Accepted Manuscript

This is an Accepted Manuscript of the following article:

Christine Schönlau, Therese M. Karlsson, Anna Rotander, Helena Nilsson, Magnus Engwall, Bert van Bavel, Anna Kärrman. Microplastics in sea-surface waters surrounding Sweden sampled by manta trawl and in-situ pump. Marine Pollution Bulletin. Volume 153, 2020, 111019, ISSN 0025-326X.

The article has been published in final form by Elsevier at http://dx.doi.org/10.1016/j.marpolbul.2020.111019

© 2020. This manuscript version is made available under the

CC-BY-NC-ND 4.0 license

http://creativecommons.org/licenses/by-nc-nd/4.0/

Microplastics in sea-surface waters surrounding Sweden sampled by manta trawl and *in- situ* pump

3

Christine Schönlau ¹ , Therese M. Karlsson ^{1,2} , Anna Rotander ¹ , Helena Nilsson ¹ , Magnus						
Engwall ¹ , Bert van Bavel ^{1,3} , Anna Kärrman ^{1,*}						
¹ Örebro University, MTM Research Centre, Fakultetsgatan 1, 701 82 Örebro, Sweden						
² University of Gothenburg, Department of Marine Sciences, Kristineberg Marine Research Station,						
45178 Fiskebäckskil, Sweden						
³ Norwegian Institute for Water Research, NIVA, Department of Environmental Chemistry, Oslo,						
Norway						
* Corresponding author: Anna Kärrman						
Email: anna.karrman@oru.se						
Telephone: +46 19 301401						

15

16 Abstract

Microplastics were sampled in open surface waters by using a manta trawl and an *in-situ* filtering pump. A total of 24 trawl samples and 11 pump samples were taken at 12 locations around Sweden. Overall, the concentration of microplastic particles was higher in pump samples compared to trawl samples. The median microplastic particle concentration was 0.04 particles per m⁻³ for manta trawl samples and 0.10 particles per m⁻³ in pump samples taken with a mesh size of 0.3 mm. The highest concentrations were recorded on the west coast of Sweden. Fibers were found in all samples and were also more frequent in the pump samples. Even higher concentrations of fibers and particles were found on the 0.05 mm pump filters. Using nearinfrared hyperspectral imaging the majority of the particles were identified as polyethylene followed by polypropylene.

27

Keywords: Plastic pollution, Polyethylene, Polypropylene, Baltic Sea, Skagerrak, Kattegat,
Microplastic sampling methods

30

31 Introduction

Microplastics in aquatic environments have become a subject of concern due to the long 32 degradation time associated with plastic products, increasing use of plastic materials, and 33 inadequate waste handling. The term microplastics is not unitary defined and can refer to 34 synthetic polymer particles with different size ranges, however, often a size less than 5 mm and 35 larger than 0.1 mm is referred to as microplastic (Hartmann et al. 2019). The annual global 36 production of plastics is reaching almost 350 million tones and more than one third is used for 37 packaging products made of polyethylene (PE) and polypropylene (PP) plastics (PlasticsEurope 38 2018). The aforementioned polymers together with polystyrene (PS) are the most frequently 39 reported types of plastic in marine samples (Hidalgo-Ruz et al. 2012). It has been estimated that 40 41 the lifetime of plastic can span centuries or even millennia (Barnes et al. 2009), although the 42 lifetime of the plastic material depends on the chemical composition of the material itself and the surrounding environment (Andrady and Neal 2009). Global assessments of floating plastics 43 in the world's oceans span from 14,400 tons to 268,940 tons and the uncertainty reflects current 44 knowledge gaps in occurrence, distribution, and environmental fate of plastics (Eriksen et al. 45

2014). It has been estimated that at least 8 million tons of plastics enter the oceans every year
from land-based sources (Jambeck et al. 2015).

Deliberately or accidentally released, plastics are transported and spread by currents and winds 48 49 and fragmented to smaller particles over time (Andrady 2011). These secondary microfragments of plastics contribute to an increasing amount of small plastic particles in our oceans 50 (Barnes et al. 2009). A mere physical threat such as entanglement, strangulation, and abrasion 51 of the gastrointestinal tract that plastic debris can pose to organisms is at hand and has been 52 reported to affect different species (Cadée 2002; Laist 1997; Mascarenhas et al. 2004). 53 Additionally, it has also been hypothesized that plastic particles can act as a vector for 54 55 transferring persistent organic pollutants (POPs) to organisms upon ingestion, after various POPs have been found on marine plastic debris (Carpenter and Smith 1972; Mato et al. 2001; 56 Teuten et al. 2007). Additionally, a risk of leaching plastic additives, monomers, oligomers, 57 and other polymer degradation products from the plastic material into the environment has been 58 recognized by researchers (Gewert et al. 2015; Teuten et al. 2009). The chemical risk that 59 especially microplastics might pose upon ingestion is however controversially debated and 60 currently not fully explored (Koelmans et al. 2016; Ziccardi et al. 2016). 61

62 The European Union has adopted a Marine Strategy Framework Directive (MSFD) to protect the marine environment (EU 2008). One of the goals is that by 2020 litter that negatively affects 63 or is likely to negatively affect marine organisms will decline. An important component 64 required to achieve this goal is the characterization of different types of litter, such as 65 microplastics, in the marine environment since that can help to understand source patterns and 66 provide a baseline for future monitoring and evaluation of preventive measures. Currently there 67 exists no standardized method for the sampling of microplastics in any environmental 68 compartment. However, a frequently used method for sampling of microplastics in surface 69 waters is the use of a neuston net or a manta trawl with the most commonly used mesh sizes 70

between 300 – 390 μm (Hidalgo-Ruz et al. 2012; Li et al. 2018). Another technique is pumping
water through filters of different mesh sizes using a stationary or submerged pump (Norén et
al. 2009; Setälä et al. 2016; Zobkov et al. 2019).

74 The Baltic Sea is one of the largest brackish waterbodies in the world which is semi-enclosed with a slow water exchange of approximately 30 years with the neighboring North Sea through 75 the Danish straits and a highly urbanized catchment area which is inhabited by about 85 million 76 people (HELCOM 2018). Due to the slow water exchange rate with the North Sea most floating 77 plastic debris can be assumed to originate from local sources of the surrounding countries. 78 Currently HELCOM is working on establishing core indicators for the assessment of marine 79 litter and it has been stated that about 70 % of the litter in the Baltic Sea are made of plastic 80 materials (HELCOM 2018). The occurrence of microplastics has been reported for many 81 marine environments globally (Cozar et al. 2014; Eriksen et al. 2014), but there is little data 82 about the occurrence and identity of microplastics in surface waters of the Baltic Sea (Gewert 83 et al. 2017; Gorokhova 2015), while several studies assessed plastic pollution in sediments and 84 beaches along the Baltic Sea (Esiukova 2017; Hengstmann et al. 2018; Näkki et al. 2019; Stolte 85 et al. 2015). 86

In this study we therefore aim to 1) study the occurrence of microplastics in surface waters of the Baltic Sea, including Skagerrak and Kattegat, 2) identify the polymer types of detected microplastic particles, and 3) compare the results of the two sampling methods employed, in order to add valuable information to the process of harmonizing sampling protocols. In addition, microplastics down to 0.05 mm particle size were analyzed for the filtering pump.

92

93

95 Materials and Methods

96 Sampling setup

Sampling was conducted in Skagerrak/Kattegat, Baltic Sea and Gulf of Bothnia in August 2014 97 using the sailing vessel 'Sea Dragon' (www.panexplore.com). The sampling started in 98 Gothenburg on the Swedish west coast on the 3rd of August and finished in Stockholm located 99 on the Swedish east coast on the 23rd of August. A total of 12 sites were sampled (Figure 1). 100 101 Sampling was conducted using two methods; a manta trawl and an *in situ* pump (see figure S1 in the supplementary material (SM)). One sampling site spanned over approximately 10 km and 102 103 3 samples were taken at each site; the first sample was taken by towing the manta trawl for 60 min at the side of the sailing vessel with a speed between 0.5 and 1.5 m/s., covering 4-5 km of 104 sea surface. The second sample was taken with the filtering pump which filtered approximately 105 20 m⁻³ of water while the sailing vessel was drifting. For the third sample the trawl was used 106 again as described above (illustration provided in the supplementary material, figure S5). A 107 108 total of 24 manta trawl samples were taken, however, due to technical difficulties at one site, 109 only 11 pump samples were collected. The sampling sites were selected primarily to give a large cross section of the waters surrounding Sweden and secondarily to match the Swedish 110 111 Meteorological and Hydrological Institute monitoring stations (smhi.se/klimatdata/oceanografi/havsmiljödata). Necessary permits for sampling national 112 waters were obtained from authorities in Sweden, Denmark and Finland. 113



114

Figure 1. Sampling locations as yellow circles (1-12), each location consists of two trawl samples and one pump sample, except sample point 5 where only trawl samples were collected (Google maps[®]). A detailed visualization of the sampling scheme is given in SM (figure S5).

118

119 Manta trawl

The manta trawl consisted of an aluminum frame with a rectangular opening with dimensions
16 cm by 61 cm, and a net with a length of 3 m and a mesh size of 333 μm. The end of the mesh

was fitted with a detachable collecting bag with dimensions 30 cm by10 cm. Immediately after 122 123 sampling the content of the trawl was rinsed with filtered sea water down into the collecting bag. The content of the collecting bag was transferred to a metal sieve with a mesh size of 300 124 um by rinsing everything with filtered sea water. Finally, the material on the metal sieve was 125 carefully transferred to glass jars by using metal tweezers and rinsing down the remaining 126 material with filtered sea water. The samples were stored in darkness at room temperature on 127 the boat prior to transport to the laboratory (3-7 days). The volume of water filtered through the 128 trawl was both calculated through multiplying the sampled distance (based on GPS coordinates) 129 with the width of the trawl times half of the height of the trawls opening area, or by using a 130 131 flow meter (KC Denmark, Silkeborg, Denmark) that was attached to the inlet of the trawl. Half 132 the height of the trawl was chosen because the trawl was often not fully submerged into the water due to wave action. 133

134 *Filtering in situ pump*

The stainless steel *in situ* pump was designed and built by KC Denmark (Silkeborg, Denmark) 135 136 in collaboration with researchers from the EU CleanSea project (Grant no. 308370). The pump is made up of a motor on top, followed by an inlet grid for water, a filter stack with room for 137 138 three filters, and a flow meter section at the bottom measuring the sampled water with high precision (for more information see figure S1 in the SM). A stack of three laser cut stainless 139 steel filters, 18 cm in diameter and with mesh sizes of 500, 300 and 50 µm were inserted in the 140 pump before each sampling. Prior to use the filters were cleaned in the laboratory with 141 laboratory detergent and rinsed with ultrapure water. Additionally, each filter was investigated 142 with a stereomicroscope for contamination, wrapped in aluminum foil and placed into metal 143 jars with a lid until sampling. The maximum flow volume of the pump is 20,000 L/h. A digital 144 flow meter records the volume exiting the filter stack and the output can be read in real time 145 with a precision of the flow data of ± 1.8 L. 146

The total sampling time for the pump at different sites was between 23 and 138 minutes. The 147 148 sailing boat was drifting during the sampling. For most of the sampling points the 50 µm filter was removed after 838-3 794 liters due to clogging. The sampled water volume for the 300 µm 149 and 500 µm filters ranged from 1 046 to 20 022 liters. The pump with the filter stack was 150 assembled right before the sampling and was put into the water at the side of the boat by a 151 hydraulic lift and a spinnaker pole with the water intake at a depth of approximately 10-20152 cm below the water surface. After sampling, the filters were carefully removed from the pump 153 and stored up-right at room temperature in metal jars prior to transport to the laboratory (3-7 154 days). A 500 µm and a 300 µm filter were left standing open on deck of the vessel for the time 155 156 of pump sampling to serve as sampling blanks.

157 Identification of microplastic particles and fibers

Samples were stored at 4 °C until analysis. Large organic material like sea grass, feathers, small 158 fishes etc. were manually picked out from the samples with tweezers, rinsed with ultrapure 159 water to avoid loss of attached particles or fibers, and transferred into empty glass jars. The 160 161 trawl samples were rinsed down with ultrapure water onto 0.3 mm pre-cleaned stainless steel filters, same type as the metal filters that were used for the pump sampling (KC Denmark, 162 163 Silkeborg, Denmark), and subjected to visual examination by stereomicroscopy. In order to compare the pump and trawl results, the counts of the 300 µm and 500 µm pump filters were 164 summarized and reported as ≥ 0.3 mm. To improve and accelerate the visual analysis of the 165 0.05 mm pump filters, the material on the filters were rinsed down with ultrapure water into 166 glass jars and the content of the glass jars was filtered through glass fiber filters (0.2 µm, 167 Whatman). The glass fiber filters were transferred to glass petri dishes and closed with a lid. 168 The preceding procedures were conducted under the fume hood to minimize sample 169 contamination from the lab. All filters were visually examined with a stereomicroscope (Stemi 170 DRC Zeiss 25x magnification (10 ocular, 2.5 lens)). The visual examinations could not be 171

172 carried out under a fume hood, but to minimize sample contamination in this step, a lab coat
173 and nitrile gloves were worn at all time. One set of filters was left standing in the laboratory as
174 a laboratory blank sample and visually investigated with a stereomicroscope as done for the
175 samples.

To qualify as anthropogenic microlitter the particles had to show an absence of organic structure 176 such as cell walls. Synthetic fibers were separated from natural fibers by having an equal and 177 even thickness throughout the entire length and a homogenous coloring, whereas natural fibers 178 such as cotton were identified as flatter in their structure. Fibers were only counted if longer 179 than 1 mm and transparent fibers were excluded. The qualitative counting of anthropogenic 180 particles was performed and calibrated between two scientists in order to agree on a protocol 181 that resulted in satisfying results. The agreed protocol was similar to other protocols described 182 in the scientific literature (Hidalgo-Ruz et al. 2012). The microplastic particles were categorized 183 based on color into blue, white, black, other plastic particles (e.g. mixed color particles) and 184 other non-plastic particles. The shape of the particles was not noted. The particle and fiber 185 186 counts of all samples were corrected for sampling and laboratory blanks by subtraction.

Further plastic polymer identification of microplastic particles and fibers was done for all first
trawl samples at each sampling location using near-infrared hyperspectral imaging (Umbio
Inspector, Sisuchema Specim, Oulu, Finland) as previously described by Karlsson et al. (2016).
To eliminate background scattering of the metal filters, the particles and fibers were transferred
with tweezers into glass petri dishes and closed with a glass lid. The petri dishes were stored in
a 4 °C refrigerator until NIR hyperspectral image analysis.

193

194

196 Results and Discussion

197 Occurrence of microplastic particles and fibers

The microscopic examination of sampling blanks and laboratory blank showed that there was 198 only a small potential for contamination of the samples during the sampling and laboratory 199 procedure. On average we found 3 fibers and no particles in all blanks. The possible 200 201 contamination with fibers of the trawl samples by rinsing down the trawl with filtered seawater 202 was negligible because the used volume for rinsing was less than 0.01% of the sampled volume. The majority of trawl samples (88%) contained microplastic particles; only 3 out of 24 samples 203 had no microplastic particles. In pump samples with a mesh size of ≥ 0.3 mm 91% of the 204 samples contained microplastic particles. The median microplastic particle concentration per 205 cubic meter (m⁻³) surface water in manta trawl samples was 0.04 microplastics m⁻³ and for the 206 corresponding mesh size (≥ 0.3 mm) using the pump 0.10 microplastics m⁻³ (Table 1). For seven 207 of the locations an additional filter with a mesh size of 0.05 mm was used during the pump 208 209 sampling. The median concentration of microplastics in this size fraction of the pump was 3.74 particles m⁻³. The concentration of microplastic particles were in general, with exception of the 210 Kattegat sample, higher in the 50 – 300 μ m fraction compared to \geq 0.3 mm size fraction of the 211 pump. The median microplastic particle concentration in the 0.05 mm pump fraction was, 212 however, not significantly higher than the total median concentration in the ≥ 0.3 mm size 213 fraction of the pump, but significantly higher than the total median concentration sampled by 214 the manta trawl (Kruskal-Wallis test: p=0.0054). 215

The maximum abundance of microplastic particles between stations was not coincident for trawl and pump samples. In pump measurements ($\geq 0.3 \ \mu$ m) the highest abundances of microplastic particles were observed in the Skagerrak/Kattegat area, while in the trawl samples the highest particle concentrations were found in the southern Baltic Proper and the western Gotland Basin. In pump samples the location with the highest abundance of microplastics also

differed according to the filter size used. For instance, for the 0.05 mm size fraction the southern 221 Baltic Proper (sample ID 3) showed the highest concentration of microplastics m⁻³ in contrary 222 to the ≥ 0.3 mm fraction that was highest in Skagerrak and Kattegat. The replicate samples 223 taken with the trawl showed a high variation, which is quite characteristic for microplastic 224 pollution, but no significant differences in microplastic counts between locations were observed 225 for the trawl samples (Kruskal-Wallis: p < 0.05, followed by Dunn's multiple comparison test: 226 p > 0.05). This emphasizes the need for replication in future studies aimed at investigating 227 differences between microplastic concentrations. 228

Table 1. Microplastic particle counts and concentrations expressed as counts per cubic meters (m³) for twelve sampling sites, using two sampling methods and different mesh sizes.

		Trawl ^a (0.3 mm)		Pump (≥ 0.3 mm)		Pump (0.05 mm)	
ID	Site	Particle count	Concentration	Particle count	Concentration	Particle count	Concentration
1	Skagerrak	3 9	0.02 0.05	8	2.59	n.s.	n.s.
2	Kattegat	2 4	0.01 0.02	11	10.5	4	3.82
3	Southern Baltic Proper	6 9	0.03 0.04	2	1.47	10	11.9
4	Southern Baltic Proper	20 29	0.16 0.16	2	0.07	n.s.	n.s.
5	Western Gotland Basin	86 24	0.46 0.13	n.s.	n.s.	n.s	n.s.
6	Bothnian Sea	5 6	0.12 0.04	0	0	0	0
7	The Quark	7 4	0.04 0.02	1	0.10	3	1.45
8	Bothnian Sea	7 9	0.05 0.05	5	0.49	n.s.	n.s.
9	Bothnian Sea	9 7	0.06 0.03	2	0.05	5	1.32
10	Northern Baltic Proper	0 1	0 0.01	2	0.10	11	8.80
11	Eastern Gotland Basin	1 0	0 0	1	0.05	n.s.	n.s.
12	Northern Baltic Proper	2 3	0.01 0.04	1	0.05	116	70.3
	Median Quartiles (1 st ; 3 rd)	6 2.75; 9	0.04 0.02; 0.06	2 1; 3.50	0.10 0.06; 0.98	5 3.50; 10.5	3.82 1.38; 10.4

231 n.s.: no sample was taken, ^a two trawl samples per site were taken

Median concentration of fibers, including natural and synthetic fibers, was 0.35 fibers m⁻³ in 232 the manta trawl samples, 2.74 fibers m⁻³ for the pump samples with a filter size of ≥ 0.3 mm, 233 and 50.4 fibers m⁻³ in 0.05 mm pump samples (Table S3 of the SM). The total median 234 concentration of fibers m⁻³ was significantly higher in pump samples for both mesh sizes 235 compared to the trawl (Kruskal-Wallis: p > 0.001). Although it has to be kept in mind that the 236 larger mesh sizes (≥ 0.3 mm) do not representatively sample fibers due to the small diameters 237 238 of fibers and attachment to biological material, therefore the data might not be completely reliable. 239

The locations with the highest amount of microplastics m⁻³ in pump samples also matched the 240 highest amount of fibers m⁻³ in pump samples (sample ID 1, 2, and 3), regardless of the used 241 mesh size. In the trawl samples the location with the highest abundance of fibers m⁻³ differed 242 from the location with highest microplastic abundance. The Bothnian Sea (sample ID 8) and 243 the Skagerrak (sample ID 1) had the highest amount of fibers m⁻³ in trawl samples. The fiber 244 counts in the trawl samples did not differ significantly among the sites (Kruskal-Wallis: p = 245 246 0.1769). The majority of fibers in pump samples and trawl samples were categorized as synthetic fibers by visual examination with the stereomicroscope (see figure S2, S3, and S4 of 247 the SM). In the 0.05 mm pump samples most fibers were synthetic and less than 15 % were 248 249 identified as natural. In one sample, at Kattegat, a slightly higher percentage (27%) of the fibers was identified as natural fibers (Figure S2). The amount of natural fibers in the samples is, 250 however, likely to be an underestimation because translucent fibers were not counted, and a lot 251 of natural fibers appear translucent. Most of the identified synthetic fibers in all samples were 252 253 black or blue, which can indicate ropes as a potential source of these fibers because these colors 254 are very common for boat ropes and fishing gear when comparing to sales items in marine stores. 255

The wind speed varied throughout the sampling period and a decline of microplastic particles in the trawl measurements could be observed with increasing wind speed. A significant negative correlation was found between the wind speed and the abundance of microplastic particles (Spearman correlation: p = 0.0021 (two-tailed); r = -0.60) (Figure 2).



Figure 2. Concentration of microplastic particles (count per m⁻³) in trawl samples plotted against
wind speed (m s⁻¹) at sample location and time. A linear trend line is inserted for visualization
of the negative trend.

264

260

265 *Microplastic characterization in trawl samples*

Particles that were identified as microplastics by visual inspection, as well as ambiguous particles, were analyzed further using near-infrared hyperspectral imaging to identify the polymer type. Microplastic particles were classified as PE, PP, PS, and polyamide (PA) or unidentified based on calibration with pristine plastic pellets of the respective polymer type as a reference material (Karlsson et al. 2016). A total of 137 particles were analyzed of which 8 particles (6%) could not be designated to a certain polymer type (unidentified polymer). The

majority of particles consisted of PE plastics (65 %) followed by PP plastics (21 %), which is 272 273 in line with other studies that have reported PE and PP plastics as the main plastic types in trawl samples (Gewert et al. 2017; Hidalgo-Ruz et al. 2012). A higher abundance of PE and PP 274 plastics has been reported also in stratified water samples (Zobkov et al. 2019) and in different 275 fish species from the Baltic Sea (Rummel et al. 2016). The spectral quality for some of the 276 particles did not allow for separation of the polymers PP and PE (PP/PE). The composition of 277 identified plastic polymers varied among the sampling locations (Figure 3). For instance, PS 278 was only found in two out of eleven trawl samples and no polyamide was found in any of the 279 samples. However, not all of the plastic particles were identified in each sample; therefore the 280 281 composition might not be directly comparable and is likely not representative for all of the samples. The number of particles for each polymer type were generally less than 10 per sample, 282 except for two samples, which also hampers further statistical evaluation as described by 283 284 Karlsson et al. (2020).



Figure 3. Identified microplastic particles from eleven individual trawl samples divided intopolymer classification (% of total).

288 Comparison of sampling devices

Although the concentration varies between different locations, the low particle counts per 289 sample, and the variation in sampled volumes make a direct comparison of the sampling 290 291 methods difficult, the pump (≥ 0.3 mm) resulted in notably higher concentrations than the trawl in 4 locations; Skagerrak, Kattegat, the Southern Baltic Proper (ID 3) and the Bothnian Sea (ID 292 8). Overall, the concentration of microplastic particles was higher in pump samples compared 293 to trawl samples in ten out of eleven sampling sites (Figure 4). In another study which compared 294 a manta trawl (333 µm) and a submerged pump (300 µm) for sampling of microlitter in the Gulf 295 of Finland, the results obtained by both devices were similar (Setälä et al. 2016). The sampling 296 297 duration was much shorter (10 min for the manta trawl) compared to the present study, and thus also the sampled water volume (10 - 139 L for the pump) was less compared to the herein 298 sampled volume (1046 - 20022 L for the pump). In the present study the highest difference in 299 microplastic concentration comparing a pump sample and the average of two trawl samples 300 from the same sampling site was 700 fold (Kattegat). The higher abundance of microplastic 301 302 particles in the Skagerrak, Kattegat and Southern Baltic Proper area in the present study were, 303 however, not exceeding reported concentrations from other studies in these areas (Bagaev et al. 2018; Norén et al. 2009). Besides a small difference in mesh size, the trawl skims the water 304 305 surface covering a larger area compared to the pump which is stationary submerged into the water surface with only a small drift during the sampling time. Therefore, heterogeneous 306 307 distribution of microplastic pollution would be better captured using the trawl method. Interestingly, the water volume sampled by the trawl was on average 180 m³ compared to the 308 average volume of 13 m³ for the pump. This is a difference of a factor of fourteen between the 309 310 sampled volumes. However, this was not reflected in microplastic counts in the samples. The amount of counted microplastic particles in trawl samples was in general less than fourteen fold 311 greater compared to pump samples. The higher volume of sampled water by trawl did not lead 312

to a higher concentration of plastic particles compared to pump samples. It has to be noted that 313 314 there is a greater uncertainty when estimating the volume sampled by the trawl compared to the volume sampled by the pump, due to the differences in submersion. Depending on the wave 315 action the trawl was not consistently submerged at the same height of its frame, this leads to a 316 greater uncertainty in the calculation of the sampled volume. Hence, the actual volume sampled 317 by the trawl is likely to be smaller than the calculated one. Therefore, the microplastic 318 319 concentrations of trawl samples might be more similar to the concentrations calculated for the pump samples. Nonetheless, both sampling devices are more suitable for surface sampling of 320 microplastics under relatively calm weather conditions. With greater wave action the trawl 321 322 tends to bounce on the water surface. Whereas a problem with the pump is that air can get sucked in when the waves are higher, which negatively affects the certainty of the sampled 323 volume. The relatively heavy weight of the device itself might be a disadvantage compared to 324 325 the trawl; two people were necessary to lift the pump into the water. The handling of the samples and the sampling itself is quite convenient for both sampling devices. Although a blank for the 326 327 trawl sampling was not taken, there might be a slightly higher risk for contamination when using the manta trawl because the rinsing procedure and transferring the sampled material from 328 329 the collecting bag into a container after sampling takes a bit longer than taking out the set of 330 filters from the pump. Another advantage of the pump is that it can be used to sample in varying depths and simultaneously collecting several size fractions, which is not possible with the manta 331 trawl. Further comparison of the two sampling devices under more controlled conditions are 332 333 presented elsewhere (Karlsson et al. 2020).



Figure 4. Number of microplastic particles per cubic meter for the trawl (n=2, 0.3 mm) and pump (n=1, \geq 0.3 mm) samples for twelve sampling sites. The mean values for the trawl samples (n = 2) from each site are presented with the standard deviations given as error bars. No pump sample was taken at Western Gotland basin. Numbers in italic shows results out of scale.

339

334

340 *Comparison to other studies*

341 Although the Baltic Sea has been declared as one of the most polluted seas in the world 342 (HELCOM 2010), there are not many studies conducted so far on microplastic pollution in the waters of the Baltic Sea that cover a large area within one sampling campaign. To date there 343 have been two studies that sampled over a larger area of the Baltic, similar to the area presented 344 here (Bagaev et al. 2018; Norén et al. 2009), whereas other studies focused on specific parts of 345 346 the Baltic Sea (Gewert et al. 2017; Gorokhova 2015; Setälä et al. 2016; Zobkov et al. 2019). The sampling techniques differed greatly among the above mentioned studies which makes a 347 direct comparison difficult and results need to be interpreted carefully. However, a comparison 348

between the conducted studies in open water of different regions of the Baltic Sea shows a wide 349 350 range of concentrations of reported microplastic particles ranging from 0.012 microplastic particles m⁻³ in trawl samples (≥ 0.3 mm) of the present study to 20,280 microplastic particles 351 m⁻³ in pump samples (20 µm mesh size) (Norén et al. 2009) (Figure 5). It has to be noted that 352 in contrary to the present study, the study of Norén et al. (2009) and Setälä et al. (2016) also 353 included black combustion particles which were the most abundant among the counted particles 354 355 in some of the samples. It is notable that with decreasing mesh size the abundance of microplastic particles increases significantly. This effect has also been reported in other studies, 356 and the present study corroborates the need for integrating smaller mesh sizes into the sampling 357 358 regimen of microplastics. Especially fibers will slip through mesh sizes that are currently in use, due to their small diameter (um-nm range) and shortness. Fibers are also more likely to 359 adhere to, for example, biological material and therefore might not be sampled representatively. 360 361 By use of a smaller mesh size a higher degree of accurate quantification of fibers is possible. An important point to consider when using smaller mesh sizes is the general composition in the 362 water phase that should be sampled. If the water phase contains a large amount of organic 363 material, a filter or net with a smaller mesh size will rapidly become clogged. In fact, this could 364 365 be observed in the present study when utilizing a 0.05 mm filter. The volume which was filtered 366 with the 0.05 mm filter was always less than the volume sampled with a \ge 0.3 mm filter due to fast clogging of the 0.05 mm filter. However, other factors than the mesh size influence the 367 detected concentrations as well, and abundances can vary several orders of magnitude even by 368 369 using the same mesh size and sampling technique (see figure 5).



Figure 5. Concentration of microplastic particles per cubic meter in surface or near surface open water in Skagerrak and the Baltic Sea presented on a logarithmic scale, reported by different studies using different mesh sizes and techniques. Striped bars represent samples that were obtained by pump sampling. a: this study; trawl samples are presented as mean values of two replicates, b: Norén et al. (2009), c: Bagaev et al. (2018), d: Gorokhova (2015), e: Setälä et al. (2016).

In a study by Gewert et al. (2017), which only focused on the Stockholm archipelago, an overall 377 median concentration of 0.6 microplastic particles m⁻³ has been reported by manta trawl 378 sampling, which is an order of magnitude greater than the median concentration of 0.04 379 microplastic particles m⁻³ found in our study by sampling with a manta trawl. The highest 380 concentrations of 7.73 and 4.93 microplastic particles m⁻³ were detected in direct proximity to 381 the city of Stockholm (Gewert et al. 2017). It should be noted that several studies have observed 382 383 that microplastic concentrations increase with decreasing distance to urban areas with pollution sources such as industry and wastewater treatment plants (TM Karlsson et al. 2018; Magnusson 384 and Norén 2014; Talvitie et al. 2015). 385

The patchiness of microplastic distribution was reflected in some replicate trawl samples from 386 387 the same site in the present study. The greatest difference with a factor of four was observed in trawl samples from the Northern Baltic Proper (ID 12). Wind, for example, has been observed 388 as an important variable for surface water sampling due to wind-induced mixing (Kukulka et 389 al. 2012) as well as currents. The inherent variation in water conditions most likely results in 390 large temporal and spatial differences in the abundance and distribution of microplastics. 391 Standardized protocols for sampling and analysis are needed as well as studies aiming at 392 assessing the microplastic concentration baseline variations. 393

394

395 Conclusions

The data reported in this study confirms that microplastic contamination is ubiquitous in 396 397 Swedish waters. It also indicates a higher accumulation on the Swedish west coast, which is known to be particularly affected by macrolitter. The present study highlights the importance 398 of using standardized methodologies in order to achieve comparable data, since the results 399 400 differed between sampling devices. Overall, the pump sampling resulted in higher detected concentrations of microplastic particles and fibers than the manta trawl sampling. It was also 401 noted that the number of detected particles and fibers increased by use of a smaller mesh size. 402 The patchiness associated with microplastic pollution is an urgent methodological challenge 403 that needs to be addressed in future scientific studies in order to allow for the assessment of 404 405 temporal and spatial trends.

406

407 Acknowledgements

This study was financed by the Swedish Environmental Protection Agency and by the Swedish
Research Council for Environment, Agricultural Sciences and Spatial Planning (C. Schönlau,

223-2014-1064). The development of the pump was conducted under the CleanSea project
European Union Seventh Framework Programme (FP7/2007–2013), grant agreement no.
308370. We would like to thank KC Denmark for technical assistance and Dr Marcus Eriksen,
5Gyres for use of the manta trawl. Pangaea Exploration and Emily Penn is acknowledged for
help during planning and execution of the sailing expedition. Lastly we are grateful to all crew
members onboard 'Sea Dragon' that participated in the Baltic Sea Expedition 2014, especially
Eric Loss and Shanley McEntee.

417

418 **References**

- Andrady AL, Neal MA. 2009. Applications and societal benefits of plastics. Philos T R Soc B
 364:1977-1984.
- 421 Andrady AL. 2011. Microplastics in the marine environment. Mar Pollut Bull 62:1596-1605.
- Bagaev A, Khatmullina L, Chubarenko I. 2018. Anthropogenic microlitter in the baltic sea
 water column. Mar Pollut Bull 129:918-923.
- Barnes DKA, Galgani F, Thompson RC, Barlaz M. 2009. Accumulation and fragmentation of
 plastic debris in global environments. Philos T R Soc B 364:1985-1998.
- 426 Cadée GC. 2002. Seabirds and floating plastic debris. Mar Pollut Bull 44:1294-1295.
- 427 Carpenter EJ, Smith KL, Jr. 1972. Plastics on the sargasso sea surface. Science 175:1240428 1241.
- Cozar A, Echevarria F, Gonzalez-Gordillo JI, Irigoien X, Ubeda B, Hernandez-Leon S, et al.
 2014. Plastic debris in the open ocean. P Natl Acad Sci USA 111:10239-10244.
- 431 Eriksen M, Lebreton LCM, Carson HS, Thiel M, Moore CJ, Borerro JC, et al. 2014. Plastic
- pollution in the world's oceans: More than 5 trillion plastic pieces weighing over 250,000 tonsafloat at sea. Plos One 9.
- Esiukova E. 2017. Plastic pollution on the baltic beaches of kaliningrad region, russia. Mar
 Pollut Bull 114:1072-1080.
- EU. 2008. Directive 2008/56/ec of the european parliament (marine strategy framework
 directive).
- 438 Gewert B, Plassmann MM, MacLeod M. 2015. Pathways for degradation of plastic polymers
- floating in the marine environment. Environmental Science: Processes & Impacts 17:1513-1521.

- 441 Gewert B, Ogonowski M, Barth A, MacLeod M. 2017. Abundance and composition of near
- surface microplastics and plastic debris in the stockholm archipelago, baltic sea. Mar Pollut
 Bull 120:292-302.
- 444 Gorokhova E. 2015. Screening for microplastic particles in plankton samples: How to
- integrate marine litter assessment into existing monitoring programs? Mar Pollut Bull 99:271-275.
- 447 Hartmann NB, Hüffer T, Thompson RC, Hassellöv M, Verschoor A, Daugaard AE, et al.
- 448 2019. Are we speaking the same language? Recommendations for a definition and
- 449 categorization framework for plastic debris. Environ Sci Technol 53:1039-1047.
- 450 HELCOM. 2010. Hazardous substances in the baltic sea. An integrated thematic assessment
 451 of hazardous substances in the baltic sea. Baltic sea environment proceedings no.120b.
- 452 HELCOM. 2018. Sate of the baltic sea second helcom holistic assessment 2011-2016. Baltic
 453 sea environment proceedings 155. Issn 0357-2994.
- 454 Hengstmann E, Tamminga M, vom Bruch C, Fischer EK. 2018. Microplastic in beach
- 455 sediments of the isle of rügen (baltic sea) implementing a novel glass elutriation column.
 456 Mar Pollut Bull 126:263-274.
- 457 Hidalgo-Ruz V, Gutow L, Thompson RC, Thiel M. 2012. Microplastics in the marine
- 458 environment: A review of the methods used for identification and quantification. Environ Sci
 459 Technol 46:3060-3075.
- Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, et al. 2015. Plastic
 waste inputs from land into the ocean. Science 347:768-771.
- Karlsson TM, Kärrman A, Rotander A, Hassellöv M. 2020. Comparison between manta trawl
- and in situ pump filtration methods, and guidance for visual identification of microplastics insurface waters. Environmental Science and Pollution Research 27, 5559-5571.
- 465 Karlsson TM, Grahn H, van Bavel B, Geladi P. 2016. Hyperspectral imaging and data
- 466 analysis for detecting and determining plastic contamination in seawater filtrates. J near
- 467 Infrared Spec 24:141-149.
- Karlsson TM, Arneborg L, Broström G, Carney Almroth B, Gipperth L, Hassellöv M. 2018.
 The unaccountability case of plastic pellet pollution. Mar Pollut Bull 129:52-60.
- 470 Koelmans AA, Bakir A, Burton GA, Janssen CR. 2016. Microplastic as a vector for chemicals
- in the aquatic environment: Critical review and model-supported reinterpretation of empirical
 studies. Environ Sci Technol 50:3315-3326.
- Kukulka T, Proskurowski G, Moret-Ferguson S, Meyer DW, Law KL. 2012. The effect of
 wind mixing on the vertical distribution of buoyant plastic debris. Geophys Res Lett 39.
- 475 Laist DW. 1997. Impacts of marine debris: Entanglement of marine life in debris including a
- 476 comprehensive list of species with entanglement and ingestion records. Marine Debris -
- 477 Sources, Impacts and Solutions Springer Series on Environmental Management 99-139.
- Li J, Liu H, Paul Chen J. 2018. Microplastics in freshwater systems: A review on occurrence,
 environmental effects, and methods for microplastics detection. Water Research 137:362-374.
- 480 Magnusson K, Norén F. 2014. Screening of microplastic particles in and down-stream a
 481 wastewater treatment plant. (IVL Report C).
- 482 www.divaportal.org/smash/get/diva2:773505/FULLTEXT01.pdf [accessed 2019-03-01].

- Mascarenhas R, Santos R, Zeppelini D. 2004. Plastic debris ingestion by sea turtle in paraiba,
 brazil. Mar Pollut Bull 49:354-355.
- Mato Y, Isobe T, Takada H, Kanehiro H, Ohtake C, Kaminuma T. 2001. Plastic resin pellets
 as a transport medium for toxic chemicals in the marine environment. Environ Sci Technol
 35:318-324.
- 488 Norén F, Ekendahl S, Johansson U. 2009. Mikroskopiska antropogena partiklar i svenska hav.
 489 www.diva-portal.org/smash/get/diva2:717410/FULLTEXT01.pdf [accessed 2019-04-03].
- 490 Näkki P, Setälä O, Lehtiniemi M. 2019. Seafloor sediments as microplastic sinks in the
- 491 northern baltic sea negligible upward transport of buried microplastics by bioturbation.
- 492 Environ Pollut 249:74-81.
- 493 PlasticsEurope. 2018. Plastics-the facts 2018. An analysis of european plastics production,
- demand and waste data. Plasticseurope (association of plastics manufacturers).
- $\label{eq:https://www.plasticseuropeorg/en/resources/publications/619-plastics-facts-2018.$
- 496 Rummel CD, Löder MGJ, Fricke NF, Lang T, Griebeler E-M, Janke M, et al. 2016. Plastic
- 497 ingestion by pelagic and demersal fish from the north sea and baltic sea. Mar Pollut Bull498 102:134-141.
- Setälä O, Magnusson K, Lehtiniemi M, Noren F. 2016. Distribution and abundance of surface
 water microlitter in the baltic sea: A comparison of two sampling methods. Mar Pollut Bull
 110:177-183.
- Stolte A, Forster S, Gerdts G, Schubert H. 2015. Microplastic concentrations in beach
 sediments along the german baltic coast. Mar Pollut Bull 99:216-229.
- Talvitie J, Heinonen M, Pääkkönen J-P, Vahtera E, Mikola A, Setälä O, et al. 2015. Do
- wastewater treatment plants act as a potential point source of microplastics? Preliminary study
 in the coastal gulf of finland, baltic sea. Water Science and Technology 72:1495-1504.
- Teuten EL, Rowland SJ, Galloway TS, Thompson RC. 2007. Potential for plastics to transport
 hydrophobic contaminants. Environ Sci Technol 41:7759-7764.
- 509 Teuten EL, Saquing JM, Knappe DRU, Barlaz MA, Jonsson S, Bjorn A, et al. 2009.
- 510 Transport and release of chemicals from plastics to the environment and to wildlife. Philos T
- 511 R Soc B 364:2027-2045.
- 512 Ziccardi LM, Edgington A, Hentz K, Kulacki KJ, Driscoll SK. 2016. Microplastics as vectors
- for bioaccumulation of hydrophobic organic chemicals in the marine environment: A state-of-
- the-science review. Environ Toxicol Chem 35:1667-1676.
- 515 Zobkov MB, Esiukova EE, Zyubin AY, Samusev IG. 2019. Microplastic content variation in
- 516 water column: The observations employing a novel sampling tool in stratified baltic sea. Mar
- 517 Pollut Bull 138:193-205.
- 518