

**Sources and fate of plastic particles in  
Northern European coastal waters**

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“By the late 1950s, as Barthes was declaring plastic a “miraculous substance,” a source of endless possibility, Carson began rethinking earlier claims about an ocean exempt from human influence by detailing the threats of nuclear waste to the seas.” (De Wolff 2014)

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## ACRONYMS

d.w.	Dry weight
EDS	Energy dispersive spectroscopy
FTIR	Fourier transform infrared (spectroscopy)
NIR	Near-infrared (spectroscopy)
NOEC	No observed effect concentration
OSPAR	The Convention for the Protection of the Marine Environment of the North-East Atlantic
PET	Polyethylene terephthalate
PMMA	Polymethylmethacrylate
SEM	Scanning electron microscopy
w.w.	Wet weight

## LIST OF PAPERS INCLUDED IN THE THESIS

This thesis is based on the following studies, referred to in the text by their Roman numerals:

I: **Karlsson, T.M.**, Kärman, A., Rotander, A. & Hassellöv, M. (n.d.). Sampling methods for microplastics >300 µm in surface waters – A comparative study between a filtering pump and a trawl. Accepted for publication in Environmental Science and Pollution Research.

II: **Karlsson, T.M.**, Vethaak, A. D., Almroth, B. C., Ariese, F., van Velzen, M., Hassellöv, M. & Leslie, H. A. (2017). Screening for microplastics in sediment, water, marine invertebrates and fish: Method development and microplastic accumulation. Marine Pollution Bulletin.122(1-2) 403-408

III: **Karlsson, T.M.**, Hassellöv, M. & Jakubowicz, I. (2018). Influence of thermooxidative degradation on the *in-situ* fate of polyethylene in temperate coastal waters. Marine Pollution Bulletin. 135, 187-194

IV: **Karlsson, T.M.**, Arneborg, L., Broström, G., Carney Almroth, B., Gipperth, L. & Hassellöv, M. (2018). The unaccountability case of plastic pellets. Marine Pollution Bulletin. 129 (1) 52--60

V: **Karlsson, T.M.**, Wilkinson, T. & Hassellöv, M. (n.d.). High concentrations of plastic particles on exposed beaches and beaches near urban areas. *Manuscript*.

## MY CONTRIBUTIONS TO THE PAPERS

I: Took part in planning and sampling. Took part in evaluations and tests during the development of an earlier prototype of the pump. Analyzed all samples and was responsible for interpreting all collected spectra. Collaboratively took part in the statistical analyses and took the lead in writing the manuscript.

II: Was responsible for planning and performing sampling, extraction and analysis of sediment, water and invertebrate biota, as well as methodological tests and development. Analyzes of samples of *Salmo trutta*. Took the lead in writing the manuscript.

III: Took part in planning and was responsible for sampling and analyzing the samples. Took the lead in writing the manuscript.

IV: Took the lead in writing the manuscript and combining the insights from the different parts of the studies. Responsible for laboratory analyses of field samples.

V: Responsible for planning, sampling and analyzes. Supported and verified all performed analyzes in the lab. Took the lead in writing the manuscript.

## OTHER PAPERS AND REPORTS NOT INCLUDED IN THE THESIS

### Scientific papers

**Karlsson, T.M.**, Grahn, H., van Bavel, B. & Geladi, P. (2016). Hyperspectral imaging and data analysis for detecting and determining plastic contamination in seawater filtrates. *Journal of Near Infrared Spectroscopy*, 24(2), 141-149.

Rist, S., Carney Almroth, B., Hartmann, N. & **Karlsson, T.M** (2018). A critical perspective on early communications of human health effects of microplastics. *Science of the Total Environment*. 626 (1) 720-726

Linders, T., Infantes, E., Joyce, A. **Karlsson, T.M.**, Ploug, H., Hassellöv, M., Sköld, M. & Zetsche, E.-M. (2018) Particle sources and their transport in stratified Nordic coastal seas in the Anthropocene. *Elementa Science of the Anthropocene*

Hartmann, N. B., Hüffer, T., Thompson, R. C., Hassellöv, M., Verschoor, A., Daugaard, A. E., Rist, S., **Karlsson, T.M.**, Brennholt, N., Cole, M., Herrling, M.P., Hess, M.C., Ivleva, N.P., Lusher, A.L. & Wagner M. (2019). Are we speaking the same language? Recommendations for a definition and categorization framework for plastic debris. *Environmental Science & Technology*. 53(3)1039-1047

Hartmann, N. B., Hüffer, T., Thompson, R. C., Hassellöv, M., Verschoor, A., Daugaard, A. E., Rist, S., **Karlsson, T.M.**, Brennholt, N., Cole, M., Herrling, M.P., Hess, M.C., Ivleva, N.P., Lusher, A.L. & Wagner M. (2019). Response to the Letter to the Editor Regarding Our Feature “Are We Speaking the Same Language? Recommendations for a Definition and Categorization Framework for Plastic Debris”. *Environmental Science & Technology*. 53 (9) 4678-4679

Schönlau, C., **Karlsson, T.M.**, Rotander, A., Nilsson, H., Engwall, M., van Bavel, B. & Kärrman, A. (n.d.). Microplastics in sea-surface waters surrounding Sweden sampled by manta trawl and in-situ pump. *Accepted for publication in Marine Pollution Bulletin*

## Technical Reports

Lachmann, F., Almroth, B.C., Baumann, H., Broström, G., Corvellec, H., Gipperth, L., Hassellöv, M. **Karlsson, T.M.** & Nilsson, P. (2017). Marine Plastic litter on Small Island Developing States (SIDS): Impacts and measures. Göteborg: Swedish Institute for the Marine Environment, University of Gothenburg.

**Karlsson, T.M.**, Kärrman, A., Rotander, A. & Hassellöv, M. (2018). Provtagningsmetoder för mikroplast >300 µm i ytvatten – En jämförelsestudie mellan pump och trål. Havsmiljöinstitutet.

Hassellöv, M., **Karlsson, T.M.** & Haikonen, K. (2018). Marint mikroskopiskt skräp. Undersökning längs svenska västkusten November 2015. Länsstyrelsen Västra Götaland.

Hassellöv, M., **Karlsson, T.M.**, Mattsson, K., Magnusson, K., Strand, J., Lenz, R., van Bavel, B. & Pettersson Eidsvoll, D. (2018). Progress towards monitoring of microlitter in Scandinavian marine environments – state of knowledge and challenges. Tema NORD. Nordic Council of Ministers

**Karlsson, T.M.** (2018). Vad säger plasten längs med Sveriges kuster om våra konsumtionsmönster? Konsumtionsrapporten.

**Karlsson, T.M.**, Ekstrand, E., Threapleton, M., Mattsson, K., Nordberg, K. & Hassellöv, M. (2019). Undersökning av mikrokräp längs bohuslänska stränder och i sediment. Naturvårdsverket.

Setälä, O., Granberg, M., Hassellöv, M., **Karlsson, T.M.**, Lehtiniemi, M., Mattsson, K., Strand, J., Talvitie, J. & Magnusson, K. (2019). Policy brief: Monitoring of microplastics in the marine environment; changing directions towards quality controlled tailored solutions rather than overarching harmonized protocols. PolitikNord: Nord. Nordic Council of Ministers 2019

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## POPULÄRVETENSKAPLIG SAMMANFATTNING

För snart tio år sedan hörde jag talas om skräpöarna i Stilla havet. De beskrevs som öar två gånger så stora som Texas, helt fulla av plast. Jag förstod snart att de här öarna inte fanns på riktigt, utan att myten om dem hade skapats genom felaktiga översättningar mellan språk. Forskare har sedan dess försökt klargöra att det inte rör sig om några öar utan snarare om en smog, eller en soppa, med små finfördelade plast-partiklar som vi hittar nästan överallt i våra hav och vattendrag.

Hur vi beskriver det spelar roll. Plast är en väldigt praktisk materialgrupp som kan hjälpa oss med de hållbarhetsutmaningar vi står inför om det används på ett genomtänkt sätt. Men för att nå ett mer hållbart användande av plast behöver vi korrekta problembeskrivningar. En del i det är att förstå var plastpartiklarna kommer ifrån. En annan del är att försöka förstå vad som händer med dem när de hamnar i havet.

Mycket av mitt arbete har de senaste åren handlat om att utveckla och testa metoder för att provta och analysera mikroplast i vatten, djur och sediment. Vi förstod dock snart att det inte finns någon perfekt metod. Istället måste vi anpassa olika metoder till de frågor vi försöker besvara och sedan vara tydliga med vilka begränsningar de bär med sig.

Jag har lagt plast i burar i havet för att se vad som händer med plasten över tid. Jag har undersökt varför vi hittar så mycket plast från industrier, plast som inte ens hunnit bli plastprodukter. Och jag har tillbringat många timmar framför mikroskop för att analysera prover från sediment, djur, vatten och stränder. Slutligen har jag jämfört det som vi ser i våra prover med det som andra beräknar borde vara där, och med det som övervakningsprogram hittar längs med våra stränder.

Det jag har sett är att även om plast i havet kan transporteras långt, så fastnar mycket långs med stränder. Våra resultat visar även att trots att många sorters plaster flyter till att börja med, så täcks de snabbt av biofilm och sjunker. Själva molekylerna i plasten förändras också, vilket bland annat leder till att de lättare går sönder och formar mindre fragmentbitar.

Det är också en del i förklaringen till att en stor del av de partiklar som vi hittar i våra fältprover är fragment, det i sin tur visar att om vi vill arbeta med att minska mängden plastpartiklar i miljön så måste vi se över hur vi använder större plastprodukter.

En del partiklar är dock tillverkade i mindre storlekar. Exempel på sådana som vi hittar i miljön är s.k. pellets och fluff - båda relaterade till produktion av plast. I en studie såg vi att miljontals pellets läcker ut varje år från en enda plastfabrik, på grund av kontinuerligt spill. Vi såg dessutom att det läckte ut vid förvaringsplatser och andra områden där de hanterades. Detta trots att det finns nationella och europeiska regelverk som, om de implementerats, ska förhindra den här sortens spill.

Det här forskningsfältet har fått alltmer uppmärksamhet under de senaste åren vilket har lett till flera lösningsfokuserade initiativ. Fokusen på plast i havet och särskilt mikroplast har dock kritiserats då vi har många andra hållbarhetsutmaningar, såsom klimatförändringar, minskad biologisk mångfald och övergödning, framför oss.

En av diskussionerna gäller vilken risk mikroplast och plast generellt innebär. Större plast har associerats med tydligare konsekvenser, framförallt i form av spökgarn - tappade fiskeutrustningar som fortsätter fånga fisk och andra marina djur. Vad det gäller mikroplast så indikerar dock mycket av de data som vi har idag att nuvarande föroreningsnivåer av mikroplast inte innebär en generell ekologisk risk. Datan är dock bristfällig och det finns områden med högre nivåer. I våra strandprover hittade vi höga koncentrationer som ligger många gånger över de nivåer som idag anses säkra. I takt med ökade föroreningsnivåer kommer de områdena troligen bli allt vanligare.

En annan del av den nuvarande debatten gäller lösningar, och resultaten från det här doktorandprojektet visar tydligt att eftersom plast kommer att fragmentera och sjunka, så är det mer effektivt att arbeta med uppstädning närmare källan än långt bort. Det är dock ännu mer effektivt att arbeta preventivt för att minska läckaget till miljön. Då en stor del av det vi hittar i miljön är plastfragment som kommer från större plastmaterial så behöver huvudfokus ligga på större plast och hur vi använder den. Det innebär att arbeta aktivt med avfallsströmmarna genom att minska konsumtionen samtidigt som vi förbättrar sophanteringen för att minska läckage.

Dagens höga, och ökande, konsumtion av plast skulle varit utmanade oavsett materialtyp och i vissa fall är plast det mest hållbara alternativet. Lösningarna blir mer effektiva genom att fokusera på den underliggande problematiken, hellre än att lägga fokus på uppstädning. Om vi gör det blir det dessutom lättare att se hur olika hållbarhetsutmaningar är sammanlänkande och hur vissa lösningar för plastskräp, som t.ex. minskad konsumtion och förbättrad sophantering, kan ha en positiv påverkan på flera hållbarhetsutmaningar.

## ABSTRACT

Plastics are integral parts of our lives and have allowed for important technological leaps within society. However, an unwanted consequence of our current consumption of plastics is marine plastic pollution and in order to reduce its impact we need to understand its sources and fate patterns. It is a threefold challenge as it requires suitable methodology, as well as in-depth studies of sources and the various processes that affect the fate of plastics. Based on comprehensive tests and evaluations, this thesis provides recommendations on suitable methodologies for sampling, extraction and identification. To further improve the understanding of the fate of plastics in the ocean, *in-situ* experiments related to oxidation and biofouling were performed. Moreover, the distributions of plastic pellets were mapped in a case study area, through field studies and calculations, to understand the spread from local point sources. The results show that floating plastics are prone to beaching and it is concluded that although plastics can be subject to long-range transport, the majority of the pollutants will be found close to the point of release. The studies also show that most floating plastics will eventually sink, due to biofouling and degradation. To provide information on diffuse sources, the evaluated methods were then applied to analyze surface waters, sediment, biota and beach materials. Most microplastics (53-100%) found in the different surveys were identified as fragments of polyethylene, polypropylene and expanded polystyrene. Since most of the microplastics therefore stem from macroplastics, any attempt to address microplastic pollution needs to have a strong focus on macroplastics. Additionally, pellets and fluff were often encountered and specific point sources related to the production of plastics were examined in an interdisciplinary case study. The study showed continuous spills of plastic pellets associated with production, transportation and storage. The study furthermore illustrated that although there is a legal framework in place, it is not being adequately enforced, which has resulted in limited responsibility and accountability for the involved actors. The studies related to fate processes illustrate why attempts to decrease plastic pollution need to be focused as close to the source as possible, since that is where prevention and mitigation measures will be most efficient. Furthermore, the results from the field studies are crucial to consider for solution-oriented initiatives. They provide important insights regarding sources and fate of plastic particles, showing that in order to decrease microplastic pollution the main focus needs to be on larger plastics and how we use them. This means working actively to decrease waste streams through a lower level of consumption, while simultaneously improving waste management strategies to prevent leakage. The increasing interest from multiple stakeholders in academia, amongst policy makers and in the civil society also emphasizes the need for empirical data and clear communication to avoid discrepancies between the perceived and the actual sources and fate of floating plastic particles.

**Keywords:** plastic pollution, polyethylene, microplastics, fate, sources, FTIR spectroscopy, method development

# BACKGROUND

Plastic pollution has reached high on the political and scientific agenda in recent years. After the introduction of the term microplastics in 2004 (Thompson et al. 2004), smaller plastic particles (<5 mm) also received increasing attention. A few years later, scientists showed that microplastics were distributed in sediment, biota, surface waters and beaches all over the world (reviewed in: Andrady 2011, Cole et al. 2011, Desforges et al. 2014 & GESAMP 2015). The majority of the early studies were focused on confirming the presence or absence of plastics but recent methodological developments have started to allow for more in-depth studies.

The public attention was followed by a demand for prevention and mitigation. There are, however, several challenges associated with prioritizing actions, especially since plastics are intricately linked to our globalized economy and everyday lives. A clear and accurate problem description is therefore crucial in order to work efficiently towards decreasing microplastics in the environment. Since most early studies focused on the absence or presence of plastics, until recently only limited data was available on compositional differences. Additionally, little was known on the fate of plastics in the ocean.

Thus, the overarching aim of the work presented here was to better understand the sources and fate of plastic particles in northern European coastal waters. The main focus was directed on plastic particles in the meso- (<25 mm) and micro (<5 mm) fractions<sup>1</sup>, but I will also touch upon the links between different sizes.

The main body of the work presented in this thesis builds on work presented in three published papers, one accepted manuscript and one manuscript that is being prepared for submission. The papers and manuscripts can all be found in the appendix of the printed thesis. The thesis is divided into four chapters and Table 1 gives an overview of the papers and how they feed into different parts of the thesis.

In order to understand the sources and fate of plastic particles, suitable methodologies to measure and identify these needed to be adapted and developed. Several methodological tests for sampling, extraction and analysis were therefore performed in **Papers I, II, III** and **V**.

In **Papers I, III, IV** and **V** a combination of field measurements, modelling and in situ experiments were performed, to further investigate the fate of plastic particles in the marine environment. The main findings and general transport patterns of plastic particles in northern European waters are detailed in Chapter 2.

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<sup>1</sup> We discuss the definitions more in-depth in Hartmann et al. 2019. There we also suggest that future studies should be defined within the size span of 1-1 000  $\mu\text{m}$  for microplastics. In this thesis I will use the conventional definition of <5 mm to be in accordance with previously published literature.

The methods developed were then applied on a variety of field samples to better understand the composition of plastic particles. In addition, the lessons learned regarding the transport and fate mechanisms were used to understand how to sample in a representative way and how to interpret the results from environmental samples. Environmental samples were analyzed in **Paper I, II, IV & V**. In order to get a more complete overview the results from the field studies were related to surveys on macroplastics, as well as to recent regional reports that have made use of material-flow analyses to assess the different sources. The insights regarding sources of plastic particles are presented in Chapter 3.

In Chapter 4 the results from the previous chapters are put into a wider context. The results are discussed in relation to risks associated with plastic pollution and how to approach prevention and mitigation solutions.

**Table 1:** Overview of the included papers and how they feed into the different sections of the thesis.

	<b>Methods</b>	<b>Fate</b>	<b>Sources</b>
<b>Paper I</b>	Compared a manta trawl and a filtering pump.  Discussed sample size, replication and suitable ways to identify plastics	Concentrations in surface waters	Compositions in surface waters
<b>Paper II</b>	Tested and adapted extraction methods for microplastics in biota and sediment		Compositions in sediment, biota and surface waters
<b>Paper III</b>	Applied a variety of methods to characterize polyethylene	Placed thermally pre-degraded plastics in cages in coastal waters and examined how the material changed during 12 weeks	
<b>Paper IV</b>		Combined field tests with drifters, calculations and field measurements to study the spreading of spills of pre-production materials in a case study area	Investigated spills of pre-production microplastics and the underlying causes of their release to the sea/to the field
<b>Paper V</b>	Compared two sampling approaches. Developed image analysis for automatic size measurements of the particles.	Concentrations on beaches	Composition on beaches and comparisons with studies on macrolitter, material flow analysis and material usage



# CHAPTER 1: METHODS FOR MEASURING AND CHARACTERIZING PLASTIC PARTICLES IN ENVIRONMENTAL SAMPLES

Early plastic pollution research built largely on lessons from planktologists. A commonly used mesh size for studying zooplankton, fish eggs, larvae and other organisms in the nekton is 0.3 mm, and consequently, this has been the cut-off size limit in many microplastic studies. While it is still frequently applied, it is becoming increasingly common to sample for smaller sizes. In terms of discussing sources and fate 0.3 mm remains a practical cut-off size. Sampling for 0.3 mm is often done with neuston nets and surface-skimming manta trawls. The identification is normally done visually with the aid of stereomicroscopy, and it is becoming more common to couple it with chemical identification techniques such as Fourier transform infrared (FTIR) spectroscopy or Raman spectroscopy (Renner 2017).

It was recognized early on that one singular method would not be adequate to assess microplastic pollution, particularly since microplastics come in many different shapes, sizes and densities (Rochman 2019). As the research field matured, other methods, specifically adapted to different matrices and research questions, were developed (reviewed in: Hidalgo-Ruz et al. 2012, Renner et al. 2017a & Prata et al. 2018). Sampling has been done in several different types of matrices including beach substrate, surface waters, biota and sediment. The sampling methods are based either on sampling a specific volume, pooling several smaller subsamples or concentrating a larger volume over a smaller surface (as in the case of the manta trawl). Due to the time-consuming analysis, small samples and few replicates are often collected. As a consequence it is not uncommon that scientists report less than 15 particles per sample (e.g. Hermsen et al. 2017, Catarino et al. 2018, Courtenes-Jones et al. 2019 & Lacerda et al. 2019). Such low concentrations do not allow for compositional analyses and requires extensive replication, to allow for spatial or temporal comparisons (**Paper I**). One associated challenge to overcome, in order to start processing larger sample volumes, is the adaptation of suitable methods for extracting microplastics from the sample matrix. Extraction methods have improved in recent years and there are now several protocols using chemical digestion, enzymatic digestion and density separation (reviewed in: Hermsen et al. 2018, Mai et al. 2018, O'Connor et al. 2019). Another challenge lies within the identification of plastic particles. Although most of the identification is still done visually, the use of chemical identification methods has also increased (Renner et al. 2017a). The multitude of methods that are used today does not allow for comparison between studies, especially since methodological quality assessments and controls are often missing or not being reported (Hermsen et al. 2018). As a result, several scientists have noted the need for harmonizing the methods used for studying microplastics (Lusher 2015, Van Cauwenberghe et al. 2015, Setälä et al. 2016 & Rochman et al. 2017). In order to allow for spatial or temporal comparisons and to provide recom-

recommendations regarding future studies, it is crucial that the strengths and limitations associated with each method, as well as the differences between methods, are better understood.

For this thesis, several different methods were developed, tested and applied for sampling and characterizing microplastics. In **Paper I**, we used a manta trawl and an *in-situ* filtering pump to compare sampling methods in field tests, to test for variation between methods and to discuss suitable sampling strategies related to replication and sample size. We also present a protocol for visual identification of microplastics and discuss FTIR spectroscopy. In **Paper II**, we developed and applied extraction methods for screening invertebrate species, water and sediment samples for microplastics. In **Paper III**, we used a wide variety of methods to characterize material properties of polyethylene exposed to different levels of thermal degradation and field exposures. In **Paper V**, we sampled different types of beaches. We also combined the visual protocol outlined in **Paper I**, with image analysis. This chapter provides reflections on the lessons learned and recommendations for future studies for different types of matrices, methods and size classes. The main focus on this section will be on methods for particles above 0.3 mm, although **Paper II** also touches upon smaller particles and several of the conclusions are useful for other size-classes. The chapter is divided into sampling methods, extraction methods, visual identification and chemical characterization

## SAMPLING AND EXTRACTION

### Matrix

All methods come with different sets of limitations; these are important to understand in order to decide on strategies for sampling and sample treatment. For sampling, it is important to consider which matrix to sample in, as well as what sampling strategy and what method to use. A common matrix to study is surface waters. The results in **Paper I**, however, illustrate that surface waters are highly dynamic environments; therefore, samples originating from there might only give a snapshot of the current status. Moreover, the results are often hard to interpret since concentrations will depend on several factors including weather (Kukulka et al. 2012) and the tow directions of the manta trawl (**Paper I**). Additionally, surface waters often have low particle concentrations compared to other compartments of the sea.

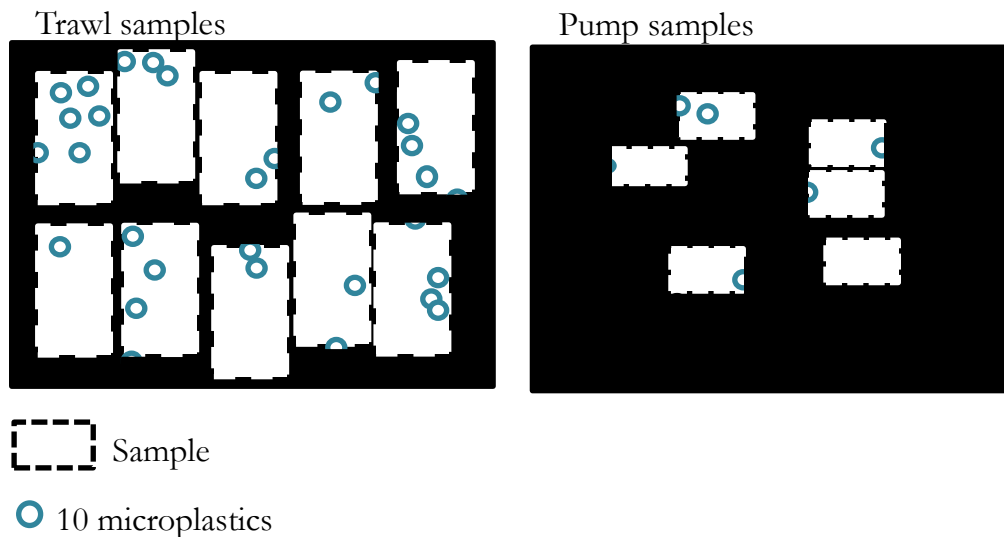
Beaches on the other hand can be expected to have higher concentrations. **Paper IV** illustrates how microplastics from local input will accumulate on beaches nearby. This was further confirmed in **Paper V** where concentrations in samples that had been pooled over a transect samples varied between 9 and 54 000 particles per kg d.w. For the samples taken in areas of the beach where litter had accumulated the highest concentration of microplastics of a size  $>300 \mu\text{m}$  was over 1 million particles/kg d.w. Although the concentrations are likely affected by a variety of factors such as tidal water, weather, topography,

substrate and vegetation, these samples show that exposed beaches are important hotspots for the accumulation of microplastics. Most of the plastic particles are, however, expected to sink. Accumulation sediment may therefore be the most suitable matrix to study plastic pollution over time, as we previously discussed in a policy brief regarding methods for studying microplastics (Setälä et al 2019).

### **Sampling strategy**

Three main strategies were used to obtain the samples for this thesis; bulk samples (i.e a smaller sample volume taken from a larger sampling matrix) (**Papers II, V**), concentrating larger volumes over a mesh (**Papers I, IV**) and pooling smaller subsamples (**Papers II, V**). Due to low concentrations and high variations, bulk sampling is likely only suitable in highly contaminated areas and for smaller size classes. Smaller samples can however be pooled, and in the screening study presented in **Paper II**, several individuals were pooled and subsampled for the biota samples. However, volumes of the subsamples in that study were on the lower end to allow for comparisons regarding concentrations or compositions. The number of particles obtained per subsample of the pooled biota (0-14 particles), means that the results are mainly useful as initial screening results. As such they do, however, confirm a widespread contamination of microplastics across several different phyla. The quantitative values should however be interpreted with care.

The results in **Paper I** further illustrate the importance of obtaining a suitable sample size in order to get enough particles per sample to decrease counting uncertainty. In Figure 1, the results from the trawl and the pump are used to illustrate that, while sampling an unknown concentration, the higher volumes achieved with the trawl give better representation of the complexity of the area. Even so, the particle numbers per sample for the trawl varied between 11 and 57. For the pump on the other hand, the particle numbers per sample varied between 0 and 13. It was concluded that for samples with less than 25 particles, the risk of obtaining false null values increases and would require extensive replication to allow for spatial and temporal comparisons.

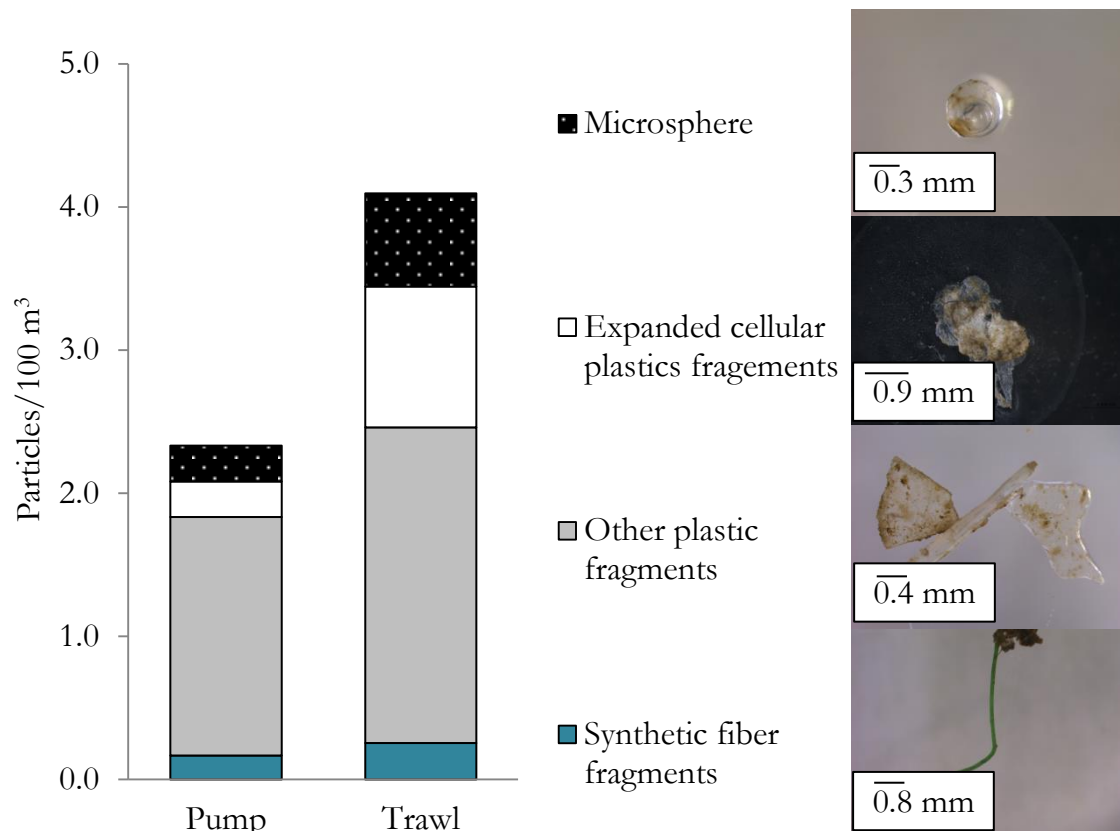


**Figure 1:** Illustration of the number of particles per replicate for the pump and the trawl. Trawl samples had a three times larger sampling volume ( $60 \text{ m}^3$ ) and therefore sampled more of the complexity compared to the pump. The pump takes longer to sample with resulting in lower sample volumes and fewer replicates. Due to the low sample volume ( $20 \text{ m}^3$ ) with the pump, the risk of obtaining false null values increased. Even with the higher trawl volume the amount of particles varied between 11 and 57 per sample.

The sampling volume should therefore be adapted for the study area as the concentration will differ dependent on where the samples are being taken. Typically, industrial areas and cities have higher concentrations (Yonkos et al. 2014, Mani et al. 2015, Hassellöv et al. 2018) but ideally pilot sampling should be done to assess the necessary sampling volume and replication. Power analysis for the trawl samples in **Paper I** showed that, in order to measure a difference between an area with an average concentration of 26 particles per sample and one with 52 particles per sample, at least 8 replicates would be necessary assuming the recorded standard deviation of 14 particles per sample to be representative. In order to measure a difference of 50 particles, on the other hand, only two replicates would be needed, assuming a similar standard deviation. To be able to do compositional comparisons the sample volumes in **Paper V** were therefore adapted to the local level of contamination, when possible, to ensure a high enough particle counts.

Regarding the choice of specific methods for sampling, it is important to know how they differ. In **Paper I**, field tests were conducted using a manta trawl and a filtering pump in order to compare methods for sampling surface waters. The tests were focused on particles above  $300 \mu\text{m}$  and showed that, since the trawl sampled higher volumes, the uncertainty related to counting statistics (number of particles per sample) was lower. Other than that, the trawl showed a higher concentration of expanded cellular plastics identified as polystyrene and air-filled microspheres identified as polymethylmethacrylate (PMMA); both of which are particles that typically float on top of the surface (Figure 2). This indicates that the trawl sampled particles floating on the surface more efficiently than the

pump. On the other hand, the sampling volume accuracy of the pump was higher and the contamination risk was lower, since only isolated parts of the pump were made from plastics. Additionally it provided a possibility of sequential filtration with filters of different sizes. All these factors are important to consider when deciding on a method as they will affect the results.



**Figure 2:** Composition for 6 replicate pump samples and 10 replicate trawl samples pooled and normalized to # of particles per 100 m<sup>3</sup> of water volume. Pictures show representative particle types for each category.

### Sample treatment

Within the research field, several methods are used to extract and isolate plastic particles from environmental samples and often adaptations have to be made for different types of matrices and polymers. For biota, plastics have been shown to be ingested by a large variety of marine species (Wright et al. 2013), and investigations of microplastic uptake are often based on stomach content analysis (Hidalgo-Ruz et al. 2012). However, it has been shown that microplastics may also be taken up via other pathways, such as the gills in crabs (Watts et al. 2014), and that translocation of smaller particles within the organisms

can occur in some cases (Browne et al. 2008). Full-body analytical approaches are therefore sometimes preferred and in **Paper II**, we tested and adapted one enzymatic method using Proteinase K that showed good recovery rates of spiked plastic particles. Since then, several other methods have been developed (reviewed in: Hermsen et al. 2018, Mai et al. 2018, O'Connor et al. 2019). The tests in **Paper II** show the importance of testing for recovery and assessing the effects the extraction protocol has on plastic particles, since a previously used protocol (also detailed in the paper) using chemical digestion showed low recoveries of spiked plastic particles and strong effects on the material; including melting, fusing and discoloration of the plastic particles.

The extraction methods for microplastics from sediments are often based on density separation through the addition of a high-density solution to the sample and the isolation of the particles that float to the surface (Claessens et al. 2013 & Nuelle et al. 2014). There are several types of methods available, but it is important to test for recovery here as well, since the plastics easily get stuck on glassware used during the separation process as noted in **Paper II**. Depending on the sediment, it can also be beneficial to apply a pre-treatment to decrease the “stickiness” and ensure efficient separation. In **Paper V**, sodium pyrophosphate was used to disperse the grains, but pre-treatments will need to be adapted to specific types of sediments.

### **Blank samples and contamination control**

Since plastics are commonly used in our everyday lives and can fragment during usage, another factor to take into account is the potential contamination of plastic particles during the sampling and sample treatment. This is often done through different types of blank- and contamination control samples. Blank samples do not contain an actual sample but are empty samples that undergo the same treatment, from sampling to analysis, as the field samples. Contamination control samples, on the other hand, can be specifically designed to test the background level of contamination in certain steps of the procedure, for example in the laboratory. In the studies presented here, the main focus was on blank samples. The results showed that, for studies of particles  $>300\ \mu\text{m}$ , blanks generally show low levels of contamination: in **Paper I**, only one particle was found among three blanks and in **Paper V** a total of 2 particles  $>300\ \mu\text{m}$  were found among 6 blanks. For studying smaller fractions, including fibers, the importance of a clean laboratory environment and blank samples is crucial, as noted in **Paper II**, where great care was taken to avoid contamination, but some contamination was still recorded. In another study, we looked at contamination levels in the lab and in the office (Rist et al. 2018). In samples that were air-exposed in the office, between 5 and 22 particles per sample were found. In the controls similar levels as noted in **Paper II** were found (average 1.7 particles per sample) (Rist et al. 2018).

## VISUAL IDENTIFICATION AND CHARACTERIZATION

It is becoming increasingly common to combine different types of spectroscopic methods, microscope-based visual identification is still the most common method for microplastic identification (Renner et al. 2017a). Although it is important to note that the human eye is quite good at sorting materials, this method has received criticism (e.g. Lenz et al. 2015, Löder & Gerdts 2015, Shim et al. 2017) since it can give both false negatives and false positives (**Papers I, V**) and is highly reliant on the experience of the researcher. Building and improving that experience requires good reference materials, clear protocols and the possibility to use other methods e.g. FTIR or Raman spectroscopy for particles that the researcher is not sure about. Still, for most published studies very little information is given as to how plastics were visually separated from other types of materials. Tests have also shown that the risk of underestimating the level of pollution is higher in smaller size fractions (Primpke et al. 2017). Therefore, unaided visual identification should rarely be considered suitable for quantitative measurements of particles below 100  $\mu\text{m}$ . For larger particles, objective protocols are key in order to compare studies. In **Paper I**, we therefore developed a protocol that can be further adapted to fit different types of microplastic studies. The analytical protocol has been developed through tests with experienced and inexperienced researchers and is divided into several categories that results in individual particle IDs for all particles. In **Paper V**, we further combined that protocol with automated image analysis of the particles to collect detailed information on particle sizes.

## SPECTROSCOPIC IDENTIFICATION AND CHARACTERIZATION

The use of polymer identification techniques is beneficial as they can increase the comparability between investigations, reduce false negatives and false positives and provide further insights into the sample composition. Even though there are other methods, such as pyrolysis GC-MS (Fries et al. 2013), the main focus here will be on the application of vibrational spectroscopy. Commonly used vibrational spectroscopy techniques are: Raman, Fourier transform infrared (FTIR) and near infrared (NIR) spectroscopy (Hidalgo-Ruz et al. 2012, Song et al. 2015). FTIR spectroscopy irradiates the sample with IR light. Some of the radiation is absorbed depending on interactions with the molecular vibrations in the material which then provides insight into the molecular structure. Several different modes and detectors are possible. Raman on the other hand uses a monochromatic light source (several different wavelengths are possible to use as light source). Similar to FTIR, the radiation then interacts with the molecules but here it creates a shift in energy for the scattered photons, which in turn provides information about the molecular vibrations. FTIR absorption depends on dipole moments and is therefore useful for detecting polar functional groups, such as carbonyl groups. The Raman signal, on the other hand, depends on changes in polarizability of chemical bonds wherefore it is useful in detecting aromatic

bonds, C-H and C=C. These differences make them complementary in studying plastic polymers (differences are further reviewed in K appler et al. 2016).

Similar to visual analyses, there are few protocols for analyzing environmental plastics with spectroscopy, and it has been emphasized that the parameters for spectral acquisition and especially spectral identification are rarely specified in scientific articles (Renner et al. 2017a). Spectra are often identified either automatically, based on similarities with reference spectra, or manually through comparing the spectra with known references and interpreting the peaks. Environmental transformations of plastics (such as oxidative weathering, hydrolysis, biofilm formation) give rise to new functional groups, chain scissoring, crystal state changes and consequently changes in the vibrational spectroscopic characteristic spectra (**Paper III**). As a result of biofilms, peaks decrease in height, broaden, and new distinct peaks appear, while broad regions characteristic of  $\text{-OH}$  and  $\text{-NH}$  groups appear (**Paper III**). Manual inspection and interpretation of the spectra, even though more time consuming than automatic identification based on library searches, allows for better identification of weathered microplastics (Renner et al. 2017b). Additives could also change the spectra to some degree. Identification of unknown particles is therefore not always (or rarely) possible with pristine polymer library matching, but it takes both environmental plastic reference libraries and some expert judgement to scrutinize the computer matching. As discussed in **Paper I**, authors often state that they have used a cut-off limit of a 60% or 80% match towards reference spectra, but without specifying which software that they use (e.g. Yang et al. 2015) or which library (e.g. Woodall et al. 2014, Avio et al. 2015, Yu et al. 2016), or which criteria/settings (e.g. Woodall et al. 2014, Yang et al. 2015, Avio et al. 2015, Castillo et al. 2016). Even if it sounds specific enough, that cut-off is therefore, in fact, rather nonspecific. Microplastics analysis using FTIR requires knowledge of polymer spectroscopy (Song et al. 2015, Mecozzi et al. 2016) and can be aided by better adapted pre-processing algorithms and analytical algorithms. One such example is the one developed by Renner and colleagues (2017b) who managed to increase the accuracy of identification from 76% to 96% compared to conventional library searches by limiting the comparison to regions with vibrational bands and applying new search algorithms (Renner et al. 2017b). It is important to note that, even with the improved identification rate, Renner and colleagues still recommend to visually double-checking the spectra after the automatic recognition.

Aside from determining the polymer type, it can be beneficial to include a characterization of the degradation of the plastics since this provides a better understanding of the material and its history. Plastics are often durable, and their longevity depends on a variety of factors including environmental factors related to light, heat and oxygen availability, material usage, and additives such as primary and secondary antioxidants and UV-stabilizers. Even so, before plastics enter the marine environment, they often have already experienced degradation, as the material starts degrading already during manufacturing and continues to degrade during its usage. It may also be exposed to degradation processes during



a period as terrestrial litter. Degradation and fragmentation can be analyzed using a wide variety of different methods as shown in **Paper III**, such as FTIR, differential scanning calorimetry (for semi-crystalline polymers), tensile strain at break, scanning electron microscopy (SEM) and image analysis.

There have been some attempts to estimate how long plastic samples have been in the environment, based on spectroscopic measurements of material degradation (Brandon et al. 2016). Subsequent discussions have, however, pointed out that the history of the material before and after it ended up in the ocean, along with the effect of different additives and other product characteristics, make estimates of the age of field-collected plastics unreliable (Andrady 2017), especially since these ageing processes are rather complex (**Paper III**). However, even if the specific age of the material may not be possible to determine from these measurements, their level of degradation still provides valuable insight into the sample composition. The observed changes in the FTIR spectra are also important to consider for the identification of plastics.

For microplastics, most techniques are still applied on a particle-by-particle basis. A drawback of this method is that it often requires that particles have to be visually identified as plastics, or suspected plastics, and then individually be tested spectroscopically. This can lead to an underestimation of the plastics in the samples (Karlsson et al. 2016). Applying hyperspectral imaging on full filters is a promising approach which combines the spatial (position on the filter) and the spectral information, and thereby can be used to identify plastics and separate them from other materials in the samples. This technique generates a big data set so it relies on the application of multivariate data analysis and data dimensionality reduction in order to distinguish plastics from the background (Karlsson et al. 2016). This approach can also be combined with newer techniques, such as focal plane array FTIR (e.g. Primpke et al. 2017), which show great promise for smaller microplastic fractions down to around 10  $\mu\text{m}$ .

## CONCLUSIONS AND RECOMMENDATIONS

Research on plastic particles in the ocean has evolved rapidly in recent years. With continuous calls for harmonization and standardized methods there has been a parallel realization that it might not be reasonable to expect a one-size-fits-all method with such a complex group of contaminants. Instead, focus has shifted towards developing and applying quality-controlled, tailored methods to fit the research question, practical limitations and the matrix in question.

Since the results will be highly influenced by the methods, the purpose of the monitoring or research study needs to be carefully designed so that suitable strategies can be used. For example, due to the high variations and short turnover times, surface waters are likely not suitable for long-term monitoring, but could instead be useful to test for differences in outflow from local point sources.

For sampling, the research presented here clearly shows that although different sampling methods can give different results it is, regardless of method, important to 1) get an adequate number of particles per sample in order to limit statistical measuring uncertainty, 2) report environmental data and characteristics, since they can affect the concentrations and 3) have enough replicates to correct for the inherent patchiness associated with the distribution and abundance of plastic particles in the environment.

For identification and characterization, it is important to realize that all methods have their inherent limitations and biases. Visual identification, supplemented by physical probing, can be very useful for larger particles ( $>100\ \mu\text{m}$ ) but does require training in order to increase accuracy and comparability. To decrease biases, it is also suggested that clear, structured protocols be used. Here, the method that was developed in **Paper I** can be a solid stepping stone towards a harmonized protocol. Similarly, for FTIR (and other spectroscopic techniques), the analyst should be trained to interpret the spectra correctly, and the protocols and algorithms used for library matching should be clearly stated. As illustrated in **Paper III**, it is also important to include the effects of degradation in the analysis of the spectra, and to use suitable pre-treatment of the samples in order to get reliable results.

## CHAPTER 2: FATE OF FLOATING PLASTIC PARTICLES

It has been estimated that 6 300 million tonnes of plastic waste have been generated up until 2015 (Geyer et al. 2017). Of those; 9% have been recycled, 12% incinerated and the remaining 79% have accumulated in landfills or in the natural environment; both on land and in the ocean (Geyer et al. 2017). In the ocean, different types of plastics will be transported in different ways. For materials with similar shape and size, higher-density material will have a more efficient sinking capacity, assuming that the material isn't air-filled, whereas light-density material will remain floating for a longer time span. The sinking rate will also depend on the size of the particle, as larger particles will have a faster sinking rate compared to smaller particles with the same density. Moreover, the sinking rate depends on the shape of the particles, which for plastics can be seen, for example, with fibers and thin films that have a higher hydrodynamic friction with the water. Additionally, plastics can be air-filled such as in the case of expanded cellular plastics which may remain buoyant even if the material density is higher than that of water. The water turbulence will also affect particles with different sinking rates in different ways (Ruiz et al. 1996). Additionally, floating plastic particles are influenced by several factors such as biofouling, degradation (**Paper III**) and aggregation (Allredge et al. 1990, Long et al. 2015) which could affect their vertical and horizontal transport. The processes affecting the transport and fate of plastic particles are, however, complex and other researchers have expressed an urgent need for a deeper understanding of them (Critchell & Lambrechts 2016, Jahnke et al. 2017, Rummel et al. 2017).

In this chapter, these processes will therefore be investigated through combining results and conclusions from studies on field samples, models and *in-situ* tests on how plastics are affected by degradation and biofilm formation. As the fate and degradation processes will differ greatly between polymer types, this chapter focuses on polyethylene, which is the most commonly used plastic polymer (Plastics Europe 2018). Most of the produced polyethylene is used in packaging (63%) (Geyer et al. 2017) but since it is a versatile polymer it is also used in toys, houseware, construction materials and electronics (Plastics Europe 2018). Polyethylene is also often specifically reported in plastic pollution studies (Andrady 2017). In the work included in this thesis, it was common both on beaches (**Paper IV, V**) and in surface waters (**Paper I, II**). Polyethylene has a density lower than 1 g/cm<sup>3</sup>, and would therefore typically float in the ocean, but polyethylene bags were found on the bottom of the sea, with signs of weathering and biofouling, already in the 1970s (Holmström 1975). Therefore, it can be assumed that the fate processes are more complicated than what may be expected from material density alone.

This chapter is divided into horizontal transport, degradation and vertical transport of plastic particles. It builds primarily on work done in **Papers III** and **IV**. But it also touches on results from analyzing field samples in **Paper I** and **V**. In **Paper III**, we investigated

the fate and degradation of plastic films in surface waters through field tests. In **Paper IV**, we combined measurements with drifters and calculations with field measurements to understand the transport pattern of plastic pellets in coastal waters.

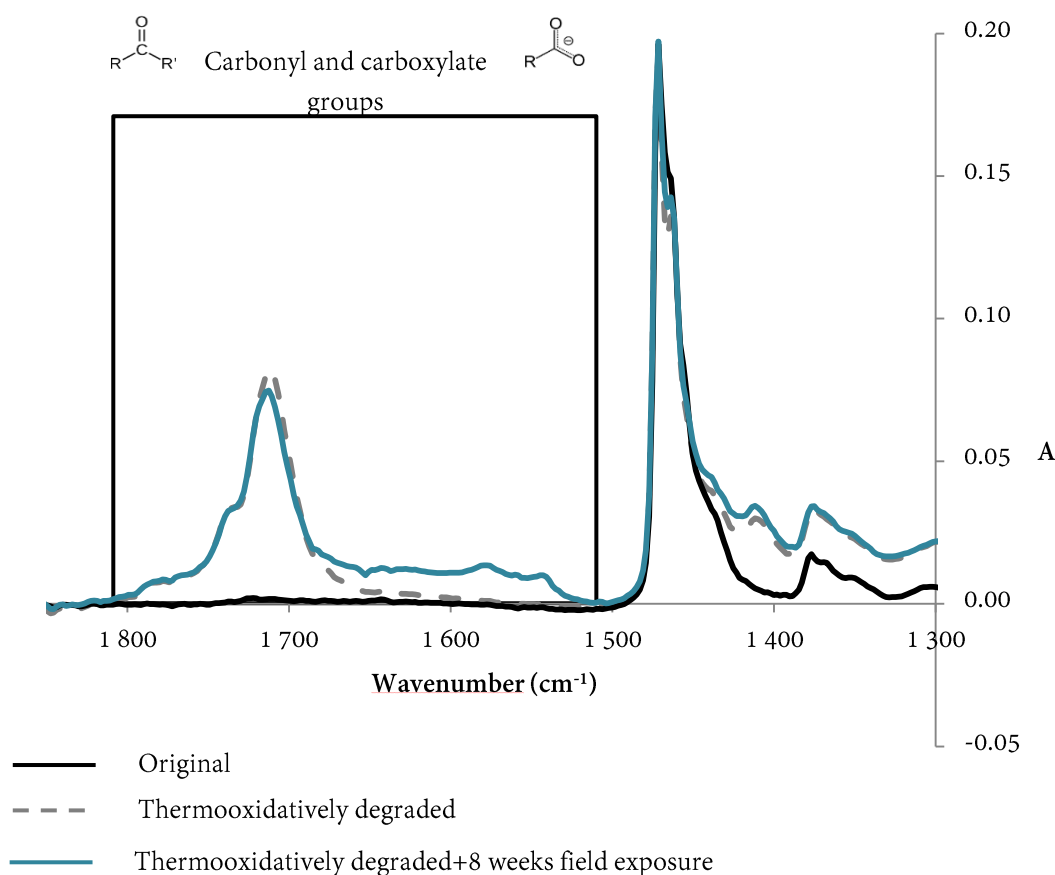
## DEGRADATION AND FRAGMENTATION

In order to understand the fate of polyethylene, it is necessary to understand the processes that can affect the material. This includes degradation and fragmentation. Although plastic degradation has been extensively studied for industrial purposes, there is a limited understanding of what happens in the field. Commercial polyethylene has a long service life because of the presence of antioxidants and stabilizers. The properties and longevity vary depending on the service conditions in different application's demands, and the usage of different additives (Gewert et al. 2015). Moreover, plastics in the marine environment, compared to plastics on land, are often subjected to lower temperatures, limited access to oxygen and lower ultraviolet (UV) radiation which can contribute to a lower rate of degradation (Pegram & Andrady 1989, Andrady et al. 1993, Muthukumar et al. 2011). Although the mineralization of plastics is expected to be a slow process, the first signs of degradation, such as reduction of tensile strain (O'Brine & Thompson 2010) and fragmentation (Weinstein et al 2016), can be seen already after 4-8 weeks.

When polyethylene is subjected to warm temperatures, ultraviolet radiation, shear stress and/or catalyst residues, a formation of alkyl macro radicals will cause degradation (Selonke et al. 2012). Oxygen reacts with the polymer and changes the chemical structure (Gewert et al. 2015) and the following reactions then lead to formation of degradation products such as ketones, aldehydes, alcohols and carboxylic acids. The degradation is dominated by the formation of carbonyl groups and vinyl species (Krehula et al. 2014). Once the plastics reach a low molecular weight distribution, the degradation products can be used by microorganisms as nutrients, thereby producing CO<sub>2</sub>, water and biomass (Albertsson et al. 1993, Chiellini et al. 1995, Prasun et. al 2014). The degradation pathways may however differ dependent on the environment in which the plastics are in, and the pathways in the marine environment are less studied than those in terrestrial matrices.

In **Paper III**, polyethylene degradation and biofouling was investigated. Polyethylene films were pre-degraded to 4 different levels of degradation, through thermooxidative degradation in a heat oven at 90° C for 0, 20, 27 and 30 days. Following thermooxidative degradation the films were cut with a scalpel into pieces of 0.8×0.8 cm. In addition, 9 larger pieces of 15×7 cm were cut from each level of degradation. The pieces were then added to stainless steel cages attached to a floating pier outside Kristineberg Marine Research station, in the Gullmar Fjord on the Swedish west coast on the 22nd of July 2016. Samples were taken after 4, 8 and 12 weeks in order to investigate their continued oxidation, biofouling and effects on material and apparent density.

Through analysis with FTIR spectroscopy it was shown that the thermal pre-degradation caused the formation of esters, peracids, acids and ketones (Figure 3). Already after 4 weeks in the water, the plastic particles started showing an additional formation of carboxylate groups and some signs of the formation of vinyl groups as well as an increase in the hydroxyl groups. As for the polyethylene films that had been exposed to the longest period of heat treatment, the carbonyl index (which is commonly used to assess polyethylene degradation (Jakubowicz 2006, Andrady 2017)) seemed to decrease. This may be a consequence of mineralization of the degradation products by microorganisms, although this assumption needs to be further investigated. The changes in the polymer spectra obtained after the combined heat treatment and field exposure were however complex. In relation to the internal CH- peak the total area for carbonyls and carboxylates also increased, a measurement that turned out to be more representative for the spectral changes. These results highlight the complexity of plastic degradation in different environments.



**Figure 3:** Degradation patterns of polyethylene as observed with FTIR. Shown here is a reference sample of the non-degraded material (black), a material that has been thermally degraded to level 4 (tensile strain at break 14%) (dotted grey line) and a material from level 4 that has been in the field for 8 weeks (blue line).

Another noticeable change was that larger pieces (15 cm) soon started fragmenting. The material that was the most degraded at the beginning of the experiment started fragment-

ing already after 2 weeks, the second-most degraded after 3 weeks and the least pre-degraded after 12 weeks. Moreover, SEM imaging revealed small cracks along the surface of the most degraded material. Similarly, the smaller pieces (5 mm) fragmented with increasing frequency with time and level of degradation. After 12 weeks, more than half of the pieces from the second-most and most pre-degraded material had fragmented. Pieces that had not been pre-degraded did not fragment but a decrease in tensile strain at break from 110% to 88% was measured after 12 weeks of exposure. Although expected, this highlights the importance of fragmentation of macroplastics in the formation of microplastics.

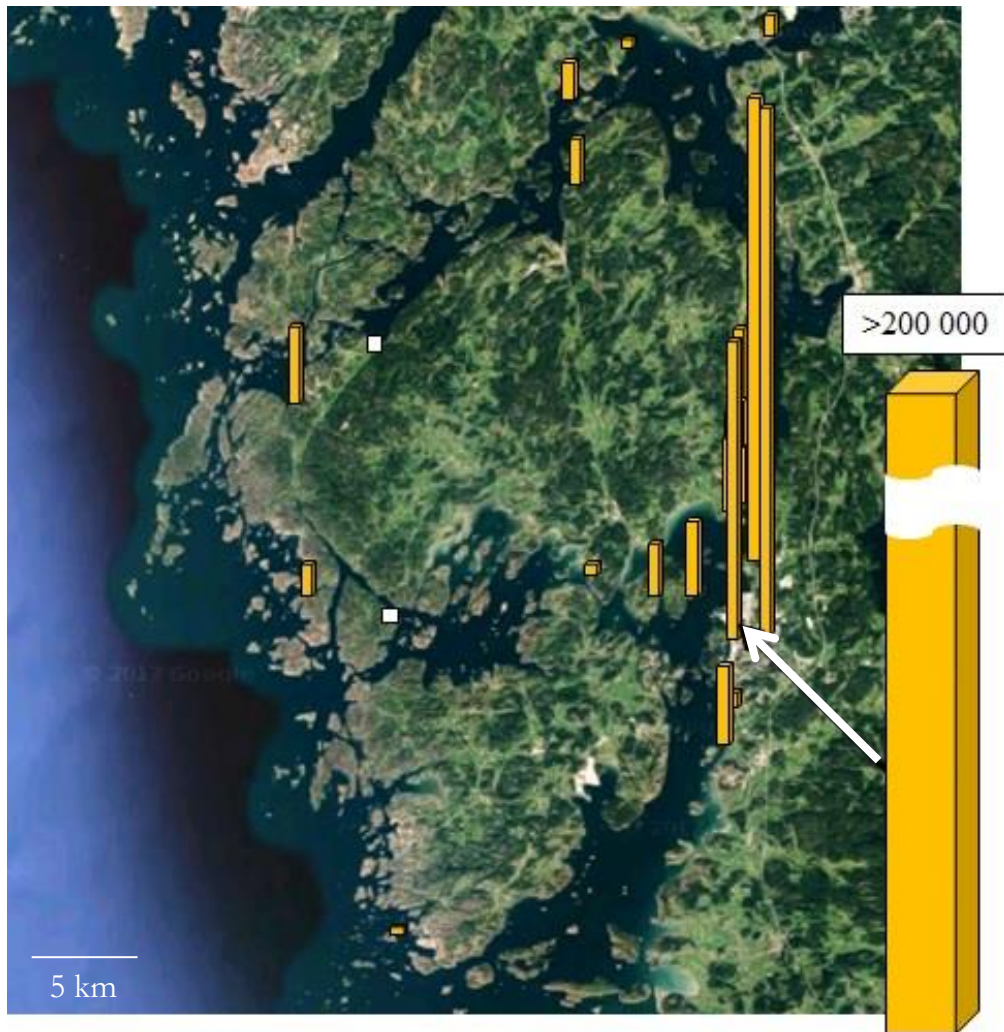
## HORIZONTAL TRANSPORT

The transportation of plastics from terrestrial to marine environments is often dependent on rain and runoff. A significant part of the transport is also expected to be via horizontal transport through rivers and other waterways (Lebreton et al. 2017). Once in the marine environment, floating plastics can be transported horizontally over long distances and their distribution often follows ocean surface circulation models (Law et al. 2010, Van Sebille et al. 2015). This long-range transport can for example be seen in plastic pellets, which have been documented in the environment since the 1970s (Carpenter et al. 1972, Carpenter & Smith 1972). Since then, pellets have been found in surface water samples and on beaches all over the world (Colton et al. 1974, Morris & Hamilton 1974, Gregory 1977, Eriksen et al. 2013, Fernandino et al. 2015) – even on beaches that are not directly in contact with petrochemical or polymer industries (do Sul et al. 2009, Fok & Cheung 2015). For the north-eastern Atlantic, simulations show that a large portion of floating particles that are released in this area will be transported to the Arctic (Cozár et al. 2017, Lachmann et al. 2017). There is, however, a discrepancy between the amounts of plastics that are expected to enter the oceans and what is found in surface waters (Thompson et al. 2004). Although there are several processes that can explain this, such as sinking and fragmentation, it can also in part be explained by beaching.

In earlier works we have applied models to show the effect of beaching (Lachmann et al. 2017). The models showed that a significant portion of particles discharged in the North east Atlantic would also get stuck along the beaches in the Skagerrak area. The stranding of litter along the shoreline of the Skagerrak region is also observed in beach litter monitoring programs along the Swedish west coast. In fact, it has long been known that due to the prevalent directions of winds and currents, plastic debris often accumulates along the exposed beaches on the Swedish west coast, from both long and short-ranged sources (Håll Sverige Rent 2014). The high concentrations shown in **Paper V** further illustrate the importance of beaching in explaining the fate of plastic particles.

The propensity for floating plastics to beach was further confirmed in a study by Critchell and Lambrechts (2016). They modelled the release of microplastics and showed that the seeding location (i.e. the location in which the particles were added) was an important factor in determining the horizontal distribution of plastic particles. They showed that if the particles were added closer to the coast, they had a lower latitudinal distribution (Critchell & Lambrecht 2016); meaning that they remained in the area nearby. For local, coastal sources, the studies on pellet release can provide further insights into the propensity of plastics to beach within the archipelago (**Paper IV**). In the study, the pellet distribution within a case study area, in the close vicinity of a plastic production plant, was examined using field measurements, drifters and modelling. The area consists of a network of islands on whose beaches the pellets could be washed up. If the pellets that leaked from the production plant had not beached within the area, a steady state of the pellet pollution level would be reached after 50 days, matching the water exchange. However, by

using drifter studies it was shown that the typical floating distance until beaching would be between 0 and 5 km. This fits well with what was observed in subsequent field studies where the number of pellets found by one person within one hour was assessed (Figure 4). These measurements and calculations highlight the importance of including beaching and remobilization trends when studying plastic distributions, especially when the sources are not situated directly at open coasts.



**Figure 4:** Relative abundance of plastic pellets, as counted on beaches in the area by one person during one hour per spot. Bars are illustrative of the relative number of pellets and the thicker bar in the front corresponds to a sample taken at the mouth of Stenunge å, where a high accumulation of pellets was observed. (Reprinted with permission from Marine Pollution Bulletin).



## VERTICAL TRANSPORT

Although floating plastics, such as polyethylene, can be transported horizontally over long distances, surface waters typically have low concentrations of plastics, as noted in **Paper I**. Some of this can be explained by beaching, as discussed in the previous section. However, models also show that a majority of the input of plastics will sink (Koelmans et al. 2017a) and experiments show that the sinking behavior will depend on several factors, including material density, particle size and shape (Kowalski et al. 2016). Plastic particles ingested by zooplankton may be incorporated into fecal pellets and sink with them to the bottom (Cole et al. 2016), but they may also be affected by (bio-)fouling and changes in material density. Due to the limited data available on these processes, scientists have expressed the need for specific knowledge on how the vertical transport is affected by degradation and biofouling (Critchell & Lambrechts 2016, Jahnke et al. 2017, Kowalski et al. 2016, Rummel et al. 2017).

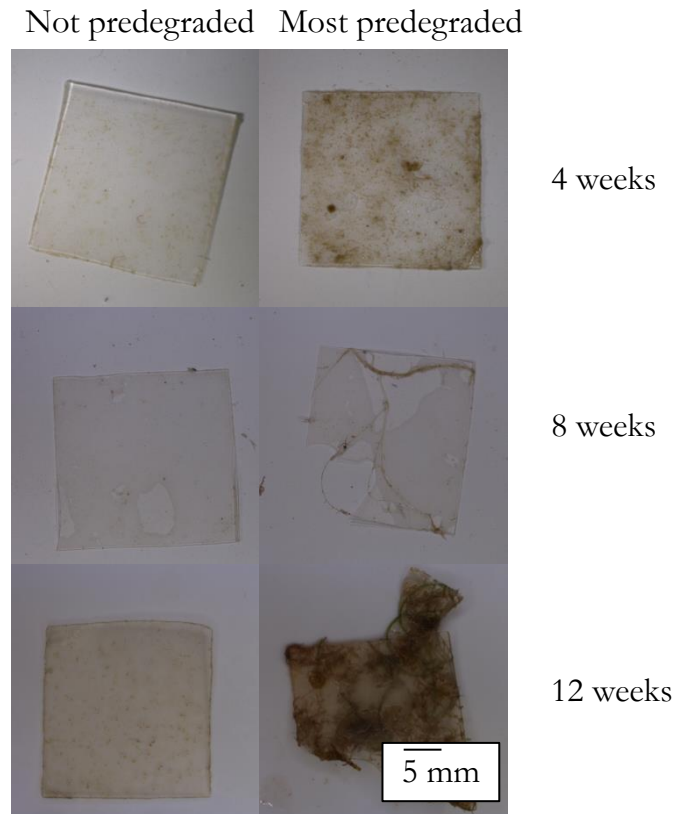
In **Paper III**, the combination of increasing levels of biofilm formation and oxidation soon resulted in more neutral buoyancy of the polyethylene pieces in the field. At the end of the experiment (12 weeks), several of the particles had sunk to the bottom of the cages. Density measurements confirmed that the uncleaned particles had increased in apparent density. Additionally, measurements of cleaned particles also revealed a slight increase in material density (Table 2).

**Table 2:** Summary of results related to biofilm, material density and apparent density for original and the most pre-degraded polyethylene after 0 and 12 weeks of field exposure (**Paper III**).

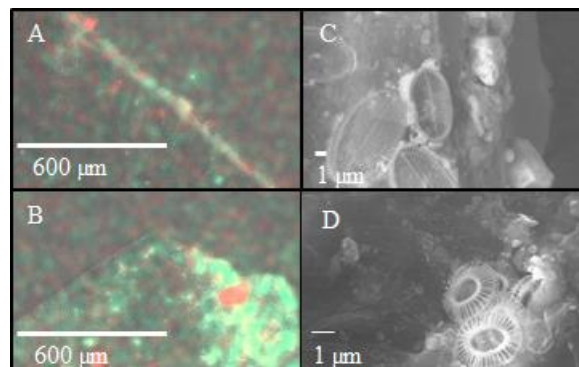
	0 weeks		12 weeks	
	Original	Pre-degraded	Original	Pre-degraded
Biofilm (% weight)	0	0	14	41
Biofilm (% coverage)	0	0	88	93
Material density (kg/m <sup>3</sup> )	919	947	920	951
Apparent density (kg/m <sup>3</sup> )	919	947	917	980

For the plastic films used in **Paper III**, biofouling continuously increased throughout the weeks with similar levels of coverage (% of surface covered) for all films but with a higher weight of the total biofilm and fouling organisms for the pre-degraded levels (Figures 5 & 6). A typical succession order was observed, with a layer of biofilm consisting of microalgae, macromolecular adhesion bacteria and other microorganisms forming first, followed by an algal mat, and then invertebrates. The formation was however seen to be faster for

the pre-degraded material, especially for the two most degraded levels. This could be because the pre-degraded material is less hydrophobic which facilitates adherence. The pre-degraded material also showed several micro-cracks on the surface, which may have created a more favorable surface for fouling organisms.



**Figure 5:** Representative pictures showing the changes throughout the field exposure for the plastic pieces that had not been pre-degraded and the most pre-degraded level.



**Figure 6:** Non- degraded (A) and pre-degraded (B) PE pieces after 12 weeks field exposure analyzed with SEM-EDS scans. The green color indicates silica, predominantly due to diatoms on the surface (C); the red color indicates calcium, which was mainly due to coccoliths (D). (Reprinted from Marine pollution bulletin (**Paper III**) with permission).

In terms of transport patterns related to material properties, it is also interesting to note that we found marked differences on beaches depending on whether they were exposed beaches, rural beaches subject to pollutants that had been transported further with cur-

rents, or beaches close to urban areas. In **Paper V**, polystyrene was, for example, almost exclusively found on urban beaches. This corresponds to our earlier measurements in the Oslo Fjord that showed higher concentrations of polystyrene closer to cities (Hassellöv et al. 2018). It could be conceived that the air in the expanded polystyrene gradually disappears, and that the particles then lose their buoyancy, and therefore are rarely found on more exposed beaches or in surface waters in more pristine areas.

As these effects may be dependent on seasonal and regional processes, the effects of biofilm formation and degradation on polyethylene should also be studied in different types of environments and during less biologically productive seasons. Moreover, different types of plastics are likely to behave in different ways. For some areas and during parts of the year where the biological productivity is high, these processes can be fast. This was seen in **Paper III**, and demonstrates why a higher sampling frequency could give even better insights into these processes. Furthermore, it would be valuable to combine a similar setup with sedimentation studies through constructing deeper cages allowing for a more controlled sampling of the particles that sediment.

## CONCLUSIONS AND RECOMMENDATIONS

A majority of all plastics ever produced still exist, and although most are still being used or are deposited in landfills, a significant portion has leaked out into aquatic environments (Geyer et al. 2017). Once there, different types of plastics will behave differently. Buoyant plastics can be transported far, as shown by pellets that have been encountered on remote islands far from petrochemical industries. But the mechanics of horizontal transport are not entirely straightforward as could be seen in **Paper V** where measurements with drifters, calculations and field measurements all confirmed that a majority of the pellets released in the case study area ended up on beaches nearby. It is, however, not known how long they then reside on the beach until they re-enter the ocean. The transportation of plastics in aquatic environments gets even more complex when other factors such as material size, density and shape are included. Even when restricted to one material class, such as polyethylene, this encompasses several different material grades with different properties, which may be further differentiated through the usage of additives. Here, we see that once the plastic ends up in the environment, processes of degradation, fragmentation and biofouling will further alter these properties. Through focusing on polyethylene, we show here that although floating plastic particles can be transported far, they are also prone to beaching and sinking. The results show that their fate is highly dependent on interactions with natural processes which need to be taken into account when discussing the fate of plastics. The results also highlighted that there is a strong interdependency between degradation, fragmentation and biofouling; meaning that a more degraded piece of plastics will fragment easier and will have more biofouling than a non-degraded piece of plastics. Studies aiming to explain transport patterns of plastic marine debris should therefore consider the history of the material, in terms of degradation and biofouling, and

the effects that these processes may have on the properties of the material and its final fate.

Taken together, the results from modelling, field tests and environmental samples show that 1) floating plastics are often likely to beach close to the source, 2) larger pieces will fragment with increasing rates as the material degrade and 3) plastics that are initially floating are likely to end up sinking due to changing material properties as a result of degradation and biofouling. These results have consequences for monitoring as well as solution oriented initiatives. Since most of the plastics that enter the marine environment could be expected to eventually beach or sediment, monitoring or cleaning initiatives focused on surface waters are only targeting a small fraction of the problem. Moreover, plastics that have sunk or fragmented will be even more challenging to retrieve and therefore solution-oriented initiatives need to focus on intercepting the waste closer to the source.

## CHAPTER 3: SOURCES OF PLASTIC PARTICLES IN NORTHERN EUROPEAN WATERS

The question “where do plastics in coastal waters come from?” can be answered in a few different ways. The most straightforward answer is probably “from us”. But in order to work efficiently with improving preventive strategies we may need to add some more complexity, especially since sources of plastic particles are almost as complex as the areas of usages are multifaceted.

This chapter aims to provide an overview of sources of plastic particles in coastal environments in northern European waters. Since different methods have different limitations, field measurements in surface waters, sediment, beaches and biota from **Papers I, II, IV and V** will be compared to macro litter found on beaches, plastic usages and material flow analyses.

### PLASTICS USAGE AND MATERIAL FLOW ANALYSES

In 2017, 348 million tonnes of plastics were produced globally. Of those, 18.5 % were produced in Europe (Plastics Europe 2018). Plastics Europe reports that 39.7% of the plastic converter demand (i.e. the processing of plastic raw material into industrial and consumer products) are for packaging and 19.8% go to construction. In addition, 10.1% go to the automotive industries, 6.2% to electrical and electronics, 4.1% to household, leisure and sports usage and 3.4% to agriculture. The remaining converter demand consists of a wide variety of products such as furniture, medical equipment and technical parts. In terms of polymer types, the most common are polyethylene (29.8%), polypropylene (19.3%), polyvinylchloride (10.2%), polyethylene terephthalate (7.7%), polyurethane (7.7 %) and polystyrene (6.6%) (Plastics Europe 2018).

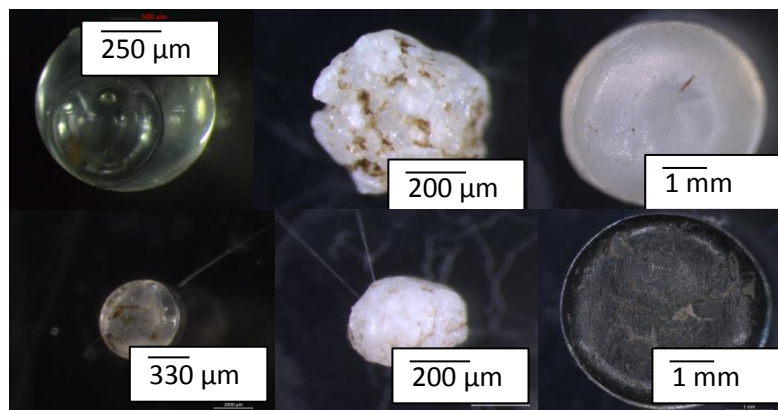
It should be noted that the usage does not directly reflect the pollution pattern since certain products are more likely to end up in the environment (Kawecki & Nowack, 2019). Additionally, there are large transnational movements of plastics through import and export. So, in order to better understand the regional pollution patterns, estimates have been made of the input of different types of microplastics for several countries in north-western Europe (Sundt et al. 2014, Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016). These types of estimates are limited, especially due to the fragmented data available, and the calculations build on sets of somewhat different assumptions. Nevertheless, they are reasonably comparable and through normalizing them to annual mass emissions per capita (in g/person) they help complement each other. The normalization to emissions per inhabitants is, however, a crude measurement and some types of emissions are likely to be more dependent on the amount of industries rather than inhabitants.

Common for the studies is that primary microplastics are less than 100 g/person/year, whereas microplastics generated from wear during usage are about seven times as high.

Most estimates did not include fragmentation from larger plastics, which could explain why estimates of microplastics fragmented from macroplastics post usage are relatively small. A more recent analysis of seven commodity polymers and their emission factors as macro and microplastics estimated that, by volume, macroplastics emissions are 12 times as high as microplastic emissions (Kawecki & Nowack 2019). It is reasonable to assume similar ratios in northern European coastal areas since the infrastructure is comparable. In regions with less advanced waste management the fraction of macroplastics can be expected to be even higher.

### Primary microplastics

Primary microplastics are defined here as manufactured plastic particles below 5 mm in size (Figure 7). This includes preproduction pellets and fluff (a smaller preproduction material, often used to make pellets), particles for cosmetics, abrasive media, paint formulation ingredients, and plastic particles used in medical predicts and in research. Taken together they are an important part of the overall release of microplastics into the marine environment.



**Figure 7:** Examples of primary plastics found in environmental samples. From left to right: microspheres, fluff and pellets.

In regional estimates, the leakage of primary microplastics in cosmetic products is, however, considered a relatively small contribution to the overall load of microplastics in the marine environment (Sundt et al., Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016). This type of particle can on the other hand be argued to be an unnecessary source, since it can easily be replaced by other materials (Fendall & Sewell 2009). Microplastics in medical products, on the other hand, might be more functionally important but are also not expected to leak much.

Another important source of primary microplastics is preproduction pellets, which enter the environment through spills during production, transport and storage. These have been found in the marine environment since the 1970s (Hays & Cormons 1974). Research has shown that, following the implementation of some preventive measures, the levels of plastic pellets in the environment decreased between 1986 and 2008 (Law et al. 2010). As seen in **Papers IV and V**, spills are however still common.

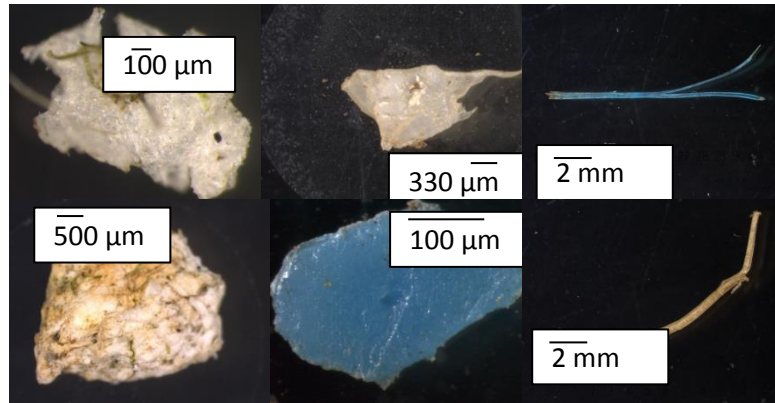
Material flow analysis and modelling have applied various emission factors (calculated estimations of leakage) to account for spills. In Norway, a factor obtained from communications between the Norwegian EPA and a factory which indicated a release of 0.04% of the produced volume, which gave an estimate of 2500 tonnes per year (Sundt et al. 2014). In Denmark, Lassen and colleagues (2014) instead used an emission factor based on a survey carried out by the Danish Plastics Federation (Lassen et al. 2014). Although only three respondents admitted to spills, Lassen and colleagues used these numbers to calculate an emission factor between 0.0005% and 0.01% of the produced plastics. The study in **Paper IV** showed a lower annual continuous release between 0.00004% and 0.0004%. This release was only from the production plant and it was noted that additional spills happen during overflows, storage and transportation and therefore the emission factor calculated by Lassen and colleagues (2014) seems realistic.

Assuming that the level of leakage of primary plastics we observed in **Paper IV** is somewhat representative for other European production plants, a rough estimate would be an annual leakage between 26 and 260 tonnes, based on overall European plastics converter demand data (PlasticsEurope 2018). If the estimates calculated by Lassen and colleagues (2014) are representative for the overall production and transportation chain, this would instead amount to an annual release of 300-6 000 tonnes due to leakages associated with production, transportation and storage of plastic preproduction materials in Europe. The data from regional estimates (Sundt et al. 2014, Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016) all show that leakages of preproduction materials from the production and transportation chain are about 30 times higher than the leakage of microplastics from cosmetics and other consumer products.

### **Secondary microplastics**

Secondary microplastics are often defined as plastic particles below 5 mm that are generated through wear and tear during and after usage (Figure 8) (Hartmann et al. 2019). According to the regional estimates three main categories of particles that occur due to wear during usage are particles associated with shipping and marinas, textiles and tires (Sundt et al. 2014, Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016).

The regional material flow analyses from Sweden indicate that particles generated through wear of boat paint, floating docks and marinas amount to 48-158 g/person/year (Magnusson et al 2016). These are not the most dominant sources of pollution to the environment overall, but since they are directly shed into the marine environment they make up an important fraction of the microplastic litter. Boat paint can be particularly troublesome since it often contains additives intended to prevent fouling such as copper, zinc and biocides.



**Figure 8:** Examples of secondary microplastics found in the environment. From left to right: expanded cellular plastics, other fragments and synthetic fiber fragments.

Textiles on the other hand, according to the regional estimates, are expected to provide a smaller source to the marine environment (0.4-10 g/person/year) (Lassen et al. 2015, Magnusson et al. 2016) since many of them are removed in waste water treatment plants (Talvitie et al. 2017). But since roughly half of the sewage sludge in Europe is used to fertilize fields or other land masses (Davis & Hall, 1997, Kelessidis & Stasinakis 2012), the fibers are still likely to end up as pollutants in the environment.

Tires are often discussed as a major source of plastic particles in the marine environment and the regional estimates range between 87-418 g/person/year (Sundt et al. 2014, Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016). The high number can in part be explained by the fact that it is relatively easy to estimate the input or loss rate. But it is also a consequence of current high levels of traffic. In addition, storm waters are often led to water recipients without any treatments (Raspati et al. 2017), further spreading the pollutants in the environment. Moreover, scrap tires are often used as filling materials for artificial turfs, which have also shown to leak into the environment (Krång et al. 2019).

Post-usage macroplastics that end up in the environment can be expected to generate increasing levels of microplastics as the material degrades, as shown in **Paper III**. For some plastics it is hard to draw a line between wear during and post usage. One example is agricultural plastics that can spread both during and after use. Annually, about 6.5 million tonnes of agricultural plastics are produced (Scarascia-Mugnozza et al. 2011) and in Europe, 3.5% of the total conversion demand are for agricultural plastics. Common usages include plastic mulch films which can rarely be recycled (Sintim & Flury 2017). Additionally, there is concern that the plastic fragments generated during usage are left in the soil (Sintim & Flury 2017). In Switzerland, Kawecki and colleagues (2019) showed that agricultural plastics are a major source of plastics to soil environments, but there are currently no reliable estimates on the leakage into marine environments, and agricultural plastics are often excluded from these types of estimates (Sundt et al. 2014, Lassen et al. 2015, Sherrington 2016, Magnusson et al. 2016).



One important example of mismanaged plastic waste is lost, abandoned or derelict fishing gear. This type of litter, often referred to as ghost nets, is known to occur in high volumes in the environment (Brown & Macfadyen 2007). They also have several documented negative effects especially related to continued fishing (Macfadyen et al. 2009). In the marine environment, fishing gear has also been shown to fragment and thus to release microplastics (Montarsolo et al. 2018). Due to the lack of reliable data it is often not included in estimates of microplastics. It was however included in the Swedish estimates where it when recalculated to microplastics/g/inhabitant, corresponds to 2.5 g per year (Magnusson et al. 2016), making it one of the smaller contributions to the overall microplastics load.

When data from Sweden, Denmark, Norway and the UK is compared and normalized to tonnes per million inhabitants, it indicates that fragmentation of particles from larger municipal solid waste items is an important source of microplastics in the marine environment even though there is a large uncertainty span (Jambeck et al. 2017, Geyer et al. 2017). Fragmentation from macrolitter is often excluded in material flow analyses due to lack of data, while estimates that included this information suggest that it is one of the main sources of microplastics. Similarly, in Switzerland Kawecki et al. (2019) showed that consumer packaging was the largest macroplastic emission to soil. This can be due to intentional and unintentional littering and due to leakages in waste management systems.

## COMPOSITION IN FIELD STUDIES

### Macroplastics

Schwarz and colleagues (2019) reviewed existing literature on the composition of plastic litter in the environment and found that while packaging and consumer products were the most common litter types found in rivers, fisheries items were dominant in marine environments. They also reported that plastics from transport, building and construction, and industry were not observed in any of the studies. This conclusion is, however, based on only two surveys, one of which had classified 93% of the plastics as “unknown” (Schwarz et al. 2019). Larger data sets such as the OSPAR marine litter surveys might be more suitable in comparing litter types, although the comparability between locations in those surveys has also been questioned (Pfeiffer 2019).

A summary of beach litter data from European cities shows that the most common litter items are plastic and polystyrene fragments, followed by lines and cigarette butts (Addamo et al. 2017). There are regional differences and in order to compare the microplastic data for the exposed beaches that we sampled in **Paper V**, the OSPAR data for those specific locations were summarized. Amongst the 56 957 objects that were registered between 2001 and 2017, 98% were plastics. Of the plastics, 38% were strings (ropes and lines with a diameter less than 1 cm) and 29% were plastic pieces of varying sizes. Other common plastic categories included; caps (5%), ropes (5%) sanitation products (3%),

industrial plastics (2%), plastic bags (2%), chips bags (2%), packaging strings (2%) and fishing equipment (aside from rope) (0.5%). Strings, ropes and fishing equipment are all likely to originate from sea-based activities and together make up more than 40% of the found objects. On a global level, abandoned, lost or otherwise discarded fishing gear is also thought to be an important contributor to marine litter, since environmental studies have shown that they make up 1-46% of the litter, dependent on location (Gilman et al. 2016), which could indicate that sea-based activities are underestimated in the material flow analyses.

A less stringent reporting system than that of OSPAR was used by Ocean Conservancy where one million volunteers participated in beach cleaning initiatives in over 122 countries and picking up 10 000 tonnes of litter in one day (Ocean Conservancy 2019). Their results showed that cigarette butts were the most common litter item, followed by food wrappers, straws/stirrers, cutlery, plastic beverage bottles, plastic bottle caps and plastic grocery bags – all single use items. Similar results have been reported on Swedish beaches closer to urban areas, where approximately 70% of the plastics that were found were characterized as single-use items, with 68% of the total litter being identified as packaging materials (Håll Sverige Rent 2014).

For macrolitter, it is therefore interesting to note that the relative composition on beaches changes along the Swedish and Norwegian west coast. Although the examples mentioned above use different protocols, comparisons of similar protocols show similar trends, such as an increased occurrence of single-use packaging closer to bigger cities whereas the northernmost part of Norway showed a higher fraction of derelict fishing equipment (Briedis et al. 2018).

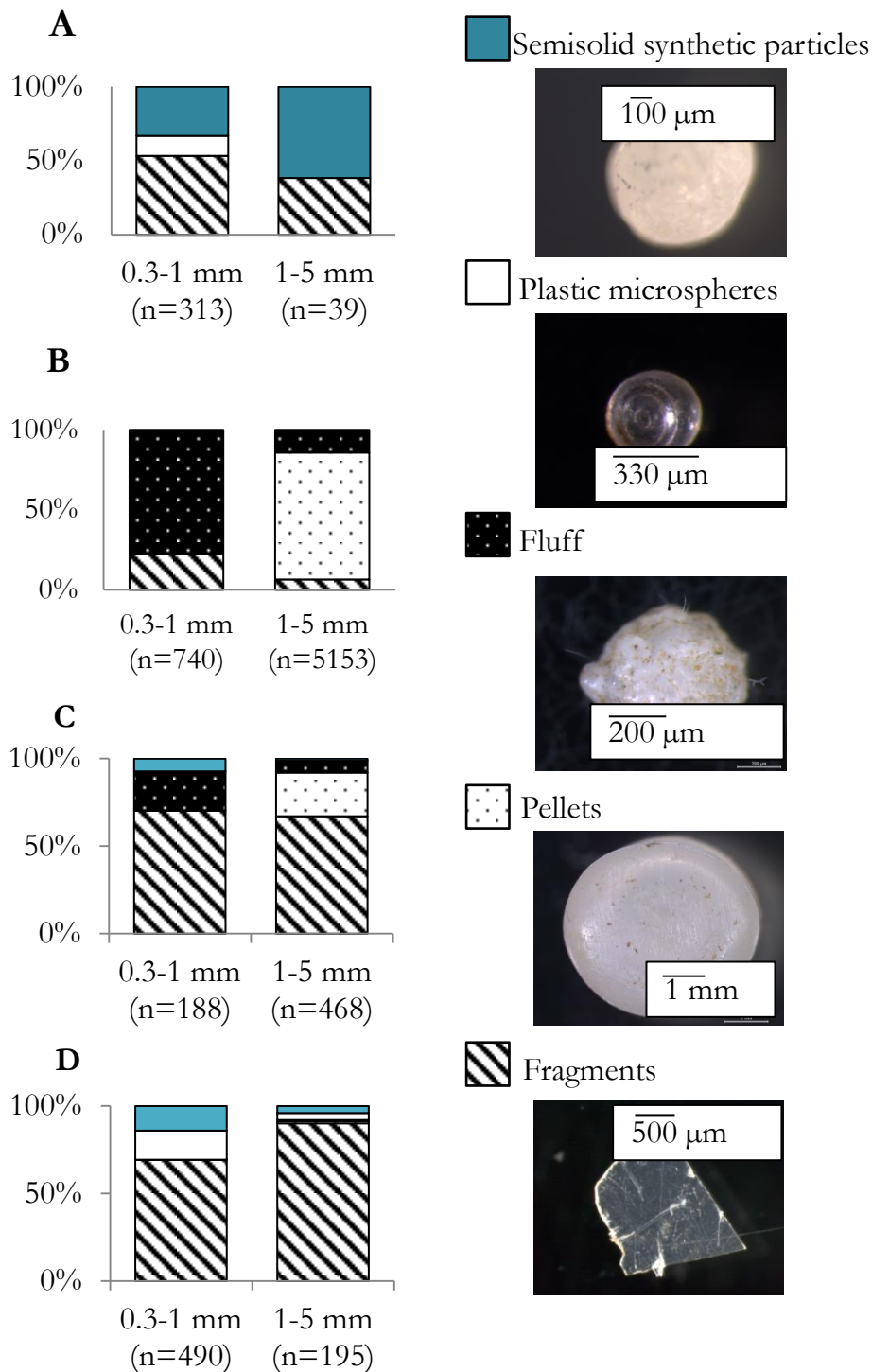
### **Microplastics**

Primary microplastics that are used in cosmetics and similar applications are often difficult to identify, as such, in environmental samples since they have several appearances. Some types of primary microplastics are, however, easier to quantify, such as preproduction pellets (also known as “nurdles”). When compared to other types of microplastics, pellets are relatively large and have a typical size of a few mm. In **Paper V**, field measurements confirmed that pellets and fluff make up important fractions on several beaches, both on those close to production plants as well as wave and wind exposed beaches located further away. **Paper IV** showed that, although there are no existing international frameworks or European laws specifically aimed at plastic pollution due to industrial spills, there are several European and national regulations and laws that are applicable. If they were to be fully implemented, they would improve prevention of leakage associated with production, transport and storage.

Aside from the pellets and the fluff identified in **Paper V**, beaches also frequently featured microspheres, spherical (or spheroid) particles which can be solid or hollow (the latter sometimes being called “micro balloons” in technical contexts). Chemical identifica-

tion in **Papers I** and **V** showed that these were either made up from PMMA or glass. Other studies have also found similar beads made up of polystyrene (e.g. Mani et al. 2014). PMMA microspheres can be used as raw material for a wide range of manufacturing applications, usually as light-weight fillers in polymer composites in aviation, furniture, lighting, construction and electronics (Mani et al. 2014). Most of the microspheres here showed no signs of degradation or tear and it is therefore plausible that they originated from spills of industrial raw materials.

A majority of the particles found in **Papers I, II** and **V**, as well as in our other environmental samples (Karlsson et al. 2016, Hassellöv 2018a, Hassellöv 2018b, Karlsson 2019) were, however, fragments (figure 9), often referred to as secondary microplastics. This indicates a diffuse and complex source pattern. The compositions differed depending on particle size and sampling location. In surface waters taken close to a smaller urban area on the Swedish west coast (Lysekil), 81% of the plastics were fragments (51% of the microplastic when paraffin was included). Of the fragments, 30% were expanded polystyrene. In the smaller fractions, PMMA microspheres were also a big fraction (18%). In surface water samples taken in Stenunge å, outside a plastics industry, the particle composition was instead dominated by pre-production pellets and fluff (**Paper IV**). In beach samples taken close to urban areas, the samples consisted of pellets, fluff and fragments; of which 30-94% were expanded polystyrene. On exposed beaches the samples also had a high percentage (50-100%) of fragments but only 0-42% of the particles were expanded polystyrene. Hollow microspheres were more common on the exposed beaches (**Paper V**), possibly due to their effective buoyancy.



**Figure 9:** Simplified particle composition in different types of samples with images of representative particle types; A) Surface waters collected with a trawl outside small urban settlement (**Paper I**) B) Surface waters collected with a trawl in a river outside a plastic production industry (**Paper IV**) C) Beach samples collected from 2 urban beaches along the Swedish west coast D) Beach samples collected from 2 exposed beaches along the Swedish west coast (**Paper V**). Number of analyzed particles denoted with n. Fragments are shown as combined results from different types of fragments including irregular, expanded cellular and synthetic fibers.

Except for the samples taken outside of the plastic industry, where preproduction materials dominated, it was noted that throughout the different studies there was a wide variety of different types of plastic particles. The particles differed in terms of size, level of degradation and shape but a majority of the particles were fragmented pieces that belonged to the main polymer groups of polyethylene, polypropylene and polystyrene. This corresponds to similar environmental studies in the literature (Schwarz et al. 2019, Wu et al. 2019).

The high percentage of polystyrene in locations close to urban areas is, however, especially interesting considering the relatively low percentage of polystyrene converter demand (6.6 % of the total European plastic converter demand). Although there could be a higher percentage total usage due to import it also illustrates that some types of plastics or plastic products can be especially prevalent due to how and where they are used.

In accordance with the field studies, the compilation of regional material flow analyses also indicated that fragmentation during and post usage would be the main source of plastics in the environment. According to those estimates, 8% of the plastics would be primary plastics such as pellets, fluff and microspheres. The environmental samples do however indicate a somewhat higher relative prevalence ranging from 19% in surface waters outside a smaller urban area to 91% in surface river waters outside a plastic industry. Although large regional differences are to be expected and the samples taken outside a plastics industry should not be treated as representative for environmental samples overall, these results illustrate that primary plastics are still an important contributor to the overall input of plastic particles in these size fractions.

As previously discussed, the composition varies depending on location, but it can also vary dependent on the matrix. Although floating particles can be expected to eventually sink as a consequence of degradation and biofouling as shown in **Paper III**, high density particles will sink more readily and therefore accumulate closer to the source. Moreover, as seen in **Paper I**, the sample composition can differ depending on sampling method. Another factor that will affect the composition in the sample is which size fraction one looks at. Figure 8 shows that pellets were rarely found in size classes below 1 mm whereas fluff and microspheres were more common in those sizes. For smaller fractions (below 300  $\mu\text{m}$ ) fibers are often the predominant microplastic type in terms of numbers (Browne et al. 2011, Desforges et al. 2014 & Lusher et al. 2014), which was also noted in **Paper II** for water, sediment and invertebrate samples.

## CONCLUSIONS AND RECOMMENDATIONS

The composition of plastics in terms of polymer types is important in risk assessments, fate predictions, and in understanding potential source patterns. Different types of estimates produce different results regarding the relative importance of different sources and large regional variation should be expected. It should also be noted that it is challenging and can at times be impossible to match a material flow analysis with the results from environmental measurements. A tire particle in the environment could for example originate from tire usage or from artificial turfs. An expanded piece of polystyrene could come from a packaging or building materials. Moreover, the compositional analysis will be different depending on size, location and matrix. Nonetheless a combination of material flow analysis and compositional studies of what we find in environmental samples will most likely provide a better-rounded picture.

Our results show that even though it has been discussed since the 1970s, industrial spills remain an important source of plastic pollutants as seen both in **Paper III** and **V**. This illustrates how limited accountability could be argued to be one of the root causes for plastic pollution. Sources of plastics in North European coastal waters are mainly diffuse which is why it is often hard to assign accountability. Even when this is possible, accountability is rarely enforced, as seen for pellet spills in **Paper III**.

In terms of comparing different sources to each other, it is clear that different approaches have different strengths and limitations. Material flow analyses can for example provide clear estimates on tire particles that can then be confirmed with environmental sampling. Studies on beach litter composition show the importance of packaging material, which is supported by modelling of material flows and the extensive uses of plastic packaging material. Microplastic analyses in field samples illustrate the relative importance of certain particles such as pellets and microspheres that can otherwise be underestimated. Microplastic analyses are also important to identify specific types of primary plastics that might otherwise be overlooked, such as the PMMA microspheres. The work presented here shows that composition results from field studies will vary depending on size, matrix, sampling location and sampling method. Studies will therefore need to be carefully designed to answer the questions at hand.

Linking microplastic studies to macroplastic studies makes sense since the majority of the plastic input to the environment is estimated to be in the form of macroplastic (Kawecki & Nowack, 2019). Amongst the smaller size fractions, the most common types of plastic particles are fragments of different shapes, colors and polymers. Material flow analyses, plastics converter demand and beach findings all indicate that the main source of plastics in the environment is packaging materials and other types of single-use plastics. The fragments are often transparent, and the majorities are polyethylene, polypropylene and expanded polystyrene, which are also the main polymers used in plastic packaging.

Other than the dominating prevalence of packaging and other single-use material, the exact relative contributions of different sources are highly complex, as illustrated by the many uncertainties in data from material flow analyses, usage and compositional analyses in the environment. The existing data does not provide enough reliability to rank the sources in terms of relative importance but there is data to confirm several main contributors to marine litter that are all 1) high production volumes, 2) have shown to have significant points of leakage to the marine environment and 3) are found in the marine environment. Seven product categories that fit these descriptions are: single-use materials (bags, cutlery, straws, cigarette butts etc.), building materials, fishing equipment, clothes (especially due to shedding of fibers during washing and wearing), agricultural plastics, tires and preproduction materials.

## CHAPTER 4: REFLECTIONS ON THE RESULTS IN A WIDER CONTEXT

Early plastic pollution studies were conducted far out at sea and created a narrative that was focused on consumer excess and the capacity of human society to affect nature (De Wolff 2014). This image of the long transport of pollutants stood in contrast to a previous notion that the oceans were too vast and too resilient for humans to affect. Plastics were already in the 1970s documented to follow the currents to areas far away from humans (Heyerdahl 1971). But the accumulation of plastic debris in ‘ocean gyres’ first started to gain public traction when Charles Moore ‘discovered’ plastic pollution in the Pacific (Moore et al 2002). Following his experiences, he and several non-governmental organizations worked hard to create change. Moore’s findings were later picked up by media and in 2004 the narrative of the “plastic island” emerged. A Russian newspaper, Pravda, talked about a ‘trash island’ in the Pacific (de Wolff 2004). The article in Pravda in turn cited a German journal; Geo, that described the phenomenon as a plastic carpet. Later analysis of the emerging narrative of the plastic island concluded “how the carpet turned into an island remains a mystery of English-German-Russian-English translation” (De Wolff 2014).

This narrative, however, grew to staggering proportions where the main story was that plastic islands were forming in the middle of the ocean, and the headlines were often accompanied with illustrative photos of piles of garbage in the water. Solution-oriented initiatives, with the aim to clean up the Pacific and remove the islands, soon followed. The problem with this narrative is that there never was such a thing as a “trash island” in the middle of the Pacific. In an attempt to clarify, Moore instead started to describe the phenomenon as a plastic soup (Moore 2011), whereas other scientists described it as plastic smog (Liboroin 2016). Both the narrative of the plastic soup and the smog lacks some of the tangibility of the garbage islands, but could be argued to hold a far more alarming narrative, as it shows that the problem is much more complex than an isolated island in the Pacific. In order to understand this complex issue, it is important that the problem is accurately described to avoid misunderstandings.

Plastic materials are important components of our society. In terms of sustainability, plastics have, for example, increased the possibility of transport of clean water to areas where it would otherwise be challenging to come by (Wisner et al. 2002, Njeru 2006), thereby improving our capacity to work with the UN sustainability goal 6 (UN nations general assembly 2015). Plastics are also important in healthcare (Andrady & Neal 2009) (goal 3) and since plastics are typically light-weight materials, they can help decrease emissions from transport (goal 13) (Hekkert et al. 2000). But the way that we use plastics today is not part of a sustainable and responsible production and consumption (goal 12), which is the reason why we can observe negative consequences, for example in the form of pollution, on land (goal 15) as well as in the ocean (goal 14). In addition, somewhat ironically, the large quantity of plastic waste has – through increased flooding and clogging of storm



water drains – a negative effect on our possibility to guarantee clean water and sanitation (Njeru 2006) (goal 6), which, in combination with some of the chemicals that are used in plastic products (Groh et al. 2018) and inadequate waste management, can in turn affect human health (goal 3). Since most plastics are made from fossil fuels, the high consumption levels also have a negative effect on the climate goals (goal 13). It therefore seems that, although plastics may be important components of creating sustainable societies, the current consumption patterns need to be altered.

As a symptom of unsustainable consumption patterns, plastic pollution differs from other symptoms, such as climate change, ocean acidification or chemical pollution, in a few ways: 1) plastic products have tangible roles in our everyday lives 2) parts of the problem are very visual, which may be why 3) there are virtually no plastic pollution deniers (Phal et al., 2019). This tangibility and the ease by which the main body of the problem can be communicated facilitate collaborations between stakeholders. Through research, citizen science, voluntary initiatives and art projects, plastic pollution has reached high on the political agenda, which recently led the UN to declare ‘war on plastic pollution’ (UNEP Newscenter 2017).

But during the past 5-10 years, as the field has grown, the focus on plastic pollution, and especially on microplastics, has been increasingly questioned. One research group even went as far as to suggest that it is a convenient distraction from other, more urgent, challenges such as climate change and overfishing (Stafford & Jones 2019a). This was followed by debates (Avery-Gomm et al. 2019, Stafford & Jones 2019b). The level of risk that microplastics constitute in the marine environment has also been debated (Burton 2017, Koelmans et al. 2017b, Backhaus & Wagner 2018, Hale 2018, Avery-Gomm et al. 2019). In this last chapter, the results outlined in previous chapters will therefore be put in to the context of the current debate on plastic pollution as a sustainability challenge, through discussing risks associated with plastic pollution as well as potential solutions.

## A FEW NOTES ON RISK

As risk is defined as the likelihood of harm based on a combination of hazard and exposure the results presented in this thesis, which are more related to exposure, are in themselves not enough to quantify the risk plastic particles in the marine environment cause. The mere presence of microplastics does not mean that they necessarily constitute a risk, since most studies indicate that high concentrations of microplastics are needed in order to cause harm (Koelmans et al. 2017, Everaert et al. 2018, Besseling et al. 2019). In surface waters, these concentrations are rarely reached as illustrated by the results in **Paper I**.

There are, however, other areas in the marine environment where the accumulation of plastic particles can reach levels of concern. In **Papers III, IV and V**, we showed that floating plastics beach and sink, and other studies have confirmed high concentrations in sediment (Karlsson et al 2019). Ballent (2016) summarized the average microplastic contamination in marine and freshwater sediment from beach studies worldwide. The averag-

es varied between 0.1 and 5 000 particles/kg d.w. The lower size cut-off limit was not reported in all of the studies but the studies that reported numbers above 500 particles/kg d.w. included samples below 100  $\mu\text{m}$  (Vianello et al. 2013, Mathalon & Hill 2014, Ballent et al. 2016). This makes our results in **Paper V** especially noteworthy as they are they are up to 10 times higher than previous studies for the pooled samples, and up to 360 times higher for the sample with the highest concentration, than existing studies even though we had a lower cut-off limit of 300  $\mu\text{m}$ . Since we saw increasing concentrations with decreasing size fractions, the concentrations are likely to be even higher below 300  $\mu\text{m}$ . Moreover, the concentrations found on beaches along the Swedish west coast in **Paper V** are also higher than current roughly estimated no-effect concentrations (Everaert et al. 2018). These high concentrations therefore show the effect of hotspots and the necessity to differentiate between different types of environments when discussing risk.

This also illustrates the discrepancy in size between field data and effect-studies. Exposures are likely underestimated since most analyses have focused on larger particle sizes than the ones that are being used to test for environmental effects (Everaert et al. 2018, Hale 2018), meaning that we still don't have an adequate understanding of current environmental concentrations. Moreover, the term microplastics covers a wide range of different types of particles with different properties, and extrapolating risk from tests on one type of particle might not provide an accurate estimation of risk for others. Additionally there are large differences between species. Liboroin and colleagues (2018) showed that no microplastics were found in 41% of the fish species that have been examined in European studies. They noted that low sample sizes were an issue in 66% of the studies, but the results still illustrates that different species will have different levels of exposure to microplastics. Furthermore, as shown in **Paper V**, the concentrations vary by several orders of magnitude depending on where the samples are taken. Taken together this shows that it is challenging, or maybe not even possible, to extrapolate current scientific data to provide a general assessment of risk, since the risk is likely to differ depending on several factors such as location and species, as well as particle size and type.

Considering that a majority of the plastic usage happens on land it is also important to study the potential risks associated with plastic particles in terrestrial environments as recent studies suggest several potential risks that can be associated with microplastics in terrestrial environments (de Souza Machado et al. 2018). Still, it is common in northern Europe to disperse sewage water sludge on fields. Modern agricultural practices also use a lot of plastics that stays in the fields, which has been identified as a major source of macro and microplastics to the environment. This illustrates the importance of including the full emission to the environment in material flow analysis, as there is nothing to suggest that it is more of a marine problem, nor is it reasonable, in most cases, to treat marine and terrestrial environments as separate entities.

When discussing risk associated with plastic pollution overall (i.e. not limited to microplastics), we need to differentiate between different types of materials and products since some are more or less likely to cause harm. This is especially clear with fishing nets. Even though some of the estimates of fishing net losses indicate that they are a comparatively small source of microplastics in particular and plastic litter in general, the consequences of ghost-fishing and entanglement are well-documented. In the Baltic, for example, the total catch of cod by lost fishing nets has shown to correspond to between 0.01% and 3.2% of the total weight of reported and landed cod in that same area. In the southwestern waters of the north-east Atlantic the ghost net fishing corresponds to between 1.46% and 4.46% of the total commercial landings of monk fish (Brown & Macfadyen 2007).

In fact, if we focus too much on microplastics in sea-food or in marine compartments far away from the sources, we risk pulling the focus away from the root of the problem: the way in which we produce, consume, use and dispose of plastics. Even though there is limited proof that – at current pollution levels – microplastics pose a risk to marine ecosystems it remains important to see what they symbolize: long-term consequences of short-term conveniences.

## SOLUTIONS TO PLASTIC POLLUTION

A wide range of strategies has been suggested, and to some extent implemented, to decrease plastic pollution. Here, I would like to examine a few of them from the perspective of the results presented in this thesis. Several suggested solutions are focused on mitigating the pollution that has already accumulated in the environment. Considering the amount of plastics that have already entered the oceans, some level of end-of-the line solutions, such as clean-up and other mitigation initiatives, might be required, but for those it is important to prioritize the more efficient approaches. Here the narrative of the plastic islands, as a false problem description, contributes towards a skewed focus. As seen in the results in **Papers III, IV and V**, floating plastics are likely to sink or get stuck on beaches and the pollution levels are often higher closer to the source. Thus, mitigation initiatives can be expected to be more efficient closer to the source of the problem.

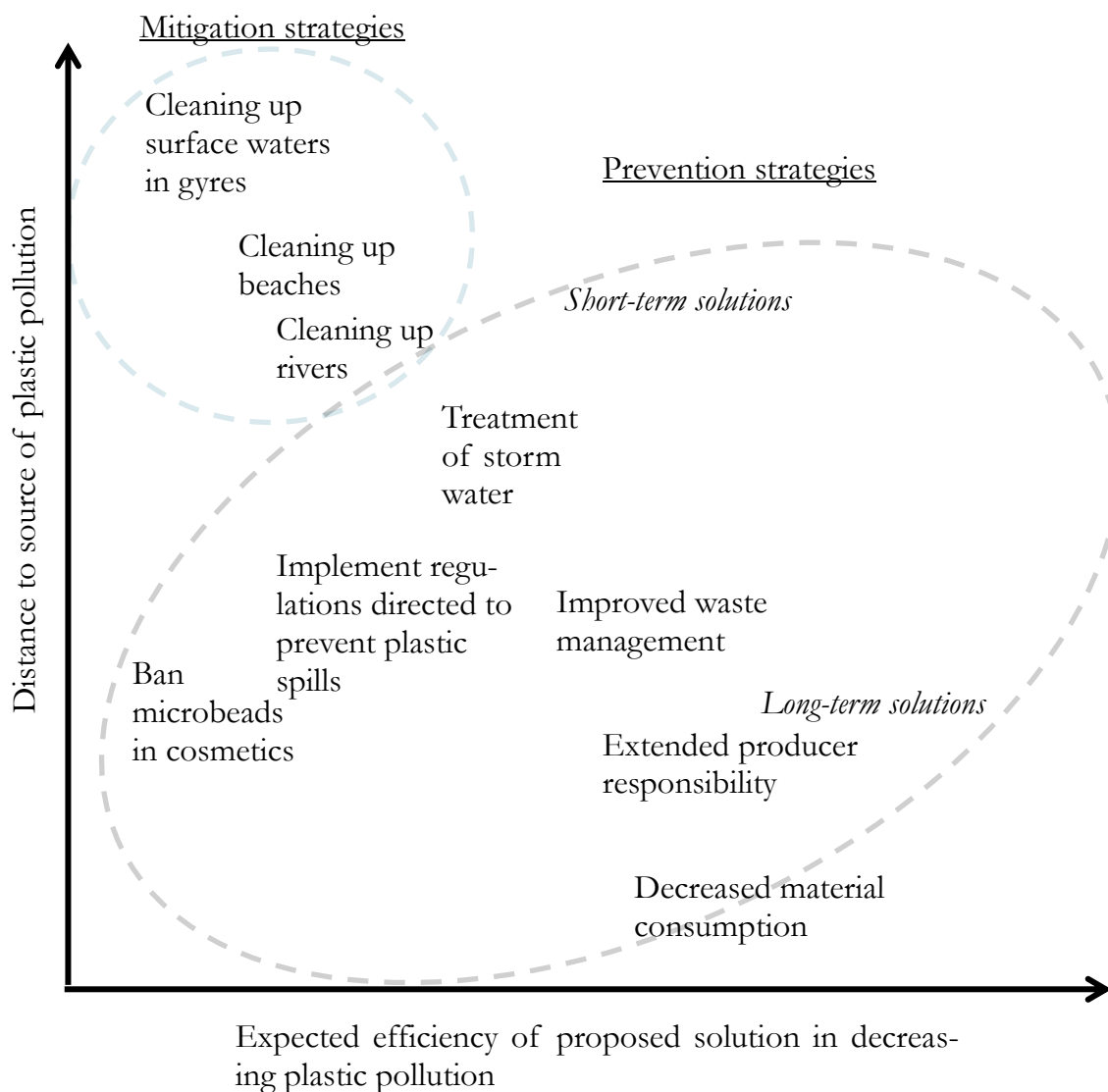
This can be further exemplified by comparing initiatives aimed at cleaning the ocean gyres to beach cleaning initiatives. The Ocean Clean-up, a project attempting to clean up floating marine litter in the Pacific, has developed a prototype. According to estimates by Boyan Slat, as published in National Geographic (Parker 2019) the device, if functional, should be able to clean up 45-70 tonnes of plastic in one year. This is comparable to the 60 tonnes of plastics that were collected in the relatively small region of Bohuslän during 2018 (Ren och Attraktiv Kust 2018). The Ocean Clean-up's future full fleet is estimated to clean 13 000 tonnes annually (Parker 2019), which could be compared to the Ocean Conservancy's annual cleanup where, in 2018, 10 000 tonnes were cleaned up from beaches in one day (Ocean Conservancy 2019). Since it is more accessible to clean up beaches, it is likely to both be more energy and cost efficient. Furthermore, Cordier and Uehara (2019) estimat-

ed that a significant cleanup effort (defined as the removal of 135 million tons of plastics from the ocean) would come at a cost equal to 1% of the global GDP.

An even more efficient approach would be to focus cleaning efforts on pathways before litter enters the marine environment, since a lot of the plastics will be transported to the ocean through a few significant pathways. One example is storm water, which often is led directly to water recipients without any treatment (Raspati et al. 2017). Other important pathways are rivers, especially those passing through urban and industrial areas (Lechner et al. 2014, Fok & Cheung 2015, Lebreton et al. 2017, Leslie et al. 2017). Thus, for mitigation measures there seems to be a clear relationship between efficiency and proximity to source. Hence cleaning up rivers (pathways) is most likely more efficient than cleaning up a beach, which in turn is more efficient than cleaning up surface waters in the gyres.

Preventive solutions would, however, be even closer to the roots of the problem than mitigation. In the case of ghost nets preventive measures have also been shown to be more efficient than mitigation measures (Brown and Macfayden 2006). Considering the challenges of access for cleaning up plastics in the environment due to long-range transportation, sinking and fragmentation, preventive measures can in general be expected to be more efficient than mitigation.

The relationships between different types of preventive measures are complex, as illustrated in the conceptual graph in Figure 10. Often, preventive measures can roughly be divided into short-term solutions such as improved waste management, or long-term solutions such as circular economy initiatives, where the long-term solutions would in general be more efficient (Macfayden et al. 2016). But not all preventive strategies will – when considering the overall amount of pollution – be more efficient than some of the mitigation strategies, even if they are directed closer to the origin of the problem. The microbead ban, for example, will be directed closely to the roots of the problems through removing an often unnecessary material from usage. But since it is directed toward a relatively low volume it is not expected to have a strong effect on the overall pollution levels. In contrast, implementation of existing regulatory frameworks to prevent plastic spills during transport, storage and production would be directed toward a larger volume, as shown in Chapter 3 where preproduction materials were identified as an important component of the overall plastic litter.



**Figure 10:** Conceptual graph showing the expected relative efficiency of some of the proposed solutions to plastic pollution, contrasted with the distance to the source of plastic pollution.

Concerning the overall composition of the microplastic pollutants, a key result repeated throughout **Papers I, II, and V**, is that a majority of the particles are fragments. Preventive strategies against microplastics therefore need to have a strong focus on how to avoid macroplastic pollution, as well as pollution caused by wear during usage, close to the point of release. Five important categories of macroplastics were identified in Chapter 3: fibers, agricultural plastics, tires, building materials and single-use materials.

Different sources might require different types of solutions. As discussed in Chapter 3, distribution of fibers could in part be remedied through re-evaluating the usage of sludge on farmland. Microplastics are, however, not likely to be the biggest risks with using sludge, since it can contain several other types of pollutants, such as polyaromatic hydrocarbons (Li & Ma 2016) and pharmaceutical residues (Magner et al. 2016). Moreover, the overall use of agricultural plastics, particularly as mulch, might need to be re-evaluated

considering the associated difficulty in retrieving and recycling that type of plastics. The leakage of tire particles, on the other hand, could be reduced through improved storm water treatment, which is also an important pathway for several different types of environmental contaminants.

A notable portion of the fragments in the studies reported here, especially closer to urban areas (**Papers I and VI**), were expanded polystyrene. Although expanded polystyrene is often used in packaging, the majority of the produced expanded polystyrene is used in construction (Plastics Europe 2018). Preventive measures aimed at decreasing the leakage of expanded polystyrene into the environment should therefore include an examination of how potential leakage and mismanaged waste could be prevented during construction work. Overall, emission models suggest that leakage from construction work could be prevented through changed routines, especially regarding waste management (Kawecki & Nowack 2019).

Related, one short-term solution that could have a strong effect on macroplastic pollution is an overall improved waste management. Improvements are continuously being made but the level of change that is necessary requires substantial advances; Jambeck and colleagues (2015) estimate that a 75% reduction in the input of plastics in the ocean could be achieved through an 85% improvement of waste management in the top 35% countries with the highest levels of mismanaged plastic waste. It is also important to note that, aside from contributing to a decrease in plastic marine debris, improving waste management would come with several further benefits. Current practices related to uncollected solid waste contribute to respiratory ailments, dengue fever, diarrhea, air pollution and flooding (as reviewed in: Hoornweg & Bhada-Tata 2012). It is also an economic issue; the global cost for municipal solid waste is expected to increase by a factor of 1.8 by 2025, compared to costs in 2012, resulting in an annual cost of 375.5 billion USD. These increases will be highest in low- and lower middle-income countries. In fact, solid waste management is already the single largest budgetary item of cities in lower income countries (Hoornweg & Bhada-Tata 2012). The multitude of benefits that would arise from an improved waste management illustrates the importance of considering the bigger perspective when discussing solutions to plastic pollution. The transboundary nature of plastic pollution in the marine environment, however, complicates governance solutions (Borrelle et al. 2017, Vince & Hardesty 2017). Especially since, in most cases, solid waste management is one of the few services that have been identified to fall entirely within the local governments' jurisdiction (Hoornweg & Bhada-Tata 2012).

The European plastics strategy has a strong focus on improved recycling (European Commission 2018), which is one aspect of improving waste management. Improved waste management can, however, be argued to be a short-term solution to the issue of plastic pollution (Löhr et al. 2017), since the underlying challenges are often associated with the high level of consumption of plastics. This is especially evident for single use

materials, which, as noted in Chapter 3, are often identified as key components in marine litter studies. Although waste management can help with the symptoms, a more long-term solution would be to decrease unnecessary use of single-use items.

There are already several incentives to reduce the usage of plastics, including bans, subsidies, taxes and behavioral interventions (Xanthos & Walker 2017, Schnurr et al. 2018, Abbott & Sumaila 2019). Different incentives are suitable for different types of plastics. Bans can for example be efficient in cases where the benefits of the materials are low and the costs are high (Abbott & Sumaila 2019). Fees on waste disposals are intended to decrease material usage but have been criticized since they require a high level of control to avoid illegal dumping (Abbott & Sumaila 2019). A more efficient way to incentivize a lower level of consumption might therefore be to introduce advance disposal fees (Abbott & Sumaila 2019). Extended producer responsibility is also often suggested as measure to incentivize greener design (Landon-Lane 2018, Abbott & Sumaila 2019). A change in material usage patterns could bring several added benefits and the world economic forum recently analyzed the material value loss for plastic packaging and found that 95% is lost after a short first-use cycle, equivalent to an annual loss of 80-120 billion dollars (Neufeld et al. 2016). They also conclude that a shift from the current linear system to a circular economy in the consumer goods sector (especially packaging) could generate a 706 billion dollars economic opportunity (Neufeld et al. 2016). To date, there is, however, limited accountability for companies involved in plastic design, production and usage. Due to the associated complexity of plastic pollution and plastic usage, it might be necessary to combine several different incitements such as hard law, soft law and corporate social responsibility in order to evoke meaningful change in creating a more sustainable use of plastics (Landon-Lane 2018). Legislation and policy need to work both with reducing the quantity of plastic waste that is generated, and the hazard of the plastics that is being used (Raubenheimer & McIlgorm 2018), thereby bringing the solutions closer to the roots of the problem.

It is important to note that replacing single-use plastics with other single-use materials is often not very efficient. Exchanging plastics for paper might, for example, seem like a good idea but life-cycle analysis show that plastic bags actually have a lower environmental impact (Lewis et al. 2010). Atmospheric emissions for plastic bags are for example 60-70% lower than for paper bags and even at zero recycling rate plastic bags contribute to less waterborne emissions than paper bags (Lewis et al. 2010). It has also been shown that plastic bags use less energy at current recycling rates (Lewis et al. 2010). Furthermore, Carrefour, a French retailer, showed that regardless of material (polyethylene, paper, biodegradable plastics) a reusable polyethylene bag was a better option than single-use carry bags; if used at least four times (cited in Lewis et al. 2010). There are however several aspects to take into consideration and a life cycle analysis on packaging used for olive oil showed that using glass would have a lower environmental impact (with respect to global warming potential, ozone layer depletion, non-renewable energy use, acidification, eu-

trophication and photochemical smog) than PET, if a high degree of the waste would be recycled (over 40%), especially if the remaining waste was assumed to go to landfill. However, using recycled PET resulted in an even lower impact (Accorsi et al. 2015). Another study (Garfi et al. 2016) showed that glass water bottles had a higher environmental impact than PET. The energy usage for glass bottles was 4900 MJ/m<sup>3</sup> of water, whereas it for PET was 1 000 MJ/m<sup>3</sup> of water. On the other hand, the study also showed that the most sustainable alternative was tap water, which was calculated to use 2-3MJ/m<sup>3</sup> water (Garfi et al. 2016).

Another alternative that is often mentioned is bio-based plastics. Here, it is important to note that they are not necessarily degradable or compostable but could work as an alternative to the usage of fossil fuel (Kershaw 2015). Some bio-based materials are made from waste, which could fit the circular economy perspective. Others are made from different crops which may be more controversial due to, among other things, their land use at a time of increasing food insecurity (Kershaw, 2015). They can also be made from starch or cellulose based materials. Life cycle assessments have shown that bio-based plastics can have a higher carbon footprint than traditional petroleum based products, even though this could be reduced through using other techniques (Boonitewanich 2014).

Although several types of action will need to be combined, it can be concluded that long-term solutions aimed at the root of the problem are often superior to short-term solutions or mitigation strategies. None of these solutions should however be considered in a vacuum and it is crucial to evaluate potential spill-over effects. These can be both positive, such as previously discussed for improved waste management, and negative, as noted for the substitution of plastics to other materials.

## CONCLUSIONS AND RECOMMENDATIONS

The complexity of the field demands input from several stakeholders in order to properly frame the associated problems and solutions. Plastic pollution engages people, providing several possibilities to work efficiently towards solutions. In order to do so, we need accurate problem descriptions to avoid fictitious trash islands and to facilitate the development of appropriate solutions.

The debate on risks associated with microplastics and other types of plastic pollution therefore brings several important perspectives. Existing data, although limited, does currently not indicate that microplastics constitute a risk at general levels of pollution. In some hotspots, such as the ones that were studied in **Paper V**, the levels do however exceed the predicted 'safe' concentrations and it seems likely that with a continued increase in pollution levels those hotspots will become more common. It is also important to consider terrestrial ecosystems since, in general, higher concentrations should be expected there. For macroplastics there are some well-documented risks for the marine environment, especially obvious in the case of ghost fishing. Discussions on risks therefore need



to be mindful of generalizations and take into consideration size and location of pollutants as well as product and material type. Moreover, if microplastics are regarded in the bigger context of plastic production and usage it is clear that microplastics are only one among several consequences, illustrating that current consumption patterns leave long-term consequences in the environment.

The work conducted in the context of this thesis shows that plastic pollution is higher closer to the source and that plastics often get stuck on beaches or sink to the bottom. It can therefore be concluded that mitigation strategies would be more efficient if implemented closer to the sources, which incidentally is also more cost-efficient. But in order to take meaningful and efficient steps towards decreasing plastic pollution we need to move even closer to the roots of the problem. In that context the results presented in **Paper IV** illustrate that there is a need for a higher level of accountability from industries. Moreover, since the main bulk of the problem consists of fragments of larger pieces of plastics the main focus needs to be on these larger items and how we use them. This means working actively to decrease the waste streams through a lower level of consumption, while simultaneously improving waste management strategies to prevent leakage.

The tangibility of plastic pollution, and to a certain degree also microplastics, can serve as guidance posts for how to approach a more sustainable culture around consumption. The current high, and increasing, levels of consumption of plastics would come with sustainability challenges regardless of material type, and in some cases, plastics remain the most sustainable option so far. Microplastics are not isolated, single issues and through focusing more on underlying issues, rather than end-of-the-line solutions, it would be easier to see the interconnectedness and how some solutions can provide several positive benefits in adjacent areas. In fact, no sustainability research can work in a vacuum and any and all proposed solutions need to be examined in a broader context. The sustainability challenges that we are currently facing are of such magnitude that we cannot afford to treat the issues separately, especially since they are often highly interconnected.

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