Abundance and composition of near surface microplastics and plastic debris in the Stockholm Archipelago, Baltic Sea

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ABSTRACT

We collected plastic debris in the Stockholm Archipelago using a manta trawl, and additionally along a transect in the Baltic Sea from the island of Gotland to Stockholm in a citizen science study. The samples were concentrated by filtration and organic material was digested using hydrogen peroxide. Suspected plastic material was isolated by visual sorting and 59 of these were selected to be characterized with Fourier transform infrared spectroscopy. Polypropylene and polyethylene were the most abundant plastics identified among the samples (53% and 24% respectively). We found nearly ten times higher abundance of plastics near central Stockholm than in offshore areas (4.2×10^5 plastics km^{-2} compared to 4.7×10^4 plastics km^{-2}). The abundance of plastic debris near Stockholm was similar to urban areas in California, USA, and the overall abundance in the Stockholm Archipelago was similar to plastic abundance reported in the northwestern Mediterranean Sea.

1. Introduction

Worldwide plastic production has increased from 5 million tons in 1950 to 322 million tons in 2015 (PlasticsEurope, 2016), and plastic debris is now ubiquitous in aquatic environments (Barnes et al., 2009). Many sources contribute to the burden of plastic pollution in marine waters. Major inputs of plastic litter from land-based sources are known to come from densely populated or industrialized areas (Gregory, 1991; Jambeck et al., 2015; Pruter, 1987), with landfills and tourism being important contributors (UNEP, 2005). Furthermore it has been shown that the effluent water of wastewater treatment plants (WWTPs) contain plastic, mainly in the form of synthetic fibers from clothing (Browne et al., 2011; Magnusson and Norén, 2014) and play a critical role in the fate and transport of microfibers in the environment (Napper and Thompson, 2016). Sea-based sources of marine litter include ships and vessels, offshore oil and gas platforms and aquaculture installations (UNEP, 2005).

Plastic litter in the environment is usually classified into different size fractions. Plastic particles exceeding 5 mm in diameter are called macroplastic (Moore, 2008). Particles < 5 mm in diameter are called microplastics and can be further distinguished between primary microplastic particles that are produced in that size range, and secondary microplastic particles formed by fragmentation of larger plastic debris (Cole et al., 2011; Derraik, 2002; Moore, 2008; Ryan et al., 2009).

Floating plastic debris in the marine environment can be degraded by reactions initiated by UV-radiation, hydrolysis and microorganisms (Gewert et al., 2015). However, in general plastic is highly durable and tends to accumulate in the environment (Barnes et al., 2009). The buoyancy of plastic debris depends on the composition, density and shape of the plastic particle, among other factors (Derraik, 2002). Plastic used in common consumer items can be buoyant or prone to sinking (Morét-Ferguson et al., 2010), but the majority of plastic debris is buoyant (Derraik, 2002). Biofilms formed on plastic particles may further modify the density of plastics making initially buoyant particles heavier and more prone to sinking (Cozar et al., 2014; Fazey and Ryan, 2016; Gorokhova, 2015; Ye and Andrady, 1991) but also more attractive for ingestion by animals such as zooplankton (Nerland et al., 2014).

Microplastics have recently become a cause for concern as ingestion has been observed in a wide range of taxa spanning from zooplankton to mammals (Browne et al., 2008; Cole et al., 2013; Derraik, 2002; Laist, 1987; Murray and Cowie, 2011; Setälä et al., 2016b, 2014; Thompson, 2004; Van Cauwenberghe et al., 2015; Wright et al., 2013). Although the effects of this exposure are not yet well understood (GESAMP, 2015), the hazard posed by microplastics has inspired initiatives aimed at reduction of plastic litter in marine environments (Moore, 2008; Storrier and McGlashan, 2006), and plastic in the global oceans was recently identified as a potential planetary boundary threat (Jahnke et al., 2017).

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The European Union's Marine Strategy Framework Directive (European Commission, 2008) sets the conditions required for European member states to achieve "good environmental status" by the end of 2020. Descriptor 10 of the directive, marine litter, states that good environmental status can only be reached when "the properties and quantities of marine litter do not cause harm to the coastal and marine environment". To reach good environmental status and to monitor future changes in the environment, it is imperative to assess the current abundance and characteristics of plastic debris in European marine waters. Although some European countries already have initiated monitoring campaigns, their efforts have mainly focused on microplastics in sediments (Claessens et al., 2011; Imhof et al., 2013; Van Cauwenberghe et al., 2013; Vianello et al., 2013) or on beaches (Ryan et al., 2009) and relatively little information regarding floating plastic debris exists from European waters.

The Baltic Sea is a semi-enclosed intracontinental sea of about 4.2×10^5 km^2 that is particularly vulnerable to pollution because of restricted water exchange with the North Sea (Feistel et al., 2008). Consequently, the Baltic Sea has often been referred to as the most polluted sea globally (HELCOM, 2010). The Swedish capital, Stockholm, is one of the largest urban areas in the Baltic Sea drainage basin, with about 900,000 inhabitants in the city and a population of about 2 million in the urbanized area (Statistics Sweden, 2016). The city of Stockholm lies within the Stockholm Archipelago, which comprises about 30,000 islands and small islets (Eriksson et al., 2004). The Archipelago is one of the top tourist destinations in Sweden with about 1,400,000 visitors in 2011 (Stockholm Visitors Board, 2011). This area is the most popular location for summer homes in Sweden (Marjavaara, 2007). In addition to being a tourist attraction and vacation area, the Stockholm Archipelago is the largest receiver of treated sewage in Sweden (Rosenberg and Diaz, 1993) and has intense ship traffic (Engqvist and Andrejev, 2003; HELCOM AIS, 2017).

Although the Baltic Sea is one of the most thoroughly studied seas in the world (Feistel et al., 2008), there are only a few studies reporting on the concentration and spatial distribution of microplastics in the Baltic Sea (Gorokhova, 2015; Magnusson, 2014; Magnusson et al., 2016; Magnusson and Norén, 2011; Norén, 2007; Norén et al., 2014, 2015; Setälä et al., 2016a; Talvitie et al., 2015). But the applied methods, i.e. samples depth, mesh size, sampling area etc. vary broadly. Some of the studies focus on specific point sources of plastic debris, which is not representative for the Baltic Sea. Hence, our knowledge regarding the distribution, abundance and composition of floating marine plastic debris in the Baltic is still limited.

In this paper we report the concentrations and compositions of floating plastic debris collected with two different types of trawls in the Stockholm Archipelago and the open Baltic Sea. Twenty-one samples were collected with a manta trawl towed behind a boat, and cover an anthropogenic disturbance gradient spanning from highly urbanized coastal areas, including harbors and a WWTP, to remote areas in the outer Stockholm Archipelago. We also report on plastics collected in four additional samples using custom-built trawls that were towed behind stand-up paddleboards by two citizen scientist outdoor adventurers who were seeking to raise awareness of pollution of the Baltic Sea by plastics.

2. Materials and methods

2.1. Sampling and study area

Our primary sampling campaign included 21 surface water samples and was conducted on the 24th (samples 1–4) and 25th (samples 5–9) of June and 7th (samples 10–12), 9th (samples 13–17) and 11th (samples 18–21) of July 2014 in the Stockholm Archipelago (Sweden). We collected samples in areas close to Stockholm city (samples 10–12), Nynäshamn (a larger commercial harbor) (samples 18–20), Trosa (a town with high leisure boat activity) (sample 21) and

Himmerfjärdsverket (sample 17), a wastewater treatment plant south of Stockholm which handles the wastewater of approximately 350,000 people (SYVAB, 2016). These areas were chosen because they are potentially important sources for plastic pollution to the marine environment and represent different types of activities. Hence, the concentration of plastics was expected to be high relative to more remote areas of the Stockholm Archipelago that were also sampled.

After our primary sampling campaign, in the summer of 2014, we were contacted by two Swedish adventurers who were planning to attempt to cross the Baltic Sea from Visby on the Swedish island of Gotland to Stockholm on standup paddleboards. The adventurers volunteered to collect surface water samples during their 210 km open water crossing to raise public awareness of plastic pollution in the Baltic Sea. We designed special lightweight trawls that could be deployed on the water surface behind the paddleboards with an 80 μm net with a round opening of 10 cm in diameter. The trawls were tested in a flume experiment in the laboratory to optimize the distance between the paddleboard and the trawl to achieve efficient sampling. Prior to their trip the adventurers received several of the custom trawls and were trained in sampling techniques and sample handling. They collected four samples from their paddleboards during their 10-day unaccompanied crossing, which occurred between the 4th and 14th of June 2015. These samples were sent to our laboratory and were separated and analyzed using the same technique as the samples collected during our primary sampling campaign.

2.2. Sampling method

Samples for the primary study were collected using a manta trawl (obtained from Marcus Eriksen, 5 Gyres institute) towed outside of the wake zone approximately 35 m behind a research vessel, harbored at the Askö Research Station in the southern Stockholm Archipelago. The 61×16 cm rectangular opening of the manta trawl was connected to a 4 m long net with a standard mesh size of 335 μm and a 30×10 cm^2 collecting bag (c.f. Hidalgo-Ruz et al., 2012; Zampoukas et al., 2010).

During sampling, the boat was traveling at a speed of 2–3 knots and its position was logged using a Garmin GPSMAP 78 GPS device (Garmin, USA). The duration of sample collection varied between 12 and 60 min (Table 1 in Supporting information) due to different surrounding factors (e.g. intense boat traffic). After each sampling event, the whole net was rinsed thoroughly with seawater from the outside starting from the opening towards the collecting bag to ensure that all the plastic debris would be collected. Then the collecting bag was deployed and rinsed with about 6–10 L of seawater and the collected samples were stored in 12 L hard plastic containers. On land, the water volume of each sample was reduced by filtering the sample through a 110 μm mesh. Then the mesh was rinsed with tap water and the sample was collected in a smaller 0.5 L container of hard plastic. The total of 21 samples were stored at 4 °C in darkness to minimize algal growth until analysis.

2.3. Separation methods

Several steps were conducted to separate the plastic particles from the biological matrix and water, using a method based on that described by Mason et al. (2016a) (Fig. 1). To avoid contamination of the samples with airborne fibers and other material, the separation was conducted under a fume hood. First larger pieces of biological material, including e.g. leaves, bugs, larger algae, wood, were picked out of the samples with tweezers and were carefully rinsed with water, which was collected back into the container to avoid loss of microplastics. Larger plastic debris was picked out and rinsed in the same way, but instead of discarding it, we counted and stored the plastic for further analysis. Then the samples were stored in the dark until denser particles had settled at the bottom of the container. Afterwards, the supernatant was filtered using a vacuum pump through a glass fiber filter (GF/F;
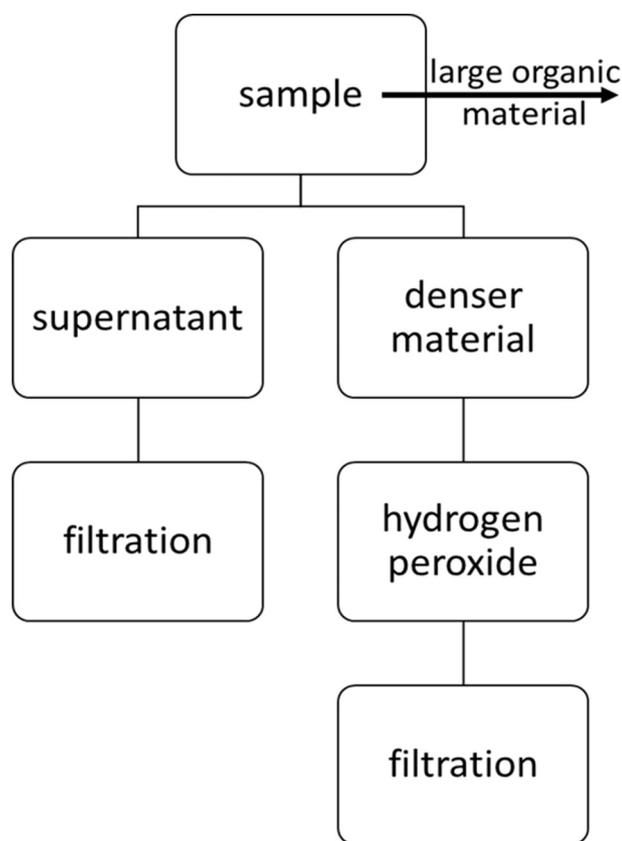
Fig. 1. Schematic overview of the separation process for isolating plastic from trawl samples.

0.75 μm; 47 mm in diameter; Whatman, Maidstone, Kent, U.K.). During this step, as much water as possible was slowly decanted to avoid pouring large non-plastic debris such as small leaves and algae onto the filter. Hydrogen peroxide (33% solution) was added to the remaining water sample containing denser material, at 1:1 volume ratio to oxidize and digest the biological material, according to methods described by Imhof et al. (2012) and Nuelle et al. (2014). The treatment of the samples continued until all visible organic material was digested or up to a maximum of 24 h. However, to not risk potential degradation of the plastics, the hydrogen peroxide concentration was lower (compared with 30% hydrogen peroxide) and the exposure time was shorter (compared with one week) than in the study conducted by Nuelle et al. (2014) and therefore not all organic material was removed within our samples. The mixture was then poured onto new glass fiber filters until a thin layer of biological and plastic debris was formed on each filter.

Subsequently all the filters were examined visually using a stereo dissecting microscope (Leica MZ6 with 32 × magnification). Particles that were visually identified as plastic were picked out by thin, pointed tweezers to a petri dish and counted. It was always double checked that only the selected plastic was transferred and no other plastic stuck to it in order to avoid an underestimation of the plastic amount. The use of hydrogen peroxide removed and bleached the organic matter which made the plastic easier to detect visually. Criteria for the visual identification of the plastic were based on other publications (Hidalgo-Ruz et al., 2012; Norén, 2007) and were: color, particularly bright and unnatural colors and same color over the whole length of a particle or fiber; no organic structures; and uniform diameter over the whole length of a fiber. Furthermore, it was possible to identify plastics by feel and texture with the tweezers. Whereas algae were very soft and tended to break easily, the plastic pieces were stiffer and did not break easily when pinched with tweezers.

2.4. Characteristics of the collected plastic

In order to characterize the plastics in terms of fiber lengths and widths, fragment sizes and color, we photographed the sample filters using a Leica DMC2900 camera coupled to a microscope (Leica WILD M10) with 20 x magnification. One area of each sample containing the most plastic pieces was chosen to be photographed. For the three samples collected in Stockholm, we additionally took pictures with a camera (Canon EOS 5D Mark III with a macro 100 mm lens) because the magnification of the microscope was too large to capture the macroplastic in these samples. Size measurements of individual fibers and fragments of plastic were made using the Fiji image analysis software (Schindelin et al., 2012). In total 248 fibers and 29 fragments were measured. The shape of the fragments varied widely and therefore the length of the short and the long side was measured and an average was calculated to characterize their size.

2.5. Plastic recovery and quality assurance

We tested for potential plastic contamination from equipment and clothing by performing a control experiment in the laboratory. To replicate conditions during the sampling we wore the same clothes as were used in the field. First we ensured that the tap water did not contain any particulates < 2 μm (measured with a Spectrex PC-2000 laser particle counter, Redwood City, USA). Then the cod-end of the trawl was rinsed into the 12 L containers. To simulate wind during the sampling, we briefly shook the sleeves of the clothes over the containers. To simulate the transport on board we placed the hard plastic containers on a shaking table for 4 h. The three blank samples were then analyzed as the other samples.

During the filtration and the visual identification, preventive measures were taken to avoid potential background contamination. This included a clean working space (in a fume hood), working with nitrile gloves and lab coats. The petri dishes for collecting the plastics were closed at all times.

Furthermore, we conducted a quality assurance test (QA) in order to obtain the recovery rate of our separation process and check for any contamination. Three replicates of plastic-spiked water samples were prepared, each with 10 pieces of plastic particles and 15 fibers. These included polyethylene pellets and fibers purchased from Goodfellow GmbH, Germany and a selection of ground common plastic consumer products of different colors and polymer types. The replicate samples were incubated in 200 ml tap water and mixed with algae from the laboratory (*Pseudokirchneriella subcapitata,* cultured in MBL medium (Nichols, 1973)). Organic matter collected in the trawl in the field was more resistant to oxidation, however we did not use organic matter from the field in our recovery experiment to avoid the possibility of contaminating the samples with plastic. The separation method was applied to these sample as described above, including filtration, digestion of organic material with hydrogen peroxide and visual sorting of the plastic pieces.

Moreover, because hydrogen peroxide has been shown to react on some types of polymers resulting in visible changes (Nuelle et al., 2014), we used a shorter exposure time and lower hydrogen peroxide concentrations. In order to ascertain that this modified hydrogen peroxide treatment was non-destructive, the method was tested with the well characterized reference plastic particles described above. Before and after the hydrogen peroxide treatment the plastic pieces were visually observed with a stereo dissecting microscope (Leica MZ6) (Nuelle et al., 2014).

2.6. Data handling and statistics

For ease of comparability with other studies, the plastic concentrations in this study are expressed both as plastic abundance per unit area and per unit volume. For the primary study the numbers of plastic

pieces km^{-2} were calculated as trawl opening width × transect length, where transect length was obtained from the GPS track recorded on the boat. For the volumetric concentration (plastics m^{-3}) we multiplied trawl opening area × 0.5 × transect length. The factor of 0.5 was applied because only the lower half of the trawl opening was submerged during sampling (Baldwin et al., 2016). Since we did not have complete GPS data from the citizen science study available, the number of plastic pieces km^{-2} were calculated as max. net width × transect length, where transect length was calculated assuming an average speed of 2.7 knots and for the volumetric concentration (plastics m^{-3}) we took the whole net opening as it was completely submerged in the water during sampling.

Other studies have hypothesized that there is a relationship between population density and number of microplastic particles (Barnes, 2005; Browne et al., 2011; Zhao et al., 2014). To test the hypothesis that plastic concentration was correlated with anthropogenic stress, we first used a distance-weighted GIS spatial layer (anthropogenic impact index) based on the concentration of potentially hazardous activities (e.g. present or historical industrial sites) in the surveyed area that was developed and described in Nyström Sandman et al. (2013). Secondly, we calculated the mean of the index within a circular buffer zone (1.7 km radius) surrounding the center coordinate of each transect (sample), which on average covered the whole transect length. We used ArcGIS 9.3 for the spatial analysis while the relationship between plastic concentration and the anthropogenic index was evaluated using linear regression where plastic density was Log-transformed and assumed to follow a normal distribution. All statistical analyses were performed in R 3.2.4 (R Core Team, 2014).

2.7. Fourier transform infrared spectroscopy

We used Fourier transform infrared spectroscopy (FTIR) to confirm the identification of the particles selected by visual sorting, as has been done by others (Doyle et al., 2011; Martins and Sobral, 2011; Nor and Obbard, 2014). The infrared spectra were recorded at $4 \, cm^{-1}$ resolution with a Bruker Vertex 70 FTIR instrument that was equipped with an HgCdTe detector. Data were recorded on both sides of the centre burst of the interferogram during forward and backward movement of the movable interferometer mirror. 200 complete interferometer scans, recorded within 13 s, were averaged for one spectrum. A zero-filling factor of 2 was used and spectra were apodized by a Blackman-Harris 3-term function. Individual particles were placed on the one-reflection diamond crystal of a Bruker Platinum attenuated reflection (ATR) accessory, which was cleaned with ethanol prior to each measurement and pressed onto the crystal with a piston. The spectrometer and the ATR accessory were continuously purged with dry air from a Balston purge gas generator 75-62 to remove ambient water vapor.

First, four plastic products of known composition (polyethylene, polypropylene, polystyrene and polyethyleneterephthalate) were used to record reference spectra (see supporting information). This selection was based on the low density of polyethylene, polypropylene and polystyrene and frequency in surface water samples (Andrady, 2011; Morét-Ferguson et al., 2010). Then, spectra of the sampled material were measured as described above. A total of 59 individual particles (24), macroplastic (8), paint flakes (2) and fibers (25) that were visible to the naked eye were used for the analysis. Smaller particles were not possible to analyze due to difficulties in handling and positioning on the ATR crystal. The obtained FTIR spectra of the samples were compared with the reference spectra (Table 3 in Supporting information).

3. Results

3.1. Plastic recovery and quality assurance

For the sampling, we used two different boats. One of them was an aluminum boat without paint on the boat hull. The other boat had a red and blue painted hull. We did not find any paint particles that could stem from our boats in our samples and therefore can exclude boat paint as a source of contamination.

The control experiment revealed that there were no particles found that could have originated from the net and hard plastic containers. However, in the replicates we found on average three fibers that might have originated from clothing worn during the sampling. Nevertheless, these blanks showed that contamination during sample handling was low.

In the three replicates of our QA test, 100% of all the added plastic particles and colored plastic fibers were recovered, indicating that our separation method works very well for those pieces. However, on average only 60% of translucent and white plastic fibers, were recovered. Furthermore, we did not find any additional fibers or particles from other sources, meaning that no contamination from the air or clothes occurred during processing in the laboratory.

The hydrogen peroxide treatment did not lead to any evident changes in the physical characteristics of the plastic (color, surface, size, and shape) after 24 h. Therefore, we conclude that the method as we applied it is non-destructive for plastic and can be used to digest the organic debris in the samples. The algae we added to the recovery experiment to simulate organic matter from the field was readily digested by the hydrogen peroxide treatment.

3.2. Characteristics of the collected plastic

We sorted the plastic pieces collected from the field into four different categories (fragments, macroplastic, paint flakes and fibers) and counted their abundance. Most of the retrieved plastic pieces were fibers, which accounted for 82% of the total number of plastic pieces we collected (Table 1 and Figs. 1–24 in Supporting information). The proportion of fibers was even higher (approximately 90%) when the three samples from near the Stockholm urban area (samples 10–12) were excluded. Macroplastic and paint flakes were almost exclusively present in the Stockholm samples (samples 10–12). These three samples also showed a relatively higher proportion of fragments compared to all the other samples (30% versus 10%).

The main colors of the retrieved fibers were blue, red, black and green (Fig. 2 and Figs. 1–24 in Supporting information). These bright and unnatural colors were easy to distinguish from other organic material. Furthermore, we identified plastic fragments in our samples. These fragments were mostly white, black and blue. All of the retrieved paint flakes were of white color.

The retrieved fibers differed widely in length (see Table 4 in Supporting information). The shortest measured fiber was 290 μm long and the longest was about 27 mm long. Only two fibers were shorter than our mesh size, with a length of 290 μm and 330 μm respectively. On average, the fibers were 2.9 mm long. The fragments had a very irregular shape and their average diameter varied between 350 μm and 5.7 mm for the plastics with at least one side longer than 335 μm (see Table 5 in Supporting information). The average diameter of the particles was 1.3 mm.

Samples from the Stockholm area (samples 10–12) contained mostly microplastics but larger plastic debris (> 5 mm) was also present (Table 1 and Figs. 22–24 in Supporting information). Compared to the other samples these three samples contained many fragments. The larger plastic fragments were dominated by translucent and white fragments, but other colors were also present. These larger fragments most likely originate from candy wrappers and other small packaging.

3.3. Identification of plastic by FTIR

Out of the 59 analyzed particles, 48 were confirmed to be plastic of a certain polymer type (Figs. 25–36 in Supporting information). Thirty-one out of the 59 analyzed particles (53%) were identified as polypropylene and 14 of the 59 (24%) were identified as polyethylene.
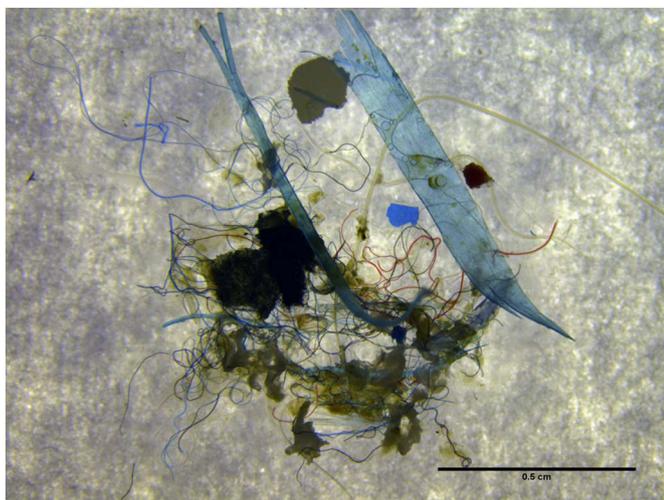
Fig. 2. Microscopic photograph of plastics retrieved from sample 19 from Nynäshamn harbor.

Only three pieces of the 59 analyzed were identified as polystyrene by FTIR spectroscopy. One example spectrum for a sample from the Stockholm archipelago identified as polypropylene is given in Fig. 3. Eight of the eleven remaining spectra show bands that indicate the polymer character of the sample but the spectra did not match any of our four reference materials. Therefore, we assume that these were of a different polymer type or a mixture. Five of these spectra show additionally bands in the range between $1728\,cm^{-1}$ and $1547\,cm^{-1}$ indicating a biofilm on top of the plastic polymer (Socrates, 2010). One spectrum shows bands typical for a biological cell spectrum without obvious contribution from non-biological polymers. The chemical composition of the last two spectra, however, could not be identified.

3.4. Abundance and spatial distribution of plastic debris

All trawl samples from the primary study contained plastic particles but the concentration varied considerably between the sample with the lowest concentration (sample 4, outer Archipelago, 1.56×10^4 plastics km^{-2} (0.19 plastics m^{-3})) and the one with the highest (sample 11, Stockholm central, 6.18×10^5 plastics km^{-2} (7.73 plastics m^{-3}), Fig. 4 and Table 1). The samples with the highest plastic concentrations were taken in Stockholm and showed an average plastic concentration of 4.21×10^5 plastics km^{-2} (5.26 plastics m^{-3}) which is > 9 times higher than in the mean concentration of 4.65×10^4 plastics km^{-2} (0.58 plastics m^{-3}) in the outer southern Archipelago (more remote/
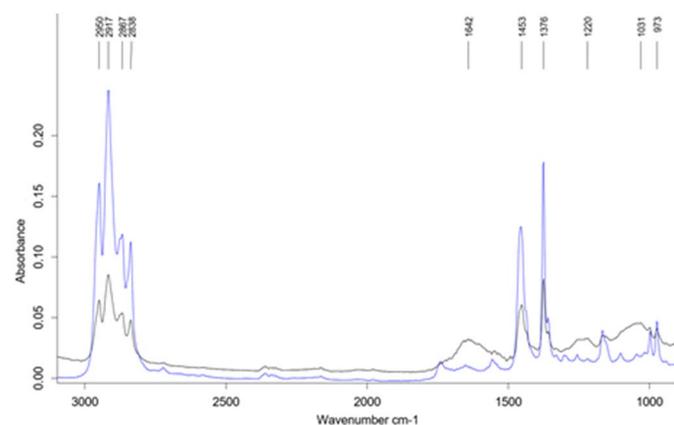

Fig. 3. Example of FTIR spectra: Reference sample of polypropylene (blue line) and sample that has been identified as polypropylene (black line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

offshore areas). The samples from Nynäshamn (sample 19) and Trosa (sample 21) harbors have 2.3 and 2.7 times higher plastic concentrations than samples from the outer southern Archipelago. Around Nynäshamn harbor we took three samples at different distances from the harbor. Sample 19 was taken within the semi-enclosed area of the harbor and had plastic concentration 2.4 times higher than sample 20 from directly outside the harbor. In turn, sample 20 had 1.8 times higher concentration than sample 18, which was taken on a transect while approaching the harbor. These results show that the plastic concentration decreases with increasing distance from the harbor. One sample (sample 17), taken at the outlet of a wastewater treatment plant, also had a concentration about 4.5 times greater compared to the outer southern Archipelago. There was a significant relationship between the anthropogenic impact index and plastic concentration ($t_{1, 19} = 5.28$, $p \leq 0.0001$, $R^2 = 0.59$ (Fig. 5A)). The relationship was however strongly driven by the samples with the highest plastic concentration (samples from Stockholm). When these samples were removed no correlation remained ($t_{1, 16} = -0.019$, $p = 0.98$, $R^2 = -0.062$ (Fig. 5B)).

The results from the citizen science study (Fig. 6) show lower concentrations of plastics further away from potential sources in the cities of Stockholm and Visby. The highest concentration was found close to Stockholm (O4) and was 6.86×10^4 plastics km^{-2} (0.87 plastics m^{-3}). This sample had a concentration that was about 8 times higher than the sample O2 (9×10^3 plastics km^{-2} (0.11 plastics m^{-3}), which was taken around Gotska Sandön, which is an uninhabited and remote island in the Baltic Sea. The sample O1, taken close to Visby, also showed an elevated concentration 4×10^4 plastics km^{-2} (0.51 plastics m^{-3}). These four samples showed a significant relationship with the anthropogenic impact index. Sample O4 from Stockholm had a lower plastic abundance by about one order of magnitude compared to the Stockholm samples (samples 10–12) of the primary study, which may reflect differences in sampling technique. For example, the traveling speed was not the same, the samples were taken in a different year and the nets had different diameters and mesh size.

4. Discussion

Our blank experiments indicated that the contamination of our samples during handling was low. Even though we used seawater to rinse the cod end, the contamination was expected to be minimal. This is due to the fact, that the used water volume to rinse the net was < 0.01% of the total filtered water volume.

The 100% recovery of particles and colored plastic fibers in our QA test demonstrates that they were readily visually identified and sorted. The lower recoveries of white and translucent fibers (60%) are likely attributable to difficulty distinguishing them from the bright background under the microscope. Other investigators have reported that most plastic particles in the marine environment are white or related (e.g., discolored, yellow, clear-white-cream) (Hidalgo-Ruz et al., 2012). The fibers and plastic particles we identified had mostly bright and unnatural colors. Therefore, we expect that white and translucent fibers were partly overlooked during the visual examination and that we underestimated the quantity of these in the samples.

Fibers are the major component of marine plastic debris we collected in the Stockholm Archipelago, this is in line with many other studies (Kanhai et al., 2017; Lassen et al., 2015; Lusher et al., 2014; Mason et al., 2016a; Sutton et al., 2016). Most of them are smaller than 1 mm, which is in agreement with Mason et al. (2016a, b) who sampled plastic from surface water using the same methodology in Lake Michigan. The diameter of all fibers was smaller than the mesh size of the net and therefore some fibers could have slipped through. Even though our manta trawl will not quantitatively collect plastic debris < 335 μm, two fibers were found with lengths of 290 μm and 330 μm respectively, which is a bit smaller than our mesh (335 μm). Such small debris was probably retained in the net because algae and other organic
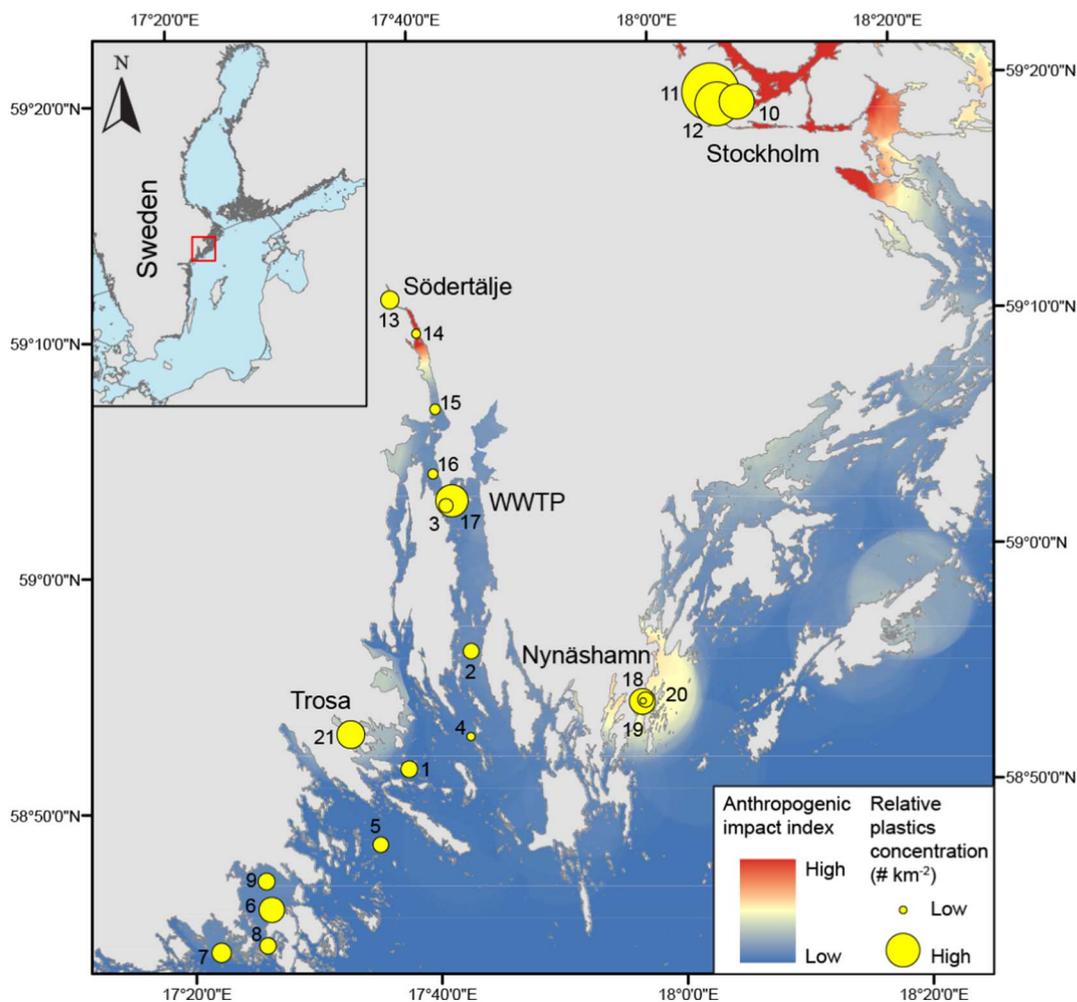
Fig. 4. Map showing sampling locations and relative near surface concentration of plastic debris from the primary sampling study in the Stockholm Archipelago, Sweden in relation to anthropogenic impact index. The anthropogenic impact index is based on the density of current or historical hazardous sites such as industries and decreases with distance from such sites. The size of the sampling points is proportional to plastic concentration with the lowest concentration being 1.56×10^4 plastics km^{-2} (0.19 plastics m^{-3}) and the highest 6.18×10^5 plastics km^{-2} (7.73 plastics m^{-3}).

Table 1

Abundance of plastic and amounts of retrieved plastic fragments, macroplastic, paint flakes and fibers, and percentage of plastic pieces that were fibers.

Sample	Plastic pieces km^{-2}	Plastic pieces m^{-3}	Total plastic	Fragments	Macroplastic	Paint Flakes	Fibers	Fibers [%]
1	4.12×10^4	0.51	51	14	0	0	37	72.5
2	3.29×10^4	0.41	76	7	0	0	69	90.8
3	2.54×10^4	0.32	53	4	1	0	48	90.6
4	1.56×10^4	0.19	56	5	0	0	51	91.1
5	4.08×10^4	0.51	120	7	0	0	113	94.2
6	9.01×10^4	1.13	219	8	0	0	211	96.3
7	5.30×10^4	0.66	125	13	0	0	112	89.6
8	4.82×10^4	0.60	146	4	0	0	142	97.3
9	5.05×10^4	0.63	105	6	0	0	99	94.3
10	2.48×10^5	3.11	441	228	28	9	176	39.9
11	6.18×10^5	7.73	407	119	12	37	239	58.8
12	3.95×10^5	4.93	474	224	24	3	223	47.0
13	6.35×10^4	0.79	70	11	0	0	59	84.3
14	1.97×10^4	0.25	79	6	0	0	73	92.4
15	2.69×10^4	0.34	114	9	0	0	105	92.1
16	2.40×10^4	0.30	63	13	0	0	50	79.4
17	2.09×10^5	2.61	300	11	0	0	289	96.3
18	2.53×10^4	0.32	74	4	1	0	69	93.2
19	1.07×10^5	1.34	300	28	1	0	271	90.3
20	4.47×10^4	0.56	122	19	0	0	103	84.4
21	1.27×10^5	1.59	303	47	1	1	254	83.8

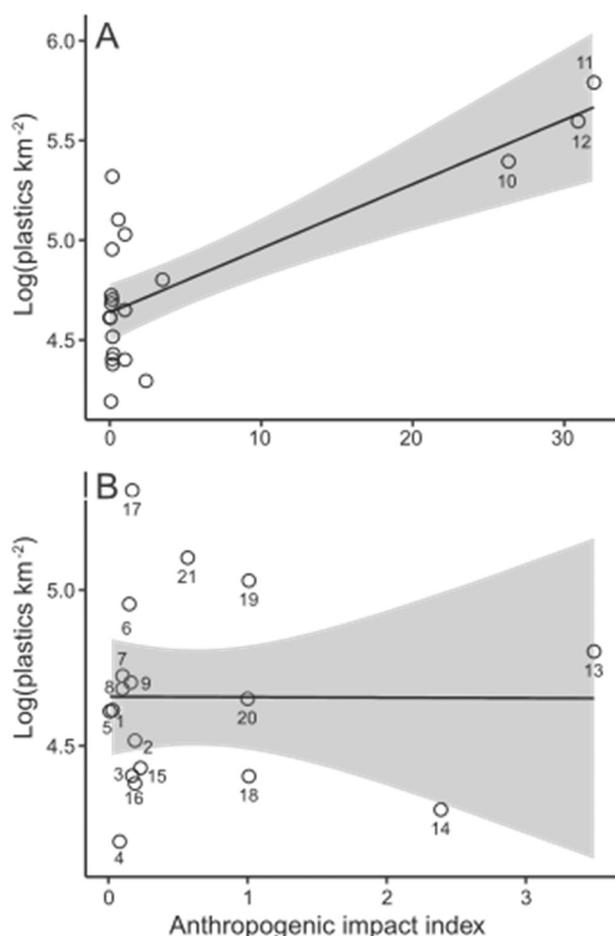**Fig. 5.** Linear regression of plastic concentration (Log10 plastics km^{-2}) as a function of modelled anthropogenic impact. The grey band around the regression line represents a 95% confidence interval. A) Including all samples; B) Without the three Stockholm samples.

debris were partly clogging the net or it might be due to inaccuracies in the size measurement with the image analysis program. Some of the fibers got entangled with each other and therefore it was difficult to distinguish very long fibers from each other when trying to measure the length. Thus, we assume that some fibers were even longer than 27 mm. Furthermore, we only measured the length of a sub-set of 248 fibers. The fragments had a very irregular shape but had at least one side longer than 335 μm.

In our study the color of fibers was predominantly blue, red, black and green and for the fragments: white, black and blue. The most common colors reported in other studies were white or related (e.g., discolored yellow, clear white-cream) (Hidalgo-Ruz et al., 2012). The difference in abundance of white fibers likely reflects different sources of plastic to the Stockholm archipelago compared to the sites reviewed by Hidalgo-Ruz, however our low recovery rate of white fibers also could contribute to the difference.

Wastewater treatment plants are known to emit plastic fibers derived from synthetic clothes and primary microplastics that come from cosmetic products (Browne et al., 2011; Fendall and Sewell, 2009; Magnusson and Norén, 2014). In one study the size of primary microplastic particles in facial cleaners was measured and a median size between 196 and 375 μm for different products was reported (Fendall and Sewell, 2009). The mesh in the trawls used in our study would not fully retain primary microplastics in this size fraction (< 335 μm) and therefore we should not expect to have quantitatively collected primary microplastics from personal care products in any of our samples. The higher amount of collected plastic in sample 17, i.e.

from the WWTP outlet is therefore rather due to fibers than due to cosmetic derived microparticles.

FTIR spectroscopy has previously been recommended to confirm the identity of plastic pieces because up to 70% of particles that visually resemble microplastics are not confirmed to be plastics by chemical analysis (Hidalgo-Ruz et al., 2012). In our FTIR analysis of 59 pieces of debris that were visually identified as plastic, 48 pieces (81%) were confirmed to be polypropylene, polyethylene or polystyrene. For the remaining 11 pieces (19%) the sample could not be identified as a certain polymer type because the spectra did not match our reference materials. However, eight of these 11 pieces showed bands typical for plastic polymers. Only one sample suspected to be plastic based on visual identification was identified as a biological cell.

Generally, some information about possible sources of plastics to the Stockholm Archipelago can be inferred by the identification of the polymer type. The most common polymer types in the samples were polypropylene and polyethylene accounting for 53% and 24% of the plastics we could identify, which is in line with other studies assessing the occurrence of plastic in surface waters (Carson et al., 2013; Hidalgo-Ruz et al., 2012; Mason et al., 2016b; Rios Mendoza and Jones, 2015). These figures are not surprising as these plastic types are positively buoyant, accumulate at the water surface and correlate well with the production volumes of polyethylene and polypropylene (PlasticsEurope, 2015). These polymers are mainly used as packaging material (PlasticsEurope, 2015), which is discarded after a short lifetime and can therefore explain the larger plastic fragments and high abundance of plastic fragments that were found in the Stockholm area. Nylon, polyester, polypropylene and polyethylene are the main synthetic materials in the manufacturing of ropes (Corbett, 2009), whereof polypropylene is the most commonly used material for floating ropes (MacKown, 1992). Other studies have identified polypropylene fibers in the aquatic environment and assume ropes as one of the main sources (Nor and Obbard, 2014). Therefore, we assume that some retrieved polypropylene pieces and especially many fibers probably originate from ropes that were used on boats.

The regressions between plastic concentrations and the anthropogenic impact index (Fig. 5A and B) indicates that the anthropogenic impact index is not an ideal proxy for plastic contamination. The correlation was strongly driven by the Stockholm samples, which contained the highest plastic concentrations by far and also have a high anthropogenic impact index. One of the reasons for the lack of correlation when the Stockholm samples are removed might be that the anthropogenic impact index is calculated using land based activities and thus does not consider loads from marine sources such as boat traffic. As many of the plastic pieces we collected were polypropylene, our hypothesis is that these stem from ropes used on boats. The largest outlier was sample 17 from the WWTP. WWTPs are known sources of microplastics (Browne et al., 2011; Magnusson et al., 2016; Magnusson and Norén, 2014; Mason et al., 2016a) which also was confirmed by our sample, but the corresponding anthropogenic impact index is low. A probable explanation for this mismatch lies in the fact that the WWTP has a catchment area of > 600 km^{-2} and processes water from approximately 350,000 person equivalents including wastewater from both standard households and large industries, and storm water which then become decoupled from the geographical position of these diffuse sources (Syvab, 2013). Moreover, the anthropogenic impact index was not specifically designed to reflect plastic pollution but rather for a broad variety of pollution to the environment from land-based point sources.

The highest plastic concentrations were found in the samples taken in Stockholm, where it is likely that land sources predominate. In areas with high leisure boat traffic (e.g. Trosa and Nynäshamn harbor) we found elevated concentrations, but still lower than the ones found in Stockholm. These results indicate that boating contributes to marine plastic debris in the Stockholm archipelago, but that land sources have a higher impact. The harbor of Nynäshamn is specifically interesting
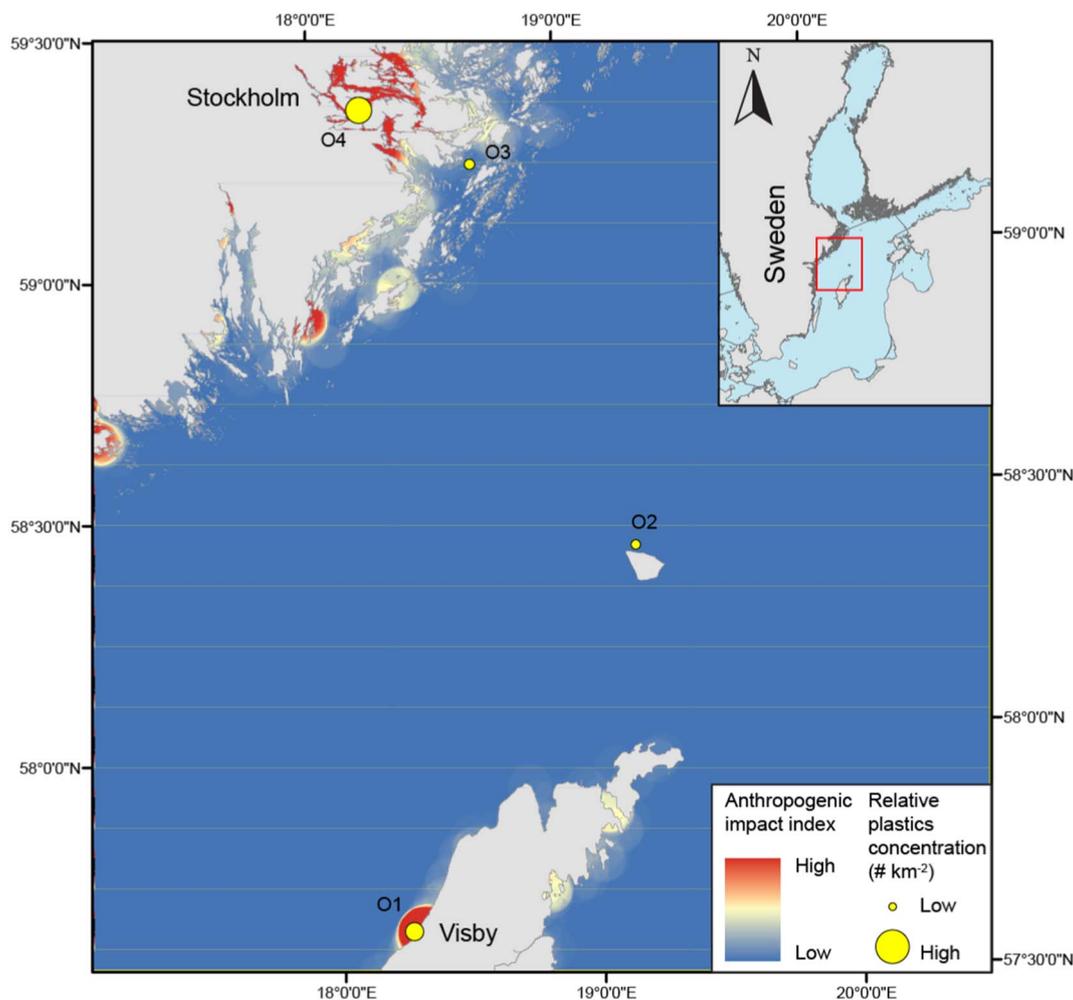
Fig. 6. Map showing relative near surface concentration of plastic debris from the citizen science study in the open Baltic Sea. The size of the sampling points is proportional to the rate of plastic collection in a purpose-built sampler towed behind stand-up paddleboards. The lowest concentration is 9×10^3 plastics km^{-2} (0.11 plastics m^{-3}) and the highest 6.86×10^4 plastics km^{-2} (0.87 plastics m^{-3}).

because in this sampling area we collected three samples at different distances from the harbor. Sample 18 was taken on a transect while approaching the harbor, sample 20 was collected right outside the harbor area and sample 19 is from the inner harbor of Nynäshamn. This gradient shows that the closer the samples were taken to the harbor, the higher the concentrations. Sample 19 shows > 2 times higher concentrations compared to sample 20 and > 4 times higher than sample 18. However, the anthropogenic impact index did not vary significantly between the samples. This also indicates that boating is a source of plastic to the marine environment that is not fully reflected in the anthropogenic impact index and that enclosed areas with point sources are more prone to higher plastic concentrations.

In our study, the plastic concentrations varied strongly depending on the sampling area but a gradient from open water to urbanized waters was evident. The mean plastic concentrations measured in the North-Western Mediterranean Sea by Collignon et al. (2012) is consistent with the values in our study (Table 2). Both studies include highly impacted areas as well as areas with a lower plastic accumulation potential. Moore et al. (2001, 2002) and Lattin et al. (2004) conducted sampling studies in areas, which are highly influenced by plastic debris. The North Pacific Central Gyre for example is an accumulation area in the Pacific Ocean influenced strongly by ocean currents (Moore et al., 2001). Higher concentrations of plastic debris originating from terrestrial sources can be found close to urban areas, sites of tourism and near river outflows (Ryan et al., 2009). This also applies to other sampling locations that are close to the urban areas of

Los Angeles and Santa Monica (USA) (Lattin et al., 2004; Moore et al., 2002). Differences in plastic concentration of our overall mean value for all 21 samples from the Stockholm Archipelago and the ones in the studies of Moore et al. (2001, 2002) and Lattin et al. (2004) might be related to the fact that we included samples from the outer southern Archipelago which supposedly is less influenced by plastic debris. The mean plastic concentration in the samples (samples 10–12) from the urban area of Stockholm, which also is considered a highly impacted area, was 5.26 plastics m^{-3} and thus in the range of these three other studies. Even though, Magnusson collected samples close to two Finnish towns, the reported plastic concentrations are more similar to the abundance in the outer Archipelago found in our study (Magnusson, 2014).

Citizen science can be a useful tool if data quality can be assured by developing clear protocols and training of volunteers (Hidalgo-Ruz and Thiel, 2015). In our citizen science study, the two adventurers were clearly instructed how to handle the sampling gear and treat the sampled material and further analysis of the samples was performed according to the same protocol used in our main study. Furthermore, we did not find plastic pieces that indicate a contamination from the two adventurers, e.g. no plastic fibers of the same kind were found throughout or in several of the samples. Thus, we feel confident to include these data.

Nevertheless, it was not possible to accurately measure the average speed and therefore we had to rely on the average speed reported by the two citizen scientists. We assume that they overestimated the average

Table 2
Plastic concentrations measured in our primary study compared to other studies.

Study	Mean value [plastics m^{-3}]	Mean value [plastics km^{-2}]	Location	Year	Mesh [µm]
Collignon et al., 2012		1.16×10^5 $(0–8.9 \times 10^5)$	North-Western Mediterranean Sea	2010	333
Moore et al., 2001	2.23	3.34×10^5 $(0.32–9.69 \times 10^5)$	North Pacific Gyre	1999	333
Moore et al., 2002	7.25		San Gabriel River	2000	333
Lattin et al., 2004	3.92		Southern California shore	2001	333
Magnusson, 2014	0–0.74		Finnish coast (Rauma and Turku)	2013	333
This study	1.37 (all samples) 3.11–7.73 (Stockholm) 0.19–1.13 (outer Archipelago)	1.10×10^5 (all samples) $2.48–6.18 \times 10^5$ (Stockholm) $1.56–9.01 \times 10^4$ (outer Archipelago)	Stockholm Archipelago	2014	335

speed and this resulted in lower plastic concentrations in those samples. In addition, the citizen science study used trawls with a smaller mesh size than those used in our main study. Consequently, we cannot directly compare the total concentrations but only the trends along the anthropogenic impact gradients. The distribution pattern of plastics in samples collected in the citizen science study was consistent with the one of our primary study. These samples also contained higher concentrations close to densely populated areas (samples O1 and O4) compared to remote areas (samples O2 and O3). The sample from Stockholm (sample O4) showed about 8 times higher concentrations compared to the sample from the most remote sampled area (sample O2). Despite the methodological differences between the two sampling campaigns, both add to our current knowledge of plastic distribution and density in the Baltic Sea; especially considering the current lack of data. Our study illustrated the potential to leverage the public's interest and awareness of plastic pollution to generate new monitoring data.

5. Conclusion

Plastics are widely distributed globally including the Baltic Sea and Stockholm Archipelago. All our collected samples contained plastics. Plastic abundance varied strongly depending on the sampling location. Sampling sites close to point sources of plastic pollution showed higher concentrations than remote areas of the outer Archipelago. FTIR spectroscopy revealed the identity of visually identified pieces. The most common polymer types were polypropylene and polyethylene, which are positively buoyant and also account for the two most widely produced plastic types.

Results from the citizen science study show similar distribution patterns in plastic concentration. Therefore, we conclude that citizen science studies work well when people are clearly instructed and are a useful tool to engage non-scientists in environmental research.

Moreover, comparisons between studies such as this one are necessary in order to identify various point sources to pollution. However, we see a strong need for standardization of sampling techniques in order for this to become possible. Currently, the methodologies for plastic sampling and identification vary greatly making comparisons hard or even impossible in some cases. Furthermore, due to a lack of advanced identification techniques especially for very small plastic pieces several errors might lead to an under- or over estimation of plastic pieces in surface water samples.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.marpolbul.2017.04.062.

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