on waste water treatment plants as pathways for microscopic anthropogenic particles to marine systems





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Preface

This report describes the results of a two year project called “Microlitter in sewage treatment systems – A Nordic perspective on waste water treatment plants as pathways for microscopic anthropogenic particles to marine systems” funded by the Marine Group (HAV) under the Nordic Council of Ministers in 2014–2015.

The aim of the project was to investigate the significance of effluent water from sewage treatment plants (STPs) as gateway for microliter and other microscopic anthropogenic particles (MAPs) to the marine and aquatic environment. Further, to investigate the occurrence of these particles both in the biotic and abiotic compartment of the receptor. STPs from Sweden, Finland and Iceland with different sewage treatment methods were included in the study. Different SPT treatments were chosen to investigate the importance of sewage treatments on microparticle retention in STPs. The report describes the methods used, results from the STP investigation and the amount of particles found in seawater, sediment and marine organisms in the receptor. Further, the report suggests a harmonized definition on particle shape for analyses which is beneficial for the international scientific community in order to facilitate comparison between studies.

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Summary

The occurrence of microscopic litter particles in the sea is a problem that has received considerable attention over the past decade. There are numerous possible sources to these microparticles and also numerous ways by which they may reach the marine environment. In order to take efficient measures to reduce the concentrations important sources and entrance routes have to be identified.

Effluent water from sewage treatment plants (STPs) is one entrance route for microlitter to the sea and other aquatic environments. The purpose of the present study has been to quantify the amount of litter particles being discharged into the sea this way and also to investigate whether elevated microlitter concentrations could be detected in water, sediment and biota in the STP recipient areas. The study was limited to particles $\geq 300 \mu\text{m}$ in water and particles $\geq 100 \mu\text{m}$ in biota and sediment.

The microlitter content was analysed in influent and effluent water at two STPs in each of Sweden, Finland and Iceland. Analyses of microlitter concentrations in water, sediment and biota were done in the recipient to one of the STPs in each country. Two major groups of microlitter were registered; microplastics and anthropogenic non-synthetic fibers (e.g. cotton).

The study showed that in the Swedish and Finnish STPs more than 99.7% of the microlitter particles $\geq 300 \mu\text{m}$ in the influent waste water were retained and were hence not discharged with the effluent water. Both these STPs were equipped with chemical and biological treatment of the waste water. The Icelandic plants, on the other hand, only had mechanical waste water treatment and no or only a limited number of microlitter particles were retained. The concentration of microplastics was between 10 and 40 particles per m^3 in treated effluent water from the Swedish and Finnish plants, and $\sim 1,500$ particles in effluent water from the Icelandic. The concentrations of non-synthetic fibers were in roughly the same range. The size of the STPs included in the study differed considerably and, as a consequence, so did the flow rate of the waste water. So in spite of similar microlitter *concentrations* in effluent waste water from all the Finnish and Swedish plants there was a larger total number of microlitter discharged from the larger STPs than from the smaller ones.

The plume of waste water coming from the discharge tube was fairly easy to detect in the Swedish and Finnish STP recipients. In both these recipients the microlitter concentrations in the plumes were found to be significantly higher than in water at the reference sites. The Swedish STP recipient water was localized in a river mouth in a heavily urbanized and industrialized area but the microlitter concentration in the waste water plume was still distinctly elevated compared to the water unaffected by the waste water.

Microlitter concentrations in the recipient to the Swedish STP were found to be considerably lower when sampling during a period with no precipitation compared to sampling during a heavy rainfall, 1.9 microplastics and 1.5 non-synthetic fibres per m³ compared to 10.5 microplastics per m³ and non-synthetic fibers too numerous to be counted. In the Finnish recipient the concentrations were on average 12.7 microplastics and 11.3 non-synthetic fibres per m³ on the first sampling occasion and 0.7 microplastic and 6.7 non-synthetic fibres per m³ on the second. The difference could not be explained by any obvious climatological factors since both samplings were carried out during periods of dry weather. The microplastic concentrations in the Icelandic STP recipient water were slightly elevated compared to the reference site, and the difference was larger for the non-synthetic fibres than for the microplastics. Concentrations of microplastics varied between 2 and 5 litter particles per m³ at both sites. The small difference between the STP recipient and the reference site was probably at least partly due to the fact that the discharge point was localized in an area with an open coastline and a very good water circulation.

Microplastics and anthropogenic non-synthetic fibres were detected in biota and sediment from the waste water recipient areas in all countries but it was not possible to trace them with any certainty to the waste water effluents.

The study shows that STP effluents are entrance routes for microplastics and other microlitter particles to the aquatic environment. If the plants are equipped with chemical and biological treatment most of the litter particles in influent waste water will be retained in the sewage sludge. This reduces the impact on the recipient water, but if the sludge for example is to be used as an agricultural fertilizer the microlitter will still be spread to the environment. Efforts to reduce the microlitter concentrations in waste water should therefore preferably be done in households and other locations where the waste water is originally being formed.

1. Introduction

The aim of the present study was to investigate whether sewage treatment plants (STPs) are important entry routes for small litter particles, so called microlitter, to the marine environment. Whether or not this is the case depends of course on the abundance of litter particles reaching the STPs, which in turn is the result of all the activities that create the waste water. But the amount of litter particles that are actually being discharged from the STP into the recipient water also heavily depends on how the waste water is treated in the plant. In this study microlitter is defined as particles <5 mm which is the definition most frequently used.

STPs are primarily designed to reduce the amount of organic matter and the nutrients nitrogen and phosphorous in waste water. The retention of particles like microlitter in the STPs is hence unintentional, but a positive side-effect of the waste water treatment.

There are within the Nordic countries large variations both in the origin of waste water being treated in STPs and in the techniques applied. In the present report, which is based on collaboration between scientist in Sweden, Finland and Iceland, the implications of these differences on the release of microlitter to the environment via STP effluents will be investigated. Two STPs were selected from each country, one representing the highest standard in the country regarding techniques for waste water treatment, and the other a representative for an "ordinary" level. The level of technology of the plants were found to vary significantly. Sweden and Finland, whose geographical positions are in densely populated areas and in enclosed sea areas, have invested in advanced treatment techniques where mechanical, chemical and biological treatment is standard. Iceland with its low density of people and the open Atlantic sea surrounding the country has at present only mechanical treatment of waste water, even at the largest STP serving the residents of Reykjavik. This large variation in waste water treatment opened up the opportunity to also study how the use of different techniques affects the content of microlitter in effluent water.

During the project period Iceland hosted a workshop from which one important outcome was a proposal to a classification system for microlitter. The system is based on, in a descending order of importance, material composition, shape and colour. The classification scheme is presented below in Figure 2 (section 6).

2. Background

Litter is not only all those visible objects that are found most everywhere in both urban and rural areas, but it also includes the minute non-visible, microlitter. Microlitter particles may be composed of fragments of larger litter objects or they may be particles that were intentionally produced in a microscopic size. What materials are included in the term *litter* may be debated, but the litter in the marine environment has been defined by the group of experts that work on litter in relation to the Marine Strategy Framework Directive (2008/56/EC): “Marine litter is any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment. Marine litter consists of items that have been made or used by people and deliberately discarded or unintentionally lost into the sea and on beaches including such materials transported into the marine environment from land by rivers, draining or sewage systems or winds. For example, marine litter consists of plastics, wood, metals, glass, rubber, clothing, paper etc. This definition does not include semi-solid remains of for example mineral and vegetable oils, paraffin and chemicals that sometime litter sea and shores” (Galgani *et al.*, 2010). Many plastic polymers are extremely resistant to degradation, and once released into the environment they might remain for decades or even centuries.

International concern over contamination of the marine environment by marine microlitter, especially plastic particles has grown very rapidly over the past ten years which is demonstrated by the large number of reports and scientific articles (GESAMP, 2015). Whereas the harmful effects of meso- and macrolitter items are well documented, e.g. marine biota being entangled or suffocated after swallowing litter items, there is not much information on the effects of microlitter to marine organisms. Micro-scale plastic particles may pose a threat to marine ecosystems both because of their content of various toxic additives like plasticisers and flame retardants, but also due to their hydrophobic nature, which allow them to concentrate chemical pollutants from the surrounding water. The particles may be consumed by marine organisms, which turns them into vectors for pollutants to the marine food webs. There are reports demonstrating direct health effects of microplastics for the consuming animals (von Moos *et al.*,

2012; Wright *et al.*, 2013; Kaposi *et al.*, 2014) indirect effects from transferred chemical pollutants (Rochman *et al.*, 2013a; Rochman *et al.*, 2013b; Besseling *et al.*, 2014a; Besseling *et al.*, 2014b) and synergistic interactions where the combined effect of microplastic and pollutants is greater than either individually (Oliveira *et al.*, 2013).

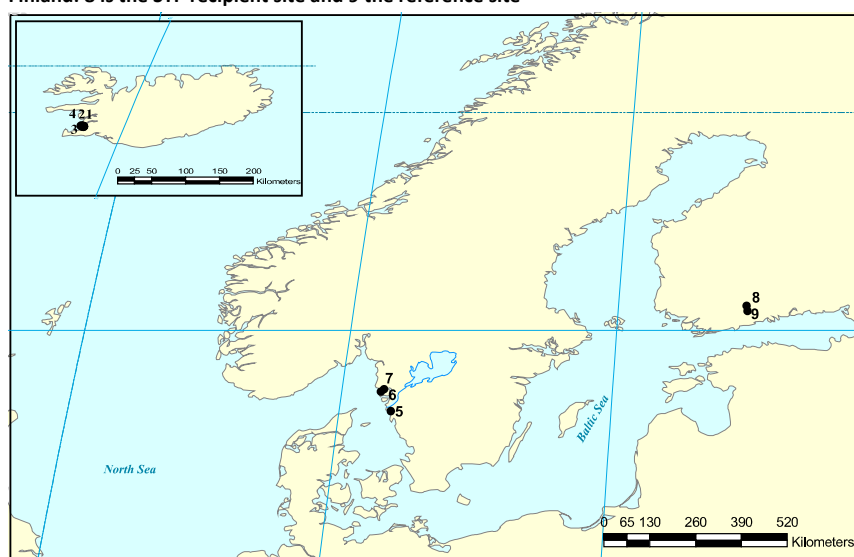
There is today a fairly good understanding of possible sources to marine microlitter. Over the past years there have been several reports on the sources and pathways for marine microlitter, and often with a specific focus on microplastics (Sundt *et al.*, 2014; Essel *et al.*, 2015; Lassen *et al.*, 2015). However, there is for the time being very limited information on the magnitude of the different sources and entrance routes. Waste water of different origin, e.g. households, industrial activities, sometimes also storm water contains microlitter particles. Microlitter has also been detected in waste water effluents from Swedish and Norwegian STPs (Magnusson, 2014; Magnusson and Norén, 2014; Magnusson and Wahlberg, 2014; Talvitie *et al.*, 2015). This shows that STPs are an entrance for microlitter to the aquatic environment (McCormick *et al.*, 2014; Yonkos *et al.*, 2014). Still, not much is known on how important this contribution is compared to other sources and input ways.

One of the key issues in the present study was to investigate the quantity and characteristics of microlitter in STP effluents and in the STP recipients. By including STPs using different techniques for waste water treatment it was also possible to see what effect this had on the amount of microlitter being discharged to the recipient waters.

3. STPs included in the study

The quantity and character of microlitter was investigated in influent and effluent waste water from six STPs in three countries, Sweden, Finland and Iceland. One of the two STPs in each country was selected for further studies of microlitter in the surface water, sediment and biota from the recipient area around the effluent discharge site. The geographical position of all STP recipient and reference sites are shown in Figure 1.

Figure 1: Sampling sites in STP recipient waters. Iceland: 1-3 are STP recipient sites and 4 the reference site. Sweden: 5 is the STP recipient site, 6 reference site 1 and 7 reference site 2. Finland: 8 is the STP recipient site and 9 the reference site



The STPs were selected to represent both a state of the art and an ordinary plant in terms of treatment techniques in each country. All STPs either had the sea or a water body in close contact with the sea as a recipient for the effluent water. The country, name, status of treatment, size and flow rate of waste water of each STP is listed in Table 1. Status of treatment is expressed as either a high or an average level for each country. The STP size is expressed as the population equivalents (PEs) of the plants.

Table 1: Presentation of the sewage treatment plants (STPs) included in the study

| STP | Person equivalent | Status of treatment within the country | Additional treatment step | Flow rates of waste water (m ³ per hour) |
|---|-------------------|--|-----------------------------|---|
| Mechanical, chemical and biological treatment of waste water | | | | |
| Ryaverket Sweden | 740,000 | Highest national standard | Disc filter 15 µm mesh size | 12,900–15,400 |
| Långevik Sweden | 14,000 | Average national standard | / | 340–440 |
| Viikinmäki Finland | 800,000 | Highest national standard | / | 10,500–17,500 |
| Kalteva Finland | 40,500 | Average national standard | / | 190–510 |
| Only mechanical treatment of waste water | | | | |
| Klettagarðar Iceland | 97,000 | Highest national standard | / | ~4,600 |
| Hafnarfjörður Iceland | 26,000 | Average national standard | / | ~1,100 |

Note: Values for flow rates of the waste water are the actual rates at the time for the sampling occasions (1) and (2) for the Swedish and Finnish STPs, whereas they are a mean values over several months for the Icelandic STPs.

3.1 Swedish STPs and STP recipient water

The Swedish STPs included in the study were Ryaverket in Gothenburg, and Långevik in Lysekil, both situated at the west coast of Sweden. The plants have mechanical, chemical and biological treatment of waste water, and in addition Ryaverket has a final step with a 15 µm disc filter. Ryaverket is the largest STP in Sweden supporting ~700,000 inhabitants as well as several industries and other activities in the Gothenburg area. The treatment performed here represents state of the art according to Swedish standards. Wastewater reaches Ryaverket through a 130 km long tunnel system and the treated water is disposed into the river Göta älv, close to its outlet into the sea. The treatment used in Långevik is less advanced and is representative for a majority of Swedish STPs. Långevik receives water from ~10,000 inhabitants and also from industries and other activities in the area around Lysekil.

Incoming untreated waste water to both Ryaverket and Långevik passes first through a series of grids where the smallest size is 2 mm. Phosphorous is eliminated by precipitation with FeSO₄ at Ryaverket and with AlPO₄ at Långevik. Nitrogen is removed in a series of steps in activated sludge tanks and with bacterial films on corrugated plastic sheets and on suspended plastic carriers. In addition Ryaverket has as a final

treatment step before the water is released into the recipient which consists of a disc filter with a mesh size of 15 μm .

The recipient of Ryaverket was selected for further investigations on waste water derived microlitter in the field. The effluent tube from Ryaverket opens in a large river, Göta älv, just before it enters the ocean. The water depth in the area of the tube is 5–10 m and there is no strong stratification of the water. This is also the harbour area of Gothenburg, the second largest city in Sweden, and beside the general urban life there are also many industries, an oil refinery and extensive shipping activities, all of which are likely sources for litter in the river.

3.2 Finnish STPs and STP recipient water

The two Finnish STPs selected for the study, Viikinmäki and Kalteva, are situated in the southern part of Finland. Viikinmäki is the largest STP in Scandinavia, treating water from 800,000 inhabitants in Helsinki and nearby cities, as well as all industrial waste waters from the area. In Viikinmäki, on average 100 million cubic meters of waste water is purified yearly. The wastewater treatment in Viikinmäki consists of 7 mm bar screening, grit removal, pre-aeration, primary sedimentation, activated sludge treatment, secondary sedimentation and a tertiary biological filtration. According to the 2012 purification results of Viikinmäki $\geq 95\%$ of BOD₇, 98% of suspended solid, $\geq 95\%$ of total phosphorus and 90% of total nitrogen are removed from the wastewater. The average water flow in Viikinmäki is 270,000 m³ per day. The treated waste water is discharged 8 km away from Helsinki at 20 m depth close to Katajaluoto sea area. Depth at the discharge site is 25 meters.

The treatment plant Kalteva in the city of Hyvinkää is responsible for purifying municipal waste waters for approximately 40,000 inhabitants living in the area. The waste water in Kalteva goes through mechanic, biological and chemical treatment. The purification percentage of suspended solids in Kalteva was 98–99% in year 2014 (<http://www.hyvinkaa.fi/hyvinkaan-vesi/jatevesien-puhdistaminen/>). After the purification process the water is discharged to the river Vantaa at a site approximately 10 meters wide and with a water depth of 2–5 meters.

Studies in the Finnish recipient waters were carried out at the recipient area right next to the discharge tube of the sewage treatment plant Kalteva. Samples were taken from the riverbank. Reference samples (water and sediment) were taken upstream from Käräjäkoski sampling site, which is used as an official reference site for water quality studies of the

river. Fish samples were taken approximately one kilometer downstream from the discharge pipe in Kalteva.

3.3 Icelandic STPs and STP recipient water

The two STPs studied in Iceland were Klettagarður in Reykjavík, and the plant in Hafnafjörður. In common with all but one STP in Iceland Klettagarður and Hafnafjörður use only the first stage treatment in settlement pools followed by a 3 mm filter before discharge.

Klettagarður was built in 2002 to serve the greater part of Reykjavík. It is the largest STP in Iceland, servicing ~160,000 inhabitants and some of the main industrial areas of Reykjavík. Sewage and drainage water are combined in the incoming waste. Waste is discharged into the sea at 5,500 m from the shore and at ~30 m depth. The last approx. 1,000 meters of the pipe has been drilled with 7 cm holes to increase mixing and dilution of the sewage. No daily or annual average volume data is available. However, a flow of ~1,500 L/s is considered typical for dry conditions, but the incoming flow may exceed the double of this in wet weather.

Hafnafjörður STP was built in 2009 and serves the town of Hafnafjörður and its surrounding industry, which includes the Alcan aluminium plant and the Promens plastic goods manufacturing plant. Hafnafjörður STP services a population of ~26,000 and all sewage and drainage water are combined.

The recipient water of the STP in Reykjavik was selected for studies of microlitter in water, sediment and biota. The effluent pipe leads out to Faxaflói bay, where it is estimated that the dilution of the sewage is considerable. Sampling of water and sediment was done at the middle part of the drilled section of the effluent tube and at the end of the tube. Reference site was chosen further out in the Faxaflói bay where limited impact was expected.

4. Sampling of microlitter

4.1 Sampling of microlitter in STP waste water

Samples of influent and effluent water to and from the STPs were sucked through filters with a 300 µm mesh size. The filters were analysed using stereomicroscopes (50 X magnification).

Sampling was carried out on influent and effluent waste water on two occasions at each of the selected STPs. In Finnish and Swedish STPs the organic content in the influent, untreated, water was much higher than in the effluent water. The organic matter clogged the filters and resulted in very different sampling volumes of influent compared to effluent water (Table 2). The organic content in effluent water from Icelandic STPs was however not very different from influent water.

Table 2: Number, volumes and methods applied when collecting samples for analyses of microlitter in *influent and effluent water from sewage treatment plants (STPs)* in Sweden, Finland and Iceland. Sampling was done at two plants in each country

| Influent water | | | | Effluent water | |
|----------------|--------------------|--------------------|--|--------------------|--|
| STP | Sampling occasions | Sampled Volume (L) | Sampling method | Sampled volume (L) | Sampling method |
| Sweden | | | | | |
| Ryaverket | 2 | 1.5–4 | Sampling in STP, filtering in laboratory | 1,000 | Pumping with submerged filter |
| Långevik | 2 | 1–2.7 | Sampling in STP, filtering in laboratory | 600–1,000 | Pumping with submerged filter |
| Finland | | | | | |
| Viikinmäki | 2 | 0.1 | Filtering in STP | 1,000 | Pumping of water through filter at STP |
| Kalteva | 2 | 0.1 | Filtering in STP | 330 | Pumping of water through filter at STP |
| Iceland | | | | | |
| Klettagarðar | 1 | 0.7–1.9 | Pumping of water through filter at STP | 0.75–1.5 | Pumping of water through filter at STP |
| Hafnarfjörður | 2 | 2.5 | Pumping of water through filter at STP | 0.25–31.2 | Pumping of water through filter at STP |

4.2 Sampling of microlitter in the STP recipients

Sampling from the recipient water around the discharge points for the waste water from STPs was carried out in 2015, one year after analyses of microlitter in the STPs. In each country only one of the two plants from the first study was selected for the recipient study: Ryaverket in Sweden, Kalteva in Finland and Klettagarðar in Iceland. The geography and the degree of influence from other polluting sources besides the STP differed considerably between the selected STP recipients in the three countries. As a consequence different methods for sampling of water, sediment and biota had to be applied.

The treated waste water from Ryaverket is discharged into the large river Göta älv just before it opens into the sea. The effluent tube is in the harbour of Gothenburg, the second largest city of Sweden and the largest port in Scandinavia, so the water is heavily influenced by other urban and industrial activities. The river reaches the sea and the Gothenburg archipelago only a few nautical miles downstream the waste water discharge point.

After the treatment process in the STP Kalteva effluent water is discharged to the river Vantaa, which is a 100 km long river in the southern Finland, starting from Hausjärvi and discharging into the Gulf of Finland in Helsinki at the bay Vanhankaupunginlahti. Altogether 1.1 million people live in contact with the river Vantaa that has a drainage area of 1,685 km². This is mainly a rural river described by the nature of its catchment area, but it is also influenced by point source pollutants, of which the most important, besides five STPs, is a mill in the upstream area in Riihimäki. In 2014 river Vantaa transferred 41 metric tonnes of phosphorus, 845 metric tonnes of nitrogen and 20 million kg of suspended solid material to the Gulf of Finland.

From Klettagarðar the effluent waste water is led through a 5,500 m long pipe into the Faxaflói bay which is open to the North Atlantic Ocean. The last approx. km of the pipe has been drilled with 7 cm holes so that the actual discharge will be spread out over a larger area. The water depth around the end of the pipe is ~35 m.

All sampling in the recipients were carried out in the area around the STP discharge tubes and in the plume of treated waste water leaving the tube. Water was sampled on two separate occasions and biota and sediment only once. A summary of the sampling data is presented in Table 3.

Since the recipient to the Swedish STP is heavily influenced by other sources for microlitter, reference water was sampled from two sites: reference 1 at a distance upstream and downstream the effluent

tube and reference 2 at a clean reference site (see 4.2.1 for further explanation).

Table 3: Locations, methods and choice of species for sampling of microlitter in water, sediment and biota in the recipient waters to treated waste water from sewage treatment plants (STPs). Data is also given for the sampling at reference locations, away from any known point sources

| Location | Water sampling/ mesh size | Sediment sampling | Biota sampling |
|---|------------------------------|--------------------------|---|
| Sweden | | | |
| Recipient water for the STP Ryaverket, in the effluent plume | Manta trawl/ 333 µm | Dredging | Dredging; blue mussels (n=17) |
| Recipient water for Ryaverket, outside the effluent plume Reference site 1 | Manta trawl/ 333 µm | Not collected | Not collected |
| Gullmarfjord Reference site 2 | Manta trawl/ 333 µm | Sediment grab | Collection by hand on rocks; blue mussels (n=15) |
| Finland | | | |
| Recipient water for the STP Kalteva | Pump with filter/ 300 µm | Portable tube sampler | Electronic fishing; bullhead (n=4), gudgeon (n=10), roach (n=4) |
| Kärjäkoski Reference site | Pump with filter/ 300 µm | Portable tube sampler | No biota collected |
| Iceland | | | |
| Recipient water for the STP Klettagarðar. Three sites round the effluent pipe end | Plankton net/ 100 µm | Sediment grab | Fishing with spinners, cod (n=1), haddock (n=5), plaice (n=2) |
| Reference site Iceland | Plankton net/ 100 µm | No sediment available | Fishing with spinners, cod (n=3), haddock (n=1) |

4.2.1 Sampling of water

Sweden: In Sweden sampling of water in the recipient of Ryaverket was done in the surface water with a Manta trawl (mesh size 333 µm). The water depth in the area around and downstream the mouth of the tube is only a few meters, so it could be expected that the plume of treated waste water will quickly reach the surface. The plume was also partly visible as an area of water with a different surface structure than surrounding water. The trawl was towed at a speed of 4 knots for 30 minutes back and forth in the plume of the water leaving the STP discharge tube. The river receives litter from a range of sources like storm water from urban life and intense industrial and shipping activities. To be able to distinguish between microlitter deriving from the treated waste water and microlitter from these other sources, additional manta trawl samples were taken at a clear distance from the outlet of the effluent tube, but still in the area that was affected by other sources (Table 3). Manta trawl samples were

also taken in the Gullmar Fjord, 100 km north of Gothenburg, which was considered as a true reference site, with no apparent point source for microlitter.

The first sampling occasion in the recipient water for Ryaverket was carried out in clear weather and after several days with no rain. Two manta trawls were done in the plume outside the STP effluent tube, and two nautical miles downstream the outlet point. On the second sampling occasion there were a series of heavy showers of rain. Again, two manta trawl samples were collected in the plume of treated waste water, and two additional trawls were made upstream the effluent point. The content in the trawls was rinsed down on 300 μm filters which were transferred to clean petri dishes and brought to the laboratory for analyses.

Finland: Sampling was carried out from the riverbank at the recipient area right next to the discharge tube of the Kalteva STP. The sampled water was pumped with a gasoline driven pump (Honda WX15) through a 300 μm filter fitted to a filter holder that was specifically constructed for field sampling of microplastics. A flow meter (Gardena water smart) was fitted to the device to measure sample volume (three replicate 500 L samples). Filters were placed in pre-cleaned petri dishes immediately after filtering, sealed and brought to laboratory for microscopy.

Iceland: In Iceland sampling was done in the recipient water for the discharge of waste water from the Reykjavik STP. Vertical sampling of the water column was performed with plankton net (radius 0.15 m; mesh size 100 μm) with a small collection tube with a filter at the bottom end. The plankton net was pulled slowly from the bottom to the surface. The collected material on net and filter was carefully washed into a plastic container. Sometimes the plankton net was partially clogged with organic material. 1–4 samples were taken from each sampling location. Even though the mesh size of the sampling equipment was smaller than the one used in the other two participating countries, a relative comparison between impact site and reference site is valid as the same sampling equipment was used in these two sites in Iceland.

Sampling of water was carried out on two occasions in 2015 in all three countries; in Sweden in August and September, in Finland in July and September and in Iceland in June and September.

4.2.2 Sampling of sediments

Sweden: Sediment samples from the recipient for discharges from Rya-verket in Sweden were collected from the area downstream the effluent tube. Efforts were made to find an area with an accumulation bottom where sediment and possibly also microlitter was likely to settle. However, no place was found where the sediment layer was thick enough to allow the use of a sediment grab. Instead sampling had to be done by dredging, which was not ideal since there was a risk that fibres from the ropes in the dredge would contaminate the samples. Two replicates were collected in the STP recipient and two replicates at a reference site.

Finland: In Finland samples were taken with a sediment sampling device from the riverbank in the area of the effluent tube, and at the reference site. The sampler is a small unique portable device with a removable Plexiglas sampling tube (496 mm x 44 mm). Sediment samples were placed into pre-cleaned white 2 L plastic containers that were immediately closed after sampling. Three replicates were collected in the STP recipient and two replicates at a reference site.

Iceland: Sediment samples were taken with a bottom grab sampler, Petite Ponar (volume of 2.4 L, sample area 152 mm x 152 mm). Two grabs were taken at the middle and at the mouth of the effluent tube and mixed into one sample for each location. No sediment was found at reference site.

4.2.3 Sampling of biota

Biota for analyses of microlitter content was collected in the STP recipients. Due to different conditions at each recipient, it was not possible to collect the same species at all three sites.

In Sweden blue mussels (*Mytilus edulis*) were selected. Animals were collected from macroalgae and rocks and stones on the bottom downstream the effluent tube and at the clean reference site. The mussels were kept in glass jars and brought to the laboratory. Fifteen animals were sampled from the STP recipient and fifteen from a reference site without any known point sources at the mouth of the Gullmar Fjord.

In the recipient area for waste water effluents from the Finnish STP in the river Vantaa three species of fish were caught: bullhead (*Cottus gobio*), gudgeon (*Gobio gobio*) and roach (*Rutilus rutilus*) (Table 4). All three are common species in southern Finland. This is especially true for bullhead and gudgeon, which are species inhabiting rivers and streams and feeding

on benthic organisms like insect larvae and small molluscs. Roach is common in brackish waters, lakes and streams, and feeds on different food items both from benthic and planktonic habitats. The preference/ability to feed on the sediment surface/benthos was obvious for all species included in this study which was proved by the fact that most individuals had ingested sand particles as well as bivalve shells in their intestines. Sampling was done downstream the discharge site by using electronic fishing. This is a better method when catching fish for studying the stomach and intestine contents compared to e.g. traps or nets. The catch resulted in four individuals of bullhead, ten of gudgeon and four individuals of roach. The fishes were brought to the laboratory in a cooling box and were then frozen. Fish samples were taken approximately one kilometer downstream from the discharge pipe.

In Iceland fish was caught with a rod and spinners in the area of the effluent tube and at a reference site. Angle was sunk to bottom and twitched until a fish was on the hook. The main species caught in the waste water recipient area were cod (*Gadus morhua*) and haddock (*Melanogrammus aeglefinus*), and in addition two individuals of plaice (*Pleuronectes platessa*). The cod was dissected directly and stomach and intestines removed, transferred to a plastic jar and frozen. The plaice was transferred to the laboratory and frozen. It was thawed at later stage for dissection where stomach and intestines were removed and transferred to a plastic jar.

5. Pre-treatment of samples

If there is a high amount of particulate matter in the water column when sampling, e.g. heavy algae blooms, leaves, insects etc. it may be difficult to detect the microlitter particles. Various methods may be used to eliminate the organic material by digestion or separate the litter particles from mineral particles by density separation. In doing this it is important to verify that the litter particles are not affected by the chemicals applied in the treatments.

5.1 Water samples from STPs and from STP recipients

Analysis of the material collected from the STP waste water was always done without any pre-treatment of the filters.

Most of the manta trawl samples from the Swedish STP recipient and the sample from the reference site could be analysed without any pre-treatment. The only exception was the samples from the recipient waste water plume on the second sampling occasion. These samples contained so much organic material that it had to be eliminated by enzymatic digestion before analyses. Most of the material seemed to consist of cellulose from toilet paper. This should be regarded as litter according to the definition (Figure 2), but in this study we limited the analyses of non-synthetic litter to include only textile fibres. The recipient filters with large quantities of material were treated according to a method developed by Mintenig (2014) with some modifications. 1 ml of the detergent sodium dodecyl sulfate (SDS) was added to the manta trawl filters in the petri dishes, which were then left in 70 °C overnight. Enzymes were added in the form of laundry detergents from Spinnrad (Bad Segeberg, Germany), Biozyme F (lipase) and SE (protease and amylase), 1 ml of each detergent, to remove unwanted organic material.

The water samples from the Finnish STP recipient could be analyzed without previous treatment.

The filtered water samples from Iceland were re-filtered in the laboratory with suction prior to analyses.

5.2 Sediment from recipients

Sediment samples from the Finnish STP recipient were treated with oversaturated NaCl with some modifications after Stolte (2015). In this method 35.7 g NaCl/100 mL was first mixed to the sediment, and after 20 min incubation in room temperature an oversaturated NaCl solution was added. The sediment slurry was let to settle and the supernatant carefully sucked through a 100 µm filter. This procedure was repeated three times to extract all plastic fragments and fibers from the sediment.

Sediment samples from the Swedish STP recipient were sieved through a mesh size of 5 mm then treated with H₂O₂ (30%) and left for 24 hours. Density separation was done with ZnCl₂ (density 1.6 g cm⁻³). The density separation procedure was similar to how it was described for the Finnish samples (see above). Samples were taken to determine the dry weight of the sediments. Special care was given to ensure that no particles that could be related to the sampling device were calculated as microlitter.

Sediment samples from Iceland were treated with saturated NaCl solution. Sediment was submerged with excess of saturated NaCl solution at room temperature in a 2 L Erlenmeyer flask and let to settle for 2 hours. Water solution was decanted to a beaker and subsequently filtered through a 300 µm filter with suction.

5.3 Biota from recipients

Blue mussels from the Swedish STP recipient water and reference site were completely digested before analyses of the microlitter content. It was hence not possible to say where in the animals the observed particles had been. The tissue of the blue mussels collected in the recipient to Ryaverket in Sweden and at the reference site was digested in the same way as the manta trawl filters with the water samples (section 5.3). The method is a modified version of that described in Mintenig (2014). The mussel tissue was carefully removed from the shells with a clean scalpel. The shell length and animal wet weight was determined and the animals were then individually wrapped in aluminum foil and put in the freezer (18 °C). The dissecting work was carried out in a ventilation fume to minimize the risk for contamination from the air. The following day the mussel were thawed and moved to individual petri dishes. The tissues were cut into smaller pieces and 0.5 g of the detergent SDS along with some ml of water was added to each petri dish and mussel. The samples were then kept at 70 °C overnight. Biozyme F and Biozyme SE were added to the

mussels, 0.5 ml of each detergent to each animal. The mussels were kept in 40 °C for two days. This procedure led to a total digestion of the mussel tissue, with exception of the foot, and it was possible to examine the presence of microlitter particles with the stereomicroscope.

A modified method of Foekema *et al.* (2013) was applied to the fish samples from the Finnish STP recipient. The frozen fish were melted at room temperature, their stomach and intestines dissected and placed to pre-cleaned glass bottles or test tubes (2% HCl, rinsed 3x with Milli-Q water). A solution of 10% KOH was added to the bottles/tubes (three times the volume of the tissue material), and the fish tissues were let to digest at 60 °C° overnight. The digested tissue was poured through 300 µm filters which were placed into pre-cleaned petri dishes, and studied under a stereo microscope. Three methodological blanks were prepared by filtering Milli-Q water under similar conditions.

The gastrointestinal tract of cod, haddock and plaice from Iceland were digested with 10% KOH solution at 60 C° for 8 hours and filtered through 300 µm filters with suction. Samples contained large amount of sediment and shells resulting in difficulties analysing samples. The samples were therefore treated similar to sediment samples, i.e. submerged in excess of saturated NaCl solution and swirled. Samples were allowed to settle for 2 hours and supernatant decanted to a clean beaker and subsequently re-filtered through 100 µm filter.

6. Analyses of microlitter

All samples, STP waste water, and water, sediment and biota from the STP recipients, were analysed visually under stereomicroscope with a magnification of x50.

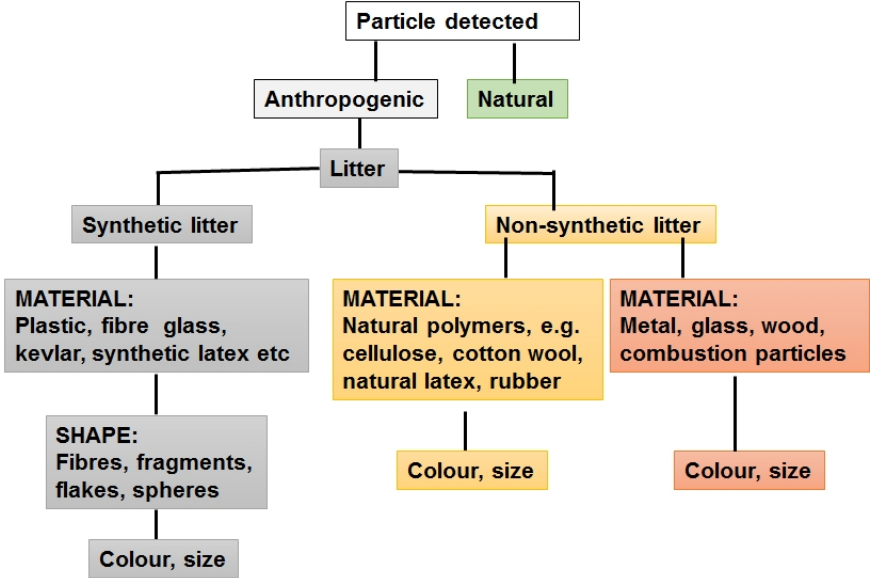
A classification scheme for the microlitter particles was constructed where particles were divided into different categories according to this material, shape and colour (Fig. 2). In the present study not all categories in the schedule were included, but we limited the analyses to the following groups:

- Microplastic particles, which were further subdivided into plastic fibres, plastic fragments and plastic flakes.
- Non-synthetic anthropogenic fibres (in this text referred to as non-synthetic fibres), e.g. textile fibres of cotton or wool.

In cases where it was difficult to distinguish between plastic and non-synthetic fibres by microscopic studies only, melting tests of individual particles were performed. The particles were transferred from the petri dish to an object glass which was held over a flame of an alcohol lamp. Plastic particles would then melt in a characteristic way that made them easy to separate from particles of other compositions.

A selection of particles from STP waste water and from the STP recipients were analysed with Fourier Transform Infrared Spectroscopy (FTIR), a microscopic technique for polymer characterization.

Figure 2: Classification scheme of microlitter as proposed of the authors to the report



7. Results on microlitter in STP waste water

7.1 Abundance of microlitter in influent and effluent water

The concentration of microplastics and non-synthetic fibres (number of particles per m³) in influent and effluent water is presented in Table 4. The concentrations were high in the influent water to the Finnish STPs, compared to incoming water to the Swedish and Icelandic plants. This might be explained by differences in the origin of the water, which in a varying degree could be e.g. residential settlements and services, industries or storm water. However, it should also be noted that all samples were snapshots of the situation and not integrated over a longer period of time, so the human activities representing the waste water at that specific time determines the sources of waste water coming into the plants.

Table 4: Microlitter particles $\geq 300 \mu\text{m}$ in influent and effluent water at STPs in Sweden, Finland, Iceland. Concentrations are presented as number of particles per m³ of influent or effluent waste water. The data are average values \pm SE (n=2 except for Klettagarðar where n=1)

| Name of STP | Sum microplastics (number particles/m ³) | | Non-synthetic fibres (number particles/m ³) | |
|----------------|--|----------------|---|-----------------|
| | Influent water | Effluent water | Influent water | Effluent water |
| Sweden | | | | |
| Ryaverket | 7,340 \pm 13 | 8 \pm 7 | 89,200 \pm 32,330 | 4 \pm 1 |
| Långevik | 12,120 \pm 6,820 | 23 \pm 1 | 66,050 \pm 36,340 | 68 \pm 33 |
| Finland | | | | |
| Viikiniemi | 100,000 \pm 43,300 | 43 \pm 36 | 250,000 \pm 23,330 | 29 \pm 26 |
| Kalteva | 91,570 \pm 28,300 | 29 \pm 10 | 193,300 \pm 146,670 | 32 \pm 28 |
| Iceland | | | | |
| Klettagarðar | 631 | 1,378 | 970 | 1,130 |
| Hafnarfjörður | 2,070 \pm 200 | 1,400 \pm 66 | 3,800 \pm 600 | 2,490 \pm 180 |

In both Icelandic STPs there were very small differences in microlitter concentrations between influent and effluent water. This means that there was no or very limited retention of litter particles in these plants. Still there were small differences, and it seems likely that these were

caused by a combination of diurnal variations in waste water litter concentration and the fact that sampling was not carried out simultaneously on influent and effluent water. The small sample volumes also add to an uncertainty in the results.

The number of litter particles leaving the STPs with effluent water can be estimated by multiplying the litter concentration with the waste water flow rate. In Table 5 is shown the number of microlitter particles reaching the water recipient per hour. The results are based on the amount of particles per m³ of waste water multiplied by the flow rate (m³ per hour).

The STPs included in the study vary considerably in size, from 14,000 population equivalent (PE) for Långevik to 800,000 PE for Viikinmäki. To the right in table 5 is shown the number of microlitter particles in effluent divided by the PE of the plants. With this “size adjusting” of the data it was revealed that Ryaverket released the lowest number of microlitter particles to the recipient followed by Kalteva and Viikinäki, thereafter came Långevik and last, with the highest number of microlitter particles per PE discharged to the recipient, the Icelandic STPs Hafnarfjörður and Klettagarðar.

Table 5: The number of microlitter particles $\geq 300 \mu\text{m}$ leaving the STPs with the effluent tube per hour expressed both as the total number of microlitter particles and as the number of particles per population equivalent (PE). Data is presented as average values of two sampling occasions except for Reykjavik where only one sampling was carried out

| Name of STP | Microlitter in STP effluent water (number particles /hour) | | Microlitter in STP effluent water adjusted to PE (number particles/hour and PE) | |
|----------------|---|----------------------|--|----------------------|
| | Microplastic particles | Non-synthetic fibres | Microplastic particles | Non-synthetic fibres |
| Sweden | | | | |
| Ryaverket | 120,100 | 54,400 | 0.16 \pm 0.14 | 0.07 \pm 0.03 |
| Långevik | 9,100 | 24,700 | 0.65 \pm 0.06 | 1.76 \pm 0.67 |
| Finland | | | | |
| Viikinmäki | 468,400 | 319,600 | 0.41 \pm 0.41 | 0.28 \pm 0.32 |
| Kalteva | 11,700 | 15,700 | 0.29 \pm 0.29 | 0.39 \pm 0.37 |
| Iceland | | | | |
| Klettagarðar | 6,348,800 | 52 224,000 | 65.2 | 53.8 |
| Hafnarfjörður | 2,232,000 | 4,104,000 | 10.9 \pm 5.7 | 65.2 \pm 45.5 |

7.2 Retention efficiency

The efficiencies by which the microlitter particles were retained in the STPs were calculated according to the formula below and are presented in Table 6.

$$\text{Retention efficiency} = ([\text{Incoming water}] - [\text{Effluent water}] / [\text{Incoming water}]) \times 100$$

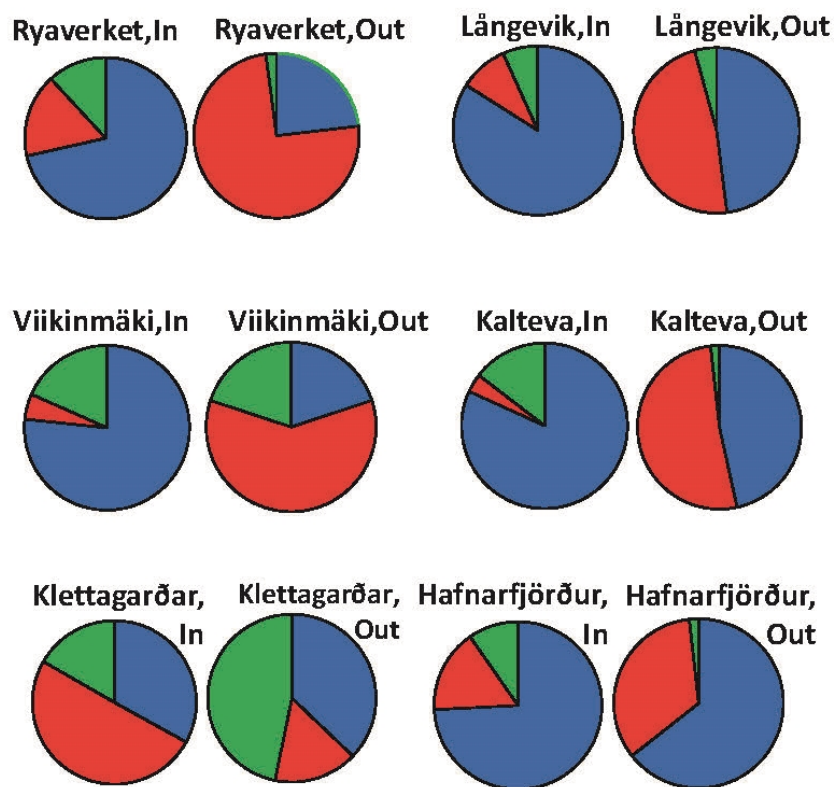
Table 6: Retention of microlitter particles $\geq 300 \mu\text{m}$ in the six STPs. Calculations are based on average values of three replicates from each sampling occasion

| Name of STP | Retention efficiency of microlitter in STPs | |
|----------------|---|----------------------|
| | Sum microplastics | Non-synthetic fibres |
| Ryaverket | 99.89 % | 99.99 % |
| Långevik | 99.71 % | 99.81 % |
| Viikinmäki | 99.93 % | 99.99 % |
| Kalteva | 99.97 % | 99.99 % |
| Klettagarðar* | (-118.23 %) | (-16.46 %) |
| Hafnarfjörður* | (50.17 %) | (57.59 %) |

Note: Most likely there was no or very limited retention in the Icelandic STPs (see section 7.1).

The microplastic particles were divided into the categories plastic fibres, plastic fragments and plastic flakes (see Figure 2). By comparing the proportion between the three categories in influent water with the proportion in effluent, it was possible to make an estimation of which of them was best retained in the STPs. A general picture is that plastic fibres (blue fields) dominated in waste water coming in to the Swedish and Finnish STPs whereas plastic fragments (red fields) were more frequent in effluent water (Fig. 3). The plastic fibres had hence been retained in the STP to a larger extent than plastic fragments. Since no retention of microplastics could be registered in the Icelandic STPs (Table 5) it could be expected that the proportion between categories of microplastics also should be the same in influent and effluent water. This was also case for the STP in Hafnarfjörður, however, in the STP in Klettagarðar the proportion of plastic fragments was lower in effluent than in influent water, the opposite from what was found in STPs in Sweden and Finland.

Figure 3: The proportion between categories of microplastic particles $\geq 300 \mu\text{m}$ in influent and effluent waste water to the six STPs. Blue indicates plastic fibres, red plastic fragments and green plastic flakes



8. Results on microlitter in STP recipient water

8.1 Microlitter in surface water

The outlet for effluent water from Ryaverket is located in the river Göta älv where it runs through Gothenburg, the second largest city in Sweden. In an attempt to separate microlitter particles from STP effluents from litter particles that had reached the river by other pathways e.g. storm water or windblown litter, trawling was done not only in the waste water plume but also at other locations away from the plume but still in the recipient area (=reference site 1 in Figure 4). A location in the Gullmar Fjord, 100 km north of Gothenburg was selected as a “clean” reference site (reference site 2). Sampling in the recipient area (in figure 4 marked as “Recipient, STP plume” and “Reference 1, outside plume”) was done at two occasions whereas sampling at reference site 2 was done only once. The first sampling occasion in the STP recipient took place during a period of dry weather and the second on a day with heavy rainfall. Sampling at the reference site took place during rainfall and should hence be compared to STP recipient data from the second sampling occasion.

The concentration of microplastics was higher in the waste water plume than outside the plume on both sampling occasions (Fig. 4), but all microlitter concentrations were higher at the second sampling. On the first occasion they were twice as high, 1.9 particles per m^3 in the waste water plume compared to 0.9 particles per m^3 outside the plume. On the second occasion the difference was larger, 10.5 microplastic particles per m^3 in the plume and 2.9 per m^3 outside the plume. Also the concentration of non-synthetic fibres was higher in the plume than outside, 1.5 compared to 0.2 fibres per m^3 . On the second sampling occasion the number of non-synthetic anthropogenic fibres (mainly textile fibres of cotton or cellulose) in samples from both the waste water plume and outside the plume were too numerous to be counted in a reasonable time.

Microplastic concentration in the Finnish STP recipient water was in the same range as in the Swedish ones at the first sampling, 12.7 microplastics per m^3 , and non-synthetic fibres were in the same range (Fig. 5). At the second sampling the microplastic concentration in the recipient

water was much lower, only 0.7 plastic particles per m³, whereas the non-synthetic fibres had a concentration of 6.7 fibres per m³ of recipient water. No microlitter particles were found at the reference station on any of the two sampling occasions.

The waste water tube from the Icelandic STP Klettagarðar had several holes drilled along its outer part and sampling was done both at the mouth of the tube and at the site of the innermost drilled holes (see section 4.2 and Fig. 1; KT2 is at the drilled part of the pipe, KT3 is at the end of the pipe and KT4 just outside the end of the pipe). On the first sampling occasion the microlitter concentrations were slightly elevated in the recipient water compared to the reference site. The concentrations were on average 5.2 microplastics and 5.1 non-synthetic fibres per m³ at the STP recipient site compared to 2.7 microplastics and 4.0 non-synthetic fibres per m³ at the reference site (Fig. 6). At the second sampling there was no difference in microplastic concentration, 2.4 microplastics per m³ in the STP recipient water and 2.6 at the reference site. The concentration of non-synthetic fibres was however higher in the STP recipient on the second sampling occasion, 26 fibres per m³, compared to 7.0 per m³ at the reference site.

Figure 4: Microlitter particles $\geq 330 \mu\text{m}$ in the waste water plume outside Ryaverket and in the same area but away from the plume (=Reference 1). Non-synthetic fibres were too numerous to be counted at sampling occasion II. Sampling was also done once at a clean reference site (=Reference 2). Data is presented as average number of litter particles/m³ of seawater \pm SE

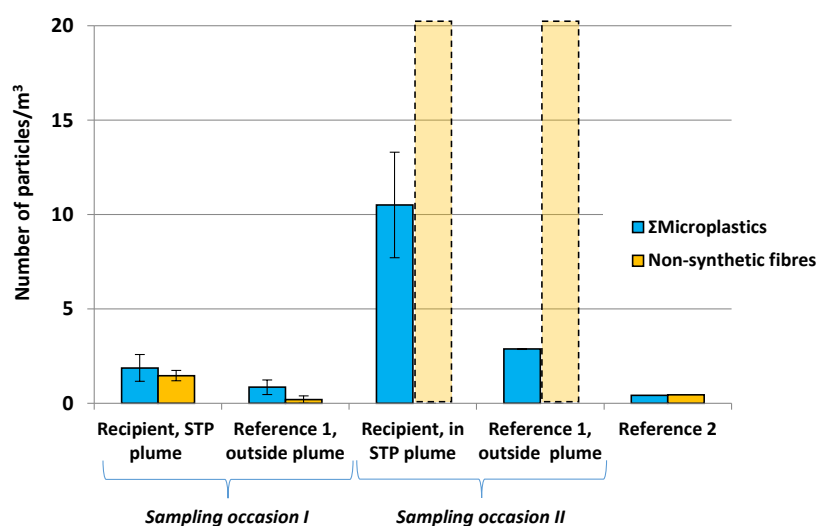


Figure 5: Microlitter particles $\geq 330 \mu\text{m}$ in the recipient for the Finnish STP Kaltava and at a reference site. Data is presented as average number of litter particles/ m^3 of sea-water \pm SE (n=3)

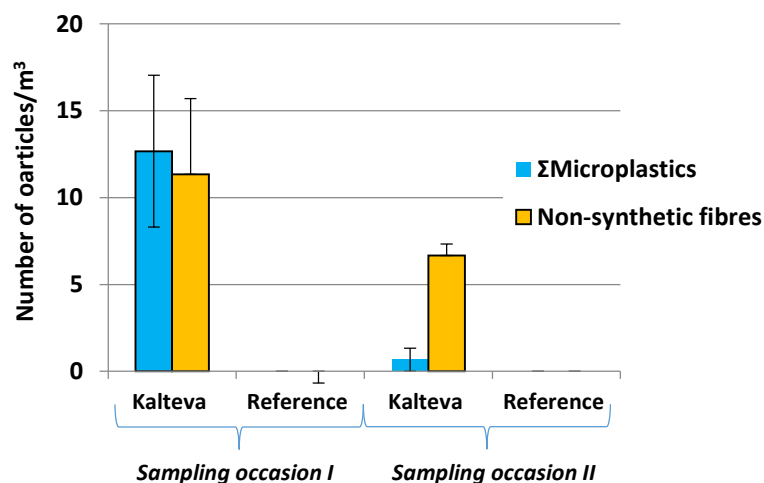
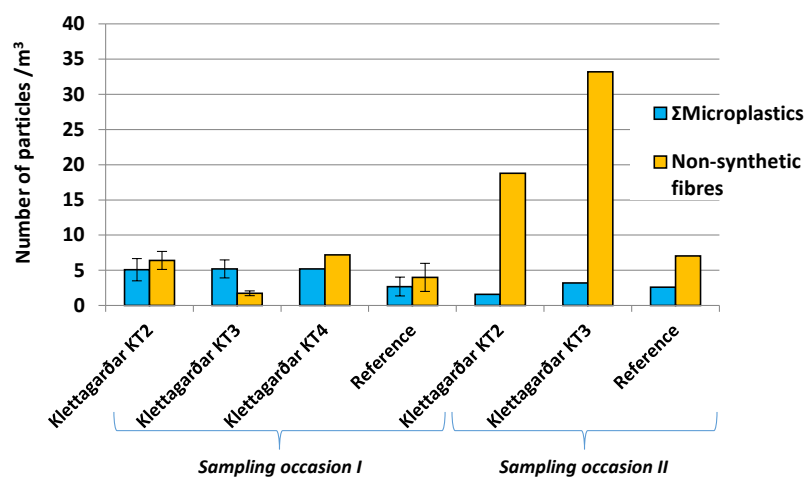


Figure 6: Microlitter particles $>100 \mu\text{m}$ in the recipient water to the Icelandic STP Kletta-garðar and a reference site. KT2 is from the (middle of the discharge tube, KT3 from the mouth of the tube), KT4 from a distance from the tube. Data is presented as average number of litter particles/ m^3 of seawater \pm SE



Note: Note the different scale compared to figures on Swedish and Finnish data.

8.2 Microlitter in sediment

The abundance of microlitter particles in sediment could be estimated only in samples from the Swedish and Icelandic STP recipient waters and from the Swedish reference site. The Finnish sediment samples were contaminated by fibres from clothing due to problems at the sampling site. During analyses it became apparent that the sampling device had been in contact with the researchers clothing and the line that was attached to the device.

Data is expressed as number of particles per dry weight of sediment. The dry weight of the sediments at the Ryaverket recipient was 38% of the wet weight; at the reference site at the Gullmar Fjord 31% of the wet weight. No dry weight/wet weight ratio was available for the Icelandic sediments, but they were made up of gravel and coarse sand so the wet weight was presumed to be similar to the dry weight. The average microplastic concentration in the sediment in the recipient water for effluents from Ryaverket Microplastic concentrations in sediments from Icelandic STP recipient water were 100 times lower than in Swedish ones, 0.005 plastic particles per g dw of sediment (Table 7). It is however difficult to compare the two sites since the Swedish sediments were fine grained and hence more likely to act as accumulation bottoms, or at least bottoms where particles could temporarily be deposited. The coarse sediment at the Icelandic site was less likely to be an area where microlitter would settle.

Table 7: Microlitter particles $\geq 100 \mu\text{m}$ in sediment at STP recipient areas and at reference sites. Data is presented as number of particles/g sediment dry weight (dw). (average values \pm SE)

| Location | Plastic fibres (no/g dw) | Plastic frag- ments (no/g dw) | Plastic flakes (no/g dw) | Sum micro- plastics (no/g dw) | Non-synthetic fibres (no/g dw) |
|---------------------------------|--|-------------------------------------|--------------------------------|-------------------------------------|--------------------------------------|
| Sweden | | | | | |
| Recipient for Ryaverket n=2 | 0.36 \pm 0.06 | 0.38.0 \pm 0.08 | 0.07 \pm 0.07 | 0.81 \pm 0.21 | Very high numbers. Not analysed |
| Gullmarfjord Reference site n=1 | 0.10 | 0.05 | 0.0 | 0.15 | 0.10 |
| Finland | Samples contaminated Samples contaminated | | | | |
| Iceland | | | | | |
| Recipient of Klettagarðar n=2 | 0.003 \pm 0.003 | 0 \pm 0 | 0.002 \pm 0 | 0.005 \pm 0.002 | 0.025 \pm 0.002 |

8.3 Microlitter in biota

Abundances of microlitter in biota may be expressed as the number of particles *per weight* or as number *per individual*. Both alternatives are practiced in the scientific literature (Cole *et al.*, 2013; Lusher *et al.*, 2013; Van Cauwenberghe *et al.*, 2015) and in the present study we have settled for the latter, *microlitter particles per individual*. Results from biota in the STP recipients in Sweden, Finland and Iceland are presented in Table 8.

Blue mussels from the recipient area for discharged waste water from Ryaverket contained more microplastics than blue mussels from the reference site in the Gullmar Fjord, on average 2.7 plastic particles per individual compared to 0.5 per individual (Table 8). The vast majority of particles were plastic fibres. In one of the mussels from the reference site was found a semi-transparent plastic fragment of a kind that was frequently found in effluents from the Swedish STPs. The number of non-synthetic fibres in blue mussels was more similar between the sites than the microplastics, 1.9 fibres per individual at the STP recipient compared to 1.3 at the reference site. The mussel tissue was completely digested prior to analyses and it was therefore not possible to say in what compartment of the living animal the litter particles had been.

For logistic reasons the mussels from the two sites were not treated identically which might affect the microlitter content. The animals from the STP recipient were transported for a longer period of time and were also subjected to a more turbulent handling than mussels from the reference site.

The biota analysed from the recipient to Kalteva in Finland were three different species of fish, bullhead (*Cottus gobio*), gudgeon (*Gobio gobio*) and roach (*Rutilus rutilus*). Of the 18 individual specimens four contained microlitter (Table 8). In total two plastic fibres, one plastic fragment and one non-synthetic fibre were found.

From the recipient to the Icelandic STP Klettagarðar, the microparticles were analyzed in the stomach and gut of three species of fish, cod (*Gadus morhua*), haddock (*Melanogrammus aeglefinus*) and plaice (*Pleuronectes platessa*) (Table 8). Plaice was only caught at the recipient site and no microparticles were found in either of the two individuals. The plaice is a bottom dwelling species and the seabed at the recipient site contained mostly gravel and stones and low amount of microparticles. This might reflect the lack of microparticles found in the plaice. Haddock and cod were caught both at the STP recipient site as well as the reference site. The haddock caught at the recipient site contained higher amounts

of both plastic fibres and plastic fragments compared to cods from the recipient site, 6.6 and 1.0 particles, respectively. The haddock is primarily a benthic feeder but it is possible that it could be feeding directly from the effluent plume. Cods from the reference site contained high amounts of plastic fragments, however the variation was large between individuals. The closeness between the recipient sites and the reference site does not imply that there should be difference between fish caught in these two sites as the results from the cod samples did not reflect what was seen in the haddock samples. There was a lower amount of plastic fibres in cod from the reference site, however, the difference was not significant or 1.0 compared to 0.7 ± 0.3 at the recipient site and reference site, respectively. It is questionable to consider the fish from the reference site feeding from different source compared to the recipient site as the sites are quite close to each other, or less than 5 km in a straight line.

Table 8: Microlitter $\geq 100 \mu\text{m}$ in biota from the STP recipient water and from reference sites. Data is presented as number of microlitter particles/individual. Figures are average values \pm SE

| | Plastic fibre (no/individual) | Plastic fragment (no/individual) | Plastic flake (no/individual) | Sum microplas- tics (no/individual) | Non-synthetic fibres (no/individual) |
|--|----------------------------------|-------------------------------------|----------------------------------|--|---|
| Sweden | | | | | |
| Blue mussels, (n=17) Re- cipient of Ryaverket | 2.5 \pm 0.6 | 0.06 \pm 0.06 | 0.1 \pm 0.1 | 2.8 \pm 0.7 | 1.9 \pm 0.48 |
| Blue mussels, (n=15) Ref- erence area | 0.4 \pm 0.2 | 0.1 \pm 0.1 | 0 | 0.5 \pm 0.2 | 1.3 \pm 0.4 |
| Finland | | | | | |
| Bullhead (n=4) Recipient of Kalteva | 0 | 0 | 0 | 0 | 0 |
| Gudgeon (n=10) Recipi- ent of Kalteva | 0.1 \pm 0.1 | 0 | 0 | 0.1 \pm 0.1 | 0 |
| Roach (n=4) Recipient of Kalteva | 0.2 \pm 0.2 | 0.2 \pm 0.2 | 0 | 0.4 \pm 0.2 | 0.2 \pm 0.2 |
| Iceland | | | | | |
| Plaice (n=2) Station KT2 Recipient of Klettagarðar | 0 | 0 | 0 | 0 | 0 |
| Haddock (n=5) Station KT3 Recipient of Klettagarðar | 1.8 \pm 0.6 | 2.2 \pm 1.0 | 4.0 \pm 1.1 | 6.6 \pm 1.5 | |
| Cod (n=5) Station KT3 Re- cipient of Klettagarðar | 1.0 | 0 | 0 | 1.0 | 1.0 |
| Cod (n=3) Reference area Iceland | 0.7 \pm 0.3 | 5.30 \pm 4.8 | 0 | 6.0 \pm 5.0 | 0 |

8.4 FTIR analyses of microparticles

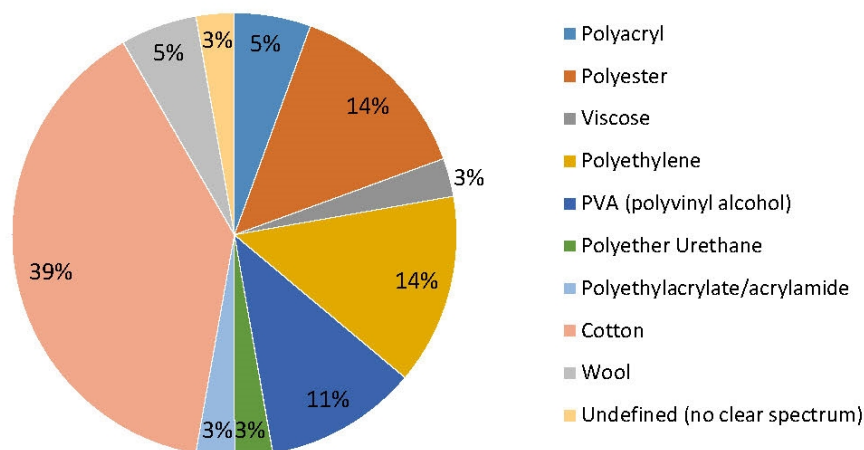
FTIR-analyses are expensive and laborious and hence only possible to do on a limited number of particles. Most particles selected for this were therefore considered to be representatives for groups of particles that were commonly found in the samples. Results from the analyses of particles from Swedish STPs are presented in Table 9. A kind of semi-transparent polyethylene particles were among the most commonly detected in STP effluents, both in this and in other studies (Magnusson and Norén, 2014; Magnusson and Wahlberg, 2014). Polypropylene was also found to be a common plastic polymer in STP effluents, both in plastic fragments and plastic fibres. All microlitter particles (microplastics and non-synthetic fibres) from the first sampling occasion at the Kalteva recipient were analysed with FTIR and the relative distribution of different polymers is presented in Figure 7.

Table 9: Results from FTIR analyses of individual microlitter particles collected in waste water in the Swedish STPs

| Particle description | Origin | Result from FTIR analyses |
|---|--|---|
| Semi-transparent fragments with irregular surface 300 x 150 x 150 µm | Effluent water Ryaverket & Långeviksverket | Polyethylene |
| Red hard flake, 300 x 300 x 50 µm | Influent water Långeviksverket, | Thermoset plastic based on aliphatic polyester resin. |
| Clear blue semi-transparent irregular fragment 100 x 50 x 50 µm | Effluent water Långeviksverket | Polypropylene |
| White, shiny, non-transparent fragment 500 x 200 x 100 µm | Effluent water Långeviksverket | Polypropylene |
| Red fibre 500 µm | Influent water Ryaverket | Polypropylene |
| Black fragment 400 x 200 x 100 µm | Effluent water Ryaverket | Polypropylene |
| Black fibre 1,000 µm | Effluent water Ryaverket | Polyethylene terephthalate (PET) |

Figure 7: Relative representation of different polymers in microlitter particles at one sampling occasion in the Kalteva recipient. Data is based on in total 34 particles

FTIR analyses from Kalteva recipient (Finland)



9. Discussion

The purpose of the present study was to investigate the importance of STPs as gateways for microlitter particles to the environment. The work was divided into two parts carried out in separate years, 2014 and 2015. The first year concentrations and composition of microlitter in waste water at six STPs in Sweden, Finland and Iceland was investigated. The second year studies were done on microlitter in the recipient water for one of the STPs from each country. The investigated particles included *microplastics* the litter group that generally is considered to be the most problematic, but also *non-synthetic anthropogenic fibres* e.g. cotton, viscose, wool, were analysed since these are commonly found in field samples.

9.1 Microlitter in STP influent and effluent water

Substantial amounts of microlitter particles were detected in incoming waste water to all the investigated STPs. The microlitter concentrations differed considerably between the countries and were highest in influent water to the two Finnish plants, followed by the Swedish ones and the lowest concentrations were found in the Icelandic STPs. The microplastic concentration in influent water was in the range of 100,000 particles $\geq 300 \mu\text{m}$ per m^3 to the Finnish STPs, around 10,000 particles per m^3 to the Swedish and roughly 1,000 microplastics per m^3 to the Icelandic STPs. Possible explanation for the difference observed between the Swedish and Finnish plants are differences in waste water origin but also the differences in sampling methods. Sampling in the Finnish STPs did not involve any suction but the samples were passing the filters by gravity only. This might improve the retention of litter onto the filters, particularly in the beginning of the sampling when filters are clean. This is supported by the fact that the sample volumes of influent water were 10–40 times higher in Sweden and Iceland where waste water was sucked through the filters compared to Finland where it was not. Without physical forcing (i.e. suction) the sample volume had to be kept relatively small, in order to prevent the filters from clogging.

The waste water treatment techniques applied were found to have a major influence on how much of the microlitter that was retained in the plants and how much that was released to the recipient with the effluent water. In the Finish and Swedish STPs, which were equipped with mechanical, chemical and biological treatment steps, >99.8%, were retained and concentrations of microplastics in the effluent water were at all occasions below 50 particles $\geq 300 \mu\text{m}$ per m^3 . The retained particles were most likely deposited in the sewage sludge. This presumption is supported by results from a previous study where a good correlation was found between the amount of microplastics lost from the influent waste water and the amount found in the sludge (Magnusson and Norén, 2014). In the Icelandic STPs, which only had mechanical waste water treatment, there seemed to be no or very low retention of microlitter since concentrations in the effluent water were in the same range as in the influent. The retention efficiency of microplastics in Swedish and Finnish plants seemed to be affected by the shape of the particles, since plastic fibres were retained to a higher extent than of plastic fragments in all the four plants.

In order to estimate the total amount of microlitter particles being discharged into the STP water recipients over time, the flow rate of the waste water had to be taken in account. When doing this it became obvious that even with the high retention efficiency observed in the Swedish and Finnish plants, the number of microlitter particles reaching the water recipient was substantial. From the smaller plants, Långevik (14,000 population equivalents, PE) and Kalteva (40,500 PE) around 10,000 microplastics $\geq 300 \mu\text{m}$ was released to the recipient water every hour, from the large STPs Viikinmäki (800,000 PE) and Ryaverket (740,000 PE) the release was 470,000 and 120,000 microplastics $\geq 300 \mu\text{m}$ per hour respectively. The number of microplastic particles leaving the Icelandic STPs, where there was no or only very limited particle retention, amounted to $\sim 6,350,000$ microplastics $\geq 300 \mu\text{m}$ per hour from Klettagarðar (97,000 PE) and $\sim 2,230,000$ microplastics per hour from Hafnarfjörður (26,000 PE). The number of non-synthetic fibres going out with the effluent water was, very roughly, in the same order of magnitude as the number of microplastics.

Important information was obtained when relating the release of microlitter via waste water effluents to the size of the STPs. The number of microlitter particles leaving the STPs was therefore divided by the population equivalent (PE) of the plants since the PE could be regarded as a proxy for the number of people connected to the plant. When analysing the PE adjusted data it was revealed that Ryaverket released the lowest

number of microlitter particles $\geq 300 \mu\text{m}$ of all investigated STPs, on average 0.16 microplastics and 0.07 non-synthetic fibres per hour and PE. This may be due to the fact that Ryaverket is equipped with an extra treatment step compared to the other Finnish and Swedish STPs. The waste water is led through a disc filter with a mesh size of $15 \mu\text{m}$ as a final treatment step before being discharged, and it could be expected that most particles larger than $15 \mu\text{m}$ would be retained in the plant. Why there still were particles as large as the ones actually detected in the effluent water may have several explanations. One is related to the size of the analysed particles. Although they are described as particles $\geq 300 \mu\text{m}$ they may not necessarily be this large in all directions, Plastic fibres, which is one important category of microplastics, may be well over $300 \mu\text{m}$ long and still have a diameter that allow them to pass through the $15 \mu\text{m}$ disc filter. There is also the possibility that there were ruptures in the filter or that not all waste water was passed through it.

The by far highest release of microlitter per hour and PE was found for the Icelandic STPs which only have mechanical waste water treatment. From Klettagarðar 65.5 microplastic particles and 53.8 non-synthetic fibres were discharged per hour and PE and from Hafnarfjörður 10.0 microplastics and 65.2 non-synthetic fibres per hour and PE.

9.2 Microlitter in the STP recipient waters

Elevated concentrations of microlitter compared to reference sites were found in the recipients to the Swedish and Finnish but not the Icelandic STP recipient waters.

The discharge point from the Swedish Ryaverket is in a large river where it runs through a heavily urbanized and industrialized area. To distinguish between STP derived microlitter and microlitter from other sources water samples were taken both in the plume of effluent waste water and in the same general area but outside the plume. This field sampling of water was done with a manta trawl and the mesh size was slightly larger than the filter size used in the STPs, $333 \mu\text{m}$ instead of $300 \mu\text{m}$. The sampling was done on two occasions, once during a period of dry weather and once during a heavy rainfall. On both occasions the microlitter concentrations were higher inside than outside the plume. It was presumed that the extra load detected in the plume derived from the waste water effluent. Water concentrations of microlitter in the recipient to Ryaverket were considerably higher during rainfall than when the weather was dry.

The rain had probably caused a higher load of litter both in the waste water by increasing the proportion of storm water coming into the STP and by increasing the general run off from land. During the rainfall a concentration of 10.5 microplastic particles $\geq 333 \mu\text{m}$ per m^3 were found in the plume, compared to 2.87 outside the plume, which might be interpreted as a contribution of ~ 8 microplastics $\geq 333 \mu\text{m}$ per m^3 from the STP effluent. During the dry weather sampling the microplastic concentration in the plume was on average 1.87 particles $\geq 333 \mu\text{m}$ per m^3 compared to 0.85 microplastics per m^3 outside the plume. This would give a contribution of ~ 1 microplastic particle $\geq 333 \mu\text{m}$ per m^3 from the waste water effluents. The concentration of non-synthetic fibres was in the same range or slightly lower than the concentration of microplastics at the dry weather sampling, whereas the concentrations in samples taken during rainfall were too high to be counted within a reasonable time. The composition of the collected material differed between the rainy and the dry weather occasions. The samples from the STP plume that were collected in the rain were full of cellulose, whereas there was hardly any cellulose in the plume sample from the dry weather sampling. This might imply that there has been an overflow of waste water due to the excess of water in connection with the rain fall, in which case cellulose from toilet paper could be expected to occur in the recipient.

Sampling of sediment and biota in the recipient of Ryaverket was only done at one location and it was therefore not possible to distinguish between the general input from this intensely urbanized area and the input from STP effluents. The concentrations in both blue mussels (*Mytilus edulis*) and sediment from the recipient water of Ryaverket were however considerably higher than concentrations in blue mussels and sediment from the reference site in the Gullmar Fjord; on average 2.8 microplastics per mussel in the STP recipient compared to 0.5 at the reference site, and 0.8 microplastics $\geq 300 \mu\text{m}$ per gram dry weight in the recipient sediment compared to 0.2 at the reference site. Some information on the origin of the litter particles may also be obtained by comparing the character of individual particles in the waste water effluents with what was found in the environment. However, none of the particles that seemed to be typical in the Swedish waste water, e.g. some semi-transparent PE particles, were detected in the sediment or blue mussels from the Ryaverket recipient area. The analysed sediment was fairly muddy which was taken as an indication that it had been collected from an area where microlitter particles could be at least temporarily deposited. But it is possible that the sampling was done too close to the discharge point to allow microlitter particles from the waste water to settle.

Microplastic concentrations in the recipient water of the Finnish STP Kalteva varied even more between the two sampling occasions than concentrations in the recipient of Ryaverlet. There were on average 12.7 microplastics $\geq 100 \mu\text{m}$ per m^3 at the first sampling occasion and 0.7 at the second. The weather was dry before and during both sampling occasions and hence does not explain the large variation in microplastic concentrations. The reason for variation is most likely a combination of occasional sampling and variations in water quality of the recipient. The most common litter type found from the recipient water from both sampling occasions was textile fibers, which indicates the impact of STP. There is likely to be an influence from other sources for microplastic at the Kalteva recipient so the relative contribution from the STP cannot be determined with certainty. Concentrations at the reference site, upstream of the Vantaa river were zero at both samplings.

In the Kalteva recipient the analysed biota included three species of fish, bullhead (*Cottus gobio*), gudgeon (*Gobio gobio*) and roach (*Rutilus rutilus*). Microplastic particles were detected in gudgeon (0.4 microplastic per individual fish) and roach (0.1 microplastic per fish) but not in the bullhead. The concentrations were lower than what was found in a study from the English Channel where microplastics were detected in all of the ten investigated species (both pelagic and demersal species) at an average concentration of 1.9 microplastic per fish (Lusher *et al.*, 2013). In that study altogether 504 specimen were analyzed and 184 individuals (36.5%) had ingested plastics. Among the fish collected from the Finnish recipient 27.7% had ingested litter. It is however possible that not all the litter particles were detected since this analyses of microplastic/microplastics in biota was the first attempt ever in Finland, and therefore must be considered as a pilot study. The method used was relatively easy, but includes some uncertainties. The fish tissue was almost completely dissolved for most of the fish specimen, but with larger fish (in this case roach) the contents of the stomach remained partly present in the sample, and may have hindered recognizing all foreign particles from the tissue material. Since the number of fish individuals analyzed per species was small, no conclusions can be made from this study on the role of feeding mode for microplastic ingestion. Common for all specimen was the presence of sand and bivalves in the stomach content, which shows that the fish were effectively feeding on the sediment.

The number of microlitter particles entering the recipient via waste water effluents from the Icelandic STPs was, as previously discussed, high compared to the Swedish and Finnish ones. From Klettagarðar the estimated number of microplastics was 6,350,000 particles $\geq 300 \mu\text{m}$ per hour and non-synthetic fibres 52,200,000 per hour. Still, the difference in water concentration of microlitter between STP recipient and a reference site was smaller than in the other countries with 3 microplastics per m^3 detected at the reference site, and 5 microplastics per m^3 at the beginning of the drilled holes in the effluent tube and at the mouth of the tube. One likely explanation to this is that the waste water from Klettagarðar is released through a 5,500 m long tube at an open coast line facing the North Atlantic Ocean and the dilution should be very rapid. In addition there was a partial dilution due to the drilled holes along the last 1,000 m of the pipe. Still, the release of waste water is done in a shallow area with a water depth of around 35 m at the mouth of the tube, so an impact on the local biota cannot be excluded. It should be noted that collection of water samples at the Icelandic recipient and reference locations was done with a plankton net with a $100 \mu\text{m}$ mesh size i.e. considerably smaller than the nets used in Sweden and Finland.

Three species of fish were analyzed from the Icelandic recipient and reference water, cod (*Gadus morhua*), haddock (*Melanogrammus aeglefinus*), and plaice (*Pleuronectes platessa*). The detected microlitter concentrations could, however, not be linked to the waste water effluents. Concentrations in fish caught in the area around the effluent tube from Klettagarðar were not generally higher than concentrations in fish from a clean reference site. In fact cod caught at the reference site contained surprisingly high concentrations, on average 6 microplastic particles per individual compared to 1 per individual in cod from the mouth of the waste water tube. On the other hand concentrations of 6.6 microplastics per fish were found in haddock from the STP recipient water. However, the fish is mobile and it cannot be excluded that fish from both sites had been feeding from the same source. The Icelandic fish, both from the STP recipient water and from the reference site, generally contained more microlitter than fish from the Finnish STP recipient water.

Microlitter concentrations in the sediments from Iceland were only analysed in the STP recipient. Values were found to be two orders of magnitude lower than what was found at the Swedish reference site. However, the data could not really be compared since they obviously were from areas with very different hydrography. The Swedish sediments were fine grained with a fairly low dry weight/wet weight ratio, indicating a bottom

where at least some sedimentation occurs. The Icelandic sediment, collected in the shallow area where the STP effluent tube is located, consisted of coarse sand and was full of gravel, which indicates that this is an erosion bottom where fine material does not settle but is immediately washed away. Therefore, it is not very likely that this should be a deposit area for microlitter.

In order to link the microlitter data from field sampled water, biota and sediment to the waste water effluents, sampling locations have to be selected with great care. The water samples must be collected in the actual waste water plume and biota should be of a species that is able to ingest litter particles and from a location where the animals have been exposed to the plume. Sediment samples should preferably be from an area where particles in the effluent waste water would have a good chance to settle. In the present study the recipient waters in the three countries were of very different character which had as a consequence that sampling could not be carried out in the same way and, when it came to biota, different species had to be selected. This meant that different sampling protocols had to be used and comparisons between the recipients in the three countries were not always possible.

In the Kalteva recipient waste water is released in a relatively narrow and shallow river (<5 m depth). The plume was easy to detect and the water sampling was done by pumping of water at one single location. The recipient water for Ryaverket is a wide river and effluent waste water could set off in several directions and also spread out over a larger area. Still, the water depth was only 5–6 m, and the plume could be expected to reach the surface quite rapidly. The sampling of water was therefore carried out in the surface with a manta trawl going back and forth in the area where the plume was estimated to be. To identify the direction of the waste water plume in the recipient water to Klettagarðar on Iceland was even more complicated since the mouth of the tube faces the open Atlantic Ocean, and the direction is highly affected by wind, waves and sea currents.

Another important factor that could affect the results is the large risk for contamination of the samples. This became obvious when analysing the Finnish sediment samples where fibres from the ropes of the sediment sampling device were so numerous that the samples had to be dismissed. The sediment sampling was carried out in the soft and muddy riverbank where the handling of the equipment became so complicated that contamination could not be avoided.

9.3 Conclusion

It can be concluded that influent waste water to STPs contained high numbers of microlitter particles. In Swedish and Finnish plants, with mechanical, chemical and biological treatment, more than 99.7% of the microparticles $\geq 300 \mu\text{m}$ were retained in the sludge. No or only limited retention took place in the Icelandic plants where the waste water was only treated mechanically by being passed through a coarse grid. In spite of the large retention efficiencies in Swedish and Finnish STPs concentrations between 8 and 40 microplastics $\geq 300 \mu\text{m}/\text{m}^3$ were found in the effluent water, and clearly elevated water concentrations were found in the STP recipients. Only slightly elevated concentrations were detected in the Icelandic recipient water, but this might have been at least partly a result of the exposed location of the waste water discharge point which most likely allowed a very rapid dilution. Microlitter particles were detected in both biota and sediment at all sampling sites but it was not possible to correlate the findings to waste water effluents.

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12. Sammanfattning

Förekomst av mikroskopiskt skräp i havet är ett problemområde som fått mycket uppmärksamhet det senaste decenniet. Marint mikroskräp kan ha många olika källor och nå havet via många olika tillförselvägar, och för att kunna vidta effektiva åtgärder måste dessa identifieras.

Utgående vatten från avloppsreningsverk (ARV) är en tillförselväg för mikroskräp till havet och annan akvatisk miljö. Syftet med denna studie har varit att kvantifiera mängden mikroskräp i obehandlat ingående och behandlat utgående avloppsvatten från ARV i Sverige, Finland och Island, två verk i respektive land. Undersökningar gjordes också i recipienten till ett av verken från varje land, och här analyserades mikroskräpmängden i vatten, sediment och biota. Storleken på analyserade partiklarna i vattnet var $\geq 300 \mu\text{m}$ medan analyserade partiklar i biota och sediment var $\geq 100 \mu\text{m}$. Mikroskräpet delades upp i två huvudgrupper, mikroplast och antropogena icke-syntetiska fibrer (t.ex. bomull).

Studien visar att i svenska och finska ARV kvarhölls mer än 99.7 % av de mikroskräppartiklar $\geq 300 \mu\text{m}$ som fanns i inkommande avloppsvatten. Dessa ARV var utrustade med mekanisk, kemisk och biologisk rening av avloppsvattnet. I de isländska verken fanns enbart mekanisk rening och här var kvarhållningen ingen/mycket obetydlig. Behandlat utgående avloppsvatten från de svenska och finska verken innehöll mellan 10 och 40 mikroplastpartiklar per m^3 , och vatten från de isländska verken $\sim 1\,500$ mikroplastpartiklar per m^3 . Koncentrationen av icke-syntetiska fibrer var i ungefär samma storlek. Eftersom det var en betydande variation i storleken på de ARV som ingick i studien, var även stora skillnader i volymen av avloppsvatten som passerade genom de olika verken. Så trots att koncentrationen av mikroskräppartiklar i utgående vatten var ungefär densamma från alla svenska och finska ARV var det betydligt fler partiklar som tillfördes recipienten från de större verken än från de mindre.

Plymen av avloppsvatten var tämligen lätt att identifiera i de svenska och finska ARV-recipienterna. I bägge dessa recipienter var koncentrationen av mikroskräp avsevärt högre än i undersökta referensområden. Den svenska recipienten utgjordes av Göta älvs mynning, ett område med omfattande urban och industriell påverkan, men koncentrationen mikroskräp var betydligt högre i själva avloppsvattenplymen än i omgivande vatten

Vattenkoncentrationen av mikrokräp i den svenska ARV-recipienten befanns vara signifikant lägre i samband med en period utan nederbörd jämfört med en period med mycket nederbörd, 1,9 mikroplastpartiklar och 1,5 icke-syntetiska fibrer per m³ jämfört med 10,5 mikroplastpartiklar per m³ och icke syntetiska fibrer alltför många för att kunna räknas. I den finska ARV-recipienten var koncentrationen i genomsnitt 12,7 mikroplastpartiklar och 11,3 icke-syntetiska fibrer vid första provtagningstillfället och 0,9 mikroplastpartiklar och 6,7 icke-syntetiska fibrer vid andra. Skillnaden kan inte förklaras med några uppenbara klimatologiska faktorer eftersom bägge provtagningarna gjordes under perioder av torrt väder. Mikroplastkoncentrationen i den isländska ARV-recipienten var något förhöjd jämfört med ett referensområde, och skillnaden var större för icke-syntetiska fibrer än för mikroplaster. Mikroplastkoncentrationen varierade mellan 2 och 5 partiklar per m³ i både ARV-recipient och referensområdet och orsaken till den lilla observerade skillnaden antas vara att utsläppspunkten för avloppsvattnet ligger i ett område med en öppen kust och mycket god vattenomsättning.

Mikroplast och antropogena icke-syntetiska fibrer hittades i både sediment och biota i ARV-recipienter i alla tre länder. Det var dock inte möjligt att säkerställa om avloppsvattnet verkligen utgjorde källan till de detekterade partiklarna.

Studien visar att utgående vatten från ARV är inkörsvägar för mikroplast och andra mikrokräpppartiklar till akvatisk miljö. Om verken är utrustade med kemisk och biologiska rening kommer merparten av partiklarna i inkommande vatten att hållas kvar i avloppsslammet. Detta minskar den omedelbara påverkan på recipientvattnet, men om slammet används t.ex. för gödsling av åkrar kommer partiklarna likväl att spridas i miljön. Åtgärder för att minska mikrokräp i avloppsvatten bör därför framförallt sättas in vid själva källan, d.v.s. i hushåll och andra platser där avloppsvattnet har sitt ursprung.

Micro litter in sewage treatment systems

The report presents results from a study on the role municipal sewage treatment plants (STPs) have as entrance routes for microplastics and other micro litter particles to the marine environment. Micro litter concentrations were analysed in waste water before and after treatment in the STPs, and in the recipient waters where the treated waste water is discharged.

Municipal waste water was found to contain a substantial amount of micro litter, but in STPs equipped with chemical and biological treatment most of the litter particles were retained in the sewage sludge. This reduces the impact on the recipient water, but if the sludge is used as fertilizer on farm land the micro litter will still reach the environment. Efforts to reduce the micro litter concentrations should therefore preferably be done in households and other locations where the waste water is originally being formed.

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