Do wastewater treatment plants act as a potential point source of microplastics? Preliminary study in the coastal Gulf of Finland, Baltic Sea

Julia Talvitie, Mari Heinonen, Jari-Pekka Pääkkönen, Emil Vahtera, Anna Mikola, Outi Setälä and Riku Vahala

ABSTRACT

This study on the removal of microplastics during different wastewater treatment unit processes was carried out at Viikinmäki wastewater treatment plant (WWTP). The amount of microplastics in the influent was high, but it decreased significantly during the treatment process. The major part of the fibres were removed already in primary sedimentation whereas synthetic particles settled mostly in secondary sedimentation. Biological filtration further improved the removal. A proportion of the microplastic load also passed the treatment and was found in the effluent, entering the receiving water body. After the treatment process, an average of 4.9 ($\pm$1.4) fibres and 8.6 ($\pm$2.5) particles were found per litre of wastewater. The total textile fibre concentration in the samples collected from the surface waters in the Helsinki archipelago varied between 0.01 and 0.65 fibres per litre, while the synthetic particle concentration varied between 0.5 and 9.4 particles per litre. The average fibre concentration was 25 times higher and the particle concentration was three times higher in the effluent compared to the receiving body of water. This indicates that WWTPs may operate as a route for microplastics entering the sea.

Key words | activated sludge process, microplastics, municipal wastewater treatment, tertiary biological filtration, textile fibres

Julia Talvitie (corresponding author)
Anna Mikola
Riku Vahala
Department of Civil and Environmental Engineering, Aalto University,
PO Box 15200, FI-00076, AALTO,
Finland
E-mail: *julia.talvitie@aalto.fi*

Mari Heinonen
Helsinki Region Environmental Services Authority HSY,
PO Box 100, FI-00066 HSY,
Finland

Jari-Pekka Pääkkönen
Emil Vahtera
City of Helsinki Environment Centre,
PO Box 500, FI-00099,
Helsinki,
Finland

Outi Setälä
Finnish Environment Institute,
Marine Research Center,
PO Box 140, FI-00251 Helsinki,
Finland

INTRODUCTION

Annual global plastic production in 2013 rose to 299 million tonnes, with a 3.9% increase compared to 2012 (PlasticEurope 2014). With growing plastic production, the amount of plastic litter in the environment is also increasing. This causes plastic litter to accumulate in various environments, including marine habitats (Andrady 2011). Marine litter consists of 60–80% plastics, most of which is quite small (<5 mm), called microplastics (Arthur *et al.* 2009).

Marine microplastic litter derives from a variety of sources, such as fragmentation of larger plastic particles, traffic, industry, and wastewater treatment plants (WWTPs). Treated municipal wastewaters contain e.g. synthetic textile fibres from washing of clothes and abrasive plastic fragments from cleaning agents (Browne *et al.* 2011; Magnusson & Norén 2014). Many marine invertebrates like bivalves, echinoderms, amphipods and zooplankton are known to ingest microplastics (Browne *et al.* 2008; Graham & Thompson 2009; Cole *et al.* 2013; Setälä *et al.* 2014). Ingested plastics can cause internal damage and also reduce feeding, disturb the digestive enzyme system and hormone balance and have an impact on reproduction (Derraik 2002). Plastics can contain harmful additives such as phthalates and flame retardants, and they may also absorb hydrophobic pollutants, such as polychlorinated biphenyls (PCBs) and DDE from the surrounding water (Mato *et al.* 2001). Microplastics may thus potentially transfer environmental pollutants to marine food webs.

The Baltic Sea is a shallow (mean depth 54 m), semi-enclosed brackish sea. It is connected to the Atlantic Ocean through the narrow Danish straits and it takes decades for all of the water to renew itself. The coastal countries around the Baltic Sea are highly industrialized, and a total of 85 million people live within its catchment area. The Baltic Sea receives nutrients and pollutants from

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agricultural, industrial and wastewater run-off, which has led to the serious eutrophication and deterioration of the water quality. Consequently, the Baltic Sea is considered to be one of the most contaminated sea areas in the world. (HELCOM 2009, 2010). Although many of the environmental problems of the Baltic Sea (e.g. eutrophication and hazardous substances) are well known (HELCOM 2010, 2014), 'plastic pollution' of the Baltic Sea has been little studied. The EU Marine Strategy Framework Directive (2008/56/EC) aims to achieve good environmental status for the EU's marine waters by the year 2020. According to this Directive, a good status in terms of marine litter will be achieved when the 'properties and quantities of marine litter do not cause harm to the coastal and marine environment'. At the moment, the goal is to clarify the quantity, quality and impacts of the marine litter and reduce the amount of litter from its present level.

Microplastics in marine environment are commonly collected with towable nets (e.g. Norén 2009; Hidalgo-Ruz *et al.* 2012; Magnusson 2014), or with submerged pumps that enable the filtration of large amounts of particles from the sea surface (Magnusson & Norén 2011). In the Baltic Sea, along the Swedish coast, according to the study of Norén (2009) the concentration of microplastics, when sampling with 20 μm plankton net, varies between 0.5–15 fibres and 0.06–105 synthetic particles per litre of sea water. In 2011 Magnusson and Norén reported the average microplastic concentrations of 0.008 fibres and 0.013 particles per litre of sea water when sampling with the filter size 300 μm and average of 4 fibres and 32 particles per litre of sea water when the filter size was 10 μm. In the west coast of Finland, the reported concentration of microplastics was up to ~0.001 items per liter of sea water, when sampling with manta trawl (mesh size 333 μm) (Magnusson 2014).

At present there is very little knowledge of the fate of microplastics in WWTPs and the microplastics concentrations on wastewater discharges. In addition, the lack of a standardized method for sampling the microplastics in wastewaters complicates the comparison of the results obtained from few studies. Browne *et al.* (2011) reported the average microplastic concentration of 1 per litre in effluent in two different tertiary-level WWTPs at West Hornsby and Hornsby Heights, Australia. The samples were collected with glass bottles (750 mL) and filtered using Whatman GFA filters. Microplastics were identified with Fourier transform infrared spectroscopy (FTIR) (Bruker I26933, Synthetic fibres ATR library). Magnusson & Norén (2014) reported the microplastic concentration of 15 per litre in influent and 0.008 per litre in effluent. The study was carried out at a small WWTP treating the wastewater of ca. 12,000 residents. The treatment process consisted of physical, chemical and biological processes, but no tertiary treatment. Incoming water was collected with a Ruttner sampler and the water was poured into a filter holder with 300 μm mesh size filter, cut from the plankton net. The effluent was sampled with the same filter holder attached into a tube and suction pump. Microplastics were counted using a stereomicroscope and individual particles were analyzed with FTIR (Bruker Tensor, ATR mode). Also Dris *et al.* (2014) reported the microplastic concentrations of 469 fibres per litre in influent and 31 fibres per litre in effluent at the Seine Centre de Colombes wastewater plant, Paris.

Aims of this study were to investigate the WWTPs as a route for microplastics entering the sea and to test the microplastic sampling method developed for wastewaters. A further objective was to obtain information about microplastic pollution levels in the Helsinki archipelago coastal Gulf of Finland.

MATERIALS AND METHODS

The study was carried out at the Viikinmäki WWTP (Helsinki Region Environmental Services Authority, HSY) and in the archipelago area (Figure 1). Viikinmäki WWTP is responsible for treating the wastewater of approximately 800,000 inhabitants in the Helsinki metropolitan area. The treated wastewater is discharged into an open sea area, at a depth of more than 20 m approximately 8 km from the southern tip of Helsinki. The wastewater treatment at Viikinmäki consists of bar screening, grit removal, pre-aeration, primary sedimentation, activated sludge treatment, secondary sedimentation and a tertiary biological filtration. The quality of treated wastewater is better than that required by the European Union's Urban Wastewater Treatment Directive (91/271/EEC) and national legislation of Finland (Government Decree on Urban Wastewater Treatment 888/2006, Finlex). In 2012, ≥95% of BOD7, 98% of suspended solids, ≥95% of total phosphorus and 90% of total nitrogen were removed from the influent of the Viikinmäki WWTP.

The amount of microplastics in the WWTP influent and the effect of the treatment process on the total amount of microplastics were studied by sampling wastewater at the beginning of the process before screening, after the primary sedimentation phase, after the secondary sedimentation phase and after the tertiary DN-post-filtration unit (Figure 1). The sampling was performed in 2012 between October and

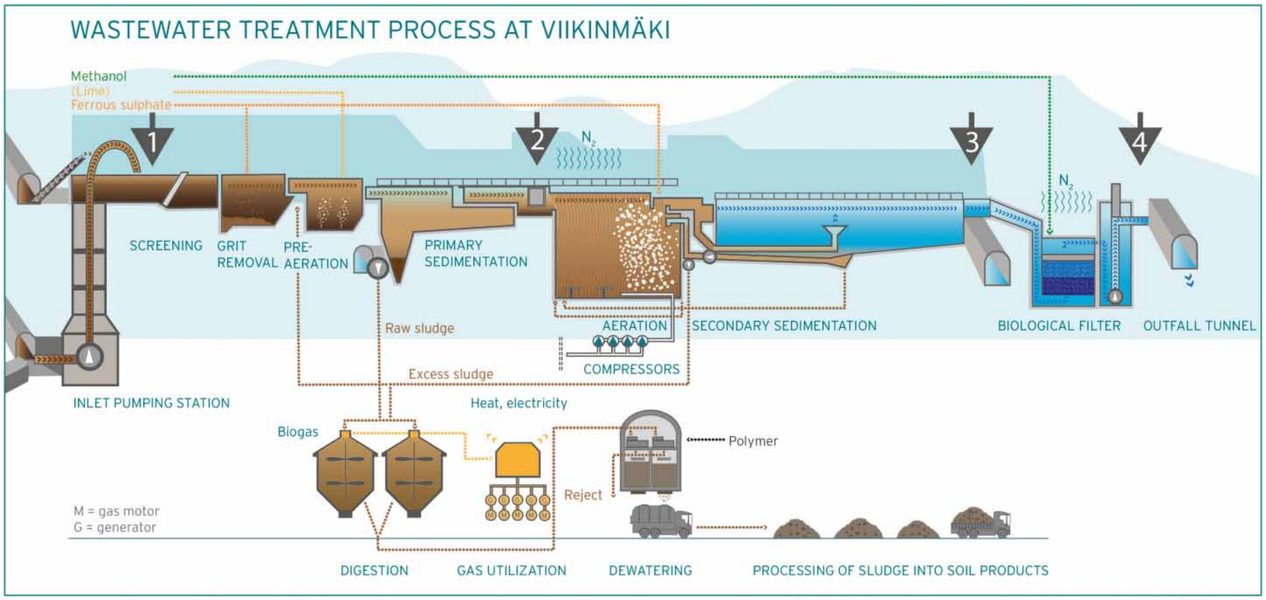

Figure 1 | Wastewater treatment process at Viikinmäki. Sampling points are marked with arrows: influent (1), after the primary sedimentation (2), after the secondary sedimentation (3) and after the DN-post-filtration unit (4).

December. Three replicates were taken from each sampling point.

The novel sampling method was developed to capture different size-fractions of the microplastics which are present in the wastewaters. The sampling method is based on separate pumping and filtration technique. The method was also used for sea water sampling as we compared the amount of microplastics in wastewater and sea water.

For the sampling process, wastewater was pumped from the wastewater stream into the filter device with an electric pump (pump drive 5206 Heidolph), with a flow rate of 1.0 ml/min. A specific filter device was designed for filtering the microplastics from the wastewater (Figure 2). Water is filtered through different mesh-sized filters: 200, 100 and 20 μm, respectively. The device consists of three transparent plastic tubes (diameter 60 mm) and screw-on plastic connectors attaching the tubes to one another. Round (diameter 80 mm) filters, cut from a plankton net, are placed into the filter device between the connectors, and the tubes are screwed tightly together using rubber o-rings (Figure 3). The largest mesh-sized filter, 200 μm, was placed on top of the device, the 100 μm filter in the middle of the device and the 20 μm filter at the bottom of the device. In the order 200 μm → 100 μm → 20 μm, the sample was filtered through the device.

The filtered sample volume depended on the used filter size and sampling point at the WWTP (Table 1).

To get information about microplastic pollution levels, the sea water samples were collected from the Helsinki archipelago (Viikinmäki VM, Suomenoja SO, 4, 18, M33, 39, 125) from August to November 2012. Suomenoja is the second WWTP in the Helsinki metropolitan area, treating the wastewater of ca. 310,000 inhabitants. Its physically, chemically and biologically treated wastewaters are conducted to the open sea area, and discharged at a depth of 20 m. To detect the possible loading effect of WWTPs, the sediment samples were collected from the wastewater discharge points (VM $n = 6$, SO $n = 4$) as well as from the reference site 59° 57.13′, 25° 7.05′ (M30 n = 6), during an R/V Muikku cruice 29.7–8.8.2013. In total, seven sea water samples and 16 sediment samples were collected (Figure 4).

The sea water samples were pumped with a petrol-driven pump (Honda WX10) and filtered using the same method as used in the WWTP. One cubic metre (1 m^3) of water was filtered through the 200 μm and 100 μm filters, while the water volume filtered through the 20 μm filter varied from 20 to 30 litres.

The sediment samples were collected using a Gemax corer sampler. The top 30 mm was removed from each sediment sample and placed into pre-cleaned containers. Microplastic litter was extracted from the sediment using concentrated saline NaCl solution (Browne et al. 2010). The sediment sample was weighed and poured into the sodium chloride solution and the mixture was stirred thoroughly for 1 minute. After mixing, the sample was allowed to settle for 20 minutes. After the settling period,

Figure 2 | The filter device.

Figure 3 | Filter is placed between the screw-on connectors.

the supernatant was filtered through a filter device, as were the water samples.

The samples were inspected using a stereomicroscope (Wild heerbrugg, magnification ×50), and the microplastics were identified and counted. The microplastics included in the study were particles and fibres with different colours. High-gloss black carbon particles were also included in the study. These anthropogenic, oil-based particles have a specific shiny blue-black appearance, which distinguishes them from other dark particles (Magnusson & Norén 2011). Dark, non-glossy particles were not included because it is not possible to characterize their material via light microscopy. Microplastics were identified through the transparent lids of the Petri dishes, but when analyzing the smallest particles from the 20 μm filter, the lid had to be removed to improve the lighting.

All equipment was rinsed thoroughly with tap water prior to sampling to avoid contamination. Potential sources of microplastic contamination include room dust and fibres from the clothing of the staff gathering the samples. To detect possible sources of contamination during transport and while performing microscopic analyses, blank samples were carried along twice, first when sampling the wastewater and then during the sea water sampling phase. The filters for blanks were prepared in the same way as for the actual sampling: they were rinsed with tap water and inspected with a microscope for possible contamination before use. All of the filters were sealed carefully in the Petri dishes and carried along during

Table 1 | The filtered sample volumes with mesh sizes of the filters and purification phase in the WWTP

Sampling point	Mesh size of the filter (μm)	Sample volume (litres)
Influent	200	0.3
	100	0.3
	20	0.3
After primary sedimentation	200	10–20
	100	10–20
	20	1
After secondary sedimentation	200	35–50
	100	35–50
	20	1
Purified wastewater	200	30–285
	100	30–285
	20	2–5

the sampling process. The water was not filtered through blank filters, and therefore, the blanks will not show the possible contamination during the sampling process. Blanks were analyzed with actual samples using a stereo microscope.

Statistical analyses

The results were statistically analyzed using analysis of variance (ANOVA). The test was applied for Viikinmäki primary clarified and effluent water data sets as well as for sediment samples.

RESULTS AND DISCUSSION

Wastewater treatment

Microplastics were found in all samples taken from Viikinmäki WWTP. As expected, the highest number of

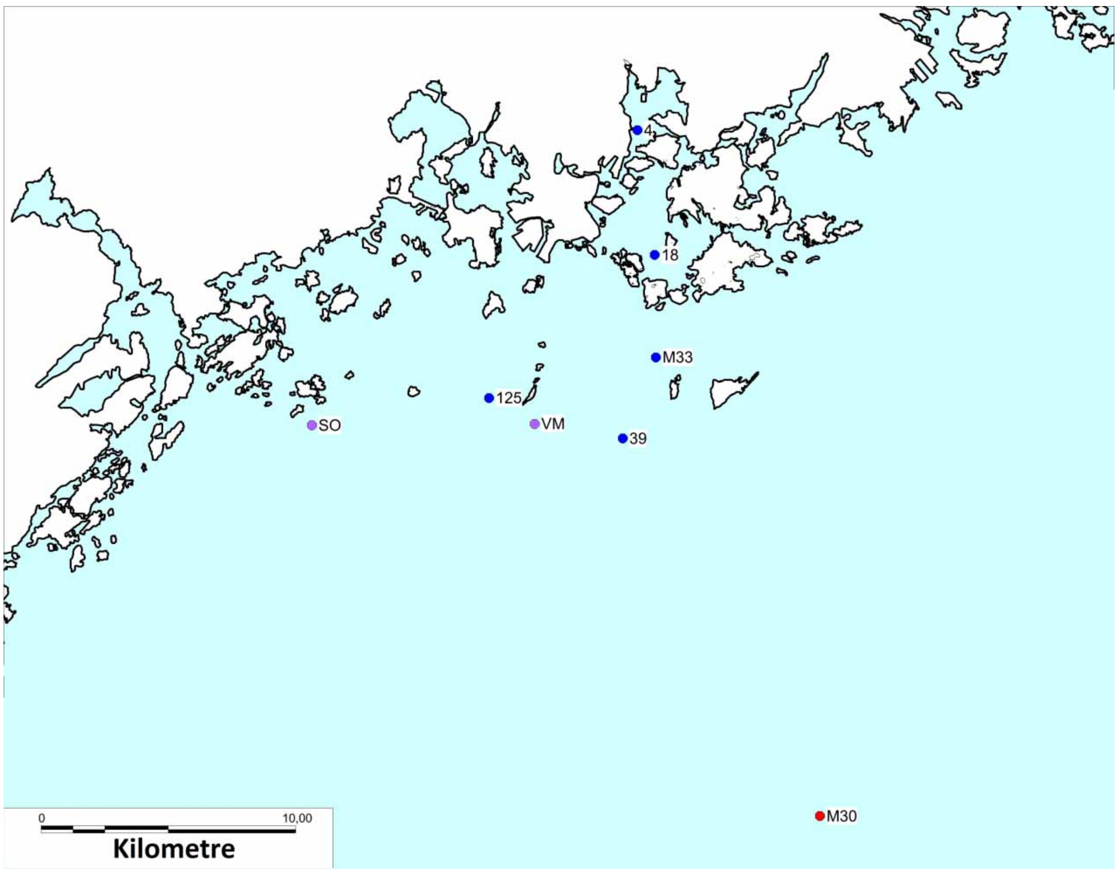

Figure 4 | The sampling sites in the Helsinki archipelago. Points (4, 18, M33, 39, 125) are the sea water samples sites, and from the points (SO, VM) both the sea water and sediment samples were collected. Point (M33) is reference site for sediment samples.

microplastics were found from the influent which contained 180 textile fibres and 430 synthetic particles per litre (Figure 5). Unfortunately, only one successful sample was taken from the influent during this study. The problem with influent samples was rapid blockage of the filters with organic litter. The first influent sampling with replicates included 1 litre of filtration. The 1 litre samples were full of organic material and identification and calculation of the microplastics with microscope became unreliable due the poor visibility. In the successful influent sample only 0.3 litre of wastewater was filtered.

After the treatment process an average of 4.9 (±1.4) fibres and 8.6 (±2.5) synthetic particles per litre of wastewater were found. The number of fibres ($F = 10.29 > 6.992$; $P < 0.01$) and particles ($F = 74.02 > 6.992$; $P < 0.01$) in the wastewater decreased significantly during the purification process (ANOVA). After the primary sedimentation wastewater contained an average of 14.2 (±0.7) fibres and 290.7 (±28.2) synthetic particles per litre of wastewater. After the secondary sedimentation the corresponding concentrations were 13.8 (±1.6) fibres and 68.6 (±6.3) synthetic particles. It seems that the fibres were settling, and captured into the sludge or filtered during the treatment process. However, a certain proportion of them also passed through the treatment process and ended up in the water environment. Our observations indicate that fibres were mostly removed already during primary sedimentation. The removal of fibres was insignificant during secondary sedimentation and biological filtration, although filtration removed fibres to a low level. A minor proportion of particles was removed in primary sedimentation and most of the removal took place during secondary sedimentation. Biological filtration removed also the particles. Due the differences in used methods (e.g. mesh size of the filter, sample volume, filtering pressure) it is impossible to reliably compare our results in detail with previous studies. However, our results are consistent with the previous studies, reporting the relatively efficient removal of microplastics from wastewater during the treatment processes (Magnusson & Norén 2014; Dris *et al.* 2014).

Sediments at discharge sites

From the discharge site of Viikinmäki WWTP 1.7 (±1.0) fibres, 1,220 (±160) black carbon particles, 7.2 (±4.9) synthetic particles and 70 (±20) ring-shaped particles per kilogram of sediment were found. In the discharge site of Suomenoja the corresponding figures were 4.7 (±3.5) fibres, 1,060 (±471) black carbon particles, 10 (±14) synthetic particles and 3.8 (±2.3) rings and at reference site 1.7 (±1.3) fibres, 346 (±186) black carbon particles, 0 synthetic particles and 1.9 (±1.5) rings (Figure 6). Only black carbon particle ($F = 22.450 > 10.044$; $P < 0.01$) and ring-shaped particle ($F = 20.676 > 10.044$; $P < 0.01$) abundances were significantly higher at the discharge site of Viikinmäki compared to the reference site (ANOVA). With fibres and synthetic particles no significant difference between the discharge site of Viikinmäki and the reference point was detected. Between the Suomenoja discharge site and the

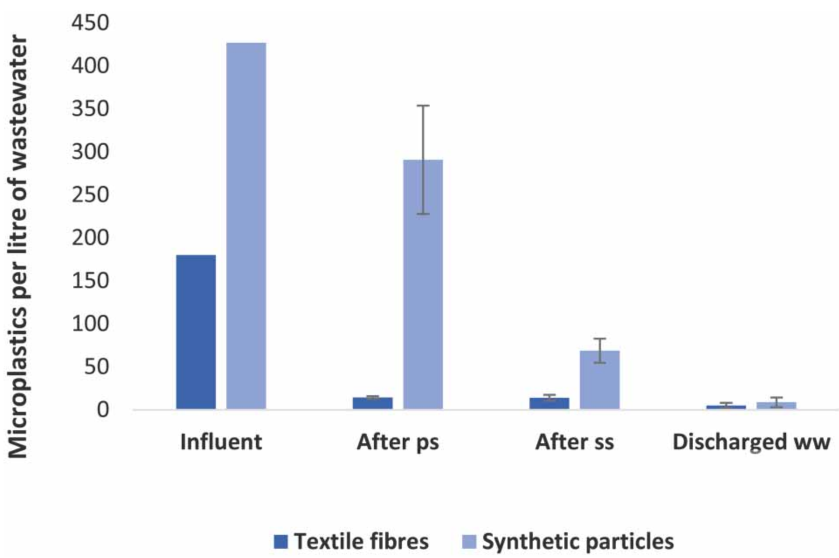

Figure 5 | The microlitter concentration in the influent ($n = 1$), after primary sedimentation (ps) ($n = 3$), secondary sedimentation (ss) ($n = 3$), and in discharged wastewater ($n = 3$). Error bars denote standard deviation.

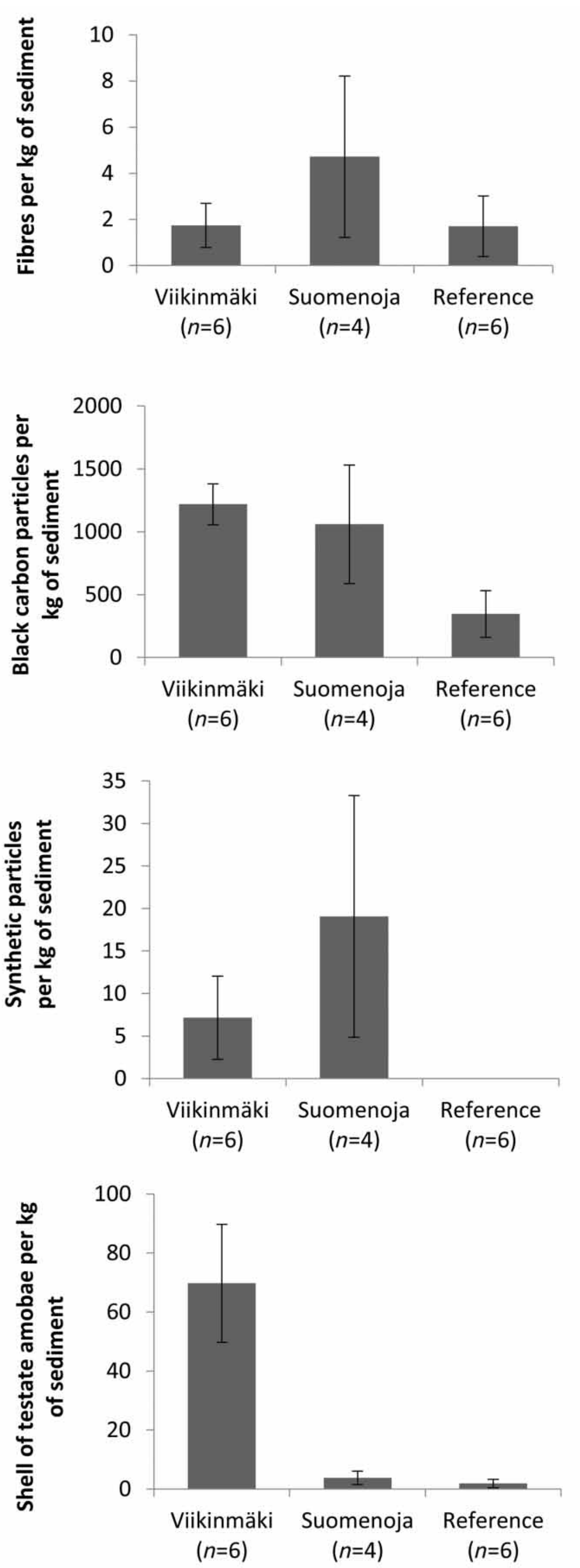

Figure 6 | Textile fibres, black carbon particles, synthetic particles and testate amoebae shell concentrations in sediment samples taken from Viikinmäki and Suomenoja WWTPs discharge sites and reference site.

reference site no significant difference was detected with any particle types or fibres.

Since not all of the microplastics were removed during the treatment process, we expected that the abundance of microplastics and would be higher in the sediment at the discharge sites of the WWTPs than at the reference site. However, we found that only the abundance of ring-shaped particles identified as the shells of testate amoebae (diameter of about 60 μm), common organisms in Viikinmäki wastewater and sludge, was significantly higher at the Viikinmäki discharge site (Figure 7). The finding suggests that at least part of the micro-sized litter has potential to settle into the vicinity of the discharge site, even when a large part of the litter is transported over a wider area. Elevated levels of black carbon particle abundances detected at discharge sites are unlikely caused by WWTPs as the particle type was not found from the wastewater. The known sources of these black particles are e.g. traffic and fossil fuel combustion (Wik & Renberg 1996; Wik & Dave 2009; Magnusson & Norén 2011). Treated wastewater from Viikinmäki and Suomenoja WWTPs are discharged into open sea areas, where rapid mixing of wastewater with large amount of sea water probably prevents microplastics accumulating in the vicinity of the discharge site.

Water quality at Helsinki archipelago

The fibre concentration in the sea water of the Helsinki archipelago varied between 0.01 and 0.65 per litre, whereas the concentration of synthetic particles varied between 0.5 and 9.4 per litre (Table 2). In comparison, Magnusson & Norén (2011) found the average of four fibres and 32 particles per litre of sea water when using the filter size 10 μm along the Swedish coast line. The samples were taken from breakwaters and bridges with filtering method similar to ours and hence the results are comparable. The concentrations obtained with 10 μm filtering are clearly higher compared to our study. One explanation for higher concentration can be the sampling locations near to land and hence near to many different land-based microplastic sources e.g. stormwater flow, traffic, industries, etc. Also, the smaller (10 μm compared to our 20 μm) mesh size of the filter may have an impact. In general, the smaller the mesh sizes in sampling, the higher microplastic concentration (Norén 2009; Magnusson & Norén 2011).

In general, the average fibre concentration was found to be 25 times greater and the particle concentration three times greater in discharged wastewater compared to sea water. This indicates that WWTPs may operate as a point

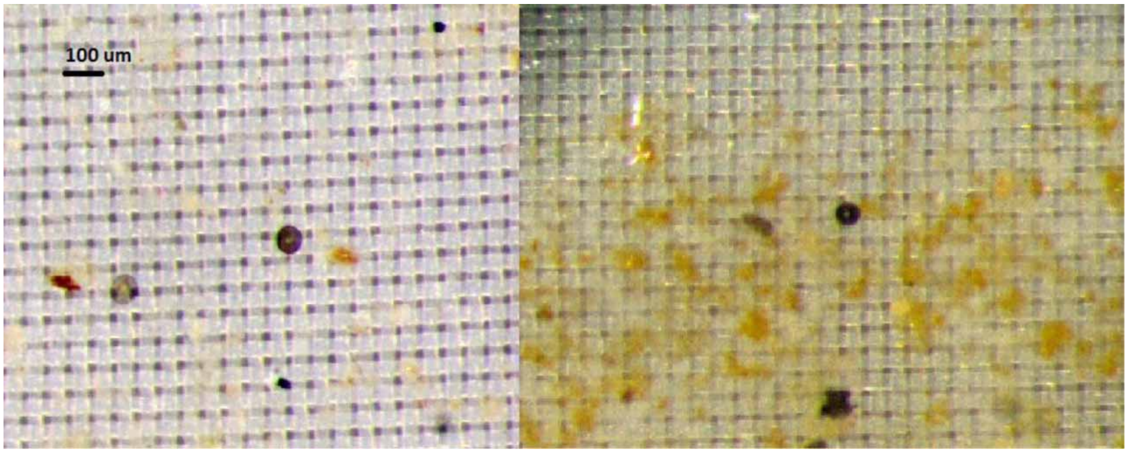

Figure 7 | Image of the shell of a testate amoebae found from a sediment near discharge site of Viikinmäki WWTP (left) and a ring found from a purified wastewater (right).

Table 2 | Microplastic abundances in different areas of Helsinki Archipelago 2012

Area	Coordinates	Fibres per litre ($n = 1$)	Particles per litre ($n = 1$)
Viikinmäki Discharge Site VM	60 ° 5.38′, 24 ° 55.063′	0.06	2.4
Suomenoja Discharge Site SO	60 ° 5.34′, 24 ° 45.678′	0.17	3.1
Vanhakaupunki Bay 4	60 ° 11.56′, 24 ° 59.386′	0.65	0.5
Helsinki Archipelago Site 18	60 ° 8.935′, 25 ° 0.121′	0.24	9.4
Helsinki Archipelago Site 33	60 ° 6.781′, 25 ° 0.175′	0.01	0.5
Helsinki Archipelago Site 39	60 ° 5.075′, 24 ° 58.774′	0.20	1.6
Helsinki Archipelago Site 125	60 ° 5.923′, 24 ° 53.133′	0.04	1.7

source for introducing microplastics into the water environment.

We did not find any microplastics from the blanks, indicating that our samples were not contaminated during the transportation and analyses. Since the blank samples did not include the filtering part, we do not know the effect of actual sampling on contamination. However, the filtering device and all other equipment were rinsed thoroughly with tap water prior to sampling to minimize possible contamination.

Applicability of the used sampling method

With the filtering device designed for microplastic sampling, any water (e.g. wastewater, sea water, sediment supernatant) can be filtered simultaneously through three different mesh-sized filters. This reduces the sampling time when different size fractions are needed. The filtering device is easy and inexpensive to build, use and carry along. Pumping water into the filter device with an electrical or gasoline driven pump is recommended if the sampled volumes are high (>20 litre). In addition, the pump and filter form a closed system and therefore contamination of the samples is unlikely during sampling.

Sediment was sampled using established techniques (Browne *et al.* 2010, 2011). Extracting the microplastics from the sediment with concentrated NaCl solution is based on density separation where lighter plastic particles are separated from the heavier sediment grains by mixing a sediment sample with a saturated NaCl solution and shaking it for a certain amount of time. The sediment samples on our study were stirred thoroughly for one minute and after mixing, the sample was allowed to settle for 20 minutes. This was enough for proper mixing and settling in our samples. However, repetition of density separation of the sample is recommended to receive more reliable results (Hidalgo-Ruz *et al.* 2012). In our study, the density separation was performed only once per sample which might have had some impact on our results.

The micro-sized litter was analyzed as inorganic material using a stereomicroscope. In addition to synthetic textile fibres and inorganic particles, the samples included organic litter. For example, some of the textile fibres were likely dyed cellulose fibres from cotton clothes. In a previous study, Magnusson & Norén (2011) found that the synthetic fibres are often straight and shiny and cross-section symmetrically round. Cellulose fibres in turn are flat, serpentine and non-glossy. Only a few fibres found in this study clearly fit into either category. The fibres found in the wastewater were usually small shreds, and it was difficult to reliably estimate the material of the fibre using only stereomicroscopy. In this study, we included all of the textile fibres in the results. Although natural fibres do not accumulate in a marine environment and will biodegrade, the non-synthetic fibres may also be a threat to marine life as cotton clothes are treated with various toxic chemicals.

More precise analyses of the litter material found in samples would give a better indication of the amounts and quality of plastics entering marine environments with the effluents. Further studies are needed to ascertain the origin and chemical composition of the observed microlitter in the discharged wastewater and coastal areas.

CONCLUSIONS

Our work provides new information on the role of wastewater treatment plants (WWTPs) as a route for microplastics entering the sea. We demonstrated that an efficient treatment process significantly decreases the concentrations of microplastics. Our observations indicate that fibres are mostly removed during primary sedimentation, although tertiary treatment further decreases the number of fibres. Most of the synthetic particle removal took place during secondary sedimentation while also tertiary treatment further removed the particles.

Although the possible microplastic loading effect of WWTPs on marine environment was not directly detected, the concentration of microplastics in discharged wastewater was higher than in the receiving water body. This indicates that WWTPs may operate as a route for microplastics to enter the water environment.

Considering that microplastics cannot be effectively removed from the water environment, future studies are needed to evaluate the sources and routes for microplastics to enter water environments, so that preventing actions of microplastic pollution can be better targeted. More knowledge is also needed to better estimate the actual role of WWTPs on marine microplastic load and their capacity to remove microplastics from wastewater.

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