## Technical University of Denmark



### **Microplastics**

Occurrence, effects and sources of releases to the environment in Denmark

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Occurrence, effects and sources of releases to the environment in Denmark

Environmental project No. 1793, 2015



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# **Contents**

Co	ntent	s	and Summary	
Pre	eface.	•••••		7
Co	nclus	1.1.1       Size definitions       33         1.1.2       Material definitions       34         1.1.3       Definition applied in this report       34         1.1.4       Primary vs. secondary microplastics       35         1.2.1       Data sources       35         1.2.2       Flow model and data management       36         Occurrence, fate and effects of microplastics in the environment       39         1.1       Occurrence of microplastics in water and sediments       39         2.1.1       Microplastics in the water column       40         2.1.2       Microplastics in sediments       44         .2       Fate of microplastics in the aquatic environment       45         2.2.1       Degradation mechanisms and rates       45         2.2.2       Transport of microplastics in the aquatic environment       46         .3       Exposure of biota to microplastics       47         2.3.1       Exposure via ingestion       51         2.3.2       Egestion or translocation       51         2.3.3       Transfer of microplastics in the food web       51         2.4       Zooplankton       55         2.4.1       Zooplankton       55         2.4.2       Benthic organisms		
Ko	nklus	sion og	sammenfatning	21
1.	Inti	roducti	on	33
	1.1	Definit	tion of microplastics	33
		1.1.1	Size definitions	33
		1.1.2		
		1.1.3		
		•		
	1.2	Metho	** **	
		1.2.1		
		1.2.2	Flow model and data management	36
2.	Occ	urrenc	e, fate and effects of microplastics in the environment	39
	2.1	Occurr	rence of microplastics in water and sediments	39
		2.1.1	Microplastics in the water column	40
		2.1.2	Microplastics in sediments	44
	2.2	Fate of		
		2.2.1	~	
				-
	2.3	Exposi		
		2.3.1		
		2.3.2		-
			-	
	2.4	Observ		
		•	<del>-</del>	
		•		
	2.5			
		Ū		
		_		59
		2.5.3		60
		0.5.4	~	03
		2.5.4		66
		0.5.5	-	
	0.6	2.5.5 Migror		
	2.6	-	plastics as a carrier for organism transportplastics as a source of microplastics in the aquatic environment	
	2.7	2.7.1	Occurrence of macroplastics in the aquatic environment	
		2.7.1	Sources of macroplastics to the aquatic environment	
		2.7.3	Long range transport of macroplastics	-
		د٠/٠٥	Long range transport or macropiastics	/ 2

		2.7.4 2.7.5	Formation of microplastics from macroplastics in the environment					
		2.7.6	Macroplastics and biota	78				
	2.8	Microp	plastics in the soil environment	78				
	2.9	Humar	n exposure to microplastics via the environment	79				
3∙			use of primary microplastics and sources of microplastics to the					
			ent					
	3.1		primary microplastics					
		3.1.1	Introduction					
		3.1.2	Primary microplastics in personal care and cosmetics					
		3.1.3	Primary microplastics as raw materials for plastics production					
		3.1.4	Primary microplastics for use in paint					
		3.1.5	Blasting abrasives.					
		3.1.6	Rubber powder and granules for artificial turfs and other applications					
		3.1.7	Primary microplastics in other applications					
	3.2		tion and releases of secondary microplastics					
	3.3	•	les of overall releases of microplastics					
4.	Fat		croplastics by sewage treatment					
	4.1		uction					
5.	4.2		ion efficiency and microplastics in the effluents					
	4.3	_	olastics in sewage sludge					
	4.4	Polyme	er composition of microplastics in sewage	106				
5.	Releases of microplastics and occurrence in the water environment in							
4.	Der							
	5.1	Use an	d releases of primary microplastics					
		5.1.1	Personal care products					
		5.1.2	Raw materials for plastics production in Denmark					
		5.1.3	Primary microplastics for use in paint					
		5.1.4	Blasting abrasives					
		5.1.5	Rubber granules for artificial turfs and other applications					
		5.1.6	Other applications of primary microplastics	119				
		5.1.7	Summary on consumption and releases of primary microplastics in Denmark	120				
	5.2	Format	tion and releases of secondary microplastics					
	J. <u>~</u>	5.2.1	Formation from tyres					
		5.2.2	Formation from textiles					
		5.2.3	Formation from paints other than marine					
		5.2.4	Formation from marine paints					
		5.2.5	Formation from road marking materials					
		5.2.6	Formation from building materials and furniture					
		5.2.7	Shoe soles					
		5.2.8	Formation from plastic kitchen utensils, scouring pads and cloths					
		5.2.9	Other sources of secondary microplastics					
		5.2.10	Summary of emission estimates for Danish sources to microplastic					
			pollution					
	5.3		microplastics in sewerage systems in Denmark					
		5.3.1	Municipal sewage					
		5.3.2	Urban runoff					
		5.3.3	Industrial sewage with direct discharges to the environment					
	5.4		s of microplastics in the sea around Denmark					
		5.4.1	Occurrence by type of microplastics and by distance to sources	146				

Appendix 1 Appendix 2 Appendix 3 Appendix 4		ix 4	Complementary information on microplastics in cosmetics					
		ix 3						
		ix 2						
		ix 1						
Ref	eren	ces		172				
8.	Abbreviations and acronyms							
_		,						
	7.2	Possible 7.2.1	e other new initiatives Catalogue of potential other new initiatives	•				
		7.1.2	Catalogue of possible new studies to be initiated by Danish authorities					
		7.1.1	Main data gaps identified					
	7.1	Possible	e new studies					
7•	Pos	sible ne	ew studies and initiatives	160				
	6.6	Initiativ	ves of industry and business organisations	158				
	6.5		Initiatives of environmental and consumer organisations					
	6.4	Regiona	al initiatives by OSPAR, HELCOM and Nordic Council of Ministers	154				
	6.3	Initiativ	ves of international organisations	152				
	6.2		ves of the European Commission and EU Member States					
	6.1		ves of Danish authorities					
6.	Ong	going in	itiatives regarding microplastics and marine litter	150				
	5.6							
	5.5							
		5.4.3	Assessments of the origin of microplastics in the Danish waters					
		5.4.2	Occurrence in the open sea and potential transport by sea currents and sediment transport	14'				

# **Preface**

#### **Background**

An increasing body of scientific studies demonstrates that small plastic particles designated as "microplastics" are widespread in the environment, accumulate in organisms and may have harmful effects on organisms and ecosystems, in particular in the marine environment. The microplastics originate from various sources: primary microplastics intentionally used in products and processes, fragmentation of larger plastic litter in the environment and releases from plastic items and coatings of products still in use. The significance of the different sources as regards the levels of microplastics found in the environment is still poorly understood. In order to improve the understanding about sources and effects and the possibilities to reduce microplastics pollution, the Government of Denmark has allocated funds to investigate the environmental impact of microplastics through the Finance Act for 2015. This study is the first of a series of projects.

#### Objectives of the study

The objectives of the study are:

- To establish an overview of the existing knowledge on microplastics for authorities and other stakeholders (literature review);
- To survey the use of primary microplastics in Denmark and assess to what extent releases of primary microplastics contribute to the levels of microplastics found in the marine environment in Denmark;
- To provide an overview of the main national and international initiatives for reduction of the use of microplastics and their occurrence in the environment, and
- To propose possible new studies and initiatives for reduction of microplastics pollution.

### **Steering group**

The project has been steered by a steering group consisting of:

- Flemming Ingerslev, Danish Environmental Protection Agency
- Kim Petersen, Danish Environmental Protection Agency
- Berit Hallam, Danish Environmental Protection Agency
- Nanna Bloch Hartmann, DTU Environment
- Fredrik Norén/Kerstin Magnusson, IVL Swedish Environmental Research Institute
- Carsten Lassen, COWI A/S.

#### Advisory group

The project has been followed by an advisory group with representations of various stakeholder organisations:

- Annette Harbo Dahl, Danish Coatings and Adhesives Association
- Henrik Beha Pedersen, Plastic Change
- Lulu Krüger, Danish Veterinary and Food Administration
- Dorthe Licht Cederberg, Danish Veterinary and Food Administration
- Hanna Løyche, Association of Danish Cosmetics, Toiletries, Soap and Detergent Industries
- Lone Munk Søderberg, The Danish Nature Agency
- Helle Fabiansen, The Danish Plastics Federation

- Aage K. Feddersen, Dansk Fashion & Textile
- Dines Thornberg, BIOFOS and Danva
- Jakob Lamm Zeuthen, Danish Chamber of Commerce
- Lone Mikkelsen, Danish Ecological Council.

#### Workshop

A workshop was held 18 August 2015 with participants from the Danish EPA, the working group, the advisory group and other invited participants.

#### **Timing**

The survey has been undertaken by COWI A/S, DTU Aqua, DTU Environment and IVL Swedish Environmental Research Institute from March 2015 to August 2015.

# **Conclusion and Summary**

This report contains a review of existing knowledge on issues related to contamination by microplastics with a focus on the use and release of microplastics in Denmark and the presence of microplastics in the surrounding waters.

The issue of pollution of the oceans with plastics, including microplastics, is not new, but has received increased international attention in recent years. There is a growing concern as to whether pollution with plastics in the long term can have greater effects on the environment than previously assumed. The concern is based on a number of factors:

- Plastics that are released into the environment can remain in the environment for hundreds of years before they finally decompose.
- Global consumption of plastics is increasing, and global emissions are likewise expected to increase, unless action is taken against emissions.
- Plastics ending up in the sea may be transported over long distances; even the most remote places on the planet are affected by plastics pollution.
- In the environment, plastic pieces degrade into smaller pieces, meaning macroscale
  plastics degrade to microscale plastics, which further fragment into nanoscale plastics.
- Microplastics are detected in organisms at all levels of the marine food chain.
- Research shows that microplastics may have effects on organisms in the environment, but our knowledge of the magnitude of these effects is limited.
- We have virtually no knowledge on the possible particle effects of nanoscale plastic particles due to their size.
- People can be exposed to microplastics via food.

The combination of continued and perhaps increasing releases of plastics into the environment, and the fact that the substances remain in the environment for centuries and are fragmented into smaller pieces over time, potentially having increasing effects on the environment, means that the microplastics problem has some characteristics of a "time bomb". The effects of current emissions will only be apparent after many years and at that time, the effects may be impossible to prevent. However, we do not know whether the "time bomb" analogy reflects the situation accurately or not.

#### What are microplastics?

In the literature, a distinction is made between the plastics at macro-, micro- and nanoscales. There is no, however, clear definition of microplastics, but in many contexts - and also in this report - microplastics are plastic pieces of a size in the range of one  $\mu m$  to 5 mm.

Furthermore, a distinction between primary and secondary microplastics is made. Primary microplastics consists of plastics which are used intentionally in sizes between 1  $\mu$ m and 5 mm. Secondary microplastics are formed by fragmentation of large plastic pieces or by wear of paint or plastic surfaces. Secondary microplastics are formed both by wear of products in use and by fragmentation from larger plastic pieces in the environment.

This report makes use of a broad definition of plastics, covering all the solid materials formed from polymers of a mainly petrochemical origin, meaning that small fragments resulting from, for example, wear of tires and paints are considered microplastics. Depending on the polymer chain length,

polymers such as polyethylene (PE) can occur as a liquid, as a wax-like substance or as a solid. Only solid polymers are classified here as microplastics, although the waxy polymer may also occur in particulate form and have similar effects.

#### The occurrence and effects in the environment

## Occurrence of microplastics in the aquatic environment

There is a huge amount of data on the prevalence of microplastics in the aquatic environment. However, investigations are generally conducted using different methods, differing ranges of particle size and expressed in different units that cannot be converted, overall making it difficult to compare results across studies. Small particles occur in larger numbers than the bigger particles, meaning that only small differences in the examined particle size ranges yield very different results. The present studies report the number of particles in different size ranges, but there are no figures on the weight of microplastics in the waters around Denmark.

For particles larger than about 300  $\mu m$ , in Danish and Swedish coastal waters as well as in the Baltic Sea and the North Atlantic, concentrations typically ranged from 0.3 to 4 particles/m³, with a trend towards higher concentrations near major cities and in inner Danish waters. However, the number of data are limited and differences between the seas must be interpreted with caution. Smaller plastic particles occur in much higher concentrations. A study which included particles down to  $\geq$ 10  $\mu$ m found 4,400 to 94,000 particles/m³ along the Swedish west coast, while a German study on particles  $\geq$ 1.2  $\mu$ m found average concentrations of granular particles and fibres of 64,000 and 88,000 particles/m³, respectively. The concentrations of microplastics  $\geq$ 10  $\mu$ m in the environment is of the same magnitude as the concentrations of microplastics  $\geq$ 10  $\mu$ m in the influent of Swedish and Norwegian sewage treatment plants, while the concentrations of particles  $\geq$ 300  $\mu$ m are more than 1000 times lower in the environment than in the influent.

The average concentration of microplastics particles  $>38~\mu m$  in sediments from marine waters around Denmark ranged from 240 particles/kg dry matter in the North Sea/Skagerrak to 1,100 particles/kg dry matter in the Belt Sea. Differences between the individual seas were not statistically significant due to the large variation within the individual seas and a low number of samples. The concentrations are, however, significantly higher than those found in studies of particles of the same size from the Belgian beaches and continental shelf of Belgium.

The number of available data from Denmark is not sufficient to establish a relationship between the potential sources and the concentration in the water column and the sediment.

#### Exposure of aquatic organisms

Microplastics have been detected in organisms at all levels of the marine food chain. The amount of ingested microplastics varies between species and location, and can vary significantly even within the same area. In the North Sea and the Great Belt microplastics have been found in the stomach, intestines and/or other tissues of seals, herring, cod, whiting and mussels, amongst others. It is well known that marine animals ingest microplastics with the food, and there are indications that certain animals ingest microplastics because they are of the same size as their regular food, such as algae. Studies also suggest that virtually all marine animals ingest microplastics, but that there is great variation between the different species in terms of the amount they ingest. Likewise, there are studies on crabs indicating that microplastics accumulate in the food chain.

#### Fate and effects of microplastics in the aquatic environment

There are a number of studies that demonstrate the harmful effects of consumption of plastics. Typical harmful effects are inner and outer lesions and blockage of the gastrointestinal tract, which can lead to a false satiation. With regard to plastics in micro- and nano-size, there are potentially three types of adverse effects associated with ingestion: (1) physical effects related to consumption similar to those found for the macro plastics (but for smaller organisms), (2) toxic responses from

the release of hazardous substances derived from the intended use in plastics or used as raw material by the production of the polymer, and (3) toxic reaction to contaminants unintentionally adsorbed to microplastics. In the absence of the ability to make field observations, researchers used laboratory experiments to investigate the possible effects. There are studies of the biological effects of microplastics in a number of categories of organisms, such as zooplankton, benthic organisms, fish and seabirds, but overall, data is limited. In laboratory experiments, it has been found that microplastics can have a significant negative impact on e.g. food uptake of crustaceans and the eating activity and weight of lugworms. Furthermore, microplastics can cause a form of inflammation in the tissue of mussels. In studies of fish, correlations between microplastics and liver stress, the formation of tumours and indications of endocrine disrupting effects have been observed. Laboratory tests often use relatively high concentrations of microplastics compared to concentrations found in the environment, and the tests only expose very few test animals of a single species compared to the number of species potentially exposed in the environment. Consequently it is uncertain to what extent the effects observed in the laboratory occur in the environment.

#### Microplastics as carriers of hazardous substances

There are a number of studies showing that microplastics in the environment contain hazardous substances and/or function as carriers of hazardous substances. There are basically two types of sources contributing to the presence of hazardous substances in microplastics: 1) substances intentionally added to the plastics or used as raw materials for the production of plastics and, 2) substances in the environment adsorbing to the surface of the plastic particles which, over time, may be absorbed into the plastic matrix.

Examples of substances found in microplastics in the environment are PBDEs and other brominated flame retardants, which have been intentionally added, or nonylphenol and bisphenol A (BPA) which are used as starting materials for the production of different types of plastics. It is likely that plasticizers such as phthalates and chlorinated paraffins as well as added biocides are present in microplastics in the environment. The concentration of flame retardants and phthalates in plastics is typically in the range of 5 to 30%, thus much higher than concentrations of hazardous substances that are absorbed from the surrounding water. The problem of spreading of dangerous substances via plastics is well known and this emission pathway has often been included in emissions inventories and risk assessments. The EU risk assessments for several phthalates, for example, show that plastics lost to the environment are the main source of releases of phthalates to the environment. Paints can likewise be a significant source for some substances. Emissions may occur through spills of plastic raw materials, formation of secondary microplastics by wear and tear during product use, and disposal and dispersion of plastic parts, which subsequently fragment into microplastics in the environment. Organisms may be exposed to hazardous substances through intake of microplastics directly, but the substances can also be spread from plastics to other matrices in the environment as well as to organisms.

Some chemicals tend to adsorb to the surface of plastics, and in some combinations of substances and types of plastics, the substances migrate into the plastic, potentially leading to higher concentrations within the plastics as compared to the surrounding environment. This phenomenon is demonstrated for a number of persistent organic pollutants (POPs) such as PCB and DDT, and uptake of POPs in plastic raw materials (pellets) lost to the environment; these pellets have been used to monitor the global dispersion of POPs for many years. It is an open question as to what extent adsorption and absorption significantly increase exposure of some organisms to hazardous substances. Correspondingly, it is uncertain to what extent the dispersion of plastics facilitates contaminant transport from highly polluted areas to areas with a low contaminant burden. The existing studies and model calculations suggest that the prevalence of microplastics may have an effect on how POPs are distributed in the environment, but there is no indication of generally increased exposure (this conclusion does not concern hazardous substances intentionally added to plastics, as mentioned above). The results do not indicate that the transport of microplastics with

ocean currents contributes substantially to the transport of POPs to, for example, the Arctic. Of critical importance in relation to controlling POPs remains avoiding their releases to the environment. However, the available study results are uncertain and partly conflicting; therefore, more data are needed for more reliable conclusions.

#### Microplastics in soil

Microplastics can end up in the soil due to application of sewage sludge and compost containing microplastics and by deterioration of outdoor plastic parts and painted surfaces. Studies show that microplastics remain in the soil for many years. Therefore, long before microplastics themselves became an issue, they were proposed as an indicator of how much sludge a soil has received. Studies of the effects of microplastics on soil-dwelling organisms have not been identified. Since many soil organisms feed in the same way as sediment-dwelling organisms tested in the aquatic environment, it is a reasonable assumption that the same exposure routes and possible effects of microplastics apply to soil invertebrates. This assumption should be further verified through specific studies.

#### **Health aspects**

People can be exposed to microplastics in a number of ways: using cosmetics containing microplastics, via the diet, through ingestion of dust in the indoor environment and in connection with the use or maintenance of plastic parts or painted/plastic surfaces. It has been beyond the scope of this review to include the possible health effects of microplastics or extract knowledge from studies, e.g. on the effects of paint and plastic dust in the working environment.

The German federal institute for risk assessment (BfR) has concluded that microplastics in cosmetics do not pose a health risk because, given their size, the particles are not absorbed via the gastrointestinal tract. In recent years, there has been a number of studies showing that microplastics may be present in food. The presence of microplastics was demonstrated in, amongst others, mussels, beer and honey. The BfR has concluded that there are insufficient data on the chemical composition, particle size and concentration of microplastics in food to make an assessment of the possible health risk of microplastics in food, and has requested the European Food Safety Agency (EFSA) to prepare a report on micro- and nano-sized plastics in food.

### Microplastics in sewage and sewage treatment plants

No studies of microplastics in sewage treatment plants in Denmark could be identified. Studies of microplastics in sewage treatment plants in Norway, Sweden and Germany show that the retention efficiency depends on the size of the particles, while the shape of the particles is of little importance. More than 99% of microplastics  $\geq$ 300  $\mu$ m end up in sludge, while for microplastics in the size range 20 to 300  $\mu$ m the corresponding number is typically only 80 or 90%. There are no studies on the retention of microplastics <20  $\mu$ m. The available studies have all counted the number of particles, whereas none of the studies has estimated the total weight of the particles.

Plastic fibres constitute the largest number of microplastics particles  $\geq 300~\mu m$ , suggesting that textiles are a major source of microplastics  $\geq 20~\mu m$  in sewage. The number of particles in the size range 20 to 300  $\mu m$  is substantially greater than the number of particles  $\geq 300~\mu m$ . Fibres in the size range of 20 to 300  $\mu m$  constitute about a third of the particles (by weight, but the small particles contribute less). The number of non-synthetic fibres are typically 5-10 times greater than the total number of microplastics particles.

Based on the available studies, it is not possible to identify how large a proportion of the non-fibre microplastics originate from intentional uses of primary microplastics (e.g. in cosmetics and plastic materials), and how much comes from deterioration/abrasion of plastic parts and paints. German investigations of the polymer composition of microplastics in waste water indicate that PE and PP represent more than 90% of the particles >500  $\mu$ m (not fibres), while PE and PVA constitute

more than the half of the smaller particles. Polymers presumably originating from painting constitute 5-10% of the total content of the smaller particles. However, the methods are still under development and the amount of investigated particles is small. Hence, the study authors note that the results should be interpreted with caution. Microplastics particles in the form of dust from tires, paint polishing and abrasion of e.g. vinyl flooring, which collectively represent a very large proportion of emissions, are typically so small that they have not been covered by the studies carried out so far. There are no studies on the degradation of microplastics in sewage treatment plants, but microplastics likely degrade only to a limited degree, and most of the microplastics end up in sludge, which is either burned or applied to agricultural soils in Denmark.

#### **Emission sources of microplastics in Denmark**

#### Use and release of primary microplastics in Denmark

Among the most important uses of primary microplastics in products in Denmark are microbeads in cosmetics, various types of microplastics in paint, small plastic particles which are used for "sandblasting" and as "EPS beads for pillows and other purposes". In addition, primary microplastics are the main raw material for the production of plastic items and rubber granules from recycled tyres are used for artificial turfs and other applications. Emissions from these uses will typically be to sewage (soil is the main release pathway for rubber granules), and some of the microplastics will end up in the aquatic environment, while most will be retained in the sewage sludge, which is partly used for agricultural purposes. Besides, within the limits of the survey, a number of applications have not been described in detail and the resulting releases have not been estimated.

The estimated releases of primary and secondary microplastics in Denmark is summarised in the table overleaf. The sources are further described below the table. With respect to emissions to the aquatic environment, the table shows the resulting emissions after sewage treatment. It is anticipated that part of the sewage is discharged untreated in case of heavy rainfall. With high retention rates in the treatment plants, the direct discharges contribute significantly to the total discharges. Some of the microplastics discharged with stormwater from paved areas end up as solid waste after cleaning of the settling lagoons. As only a part of stormwater sewers are equipped with settling lagoons, it is estimated that on average only 10-20% of the microplastics in stormwater is retained.

The total quantities discharged to sewage are estimated at 2,000-to 5,600 t/year. The main sources are estimated to be tyres and textiles, but many other sources may contribute significantly.

The amounts are indicated in t/year, and there are not sufficient data available to estimate the number of microplastics particles/year. Particles in plastics raw materials and rubber granules are relatively large in relation to other particles that would typically occur as dust/powder. Consequently, plastic raw materials and rubber granules would represent a significantly smaller proportion if the discharges were stated in numbers of particles.

Emissions estimated occur to the immediate recipient, and there is no basis for calculating the extent to which the particles are spread further within the environment, and what effects they might have. Particles from tires, paints, vinyl coatings (PVC) and textile fibres typically have a density above unity and must hence be expected to be less widespread in the environment than polyethylene in cosmetics, a large proportion of plastics raw materials, EPS beads, polyurethane foam and microplastics paint that all have a density below unity. There is no basis to assess whether some types of microplastics have a greater potential for adverse environmental effects than others, aside from the chemical effects of hazardous substances that are present in some of the particles (flame retardants, plasticizers, etc.).

**TABLE 0**SUMMARY OF EMISSIONS OF PRIMARY AND SECONDARY MICROPLASTICS IN DENMARK (EXCL. FORMATION FROM MACROPLASTICS IN THE ENVIRONMENT)

	Total emis- sion t/year	% of total (average)	Emission to sewage treat- ment plants (STP) t/year	Ultimate emission to the aquatic environment * t/year	% of total ultimate emission to the aquatic environment (average)		
Primary microplastics							
Personal care products	9-29	0.2	10-22	0.5-4.4	0.1		
Raw materials for plastics production	3-56	0.3	3-56	0.1-4.5	0.1		
Paints	2-7	0.1	2-7	0.3-1.8	0.1		
Blasting abrasives	0.05-2.5	0.01	0.03-1.3	0.03-1.4	0.04		
Rubber granules	450-1,580	10.5	20-330	1-20	0.6		
Other applications	?	?	?	?	?		
Total, quantified primary microplastics	460-1,670	11	35-416	2-31	0.9		
Secondary microplas	stics						
Tires	4,200-6,600	56	1,600-2,500	500-1,700	60		
Textiles	200-1,000	6.2	200-1,000	6-60	1.8		
Paints (excl. ship paints)	150-810	4.9	14-220	6-149	4.2		
Ship paints	40-480	2.7	0-50	21-240	7.1		
Road markings	110-690	4.1	40-260	10-180	5.1		
Building materials of plastics	80-480	2.9	30-150	4.5-37.5	1.1		
Footwear	100-1,000	5.7	40-380	10-260	7.3		
Cooking utensils, scouring sponges and cloths	20-180	1.0	20-180	1-50	1.4		
Other uses	100-1,000	5.7	20-500	8-375	10		
Total, secondary microplastics (rounded)	5,000- 12,200	89	2,000-5,200	600-3,100	98		
Total quantified microplastics (rounded)	5,500-13,900		2,000-5,600	600-3,100	99		

<sup>\*</sup> Indicates the resulting emissions after sewage treatment.

**Cosmetics** - Microplastics beads have typically been used in cosmetics as an abrasive to provide a scrubbing effect. The primary application areas are scrubbing face creams, shower gel and hand cleaners. These applications are thoroughly described in the literature. Micro-coloured plastics are also used to some extent to achieve a colour effect in, for example, toothpaste. Some types of microplastics are also used for obtaining a glitter effect in cosmetic products. The most commonly used

polymer is polyethylene (PE). Polyethylene and other polymers of shorter chain length, rendering the polymers liquid or waxy, are apparently applied in a variety of cosmetic products, but these uses are beyond the scope of the definition of microplastics used in this study and were not further investigated.

A European study compiled by various players in the cosmetics industry has estimated the total content of microplastics in scrubbing and cleaning products sold in Denmark at 29 tonnes in 2012. The estimate does not include microplastics in toothpaste or other types of products where microplastics are added to achieve colour and glitter effects.

According to information provided by the Danish trade association SPT, consumption of microplastics has been declining as the microplastics have been replaced in many products. The consumption in 2014 is therefore lower than estimated for 2012. According to a survey among members of the association, microplastics will be phased out in almost all cosmetic products in 2017, except for certain hand cleaners. However, according to available information, the industry is also working on the phase-out of microplastics in hand cleaners. There is no known consumption of microplastics in toothpaste sold in Denmark today. Taking into account that the consumption in 2012 is uncertain and may be underestimated, the total content of microplastics in cosmetic products sold in Denmark in 2014 is estimated at 9-29 tonnes. Ninety percent of this amount is assumed to be washed off and ends up in sewage. Since 2013, an agreement has been established within the European trade association Cosmetics Europe not to support the use of microplastics in cosmetics. The same view is shared by the Danish trade association.

Plastics raw materials - Plastics raw materials for the manufacture of thermoplastics in Denmark are imported as plastic pellets or powder. Virtually all solid plastic materials fall within the definition of microplastics. During plastics manufacture the raw materials are melted, so that the finished articles do not contain any microplastics. With consumption of approximately 550,000 t/year, this is by far the largest use of primary microplastics. Emissions occur either directly to the environment or via the effluent and may happen during transport, loading/unloading and storage, whereas emissions from the production process are considered negligible. There is no requirement in the standard conditions for environmental permits for plastics manufacturing companies concerning spills of plastic materials. Foreign studies have shown that emissions of plastic materials can be significant and most microplastics in the river Danube are considered to be plastics raw materials that have been discharged into the river. A significant proportion of the releases take place in the production and loading of the raw materials, and these processes do occur outside of Denmark.

Under the auspices of the Danish Plastics Federation, the "Operation Clean Sweep" programme focusses on methods for reducing waste. However, currently there are only 9 out of 250 plastic processing companies in Denmark that have joined the programme. Data on spill percentages collected via the Danish Plastics Federation from companies that have joined Operation Clean Sweep indicate that loss to sewage from within the companies' area (incl. unloading from trucks that deliver raw materials) accounts for a maximum of about 0.001% of raw material consumption. This percentage is far lower than the "worst case" default emission factor developed by the OECD for loss to waste water.

It is unknown to what extent the average emissions of all plastics companies in Denmark exceed this figure, but it is assumed that the average would not be more than 10 times higher than the highest values indicated in the Danish Plastics Federation survey. On the basis of available data, the total releases of microplastics to waste water are estimated at 3-56 t/year.

**Paint** - Particles of microplastics are widely used in building materials in order to reduce the need for pigment, reduce the density and provide special surface properties. In addition to microplastics,

as defined in this report, wax particles are used to provide surface structures. The total consumption in Denmark in 2014 is estimated at about 254 tons for a total of 64,000 tons of paint by the Danish Paint and Adhesives Association (DFL). When the paint cures, the plastic particles become an integrated part of the paint matrix. During abrasion, the microplastics particles remain an integrated part of the paint particles, which are described further under secondary microplastics. When using wall paint, about 1% of the uncured paint ends up in sewage due to cleaning of brushes and other tools. It is uncertain as to what extent microplastics in sewage combine with the binder in the paint, or whether they are released and suspended in the waste water as free particles. On this basis, the total emission of primary microplastics to sewage containing paint is estimated at 2-7 tonnes. When using paint outdoors, some paint may be spilled on the soil surface. The spilled paint will likely cure, meaning that there is no further transport of the free particles, but the paint as such can contribute to emissions of secondary microplastics.

**Blasting abrasives** - There is little information available about the use of microplastics as blasting abrasives for cleaning surfaces. One study shows that many different types of plastics are used for various purposes as blasting abrasives in Denmark. The blasting abrasives are typically re-used 10-20 times. The total consumption of plastic-based blasting abrasives in Denmark is estimated at 5-25 t/year. The vast majority of applications are indoors; thus, loss to sewage is expected to be limited. However, some applications may partly take place outdoors, i.e. on ships and off-shore installations. In such cases, a direct emission to the environment may occur. Based on the available data, a rough estimate predicts emissions to the aquatic environment in the range of 0.1 to 1.3 t/year each.

**Rubber granules** - The size of rubber granules from recycling of tyres varies between 0.7 and 3 mm, which thereby classifies the rubber granules as primary microplastics as defined in this report. The granules are used as infill for artificial turfs for e.g. football and rugby and golf fields. Combined with a binder, the granules are used for playgrounds and running lanes and a significant quantity is used for polymer modified asphalt. Wear of artificial turf fields and other areas with rubber coating will release rubber granules used in the fields and furthermore, secondary microplastics are formed and released from synthetic grass fibres. There is high uncertainty about the extent to which microplastics released from such areas get into drainage and sewerage systems. The total quantities added to sewage are estimated at 20-330 t/year. It should be noted that the releases of rubber granules are quite small as compared with the releases of particles from the tyres before they are recycled.

Cleaning and maintenance products - According to the trade association SPT, there is no information on the use of microplastics in cleaning and care products apart from hand cleaners, which are included in the estimate for cosmetics. Additionally, no specific information be found on this application in the literature. Nonetheless, it is mentioned as a possible application, and it cannot be ruled out that there may be some applications where microplastics are used as scrubbing media in maintenance products in the same way as used in cosmetics.

**Other applications** - For a number of other applications of primary microplastics, it has not been possible to describe the use and releases in detail within the scope of this survey. These are beads of expanded polystyrene for furniture and cavity wall insulation, and toner for laser printers, as well as plastic beads used in certain industrial dishwashers, medical purposes, and research purposes, and microplastic used as specialty chemicals. The total amounts used for a few of the application areas may be significant, but the resulting releases to the environment likely account for a limited part of the total releases.

# Formation of secondary microplastics from articles in use and painted/plastic coated surfaces

There is a variety of sources of formation of secondary microplastics. The most important sources are considered to be wear on tires and road markings, wear and tear of clothes and other textiles made of synthetic fibres, wear and polishing of paint, wear of large surfaces of plastics e.g. vinyl flooring and roofing, and wear of shoe soles and kitchen utensils.

Emissions to the aquatic environment come from municipal sewage treatment plants, via stormwater and urban run-off or directly to the aquatic environment, for example by activities in harbours or on ships. The particles formed by the wear of tires, plastic materials (e.g. vinyl flooring) and sanding of paint are typically less than 10  $\mu$ m and therefore are not subject to the investigations that have been made of microplastics in the environment and sewage. This may be a possible explanation as to why particles from these significant sources are not mentioned in greater detail in the studies of polymer composition of microplastics in sewage and the environment.

**Tires and road markings** - Particles abraded from tires and road markings are included in road dust, which partly is removed via waste water and partly spread to the surrounding soil and surface water. The total formation of particles from tires is estimated at 4,200 to 6,600 t/year; of this, 1,600-2,500 is released to sewage, thus forming the largest single source of releases of microplastics to the environment. The estimate is relatively certain. Emissions from the abrasions of road markings are estimated at 10-180 tons, thus also forming a significant source.

**Textiles** - Significant amounts of fibres in the microplastics range are formed and released from textiles due to wear and tear of the products during normal use and during washing. Synthetic fibres, most likely originating from textiles, constitute a large proportion of microplastics  $\geq 20~\mu m$  in sewage treatment plants. These fibres also form a significant proportion of microplastics in coastal waters. The data are limited, but based on a comparison of data on the release by laundry with knowledge of the amount of fibres in the influent of sewage treatment plants, it is possible to estimate the probable magnitude of the formation of microplastics. The emissions of synthetic fibres to sewage treatment plants are estimated at 200-1,000 t/year.

**Paints (except ship paints)** - Microplastics particles derive from abrasion of the paint as well as from sanding/scraping off paint for maintenance of the painted surfaces. There will primarily be releases from paints used outdoors, and the survey has focused on these paints. Total emissions are estimated at 150-810 t/year, of which 14-220 t/year are estimated to be discharged to sewage treatment plants via runoff from paved areas.

**Marine paints** - Microplastic particles are formed from abrasion and maintenance of marine paints used for recreational boats and larger vessels. A significant proportion of the releases occur directly to aquatic environments. The total releases to aquatic environments from the use of paints on recreational boats is estimated at 5-50 t/year, while some 16-190 t/y is estimated to be released from the use of paints for larger vessels. In addition, microplastics may possibly be formed by the releases of self-polishing antifouling paints when the vessels are in the water, but no information on this possible source is available.

**Building materials of plastics** - Plasticised as well as hard PVC makes up the majority of the plastic building materials subject to deterioration and weathering. There is some knowledge available about deterioration of PVC from experiences in estimating the release of phthalates and heavy metals used in PVC. The main sources are believed to be flooring, roofing and gutters. Emissions of microplastics from building materials to sewage are estimated at a total of 30 to 150 t/year.

**Footwear** - soles of footwear are typically made of PVC, polyurethane or synthetic rubber. During wear on the soles, microplastics particles are formed. Estimates of release of plastics from PVC soles exist. On this basis, the total releases to sewage and storm water are estimated at 30 to 430 t/year.

**Cooking utensils, scouring pads and cloths** - Wear and tear in tools, scouring pads and plastic cloths used in kitchens and bathrooms may cause a release of microplastics directly discharged to sewage. Scouring pads and synthetic cloths are believed to be the major sources, while the wear of kitchen utensils is estimated to be less significant. Based on the available information, the total emission to sewage is estimated at 40-380 t/year.

Other sources - There are a number of other possible sources of formation of secondary microplastics for which it has not been possible to establish an estimate within the limits of this survey. The most important potential sources are considered to be handling of plastics materials and articles during industrial and professional use, nets and other fishing tools, polyethylene foils used in agriculture, polymer modified bitumen, biowaste, paper recycling, shredders, and food waste shredders on ships. Based on the experience of foreign studies, it is estimated roughly that the total releases from these sources would be in the range of 100-1000 t/year.

#### Formation of microplastics from macroplastics in the environment

Microplastics are formed by fragmentation of larger plastic pieces dispersed in the environment. Considerable knowledge exists about types of plastic found on beaches, and hence much knowledge is available about the sources of macroplastics in the environment.

It is estimated that in the nineties about 20,000 tons of waste were dumped into the North Sea, of which a large part is plastic waste. Of this, approximately 15% ends up on beaches while the rest is expected to float around, during which time it is partially fragmented before it ultimately sinks to the bottom. There are no specific inventories for plastics and no estimations of the rate and to what extent the macroplastics floating or at the bottom are fragmented into microplastics. The timeframe for a complete degradation (mineralization) of plastics may be many hundreds of years. Available data for fragmentation indicate that many types of macroplastics are fragmented to a certain degree within a timeframe of years or decades.

A substantial portion of the plastic pieces in the marine environment therefore fragment before they are removed from beaches or before they are covered by sediment. There are no model calculations estimating the formation of microplastics based on the occurrence of macroplastics in the environment. A Norwegian survey provides what the authors of the survey would term a "qualified guess" with respect to formation of microplastics formed through the fragmentation of macroplastics in the aquatic environment of Norway. The estimates show a total amount of 360 to 1,800 t/year from the main sources, but the actual uncertainty of the estimate is likely to be greater than the range suggests. Nonetheless, the estimate suggests that the fragmentation of macroplastics in the environment is likely to contribute significantly more than the emissions of primary microplastics, whereas it is more uncertain as to whether the quantities formed in the environment are of the same magnitude as the releases of secondary microplastics. Since there are no data allowing better estimates than this "qualified guess", no attempt has been made to prepare another "qualified guess", and the present study refers to the Norwegian estimate.

# Assessment of sources of microplastics in the marine environment of Denmark

Microplastics in Danish waters originate from local land-based sources, shipping and other seabased sources, fragmentation of plastic litter in the environment, or it may enter the waters by ocean currents. There are no measurements or models which can be used to indicate the importance of the different sources. Likewise, models describing the final fate of microplastics supplied or produced in the marine environment of Denmark are lacking. The present studies of microplastics in the marine environment contain limited information on possible sources.

There is a general tendency to find higher concentrations of microplastics in coastal waters near major cities and higher concentrations in the inner Danish waters than in the surrounding seas, indicating that the microplastics originate from more local sources than from inflow from neighbouring seas. However, the number of measurements is too small to draw firm conclusions, which would also require detailed modelling of emissions, transport and the formation of microplastics in the waters. In one case, high concentrations of microplastics were found close to a Swedish plastics producing company, illustrating that there might be high concentrations near point sources (the company has installed release reduction measures since then). Furthermore, a recent study indicates that microplastics at one of the investigated stations likely consisted of spilled plastic materials.

A Swedish study (on particles >300  $\mu$ m) has found higher concentrations in the Sound (Øresund) than at the other measuring points along the Swedish coasts of Kattegat and the Sound. German studies (on particles >100  $\mu$ m) found higher concentrations in the Kattegat and Baltic Sea south of Lolland Falster than in the North Sea. Swedish studies have also found higher concentrations in the waters close to larger cities than in waters close to smaller towns. Sewage treatment plants are evidently a source of high local concentrations around the outlet, suggesting that local sources are the greatest contributors for coastal waters at least. Studies of coastal waters found a very high incidence of fibres, indicating that sewage treatment plants as well as fishing gear could be a significant source. There were no studies in which the thickness of the fibres and the polymer composition are reported, which otherwise could help to determine if the sources were textiles or fishing gear. The present results should generally be interpreted with caution because of the limited data. The studies from the open sea are not extensive enough to identify possible sources, and there is no basis for assessing whether the open sea represents a net addition or removal by ocean currents.

A recent German study has shown that for the particles  $>500 \mu m$ , the most abundant polymers are polyurethane (PUR, average 51%), polyethylene (PE, 29%) and polypropylene (PP, 17%). PE and PP are the plastics used in the greatest quantities, have a density above unity, and are widely used in packaging; hence, their presence as microplastics in the ocean is not surprising. The abundance of PUR is more surprising. Foamed polyurethane is used for mattresses and upholstered furniture as well as for insulation purposes, and PUR may furthermore be used in some paints, but it is not evident how the PUR is released in such quantities that it is the dominant polymer for the larger particles. The methods for investigating polymer composition of the microplastics are not well developed, and results are only available from one study which is still not published. However, the study demonstrates that more investigations of polymer composition of the microplastics would be a valuable contribution to a better understanding of the sources and fate of microplastics in the ocean.

#### Catalogue of possible initiatives by national authorities in Denmark

Based on an analysis of the main data gaps and an assessment of the most obvious opportunities to reduce emissions of microplastics, a catalogue of possible new initiatives for implementation by national authorities in Denmark has been prepared. The proposals are an expression of the authors' view after obtaining feedback from the project's advisory group.

The catalogue of potential studies focusses on studies which could be undertaken by Danish national authorities and which are important for any new measures to limit emissions of microplastics. They relate to degradation of microplastics in sewage treatment plants, possible effects of microplastics in sewage sludge used for agricultural purposes, investigations of microplastics in food in Denmark, and investigation of effects of plastics on the nanoscale.

Within the framework of the OSPAR and HELCOM conventions, action plans for marine litter including microplastics have recently been developed. The action plans include a large number of initiatives in order to provide more knowledge on pollution prevention by macro- and microplastics and can serve as a basis for initiatives at regional and national levels. A reconciliation between the proposals for new initiatives in Denmark with the initiatives already taken in relation to the marine conventions has been attempted.

# Konklusion og sammenfatning

Denne rapport indeholder en gennemgang af den eksisterende viden om problemstillingerne i relation til forurening med mikroplast med fokus på anvendelse og udslip af mikroplast i Danmark og forekomsten af mikroplast i de omkringliggende farvandsområder.

Problemstillingen omkring forurening af havene med plast, og herunder mikroplast, er ikke ny, men den har de senere år internationalt fået øget opmærksomhed og der er en stigende bekymring for, om forureningen med plast på længere sigt har større effekter på miljøet end hidtil antaget. Bekymringen tager sit udgangspunkt i en række forhold:

- Plast, som frigives til miljøet, kan opholde sig i miljøet i mange hundrede år, før det endeligt nedbrydes.
- Det globale forbrug af plast er stigende, og de globale udslip til miljøet forudses også at være stigende med mindre der gribes ind over for udslippene.
- Plast, som ender i havet, kan transporteres over store afstande og selv de fjerneste steder på kloden er påvirket af forurening med plast.
- I miljøet neddeles plaststykkerne til mindre stykker dvs. plast i makrostørrelse nedbrydes til mikroplast, der igen nedbrydes til plast i nanostørrelse.
- Mikroplast er fundet i organismer på alle niveauer i den marine fødekæde.
- Der er resultater, der viser, at mikroplast kan have effekter på organismer i miljøet, men vores viden om omfanget af disse effekter er begrænset.
- Vi har stort set ingen viden om mulige særlige partikeleffekter som plastpartikler i nanostørrelse muligvis kan have som konsekvens af deres størrelse.
- Mennesker kan udsættes for mikroplast via fødevarer.

Kombinationen af fortsatte og måske stigende udslip, at stofferne bliver i miljøet i århundreder og over tid fragmenteres til mindre stykker, som kan have stigende effekter på miljøet, betyder at mikroplastproblematikken har elementer af en "tidsindstillet bombe", hvor effekterne af de udslip, der sker i dag, først ses om mange år, hvor effekterne kan være umulige at begrænse. Om det faktisk forholder sig sådan, ved vi endnu ikke.

#### Hvad er mikroplast?

Der skelnes i litteraturen mellem plast, i makro-, mikro- og nanostørrelse. Der er dog ikke en fast definition af mikroplast, men i mange sammenhænge - og også i denne rapport - er mikroplast fastsat til plaststykker af en størrelse på 1  $\mu$ m til 5 mm.

Der skelnes mellem primær og sekundær mikroplast. Primær mikroplast er plast, som tilsigtet anvendes i størrelser mellem 1  $\mu$ m til 5 mm. Sekundær mikroplast er dannet ved fragmentering af større plaststykker eller ved slitage af maling- eller plastoverflader. Sekundær mikroplast dannes både fra produkter i brug og fra større plaststykker, som er spredt i miljøet.

I denne rapport anvendes en bred definition af plast, som dækker alle faste materialer dannet ud fra polymerer af fortrinsvis petrokemisk oprindelse, hvilket betyder, at små fragmenter fra slitage af eksempelvis maling og dæk henregnes til mikroplast. Polymerer som f.eks. polyethylen (PE) kan, afhængig af polymerkædens længde, optræde som flydende, voksagtige og faste. Det er kun de faste,

som her henregnes til mikroplast, selvom det ikke kan afvises, at de voksagtige også kan optræde som partikler og måske have lignende effekter.

### Forekomst og effekter i miljøet

#### Forekomst af mikroplast i vandmiljøet

Der foreligger et stort datamateriale om forekomsten af mikroplast i vandmiljøet, men undersøgelserne er generelt foretaget med forskellige metoder, med forskellige opdelinger i partikelstørrelse og angivelser i forskellige enheder, der ikke kan omregnes, hvilket gør, at det er vanskeligt at sammenligne resultater på tværs af undersøgelserne. Små partikler optræder i langt større antal end relativt store partikler, hvilket gør, at bare små forskelle i de undersøgte partikelstørrelsesintervaller giver meget forskellige resultater. De foreliggende undersøgelser rapporterer antallet af partikler i forskellige størrelsesintervaller, mens der ikke findes opgørelser af massen af mikroplast i havområderne omkring Danmark.

For partikler større end ca. 300 µm er der i danske og svenske kystvande samt undersøgelser i Østersøen og Nordatlanten typisk fundet koncentrationer i intervallet 0,3 - 4 partikler/m³, med en tendens til højere koncentrationer nær større byer og i de indre danske farvande. Datamaterialet er dog spinkelt og forskelle mellem farvandsområder skal fortolkes med forsigtighed. I undersøgelser, hvor der er inddraget mindre plastpartikler, er der fundet langt højere koncentrationer, og i en undersøgelse som inddrog partikler ned til ≥10 µm blev der fundet 4.400-94.000 partikler/m³ langs den svenske vestkyst, mens en tysk undersøgelser af partikler ≥1.2 µm fandt gennemsnit på 64.000 og 88.000 partikler/m³ af henholdsvis granulære partikler og fibre. Koncentrationerne af mikroplast ≥10 µm i miljøet er af samme størrelse som koncentrationerne af mikroplast af denne størrelse i indløb til svenske og norske renseanlæg, mens koncentrationerne af partikler >300 µm er mere end 1000 gange mindre i miljøet end i indløb.

Gennemsnitskoncentrationen af mikroplastpartikler  $>38~\mu m$  i sedimenter fra farvandsområderne omkring Danmark varierede fra 240 partikler/kg tørstof i Nordsøen/Skagerrak til 1.100 partikler/kg tørstof i Bælthavet. Grundet stor variation inden for de enkelte farvandsområder og et lavt antal prøver var de fundne forskelle mellem de enkelte farvandsområder ikke statistisk signifikant. Koncentrationerne er dog signifikant højere end de, som er fundet i undersøgelser af partikler i samme størrelse fra belgiske strande og fastlandsoklen ud for Belgien.

Antallet af tilgængelige data fra Danmark er ikke tilstrækkelige til at pege på en sammenhæng mellem mulige kilder og koncentrationen i vandfasen og sedimentet.

#### Eksponering af organismer i vandmiljøet

Mikroplast er påvist i organismer på alle niveauer i den marine fødekæde. Mængderne af indtaget mikroplast varierer mellem arter og områder og kan variere betydeligt selv inden for samme område. I Nordsøen og i Storebælt har man fundet mikroplast i maven, indvoldene og/eller vævet af bl.a. sæler, sild, torsk, hvilling og muslinger. Det er velkendt, at marine dyr indtager mikroplast sammen med den føde, de spiser, og der er indikationer på, at dyrene spiser mikroplasten af den årsag, at den har den samme størrelse som deres normale føde, som f.eks. er alger. Studier tyder ligeledes på, at stort set alle marine dyr indtager mikroplast, men at der er stor variation mellem de forskellige arter hvad angår mængden, de indtager. Ligeledes er der studier på bl.a. krabber som indikerer, at mikroplast akkumulerer i fødekæden.

#### Skæbne og effekter af mikroplast i vandmiljøet

Der foreligger en række studier, der påviser skadelige effekter af indtagelse af makroplast så som indre og ydre læsioner og blokering af fordøjelseskanalen, som kan føre til en falsk fornemmelse af mæthed. Hvad angår plast i mikro- og nanostørrelse er der potentielt tre typer af skadelige virkninger forbundet med indtagelse: (1) fysiske virkninger relateret til indtagelse, som ligner dem, der er fundet for makroplast (men på mindre organismer), (2) toksisk respons fra frigivelse af farlige stof-

fer som stammer fra en tilsigtet anvendelse i plasten eller ved produktionen og, (3) toksisk reaktion på forureningsstoffer, der er adsorberet til mikroplasten. I mangel på muligheden for at lave feltobservationer har forskere brugt laboratorieeksperimenter til at undersøge mulige effekter. Der foreligger undersøgelser af biologiske virkninger af mikroplast på en række kategorier af organismer, såsom zooplankton, bundlevende organismer, fisk og havfugle, men samlet findes der stadig et meget begrænset datamateriale. I laboratorieeksperimenter har man fundet, at mikroplast kan have en signifikant negativ indflydelse på bl.a. krebsdyrs fødeoptag, sandormes spiseaktivitet og vægt samt, at mikroplast kan forårsage en form for betændelsestilstand i vævet hos blåmuslinger. I studier på fisk er der set sammenhænge mellem mikroplast og lever-stress og dannelsen af tumorer samt indikationer på hormonforstyrrende effekter. I laboratorieforsøg anvendes der ofte relativt høje koncentrationer sammenlignet med de koncentrationer, der findes i miljøet, og forsøgene omfatter kun få testorganismer sammenlignet med de mange forskellige arter, der findes i miljøet. Det er derfor endnu usikkert, i hvilken grad de observerede effekter i laboratoriet forekommer i miljøet.

#### Mikroplast som bærer af farlige stoffer

Der foreligger en række undersøgelser, der viser, at mikroplast i miljøet indeholder og/eller er bærer af farlige kemiske stoffer. Der er grundlæggende to typer af kilder til forekomsten af farlige stoffer i mikroplast: 1) stoffer, som tilsigtet er tilsat plasten eller er anvendt som udgangsstof ved produktionen af plasten og, 2) stoffer, som i miljøet binder sig til overfladen af plastpartiklerne og i mange tilfælde over tid videre optages i plasten.

Eksempler på påviste stoffer i mikroplast i miljøet er PBDE'er og andre bromerede flammehæmmere, der tilsigtet er tilsat samt nonylphenol og bisphenol A (BPA), som anvendes som udgangsstoffer ved produktion af forskellige typer plast. Blødgørere som phthalater og chlorparaffiner og tilsatte biocider vil utvivlsomt også kunne optræde i mikroplast i miljøet. Koncentrationen af flammehæmmere og phthalater i plast er typisk i størrelsen 5-30% og langt højere end de forekomster, der kan være af farlige stoffer, der optages i plasten fra det omgivende vand. Problemstillingen om spredning af farlige stoffer med plast, der tabes til miljøet, er ikke ny, og denne spredningsvej har typisk indgået i udslipsopgørelser og i forbindelse med risikovurderinger. EU's risikovurderinger for flere phthalater regner eksempelvis med, at plast spredt i miljøet er den største kilde til udslip af phthalater til miljøet og også malinger kan være en væsentlig spredningsvej for nogle stoffer. Udslip kan ske i form af spild af plastråvarer, dannelse af sekundær mikroplast ved slitage af produkter ved brug og bortskaffelse og spredning af plastdele, som efterfølgende fragmenteres til mikroplast i miljøet. Ved indtag af mikroplasten kan organismer eksponeres for de farlige stoffer, men stofferne kan også spredes fra plasten til andre matricer i miljøet og derfra videre til organismerne.

Nogle kemiske stoffer har tendens til at adsorbere til overfladen af plast og for nogle stoffer og plasttypers vedkommende optages de i plasten, og der kan derved ske en opkoncentrering så koncentrationerne bliver meget højere i plasten end i det omgivende miljø. Dette er påvist for en række persistente organiske miljøgifte (POP-stoffer) som PCB og DDT, og optag af POP-stoffer i plastråvarer (pellets) tabt til miljøet har i mange år været brugt til at monitere den globale udbredelse af POPstoffer. Det åbne spørgsmål er, i hvilket omfang dette optag på en væsentlig måde øger nogle organismers eksponering for stofferne, og i hvilken grad optag i plastpartikler kan resultere i spredning af stofferne fra områder med høj lokal belastning til områder med lavere belastning. De undersøgelser og modelberegninger, der foreligger, tyder på, at forekomsten af mikroplast vil kunne have en vis effekt på, hvordan POP-stofferne fordeler sig i miljøet, men der er ikke noget der tyder på, at det generelt giver øget eksponering (konklusionen vedrører ikke de farlige stoffer, som tilsigtet er tilsat til plasten i høje koncentrationer, som nævnes ovenfor). Resultaterne tyder heller ikke på, at transport af mikroplast med havstrømme bidrager væsentligt til transporten af POP-stoffer til eksempelvis de arktiske egne. Det centrale i relation til POP-stoffer er stadig at undgå, at de overhovedet spredes til miljøet. Der er dog noget usikkerhed og lidt modstridende resultater, så flere resultater er nødvendige for at kunne komme med mere sikre konklusioner.

#### Mikroplast i jordmiljøet

Mikroplast kan ende i jordmiljøet ved udbringning af slam og kompost indeholdende mikroplast og ved slitage af udendørs plastdele og malede overflader. Undersøgelser viser, at mikroplasten forbliver i jorden i mange år, og mikroplast i jorden har - længe før mikroplast i sig selv blev en problematik - været foreslået som en indikator for, hvor meget slam en jord var blevet tilført. Der er ikke fundet undersøgelser af effekterne af mikroplast på dyr, som lever i jorden. Da mange dyr i jorden ernærer sig på samme måde som sedimentlevende dyr undersøgt i vandmiljøet, må det i første omgang antages, at der vil kunne være samme eksponeringsveje og mulige effekter af mikroplast på jordlevende dyr. Denne antagelse bør dog nærmere verificeres gennem konkrete undersøgelser.

#### Sundhedsmæssige aspekter

Mennesker kan udsættes for mikroplast ved brug af kosmetik med mikroplast, gennem føden, gennem indtagelse af støv i indemiljøet og i forbindelse med brug eller vedligeholdelse af plastdele eller malede/plastbelagte overflader. Det har været uden for rammerne af denne gennemgang at se på mulige sundhedsmæssige effekter af mikroplast, og i hvilken grad, der eksempelvis kunne uddrages viden fra undersøgelser af eksempelvis effekter af maling- og plaststøv i arbejdsmiljøet.

Det tyske føderale institut for risikovurderinger (BfR) har vurderet, at mikroplast i kosmetik ikke udgør en sundhedsmæssig risiko, fordi plastpartiklerne har en størrelse, så de ikke optages via tarmen. Der er i de senere år kommer en række undersøgelser, der viser at mikroplast kan forekomme i fødevarer. Mikroplast er således påvist i bl.a. muslinger, øl og honning. Det tyske føderale institut for risikovurderinger har vurderet, at der ikke er tilstrækkelige data vedrørende den kemiske sammensætning, partikelstørrelse og koncentration af mikroplast i fødevarer til at foretage en vurdering af mulige sundhedsmæssige risici af mikroplast i fødevarer, og har opfordret den Europæiske Fødevarautoritet (EFSA) til at udarbejde en redegørelse om plast i mikro- og nanostørrelse i fødevarer.

#### Mikroplast i spildevand og i renseanlæg

Der er ikke fundet undersøgelser af mikroplast i renseanlæg i Danmark. Undersøgelser af omsætningen af mikroplast i renseanlæg i Norge, Sverige og Tyskland viser, at tilbageholdelseseffektiviteten er afhængig af størrelsen af partiklerne, mens formen af partiklerne er uden væsentlig betydning. For mikroplast  $\geq 300~\mu m$  ender mere end 99% i slamfasen, mens det for mikroplast i størrelsesintervallet 20 til 300  $\mu m$  typisk kun er 80 til 90%, der ender i slamfasen. Der er ingen undersøgelser af tilbageholdelsen af mikroplast < 20  $\mu m$ . De tilgængelige undersøgelser har alle opgjort antallet af partikler, mens der ikke er undersøgelser hvor den samlede vægt af partiklerne er beregnet.

Plastfibre udgør det største antal mikroplastpartikler ≥300 µm, hvilket indikerer, at tekstiler mængdemæssigt er en meget væsentlig kilde til mikroplast i spildevandet. Antallet af partikler i størrelsesintervallet 20 til 300 µm er væsentligt større end antallet af partikler ≥300 µm og for de disse partikler udgør fibrene typisk omkring en tredjedel af partiklerne (men vægtmæssigt bidrager de små partikler mindre). Antallet af ikke-syntetiske fibre er typisk 5-10 gange større end det samlede antal af mikroplastpartikler. Det er på baggrund af de foreliggende undersøgelser ikke muligt at pege på, hvor meget af den del, der ikke er fibre, der stammer fra tilsigtede anvendelser af primær mikroplast (f.eks. i kosmetik og plastråvarer), og hvor meget der stammer fra slitage/bearbejdning af plastdele og maling. Tyske undersøgelser af polymersammensætningen af mikroplast i spildevand peger på, at PE og PP udgør mere end 90% af partiklerne >500 µm (ikke fibre), mens PE og PVA udgør mere end halvdelen af de mindre partikler. "Malingslignende" polymerer udgør 5-10% af det samlede indhold af de mindre partikler. Metoderne er dog stadig under udvikling og mængden af undersøgte partikler er lille, så forfatterne påpeger, at resultaterne skal tolkes med forsigtighed. Mikroplastpartikler i form af støv fra dæk, afslibning af maling og slid på eksempelvis vinylgulve, som samlet udgør en meget stor del af udledningerne, er typisk så små, at

de ikke vil være omfattet af de undersøgelser af mikroplast i spildevandsrenseanlæg og i miljøet, der hidtil er foretaget.

Der er ingen undersøgelser af nedbrydning af mikroplast i renseanlæg, men mikroplasten nedbrydes formentlig kun i meget begrænset omfang, og hovedparten af mikroplasten vil derfor ende i slammet, som i Danmark enten brændes eller udbringes på landbrugsjord.

#### Kilder til udslip af mikroplast i Danmark

#### Anvendelse og udslip af primær mikroplast i Danmark

Blandt de mængdemæssigt væsentligste anvendelser af primær mikroplast i produkter i Danmark er mikroperler i kosmetik, forskellige typer af mikroplast i maling og små plastpartikler som anvendes til blæserensning. Herudover anvendes primær mikroplast i stort omfang i produktionen af plastemner og gummigranulat fra genanvendelse af dæk anvendes til blandt andet kunstgræsbaner og legepladser. Udslip fra anvendelserne vil typisk være til spildevand (jord er dog den vigtigste vej for gummigranulat), hvorfra en del vil ende i vandmiljøet, mens hovedparten ender i spildevandsslam, som delvist anvendes til jordbrugsformål. Der er herudover en række anvendelser, som det inden for projektets rammer ikke været muligt at estimere forbruget af og de tilhørende udslip.

De samlede opgørelser af kilder til udledninger af primær og sekundær mikroplast i Danmark er vist i nedenstående tabel o. Kilderne beskrives nærmere under tabellen. For udledninger til vandmiljøet er der i tabellen opgjort de resulterende udledninger til vandmiljøet efter spildevandsrensning. Der er regnet med, at en del af spildevandet udledes urenset i forbindelse med kraftige regnhændelser. Med den høje tilbageholdelsesgrad i renseanlæggene, vil de direkte udledninger kunne bidrage væsentligt til de samlede udledninger. For mikroplast, der udledes med regnvandsbetingede udledninger fra befæstede arealer, vil der være en del, som ender som fast affald i forbindelse med oprensning af sedimentationsbassiner. Da der kun er en del af de separatkloakerede arealer der er forsynet med sedimentationsbassiner er der regnet med, at det i gennemsnit kun er 10-20% af mikroplasten i de separatkloakerede udledninger, der tilbageholdes.

De samlede udledninger til spildevand er anslået til 2.000-to 5.600 t/år. De største kilder vurderes at være dæk og tekstiler, men der er en række kilder som kan bidrage med betydelige mængder.

Mængderne er opgjort i tons pr. år, og der er ikke et tilstrækkeligt datagrundlag til at beregne antal partikler pr. år. Partikler i plastråvarerne og gummigranulat er relativt store i forhold til andre partikler, som typisk vil forekomme som støv/pulver, og plastråvarerne og gummigranulat ville repræsentere en markant mindre andel, hvis udledningerne alternativt blev angivet i antal partikler. Udledningerne er til den umiddelbare recipient, og der er ingen grundlag for at beregne, i hvilken grad partiklerne vil kunne spredes videre i miljøet, og hvilke effekter de vil kunne have. Partikler fra dæk, maling, vinylbelægninger (PVC) og tekstilfibre vil typisk have en vægtfylde over én, og må dermed forventes i mindre grad at spredes i miljøet end polyethylen i kosmetik, en stor del af plastråvarerne, EPS kugler, polyurethanskum og mikroplast i maling, som har en vægtfylde under én. Der er ikke basis for at vurdere, om nogle typer mikroplast har større potentiale for effekter på miljøet end andre, bortset fra effekter af farlige kemiske stoffer, som forekommer i nogle af partiklerne (flammehæmmere, blødgørere, mm).

**TABEL 0**OVERSIGT OVER SAMLEDE UDSLIP AF PRIMÆR OG SEKUNDÆR MIKROPLAST I DANMARK (EXCL. DANNELSE AF MIKROPLAST FRA MAKROPLAST I MILJØET)

Produktgruppe	Samlet udslip ton/år	% af sam- let udslip (gennem- snit)	Udledning til renseanlæg ton/år	Resulterende mængder der ender i vand- miljøet, ton/år*	% af samlet udslip til vandmiljøet (gennemsnit)			
Kilder til emission af primær mikroplast								
Personlig pleje	9-29	0,2	10-22	0,5-4,4	0,1			
Plastråvarer	3-56	0,3	3-56	0,1-4,5	0,1			
Maling	2-7	0,1	2-7	0,3-1,8	0,1			
Blæsemidler	0,05-2,5	0,01	0,03-1,3	0,03-1,4	0,04			
Gummigranulat	450-1.580	10,5	20-330	1-20	0,6			
Andre anvendelser	?	?	?	?	?			
Samlet, primær mikroplast	460-1.670	11	35-416	2-31	0,9			
Kilder til emission af	sekundær mik	roplast						
Dæk	4.200-6.600	56	1.600-2.500	500-1.700	60			
Tekstiler	200-1.000	6,2	200-1.000	6-60	1,8			
Maling (ekskl. skibsmaling)	150-810	4,9	14-220	6-149	4,2			
Skibsmaling	40-480	2,7	0-50	21-240	7,1			
Vejstriber	110-690	4,1	40-260	10-180	5,1			
Byggematerialer af plast	80-480	2,9	30-150	5-38	1,1			
Fodtøj	100-1.000	5,7	40-380	10-260	7,3			
Køkkenredskaber, skuresvampe mm.	20-180	1,0	20-180	1-50	1,4			
Andre anvendelser	100-1.000	5,7	20-500	8-375	10			
Samlet, sekundær mikroplast (afrun- det)	5.000- 12.200	89	2.000-5.200	600-3.050	98			
Samlet i alt (af- rundet)	5.500-13.900		2.000-5.600	600-3.100	99			

<sup>\*</sup> Angiver udslip efter forudgående rensning.

Kosmetik - Mikroplastperler har typisk været brugt i kosmetik som slibemiddel til at opnå en skrubbende effekt og mikroplast er især anvendt i skrubbecremer til ansigtet, "shower gel" og håndrensemidler. Anvendelserne er grundigt beskrevet i litteraturen. Farvet mikroplast bruges også i et vist omfang til at opnå en farveeffekt, eksempelvis i tandpasta, og visse typer mikroplast anvendes til at give en glimmereffekt. Den mest anvendte polymer er polyethylen (PE). Polyethylen og andre polymerer af kortere kædelængde, som er flydende eller voksagtige, anvendes tilsyneladende i en lang række kosmetikprodukter, men disse anvendelser er uden for definitionen af mikroplast anvendt i denne undersøgelse og er ikke nærmere undersøgt.

En europæisk undersøgelse udarbejdet af forskellige aktører inden for kosmetikindustrien har estimeret det samlede indhold af mikroplast i skrubbe- og renseprodukter solgt i Danmark til 29 tons i 2012. Estimatet omfatter ikke mikroplast i tandpasta samt i andre typer af produkter, hvor mikroplast er tilsat for at opnå farve- og glittereffekter. I følge oplysninger fra brancheforeningen SPT har forbruget været faldende idet mikroplast er erstattet i mange produkter, og det er derfor lavere i 2014 end estimeret for 2012. I følge en undersøgelse blandt foreningens medlemmer vil mikroplast være udfaset i 2017 i stort set alle kosmetikprodukter, men forventes stadig at anvendes til visse håndrensemidler. I følge oplysninger fra branchen arbejdes der også på udfasning af mikroplast i håndrensemidlerne. Der er i dag ingen kendt anvendelse af mikroplast i tandpasta solgt i Danmark. Under hensyntagen til, at forbruget i 2012 er usikkert bestemt og muligvis er underestimeret, er det samlede indhold af mikroplast i kosmetikprodukter solgt i Danmark i 2014 estimeret til 9-29 tons. Af dette antages 90% af blive vasket af og ende i spildevand. Siden 2013 har der i den europæiske brancheforening Cosmetics Europe været enighed om ikke at støtte brugen af mikroplast i kosmetik, og det samme synspunkt støttes af den danske brancheforening.

**Plastråvarer** - Plastråvarer til fremstilling af plastartikler af termoplast i Danmark importeres som plastpiller (pellets) eller pulver og stort set alle faste plastråvarer falder ind under definitionen af mikroplast. I forbindelse med plastfremstillingen smeltes råvarerne sammen, så de færdige artikler vil ikke indeholde mikroplast. Med et forbrug på ca. 550.000 tons/år er det langt den største anvendelse af primær mikroplast. Udslip, enten direkte til miljøet eller mere typisk til spildevand, vil primært kunne ske i forbindelse med transport, omlastning og oplagring, hvorimod udslip fra selve produktionsprocessen vurderes at være ubetydelig. Der er ingen krav i standardbetingelser til miljøgodkendelser for plastproducerende virksomheder vedr. udslip af plastråvarer. Udenlandske undersøgelser har påvist, at udslip af plastråvarer kan være betydelige og den største del af mikroplast i Donau vurderes at være plastråvarer, som er udledt til floden. En væsentlig del af udslippene vil dog foregå i forbindelse med produktion og udlastning af plastråvarerne og disse processer foregår ikke i Danmark.

I regi af brancheorganisationen Plastindustrien forsøger man med programmet Operation Clean Sweep at sætte fokus på metoder til nedbringelse af spild, men der er i skrivende stund kun 9 ud af 250 plastforarbejdende virksomheder i Danmark, der har tilsluttet sig programmet. Data om spild-procenter indsamlet via Plastindustrien fra virksomheder, som har tilsluttet sig Operation Clean Sweep indikerer, at tab til spildevand inden for virksomhedens område (inkl. aflastning fra lastbiler, som bringer råvarerne) maksimalt drejer sig om 0.001% af råvareforbruget. Dette er langt lavere end "worst case" standard emissionsfaktorer udviklet af OECD for tab til spildevand. Det vides ikke, hvor meget højere de gennemsnitlige emissioner fra alle plastvirksomheder i Danmark vil være, men det anslås, at gennemsnittet næppe er mere end 10 gange højere end de højeste værdier angivet i Plastindustriens undersøgelse. På grundlag af de foreliggende data er de samlede udslip af mikroplast til spildevand anslået til 3-56 tons/år.

**Maling** - Partikler af mikroplast anvendes i vidt omfang i bygningsmaling til at mindske behovet for pigment, mindske vægtfylden, og give særlige overfladeegenskaber. Ud over mikroplast, som defineret i denne rapport, anvendes i vid udstrækning vokspartikler til at give overfladestrukturer. Det samlede forbrug i Danmark i 2014 er af brancheforeningen DFL anslået til ca. 254 tons i samlet 64.000 tons maling. Når malingen hærder op, vil plastpartiklerne indgå som en del af den samlede malingmatrice og ved slid og afslibning vil mikroplasten indgå som en integreret det af malingpartiklerne, som omtales videre under sekundær mikroplast. Ved brug af vandmalinger vil omkring 1% af den uhærdede maling ende i spildevand i forbindelse med rengøring af pensler og andet værktøj. Det vides ikke i hvilken grad mikroplasten i spildevandet vil gå i forbindelse med binderen i malingen eller vil kunne frigives og suspenderes i spildevandet som frie partikler. På det foreliggende grundlag er de samlede udslip af primær mikroplast til spildevand med maling anslået til 2-7 tons. Ved anvendelse udendørs vil en del af malingen spildes til jord, men den tabte maling vil formentlig

hærde op, så der ikke ad den vej er en videre transport af de frie partikler i malingen, men at hele malingen som sådan kan bidrage til udslip af sekundær mikroplast.

**Blæsemidler** - Der er i litteraturen meget få oplysninger om brugen af mikroplast som blæsemidler til afrensning af overflader. Undersøgelsen viser, at blæsemidler af mange forskellige typer af plast anvendes til forskellige formål i Danmark. Blæsemidlerne genbruges typisk 10-20 gange. Det samlede forbrug af plastbaserede blæsemidler i Danmark er anslået til 5-25 tons. Langt de fleste anvendelser er indendørs, hvor der regnes med at være et meget begrænset spild til spildevand. Der er dog også anvendelser, som delvist vil kunne foregå udendørs på skibe og offshore installationer, hvor der vil kunne være et direkte spild til omgivelserne. Ud fra de foreliggende data anslås groft, at der vil kunne være et tab til vandmiljøet i størrelsen 0,1-1,3 tons/år.

**Gummigranulat** - Størrelsen af gummigranulat fra genbrug af dæk varierer mellem 0,7 og 3 mm, som betyder at gummigranulat henregnes til primær mikroplast, som det er defineret i denne rapport. Granulatet anvendes som fyld (infill) i kunstgræsbaner til fodbold, rugby og golf. Kombineret med et bindemiddel, anvendes granulatet blandt andet til legepladser og løbebaner og der er desuden et betydeligt forbrug til polymer-modificeret asfalt. Slid på kunstgræsbaner og andre områder med gummibelægning vil frigive gummigranulat anvendt på banerne, og desuden vil der dannes sekundær mikroplast, som frigives ved slid på de syntetiske græsfibre. Der er stor usikkerhed om, i hvilket omfang mikroplast, som frigives fra banerne, ender i dræn og kloaksystemer. De samlede mængder, der tilledes spildevand, er anslået til 20-330 t/år (inkl. sekundær mikroplast). Det skal bemærkes, at udledninger fra anvendelsen af gummigranulat er små sammenlignet med udledningerne af partikler fra dækkene, før de genanvendes.

**Rengørings- og plejemidler** - Brancheforeningen SPT har ingen oplysninger om brugen af mikroplast i rengørings- og plejemidler bortset fra håndrensemidler som indgår i opgørelsen for personlige plejemidler. Der er heller ikke fundet konkrete oplysninger om denne anvendelse i litteraturen, men den er nævnt som en mulig anvendelse, og det kan ikke afvises, at der kan være en vis anvendelse, hvor mikroplasten anvendes som skrubbemiddel i plejemidler på samme måde som i kosmetik.

**Andre anvendelser** - Der er en række anvendelser af primær mikroplast, som det inden for rammerne af dette projekt ikke har været muligt at beskrive i detaljer. Det drejer sig om kugler af ekspanderet polystyren til møbler og hulmursisolering, toner til laserprintere, plastkugler som anvendes i visse industrielle opvaskemaskiner, medicinske formål, forskningsformål og mikroplast anvendt som specialkemikalier. De samlede mængder anvendt til enkelte af anvendelsesområderne kan muligvis være betydelige, men de resulterende udslip vil formentlig udgøre en begrænset del af de samlede udslip.

# Dannelse af sekundær mikroplast fra artikler i brug og malede/plastbelagte overflader

Der er en lang række kilder til dannelse af sekundær mikroplast, hvoraf de vigtigste vurderes at være slid på dæk og malede striber på veje, slid på tøj og andre tekstiler af syntetiske fibre, slid og afslibning af maling, slid på store overflader af plast som eksempelvis vinylgulve og tagbelægninger, slid på skosåler og slid på kunstgræsbaner. Udslip herfra vil være til kommunale renseanlæg, til vandmiljøet via regnvandsbetingede udløb og overfladeafstrømning eller direkte til vandmiljøet eksempelvis ved aktiviteter på havne eller skibe. Partiklerne, som dannes ved slid på dæk og plastmaterialer (f.eks. vinylgulve) samt ved slibning af maling, vil typisk være mindre end 10  $\mu$ m og dermed ikke være omfattet af de undersøgelser, der er lavet af mikroplast i miljøet og spildevand. Det er en mulig forklaring på, at partikler fra disse væsentlige kilder ikke mere markant viser sig i de undersøgelser, der er af polymersammensætningen af mikroplast i spildevand og miljøet.

**Dæk og vejstriber** - Partikler, som afslides fra dæk og vejstriber indgår i vejstøv, som dels afledes med spildevand, dels spredes til det omgivende jord og overfladevand. Den samlede dannelse af

dækpartikler er anslået til 4.200-6.600 t/år, og vurderes at være den største enkeltkilde til spredning af mikroplast til miljøet. Udslippet er relativt sikkert bestemt. De samlede udslip til vandmiljøet fra brug af vejstriber anslås til 10-180 tons, og vurderes dermed også at være en betydende kilde.

**Tekstiler** - Der dannes væsentlige mængder af fibre i mikroplaststørrelse fra slid på tekstiler, dels ved brug, dels i forbindelse med tøjvask. Syntetiske fibre, som mest sandsynligt stammer fra tekstiler, udgør en meget stor del af mikroplast ≥20 µm i renseanlæg og er også påvist at udgøre en væsentlig del af mikroplast i kystnære farvande. Der foreligger et meget begrænset datamateriale, men ved at sammenholde de begrænsede data om udslip ved tekstilvask med viden om mængden af fibre i spildevand, der tilføres renseanlæg, er det muligt at anslå en sandsynlig størrelsesorden på dannelsen af mikroplast. På det foreliggende grundlag anslås mængderne af syntetiske fibre, der tilføres renseanlæg, at være 200-1.000 t/år.

**Maling (bortset fra skibsmaling)** - Der dannes mikroplastpartikler ved slitage på maling og ved afslibning/afskrabning af malingen i forbindelse med vedligeholdelse af de malede overflader. Der vil primært kunne ske udslip til miljøet fra malinger anvendt udendørs og opgørelsen fokuserer derfor på disse malinger. De samlede udslip anslås til 150-810 t/år hvoraf 14-220 t/år anslås at tilføres renseanlæg via afstrømning fra befæstede arealer.

**Skibsmaling** - Mikroplast partikler kan dannes ved slid og vedligeholdelse af marine malinger, der anvendes til lystbåde og større skibe. En væsentlig del af udledningerne vil være direkte til vandmiljøet. De samlede udledninger til vandmiljøet fra anvendelse af maling på lystbåde anslås til 5-50 t/år, mens i størrelsen 16-190 t/år anslås at blive udledt fra brugen af maling til større fartøjer. Desuden kan mikroplast muligvis dannes ved frigivelse af selvpolerende antibegroningsmalinger, når fartøjerne er i vandet, men der findes ingen oplysninger om denne mulige kilde.

**Byggematerialer af plast** - PVC, såvel blødgjort som hård PVC, udgør hovedparten af de byggematerialer af plast, som vil kunne udsættes for slid og forvitring. Der foreligger nogen viden, som er frembragt i forbindelse med at estimere udslip af phthalater og tungmetaller, som anvendes i PVC. De væsentligste kilder vurderes at være gulvbelægninger, tagbelægninger og tagrender mm. af PVC. De samlede udslip af mikroplast fra byggematerialer til spildevand er anslået til 30-150 t/år.

**Fodtøj** - Såler på fodtøj er typisk lavet af PVC, polyurethan eller syntetisk gummi. Ved slid på sålerne dannes der mikroplastpartikler. Der foreligger nogle estimater for frigivelse af plast fra såler af PVC og på det grundlag er de samlede udslip til spildevand og vejvand estimeret til 40-380 t/år.

**Køkkenredskaber**, **skuresvampe og klude** - Slid på redskaber, klude og svampe af plast anvendt i køkkener og på badeværelser vil kunne resultere i en afgivelse af mikroplast, som direkte afledes til spildevand. De største kilder vurderes at være skuresvampe og syntetiske multiklude, som det er forsøgt at udarbejde et estimat for, mens slid på køkkenredskaber vurderes at udgøre mindre end disse to kilder. På baggrund af den tilgængelige viden er de samlede udslip til spildevand anslået til 20-180 t/år.

**Andre kilder** - Der er en lang række andre mulige kilder til dannelse af sekundær mikroplast, som det inden for rammerne af denne undersøgelse ikke har været muligt at undersøge nærmere. De vigtigste vurderes at være industriel og professionel bearbejdning af materialer og artikler af plast, fiskenet og andre fiskeredskaber, polyetylenfolier anvendt i landbruget, polymermodificeret bitumen, bioaffald, papirgenanvendelse, bilfragmenteringsanlæg (shredderanlæg) og udstyr til fragmentering af madaffald på skibe. På basis af erfaringerne fra udenlandske undersøgelser, anslås det groft, at de samlede udledninger fra disse anvendelser vil være i størrelsen 100-1.000 t/år.

På basis af erfaringerne fra udenlandske undersøgelser vurderes det groft, at de samlede udledninger vil være inden for intervallet 100-1000 tons/år.

#### Dannelse af mikroplast ud fra makroplast i miljøet

Mikroplast dannes i miljøet ved fragmentering af større plaststykker, som er spredt i miljøet. Der findes meget viden om, hvilke typer af plast, som skyller op på strandene, og dermed meget viden om, hvilke kilder der er til makroplast i miljøet. Det er vurderet, at der i 1990'erne blev dumpet omkring 20.000 tons affald i Nordsøen, hvoraf en stor del antages at være plastaffald. Det er vurderet, at 15% af dette ender oppe på strandene, mens den øvrige del flyder rundt og synker til bunden efter delvist at være blevet fragmenteret. Der findes ingen specifikke opgørelser for plast og ingen beregninger af med hvilken rate og i hvilke mængder, mikroplast dannes ud fra makroplasten når den flyder eller ligger på bunden. Tidsperspektivet for en total nedbrydning (mineralisering) af plast kan være mange hundrede år, men de tilgængelige data for fragmentering, indikerer, at mange typer af makroplast i et vist omfang fragmenteres inden for et tidsperspektiv af år eller tiår.

Der må derfor regnes med, at en væsentlig del af de plaststykker, som ender ude i havmiljøet, faktisk når at fragmentere inden de fjernes fra strandene eller dækkes af sedimentet. Der er ikke fundet modelberegninger, der ud fra viden om forekomst af makroplast i miljøet, estimerer mængden af mikroplast, der dannes. En norsk opgørelse kommer med, hvad forfatterne betegner som et "bedste gæt" på, hvor meget mikroplast der kan dannes fra fragmentering fra makroplast i vandmiljøet omkring Norge. Opgørelsen når frem til en mængde på 360-1.800 t/år fra de vigtigste kilder, men den reelle usikkerhed på estimatet vurderes at være større end intervallet antyder. Det viser dog, at dannelse af mikroplast fra makroplast i miljøet formentlig er væsentlig større end udledningerne knyttet til brugen af primær mikroplast, mens det er mere usikkert om der er tale om lige så store mængder som udledningerne af sekundær mikroplast. Da der ikke er et datamateriale, der tillader bedre estimater end dette "bedste gæt", er det i nærværende undersøgelse ikke forsøgt at etablere et selvstændigt "bedste gæt", men at referere til den norske undersøgelse.

#### Vurdering af kilder til mikroplast i havmiljøet omkring Danmark

Mikroplast i havmiljøet omkring Danmark vil stamme fra lokale landbaserede kilder, skibsfart og andre havbaserede kilder, fragmentering af plastaffald i miljøet, eller det kan være tilført udefra med havstrømme. Der findes ingen målinger eller modeller, som kan pege på betydningen af de forskellige kilder, og der er ingen modeller, der beskriver den endelige skæbne af mikroplasten, som tilføres til eller dannes i havmiljøet omkring Danmark. De foreliggende undersøgelser af mikroplast i havmiljøet giver kun meget begrænsede oplysninger om mulige kilder.

Der ses en generel tendens til højere koncentrationer af mikroplast i kystnære lokaliteter nær større byer og højere koncentrationer i de indre danske farvande end i de omgivende havområder, som kunne tyde på at mikroplasten i højere grad skyldes lokale kilder end fra indstrømning fra tilgrænsende havområder. Antallet af målinger er dog endnu for beskedent til at udarbejde sikre konklusioner, og der vil ligeledes være behov for mere detaljerede modelleringer af udledninger, transport og dannelse af mikroplast i farvandene. I en svensk undersøgelse er der fundet høje koncentrationer af mikroplast ud for en plastproducerende virksomhed, som viser, at der vil kunne optræde høje koncentrationer nær punktkilder (virksomheden har siden fået installeret rensningsforanstaltninger). En ny undersøgelse peger endvidere på, at mikroplast på en enkelt af de undersøgte stationer sandsynligvis var plastråvarer, som var spildt.

En svensk undersøgelse (af partikler >300 µm) har fundet højere koncentrationer i Øresund end på de øvrige stationer langs de svenske kyster i Kattegat og Øresund, og tyske undersøgelser (partikler >100 µm) finder større koncentrationer i Kattegat og Østersøen syd for Lolland-Falster end i Nordsøen. Svenske undersøgelser har desuden fundet større koncentrationer i havet ud for større byer end i områder ud for mindre byer, og renseanlæg er påvist at være en kilde til lokale høje koncentrationer omkring udløb. Dette kunne pege på, at det, i hvert fald i kystnære farvande, er lokale kilder, der er af størst betydning. Der er ved undersøgelserne i kystnære farvande fundet en meget stor forekomst af fibre, som kunne tyde på at renseanlæg kunne være en væsentlig kilde, men også fiskeredskaber bidrager til forurening med fibre. Der er ikke fundet undersøgelser, hvor tykkelsen af

fibrene og polymersammensætningen er rapporteret, hvilket ellers kunne hjælpe til at bestemme, om kilderne var tekstiler eller fiskeredskaber. De foreliggende resultater skal generelt fortolkes med forsigtighed på grund af det begrænsede datamateriale. Undersøgelserne fra de åbne havområder er ikke omfattende nok til at pege på mulige kilder, og der er ikke grundlag for at vurdere, om der til de åbne havområder er en nettotilførsel eller fraførsel med havstrømme.

En nylig tysk undersøgelse har vist, at for partikler >500 µm er de mest forekommende polymerer polyurethan (PUR, gennemsnit 51%), polyethylen (PE, 29%) og polypropylen (PP, 17%). PE og PP er de plasttyper, der både anvendes i de største mængder, har vægtfylde over 1 og anvendes i udstrakt grad til emballage, så det er oplagt, at de forekommer som mikroplast i havet. Men det er ikke umiddelbart oplagt, ud fra analysen af mulige kilder, at pege på, hvorfor polyurethan optræder i så store mængder. Opskummet polyurethan anvendes til madrasser og polstermøbler samt til bygningsisolering, og polyurethan anvendes desuden i maling, men det er ikke klart, hvorledes plasten skulle spredes til miljøet i så store mængder, at det er den dominerende plasttype for de større partikler. Der er tale om nye metoder, og der kun foreligger kun en enkelt undersøgelse, som endnu ikke er publiceret, men resultaterne viser, at undersøgelser, hvor der ses nærmere på polymersammensætningen af mikroplasten, vil være et værdifuldt bidrag til at få en bedre forståelse af kilderne til mikroplast i havet og skæbnen af denne.

#### Katalog over mulige initiativer af nationale myndigheder i Danmark

På grundlag af en analyse af de vigtigste mangler i den eksisterende viden og en vurdering af de mest oplagte muligheder for at begrænse udslip af mikroplast, er der udarbejdet et katalog med forslag til mulige nye initiativer, som kan iværksættes nationalt i Danmark. Forslagene er alene udtryk for forfatternes umiddelbare opfattelse efter at have indhentet kommentarer fra projektets følgegruppe.

Kataloget over mulige undersøgelser fokuserer på undersøgelser, som kunne iværksættes af nationale danske myndigheder og som er af betydning for eventuelle nye tiltag til begrænsning udslip af mikroplast. De vedrører omsætning af mikroplast i renseanlæg, mulige effekter af mikroplast i slam fra renseanlæg som anvendes til jordbrugsformål, undersøgelse af mikroplast i fødevarer i Danmark, og undersøgelse af effekterne af plast i nanostørrelse. Der er i regi af OSPAR og HELCOM for nylig udarbejdet handlingsplaner for marint affald, herunder mikroplast, og der er i den sammenhæng igangsat en lang række initiativer, som skal give mere viden om, hvad der kan gøres for at forhindre forurening af havene med makro- og mikroplast, og danne grundlag for yderligere initiativer på regionalt og nationalt niveau. Det er forsøgt ved forslag til nye initiativer i Danmark at afstemme forslagene med de initiativer, der allerede er taget i relation til havkonventionerne.

# 1. Introduction

### 1.1 Definition of microplastics

#### 1.1.1 Size definitions

Different classifications of plastic debris based on their physical dimensions have been suggested and used in the scientific literature, reports and international organisations.

For example, Arthur *et al.* (2009) and The Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) (GESAMP 2015) have defined microplastics as a size range between 333  $\mu$ m-5 mm and 1 nm-5 mm, respectively. Some of the proposed size limits are based on pragmatic rather than scientific concerns and are determined by the techniques used for field sampling of microplastics. For example, some studies refer to a lower size limit for microplastics of 333  $\mu$ m which is based on a commonly used net mesh size for collection of plankton and debris in the water column (Arthur *et al.* 2009).

No official definition of microplastics has been adopted (Hidalgo-Ruz et~al., 2012), but definitions for what could be termed "micro-", "meso-", "macro"- and "mega-"plastic debris have been proposed and generally accepted according to the US EPA (2011). "Micro-", "meso-", and "macro"-plastic debris would be defined as <5 mm, 5–20 mm and >20 mm diameter, respectively, whereas "mega-" plastic debris would be >100 mm (US EPA 2011).

The definition used by GESAMP (2015) is the only currently proposed definition that also covers what has elsewhere been termed "nanoplastics". In 2011, the European Commission came forward with a proposed definition of nanomaterials which defines these as single particles, aggregates or agglomerates for which 50% or more of the nanomaterial in a number size distribution is within the range of 1 - 100 nm (European Commission 2014). In order not to confuse "nanoplastics" with "nanomaterials", in this report the term "plastics in the nano range" is used. A second term besides microplastics, which sometimes occurs in the literature, is "neuston" plastic which is defined as particles  $\geq$ 500  $\mu$ m, referring to particles that have been broken down to a small size and are now floating just at or below the surface of the water and that are caught in nets designed to catch surface plankton.

Within the context of this report, we classify debris with sizes of 1  $\mu m$  - 5 mm as microdebris. On this basis it follows that plastic particulates with these physical dimensions are considered to be microplastics.

#### Microplastics as a sub-category of marine (micro) litter

Microplastic debris is one out of several types of man-made anthropogenic litter found in the environment, including marine, freshwater and terrestrial compartments. Marine litter, in particular, has received increasing scientific, public and regulatory focus over the past decades. A general definition of marine litter has been put forward by the United Nations Environment Programme (UNEP):

#### **Definition of marine litter (UNEP 2005)**

"Marine litter is any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment. Marine litter consists of items that have been made or used by people and deliberately discarded into the sea or rivers or on beaches; brought indirectly to the sea with rivers, sewage, storm water or winds; or accidentally lost, including material lost at sea in bad weather" (UNEP 2005)

Marine litter consists mainly of plastic items (UNEP 2005). In a 2002 review of data on marine litter composition it was found that 32-92% of marine litter is plastics with an average of ~ 70% (Derraik 2002). This data was mainly based on beach and shoreline field sampling. The same pattern is reflected in the monitoring data of marine litter in the OSPAR region (the North East Atlantic), where 75% of litter on beaches was found to be plastic (OSPAR Commission 2007).

#### 1.1.2 Material definitions

In this report, we use a broad definition of "plastics" as organic, solid materials based on a matrix of synthetic polymers most commonly derived from petrochemicals but they may be partly natural. The scope is similar to the scope of a recent assessment of sources of microplastics in Norway (Sundt *et al.* 2014); the group of materials includes the traditional plastic materials, synthetic textile fibres, synthetic rubbers, as well as cured paints, fillers and similar products based on binders of synthetic polymers.

#### Other polymeric materials which have been considered

Synthetic waxes are generally low molecular weight polymers (400 to ca. 10,000 dalton) with ethene polymers being the largest volume product (Kirk Othmer 2000). Polymer waxes are popular gellants for personal care and cosmetic products and used in many technical products, and are available as powder, flakes or granules as further described in section 3.1.2. According to Leslie (2014), polyethylene waxes are nondegradable, water insoluble, solid materials with melting points well above maximum sea temperatures, and thus fall under the definition of microplastics.

A recent survey of the use of microplastics in Germany (Essel *et al.* 2015) includes waxes in the survey of microplastics, with reference to the review by Leslie (2014). The uses are briefly described in section 3.1.4 (paints) and section 3.1.7 (other applications).

In this review the waxes are not considered microplastics.

**Binders in paint** - Paints are dispersions of sub-micrometre polymer particles. The polymer particles are designated binders. Acrylic polymeric nanoparticles are the basic binder in conventional paints. The particles are non-aggregated and app. 50 nm - 200 nm in size (Tønning and Poulsen 2007). The polymeric binder particles can only be handled in suspension. Upon curing, the particles bind together to form the polymeric network that constitutes a dry paint. It cannot be ruled out that binders can form microplastics particles under certain circumstances, but the binders themselves would not fall within the definitions of microplastics used here.

#### 1.1.3 Definition applied in this report

Based on the above definitions and classifications, we have determined that the definition of microplastics applied in this report is:

#### Definition of microplastics used in this report

Persistent, solid particulates composed of synthetic or semi-synthetic polymers and physical dimensions of 1  $\mu$ m - 5 mm originating from anthropogenic sources.

At the same time, however, we acknowledge (and address where relevant) that:

- 1. This size limits are not rigidly defined or based on scientific evidence but are rather set based on commonly used definitions with the purpose of clearly framing the scope of this report.
- 2. Within the defined size range, larger sized particles may possess properties and behave very differently from smaller sized particles.
- 3. The lower size limit of 1 µm was chosen to purposely exclude plastic debris in the nanometre size range from this study. Particles in the nano-size range are known to interact with their surroundings and behave differently compared to their larger sized counterparts of the same material. Due to their large surface-to-volume ratio, they also cause novel effects in living organisms. Again, this lower size limit is not based on specific scientific data and we will occasionally draw on information from studies on plastic particles below this size limit in this report.
- 4. Mesoplastics and macroplastics may be significant sources of the smaller microplastic particle fractions through weathering (such as abrasion and UV degradation) and disintegration of the polymer structure.
- 5. The materials are not rigidly defined and other polymer types, e.g. biopolymers and waxes, may exert some of the same properties as the microplastics addressed here.

#### 1.1.4 Primary vs. secondary microplastics

We propose to use the following definition in accordance with the definition applied by the international GESAMP Working group on "Sources, fate and effects of micro-plastics in the marine environment – a global assessment" (GESAMP 2012; 2015):

- Primary microplastics: Microplastic particles intentionally produced for direct use
  e.g. in cosmetics and abrasives, or as raw materials for production of larger plastic
  items;
- Secondary microplastics: Microplastic particles originating from the fragmentation of larger plastic items by use, waste management or in the environment.

A recent Norwegian assessment suggests distinguishing instead between **primary sources:** the direct input to the environment of micro-sized plastic particles from human activities, and **secondary sources:** the breakdown and defragmenting of macroplastic litter to microplastics in the ocean (Sundt *et al.* 2014). Compared to the present assessment, the sources covered by the two assessments are the same, but the grouping of the sources is different.

#### 1.2 Methodology applied in this study

#### 1.2.1 Data sources

Data for the review and survey have been collected from the literature and direct enquiries to Danish trade organisations and individual companies.

The literature search has included literature on microplastics and marine litter from following data sources:

- The Danish EPA and the Danish Nature Agency
- International organisations: Nordic Council of Ministers, HELCOM, OSPAR, UNEP, IMO, OECD, and the European Commission
- Environmental authorities in Denmark, Norway, Sweden, Germany, the Netherlands, Belgium, Germany, the UK and Austria
- PubMed and Toxnet databases for identification of relevant scientific literature
- Production and external trade statistics from Eurostat's databases (Prodcom and Comext)
- · Chemical information from the ICIS database
- Proceedings and posters from meetings regarding the subjects.

Direct enquiries were also sent to Danish trade organisations and a few key market actors in Denmark. Data have been collected from member companies by the Association of Danish Cosmetics, Toiletries, Soap and Detergent Industries, the Danish Plastics Federation and the Danish Coatings and Adhesives Association.

Information on ongoing activities have been collected form the project's advisory group and from websites of Danish National authorities, relevant NGOs (environmental, consumer and industry), international organisations, and information obtained from various meetings and seminars.

## 1.2.2 Flow model and data management

For the calculations of releases to the environment, a flow model as illustrated in Figure 1 has been applied. The microplastics may be released to surface water (aquatic environment) by four possible pathways: direct release (e.g. activities on harbours), by urban run-off from areas with separate sewer systems, by sewage by-passing the sewage treatment plants e.g. by heavy rainfall, and by discharges from sewage treatment plants. Emissions are calculated using emission factors for emissions from the initial pathways (e.g. sewage) and distribution factors for urban runoff and sewage reaching the municipal sewerage system.

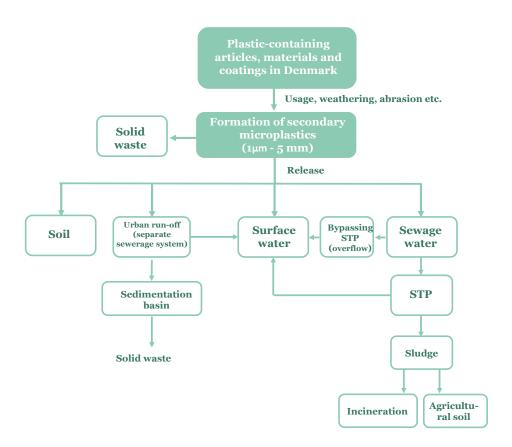


FIGURE 1 CONCEPTIONAL FLOW MODEL DEPICTING THE RELEASE PATHWAYS AND FATE OF SECONDARY MICROPLASTICS IN THE ENVIRONMENT.

Most estimates in the report regarding the expended quantities of primary microplastics and the formation and releases of secondary microplastics are based on incomplete sets of data. For none of the estimates can the uncertainty on the estimate be calculated from basic data using conventional statistical methods. Uncertainties are consequently estimated by the authors based on a limited data set and "expert estimates". The estimates are represented by a 90% "confidence" interval, which represents the range within which the authors estimate that the true value will be with a probability of 90%; i.e. for 10% of the estimates, the "true value" would likely fall outside the indicated range. Arithmetically, the data management of ranges is not completely consistent, but considering the high uncertainties, it has been decided not to apply more consistent and time consuming data management tools such as Monte Carlo modelling. The data presented in ranges are in general rounded to two significant figures of the upper value of the range (e.g. 0.1-2.2).

# 2. Occurrence, fate and effects of microplastics in the environment

## 2.1 Occurrence of microplastics in water and sediments

Microplastics have been demonstrated to occur in the marine environment all over the globe. To estimate the quantities of microplastics and other microlitter in the marine environment is a challenge, not least because the distribution is highly variable on several levels. There are differences between sea areas on a global scale (Cózar *et al.* 2014; Eriksen *et al.* 2014), between hot spots and more remote areas along a coast line (Claessens *et al.* 2011), and between different compartments in the marine environment (surface water, water column, and sediment).

The microplastic distribution is affected by sea currents and meteorological conditions (Collignon *et al.* 2012; Kukulka *et al.* 2012), but also by intrinsic characteristics of the microplastic particles themselves. Factors like polymeric composition, additives, particle morphology, and also the degree of biofouling on particle surfaces may have a great impact on where in the marine environment the microplastics appear (Morét-Ferguson *et al.* 2010). All of these aspects lead to problems in deciding where, when and how to sample in order to obtain data that can be extrapolated to come to more general conclusions. Caution should also be taken when referring to older field data since there seems to have been a shift in the dominating sources to marine microplastics over the past decade. In the 1970s, primary plastics (industrial plastic pellets) made up a more important part of the marine microplastics (Carpenter and Smith 1972) than it does today. The concentration of primary plastics in the water column have decreased over the past decades, and the amount of primary plastics in North Sea fulmars have decreased by 75% since the 1980s (van Franeker and Law 2015) (Morét-Ferguson *et al.* 2010).

The occurrence of microplastic particles in the sea is a relatively new area of concern and there is still no consensus on what techniques should be applied for sampling and analysis. A variety of different strategies have been used, which make it complicated to estimate field concentrations and to compare microplastic abundances and composition between areas and over time. Hence, there is a definite need to develop standardized methods for monitoring of microplastics and also to formulate indicators for defining the environmental effects they have. Initiatives to start this work have been taken at various national and international levels within Europe (see section 6).

Most field studies of marine microplastics have focused on sampling of surface water. Sampling has often been carried out with manta trawls which are designed to collect particles ≥333 µm in the top ten centimetres of the water column. Since the density determines the particle distribution in the water column, and between water column and sediment, there is a risk that if all sampling is done by manta trawl, only particles made of polymers with a density lower than water will be collected, e.g. polypropylene (PP), low-density polyethylene (LDPE) and high-density (HDPE). Particles with a density greater than seawater, e.g. polyvinyl chloride (PVC) and nylon would then be excluded from the results (Table 1). Particles smaller than the mesh size of the manta trawl would also be left

out, which is a serious limitation, since it has been shown that harmful effects on marine biota may also be triggered by particles considerably smaller than  $333 \mu m$ .

TABLE 1
THE DENSITY OF SEA WATER AROUND THE DANISH COAST AND OF PLASTIC POLYMERS COMMONLY FOUND IN THE MARINE ENVIRONMENT

	Density of seawater and plastic polymers g/cm ***
West coast of Denmark sea water	1.03
Kattegat sea water	1.01-1.03
The Belt Sea water	1.01 (Feistel <i>et al.</i> 2010)
Polypropylene	0.90-0.91*
Low density polyethylene (LDPE)	0.91-0.94*
High density polyethylene (HDPE)	0.92-0.99*
Polystyrene (PS) **	1.04-1.13*
Acrylic fibres (most used fibres in textiles)	1.16 ****
Polymethyl methacrylate (PMMA, Plexiglas)	1.17-1-20*
Polycarbonate	1.20*
Polyurethane	1.30*
Polyamide /Nylon	1.06-1.39*
Polyethylene terephthalate (PET, Dacron)	1.41*
Polyvinyl chloride (PVC)	1.39-1.43*
Epoxy resin	0.96-2.10*
Polytetrafluorethylene (PTFE, Teflon)	2.28-2.29*

<sup>\*</sup> Source: Polymer Data Handbook 1999

## 2.1.1 Microplastics in the water column

Sampling of microplastics in the water column is generally done by collecting suspended particles on some sort of filter material. This could be done in several ways, e.g. by trawling, by pumping water through a filter or by collecting water in a container and pouring it over a filter. Independently of what method is being used the mesh size of the filter or the trawl will determine the lower cut off size for particles being collected. As can be seen in Table 2, which summarizes data on microplastic abundances in the water column from the North Sea, Baltic Sea, and other sea areas around the world, a variety of cut off sizes have been applied in different studies. This makes it complicated to compare the results and highlights the importance of developing standardized sampling methods.

As shown in Table 2, microplastic particles larger than ~300 µm generally occur in concentrations of less than one to a couple of particles per m³. This is true for European sea areas (Magnusson and Norén 2011; Cole *et al.* 2014; Mintenig 2014) and elsewhere (Lattin *et al.* 2004; Desforges *et al.* 2014; Song *et al.* 2014).

<sup>\*\*</sup> Expanded polystyrene (EPS), a material frequently found in the sea, is filled with air bubbles and hence has a density much lower than solid PS.

<sup>\*\*\*</sup> Indicates the density off the basic polymer; the density of the final plastics also depends on the additive load and plastics with a high load of pigments and fillers may have significantly higher densities than the basic polymer.

<sup>\*\*\*\*</sup> Textile Learner RF2015.

Microplastics <300  $\mu$ m are much more abundant than those >300  $\mu$ m. In a study where parallel samplings of surface water were carried out with a 300  $\mu$ m filter and a 10  $\mu$ m filter, the microplastic concentration was found to be 0-1.5 m³ for particles >300  $\mu$ m and 4,400 – 94,000 per m³ for particles >10  $\mu$ m to (Norén *et al.* 2014). Dubaish and Liebezeit (2013) analysed plastic particles >1.2  $\mu$ m in surface water from the Jade System in the southern North Sea, and found that concentrations of granular plastic particles amounted to an average of 64,000 per m³ and plastic fibres to an average of 88,000 per m³.

Close to point sources, the concentrations of microplastics may be particularly high. In the harbour of a Swedish production plant for polyethylene (PE) pellets, the concentration of plastic particles  $>80 \mu m$  was found to be  $102,550 \text{ per m}^3$  of sea water (Norén 2007).

Data from the Arctic waters are very limited, but there is one study reporting on fairly high concentrations of microplastics trapped in the Arctic Sea ice (Obbard  $et\ al.\ 2014$ ). As the ice is formed it concentrates particles from the water, and if microplastics are present they will also be trapped in the ice. Between 38-234 particles <2 mm were found per m³ of ice, and as it melts, microplastics are released into the water and may be taken up by species in the Arctic food web.

TABLE 2 A SELECTION OF REPORTED VALUES OF MICROPLASTIC CONCENTRATIONS IN SEA WATER IN DIFFERENT PARTS OF THE WORLD. THE APPLIED CUT OFF SIZE IS NOTED, AND UPPER SIZE LIMIT IS  $_5$  MM UNLESS SOMETHING ELSE IS STATED. ABUNDANCES ARE EXPRESSED AS NUMBER OF MICROPLASTIC PARTICLES PER  $_{\rm m}$ 3 OF SEAWATER.

Geographical area	Cut off size/ range of size	Abundance particles/m³	Reference
Danish coastal waters  North Sea  Kattegat  The Belt Sea  Plastic particles but not plastic fibres	>100 μm	0.39±0.19 3.54 1.44	Mintenig 2014 See Figure X
Swedish west coast close to PE production plant	>80 µm	~102,550	Norén 2007
Swedish coast, close to the shore  • Kattegat  • The Sound  • The Baltic	>300 µm	1.08±0.22 4.0 0.56±0.40	Magnusson and Norén 2011 See Figure 2
Swedish west coast Plastic particles but not fibres	≥10 µm ≥300 µm	4,400-94,000 0-1.5	Norén <i>et al</i> . 2014
The Gulf of Finland  Turku harbour  Archipelago  Off shore	>333 µm	0.73 0.25±0.07 0.48	Magnusson 2014a
North Sea coast of Germany	>1.2 µm Granular particles Plastic fibres	64,000±194,000 88,000±82,000	Dubaish and Liebezeit 2013
Western English Channel	>500 µm	0.27	Cole <i>et al.</i> 2014
NE Atlantic (not the North Sea)	250 - 1,000 μm	2.46	Lusher et al. 2014
NE Pacific Ocean	>62 µm	279 ± 178	Desforges et al. 2014
NE Pacific Ocean	>500 µm	0.004-0.19	Doyle et al. 2011
Korea			Song et al. 2014

Geographical area	Cut off size/ range of size	Abundance particles/m³	Reference
Plastic particles, not paint fragments	>50 μm >333 μm	1,143±3,353 47±192	
Korea Paint fragments (alkyds and polyacrylat/polystyrene)	>50 μm >333 μm	196±121 0.88±0.81	Song <i>et al.</i> 2014
California, USA	>333 µm	~3.9	Lattin et al. 2004
Arctic Sea ice	<2 mm	38-234 parti- cles/m³ ice	Obbard <i>et al</i> . 2014

There are a few available reports on microplastic particles in or close to Danish marine waters. Water samples from low to moderately urbanized areas along the Swedish south west and south coasts, taken very close to the shore, contained 0-4.0 microplastic particles >300  $\mu$ m per m³ (Figure 2, Table 2) (Magnusson and Norén 2011). The concentration was highest at the sample station in the Sound, 4.0 microplastics per m³ (station E in Figure 2). This station was also located in the most populated area among the sample stations. The Kattegat stations (stations A-C) had 1.08±0.22 microplastics per m³, and the lowest concentrations were found at the Baltic sampling stations F-I, with 0.56±0.40 particles per m³. Plastic fibres made up >80% of the microplastic particles at the sampling stations in the Kattegat and the Sound, and 36% at the Baltic stations.

In a study by Mintenig (2014), sampling of microplastics (not including plastic fibres) in surface water was carried out in the North Sea and the Baltic Sea, and a cut off size of 100  $\mu$ m was used (Figure 3). The highest concentration by far, 3.54 microplastics per m³, was found at a location north west of Skagen (Fig. 3 station 15, Table 2). The concentrations along the German and Danish North Sea coast were 0.39±0.19 microplastics >100  $\mu$ m per m³, with the highest concentration, 1.3 particles per m³, at station 12. The only analysed sample from the Baltic Sea was taken in the south west part and had a concentration of 1.44 microplastics >100  $\mu$ m not including plastic fibres. Due to problems with identification, plastic fibres were not included in this study, so it is likely that the total concentrations of microplastics (based on numbers) actually were considerably higher. In several studies plastic fibres have been found to make up 80-95% of the total number of microplastics (Magnusson and Norén 2011; Lusher *et al.* 2014).

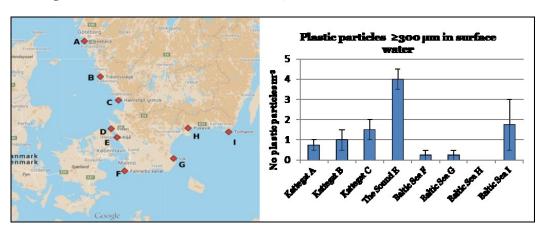


FIGURE 2 ABUNDANCE OF MICROPLASTIC PARTICLES ≥300  $\mu$ M IN SURFACE WATER. SAMPLES WERE TAKEN CLOSE TO THE SHORE ALONG THE SWEDISH WEST AND SOUTH COAST (MEAN VALUES ±SE, N=2). CONCENTRATIONS ARE EXPRESSED AS NUMBER OF PARTICLES/m³. THE LOCATIONS OF THE SAMPLING STATIONS ARE SHOWN IN THE MAP TO THE LEFT (DATA FROM MAGNUSSON AND NORÉN 2011). AT STATION D, PARTICLES ≥300  $\mu$ m WERE NOT SAMPLED.

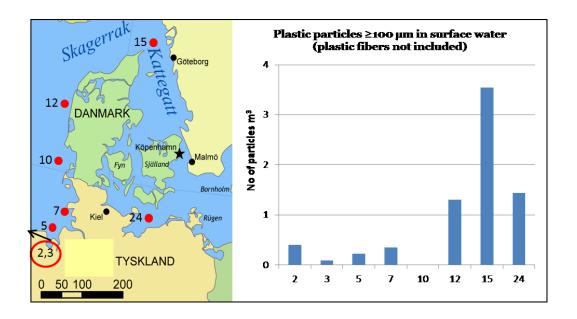


FIGURE 3 SURFACE WATER ABUNDANCE OF MICROPLASTIC PARTICLES>100  $\mu\text{M}$ . PLASTIC FIBRES WERE NOT INCLUDED. SAMPLES WERE TAKEN FROM THE GERMAN BIGHT IN THE NORTH SEA TO THE MECKLENBURG BIGHT IN THE BALTIC SEA. THE RED MARKS IN THE MAP ARE THE APPROXIMATE POSITIONS OF THE SAMPLING STATIONS. STATIONS 2 AND 3 WERE LOCATED OUTSIDE THE FRISIAN ISLANDS (NOT SHOWN IN THE MAP). DATA FROM MINTENIG (2014).

A survey of microscopic litter  $\geq$ 10  $\mu$ m in surface water was conducted north of Jutland along a transect between Flødevigen in Norway and Hirtshals in Denmark (Figure 3) (Norén and Naustvoll 2010). Sampling was done at eight stations, and the mean concentration was found to be 1,700 particles per m³. The highest concentration, 6,000 particles per m³, was found at the station closest to Denmark (marked 52 Nm in Figure 4). The salinity at this station indicated that the sampled surface water was of North Sea origin. Blue and red particles were the most abundant of the sampled microplastic particles. The same kind of blue and red particles, in the size range of 10-100  $\mu$ m, have also been found to be abundant in Swedish coastal waters and are suspected to derive from antifouling paint from ships (Norén *et al.* 2014). In the study by Lusher *et al.* (2014) in the Northeast Atlantic blue particles were as well more common than particles of other colours (37.7%).

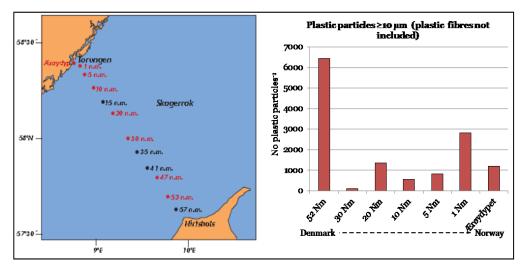


FIGURE 4 ABUNDANCE OF MICROPLASTICS  $\geq$ 10  $\mu m$  IN SURFACE WATER ALONG A TRANSECT BETWEEN NORWAY AND DENMARK. THE NAMES OF THE STATIONS CORRESPOND TO THE DISTANCE IN NAUTIC MILES (NM) FROM AEREDYPET, THE SAMPLING STATION CLOSEST TO THE NORWEGIAN COAST. ABUNDANCES EXPRESSED AS NUMBER OF PARTICLES/m³. DATA FROM NORÉN & NAUSTVOLL (2010)

The plastic composition of pelagic microparticles in North East Atlantic/Baltic Sea waters is at present best investigated in the study by Mintenig 2014 (Figure 5). The study included, in all, 28 stations. At all of them, all particles defined as *visible particles* (>500  $\mu$ m) were analysed for plastic composition, whereas particles <500  $\mu$ m were only analysed at the eight stations marked in Figure 5. The analyses were done with Fourier transform infrared spectroscopy (FTIR). For a more detailed description, see Mintenig 2014.

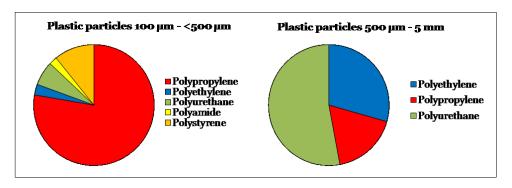


FIGURE 5
THE PLASTIC COMPOSITION OF PELAGIC MICROPARTICLES IN NORTH EAST ATLANTIC/BALTIC SEA WATER. DATA FROM MINTENIG (2014). THE CHART TO THE LEFT INCLUDES RESULTS FROM 28 SAMPLING STATIONS WHEREAS THE CHART TO THE RIGHT INCLUDES THE STATIONS SHOWN IN FIGURE 3 WITH THE EXCEPTION OF STATION 12. UNLIKE THE SITUATION AT THE OTHER SAMPLING STATIONS, PARTICLES >500 µM AT STATION 12 WERE COMPLETELY DOMINATED BY POLYETHYLENE PARTICLES. DATA FROM MINTENIG (2014).

Grouping together all particles >500  $\mu$  from 27 of the 28 stations (not station 12) it was found that 51.4% of the collected microplastic particles >500  $\mu$ m consisted of polyurethane (PUR), 28.6% of PE, and 17.1% of PP. Particles <500  $\mu$ m were more difficult to analyse, but of those which were successfully determined, 82.0% consisted of PP, 9.9% of polystyrene (PS), 6.3% of PUR, 0.9% of PE and 0.9% of polyamide (PA). The particle composition at station 12 differed from the others and data from here was therefore presented separately. Station 12 was situated in the North Sea off the Danish west coast and the microplastic fraction>500  $\mu$ m consisted of 97.4% PE particles and 2.6% PUR particles. The analysed particles <500  $\mu$ m from station 12 were made up of PP (60.7%), PS (14.3%), PE (10.7%), PA (7.1%) and PUR (7.1%). The data are from a report and not a peer-reviewed scientific publication and should likely be interpreted with caution.

The particles detected in the Arctic Sea ice were found to be made up of polyester, polyamide, polypropylene, polystyrene, acrylic and polyethylene (Obbard *et al.* 2014). The most prevalent synthetic particles were rayon, which is a cellulose-derived polymer.

## 2.1.2 Microplastics in sediments

Microplastic particles that are denser than seawater, either because they are made of denser plastics or because biofouling has made them heavier, are likely to be found in the sediments. It is also possible that microplastics in the photic zone become entangled in the marine snow and sediment to the bottom in association with these aggregates. The importance of this process has, however, not yet been investigated.

Data on microplastics in sediments in and close to Danish coastal waters, as well as data from other sea areas, are presented in Table 3. There is at present no available data on microplastics in Arctic sediments.

The amount of microplastics discovered in Danish coastal sediments is fairly high compared to what has been detected in sediments from the North Sea coast of Germany and Belgium (Claessens *et al.* 2011; Liebezeit and Dubaish 2012; Strand *et al.* 2013). Sediments from the Dutch coast had on average higher concentrations, with an extreme value, 3,305 particles/kg dry weight, in sediments

from the Rhine estuary (Leslie *et al.* 2013). The highest concentrations in Danish waters were found in the Belt Sea at 1,100 microplastic particles/kg dry sediment. In Skagerrak, Kattegat and the Baltic Sea areas average concentrations were between 240 and 350 microplastics per kg dry sediment. However, the variation within these sea areas was very large, and the differences in average values were not statistically significant (Table 3).

TABLE 3 A SELECTION OF DATA ON MICROPLASTIC CONCENTRATIONS IN SEDIMENTS. THE APPLIED CUT OFF SIZE IS REPORTED AND THE UPPER SIZE LIMIT IS  $_5$  mm UNLESS SOMETHING ELSE IS STATED. THE ABUNDANCES ARE EXPRESSED AS THE MEDIAN NUMBER OF PARTICLES PER kg DRY SEDIMENT ( $_{75}\%$  CENTRAL RANGE) OR ±SD.

Geographical area	Cut off size/range of particle size	Abundance  No. plastic particles/kg dry sediment	Reference
<ul> <li>Danish coastal waters</li> <li>North Sea &amp; Skagerrak</li> <li>Kattegat</li> <li>Belt Sea</li> <li>Baltic Sea</li> </ul>	>38 µm	100 (75 – 268) 120 (60 – 195) 380 (280 – 1,090) 335 (145 – 543)	Strand et al. 2013
Germany East Frisian Islands	>1.2 μm	210 plastic granules 461 plastic fibres	Liebezeit and Du- baish 2012
Dutch coast	>1 μm	3,305±295 455 (390 – 520) 770	Leslie et al. 2013
Belgian coast      Harbours     Continental shelf     Beaches	>38 µm	167±92 97±19 93±37	Claessens et al. 2011
Belgian beaches  High-water mark  Low-water mark	5 – 1,000 μm	17.6±9.4 9.2±5.0	Van Cauwenberghe <i>et al.</i> 2013a
Lagoon of Venice	32 – 1,000 μm	1,445±458	Vianello <i>et al.</i> 2013

## 2.2 Fate of microplastics in the aquatic environment

## 2.2.1 Degradation mechanisms and rates

Microplastics particles in the ocean are to some extent formed from macroplastics and further degraded into plastics in the nano range and mineralised by some of the same mechanisms. The overall information on the mechanisms of degradation of plastics and the formation of microplastics from macroplastics is described in section 2.7.4 whereas this section focuses on the fate of microplastics.

In the oceans, the formation and degradation of microplastics is known to be influenced by a combination of environmental factors and the properties of the polymer, but there is a general lack of research available on weathering and fragmentation of microplastics in the marine environment and how the combined effects of photo-oxidation, fragmentation, mechanical abrasion and additive chemicals affect the formation of microplastics (GESAMP 2015). When it comes to degradation mechanisms and degradation rates of microplastics, it is important to clarify that degradation in this context is synonymous with destruction of the polymer chain and a reduction of the molecular weight of the plastic material, and thus implies some degree of mineralization (GESAMP 2015).

Microplastics may undergo degradation, generally via biodegradation, where microbial colonies utilize the carbon in the polymer, which is converted into  $\mathrm{CO}_2$  and incorporated into the marine biomass (Andrady 2011). According to Andrady (2011), there are basically no data on mineralisation of microplastics in the environment and the existence of nano-scale plastic in the ocean has yet to be reported (GESAMP 2015). The weathering and fragmentation rates of plastics vary depending on where they are located in the environment. When plastic is present in aquatic environments, the rate of degradation is retarded, as a result of lower oxygen concentrations and lower temperatures than compared to when present in air or on land (e.g. beaches) (Andrady 2011). For plastics on beaches, the fragmentation rates are relatively rapid, whereas they are several orders of magnitude slower for plastics floating in water, in the mid-water column or in the marine sediments. The degradation on beaches depends on factors such as UV radiation, sample temperatures and mechanical abrasion attained by the beach litter, whereas the degradation of plastic in different compartments of the marine environments depends on the plastic and the water temperatures. Additives could also prolong the degradation time of plastics and fouling of the plastic could result in an even slower degradation rate, as biofilm can form a protective layer against UV radiation.

In general, no degradation is expected in aphotic (dark) and cold sediment environments, but virtually no information is available on the fate of plastics in aphotic marine sediments, according to GESAMP (2015).

More knowledge on the long-term fate of microplastics, and data addressing the extent to which the fragmentation of the microplastics under environmental conditions results in billions and billions of sub-micrometre particles, are considered crucial for the assessment of the potential long-term effects of the pollution of the environment with both macro and microplastics.

## 2.2.2 Transport of microplastics in the aquatic environment

Due to their buoyancy and durability, plastics tend to accumulate on coastlines, in oceanic gyres and in enclosed seas (e.g. Mediterranean) where surface water is retained for long periods of time (Ryan *et al.* 2009; Zarfl and Matthies 2010; Goldstein *et al.* 2012; Eriksen *et al.* 2013; Faure *et al.* 2012, GESAMP 2015; Eriksen *et al.* 2014).

There is a significant small-scale variability within these regions when it comes to spatial distribution of microplastics with variations of orders of magnitude in concentration within tens of kilometres. This phenomenon is due to factors such as wind-driven currents and centimetre-scale turbulent motion. One aspect that current attempts at modelling the fate of microplastics have not taken into consideration is the fact that microplastics can be taken up and retained by marine organisms for varying periods of time, which can potentially transport microplastics across significant distances (GESAMP 2015).

When it comes to the horizontal distribution of microplastics in the water column, Woodall *et al.* (2014) have found that the amounts of microplastics observed on the deep sea floor is about four times the amount observed at the surface. It is well known that the fate and behaviour of microplastics depends on 1) the size distribution, shape and the specific type of polymer, 2) whether it is a light- or high density type (Lusher *et al.* 2014), and 3) the oceanic currents that are known to vary with depth. Sinking rates of marine particles have been reported to vary between 10 to 150 metres per day (GESAMP 2015). Light-density plastics have positive buoyancy, and therefore can be found in the surface layer. Buoyancy can vary over time and several studies have furthermore suggested that fouling on plastics can cause negative buoyancy (Moore *et al.* 2001; Derraik 2002; Thompson *et al.* 2004; Barnes *et al.* 2009; Browne *et al.* 2007), while light-density plastics may sink anyway. Upon sinking, the particles encounter lower temperatures and limited light, which provide poor environments for microorganisms i.e. conditions that may negate the fouling and cause the plastics to re-emerge up into the surface layers. A study by Morét-Ferguson *et al.* (2010) confirms fouling occurrence on microplastics, but microplastics tends to be present near the surface (Cole *et al.* 2011), which contradicts heavy fouling. High-density plastics are most likely found near or in the

benthos (Cole *et al.* 2011) as shown by deep sea sediment sampling (Van Cauwenberghe *et al.* 2013b) or near the point of entrance to the environment e.g. estuarine waters.

In addition to buoyance, wind-driven mixing is an important factor for the fate of plastic particles. The turbulence created by wind-driven mixing of the surface layer causes the plastics to migrate downward, which is why surface sampling can underestimate the amounts of microplastics (Eriksen *et al.* 2014). Lattin *et al.* (2004) suggests that only a small amount of turbulence is needed in order for the plastics to re-suspend into mid-water, as observations after a storm showed an increase of density in the mid-water and a decrease close to the bottom. Even high-density plastics have been documented to remain suspended after entering oceans as a result of turbulence. Additionally, large oceanic currents play a major role in the plastics' environmental fate. Accumulation zones coincide with the great currents (Eriksen *et al.* 2013) such as the Kuroshio current flows, found to be important in regard to the transport and distribution of plastic debris across the North Pacific Ocean (Yamashita and Tanimura 2007).

An investigation of spatial distribution and occurrence of microplastics in sediments from Norderney found no correlation between the amount of microplastics comprised of larger plastic debris, and further found that the microplastics were distributed rather homogenously (Dekiff *et al.* 2014). This finding suggests a deposition of marine microplastics on the beach sediment. Lusher *et al.* (2014) found that plastic particles are widespread throughout the subsurface layers of the Northeast Atlantic Ocean. Concentrations in sediments were found to be higher in sediments in the Lagoon of Venice, Italy, in the Mediterranean Sea (Vianello *et al.* 2013) compared to sediments along the Belgian coast (Claessens *et al.* 2011), which corresponds well with the fact that the Mediterranean Sea is an enclosed sea and therefore would be expected to have higher concentrations. This conclusion has also been confirmed by the modelled distribution by Eriksen *et al.* (2014) that showed very high concentrations of microplastics in the Mediterranean Sea.

Numerous attempts have been made to model the plastics' fate and behaviour. Eriksen *et al.* (2014) modelled the global distribution of plastic debris and found that there are great losses of microplastics, suggesting that large proportions of microplastics are stranding on seashores, degraded or sinking to the sea floor (see further description in section 2.7.4). The model included a vertical correction to account for wind-driven turbulence and other hydrodynamic processes such as downwelling of microplastic particles at convergence zones. If the model predictions are reliable, it appears that sediment sampling and mid-water sampling are very important, as surface water sampling alone would underestimate the occurrence of buoyant microplastics in particular (GESAMP 2015).

## **2.3** Exposure of biota to microplastics General introduction to exposure

The extent of adverse impacts of microplastics on biota depends on the levels of exposure as well as the adverse effects that microplastics could potentially cause biota, if exposure takes place. Plastics in the micro and nano range can be taken up in aquatic organisms by oral ingestion or through the gills (Watts *et al.* 2014). For example, indiscriminate feeders, such as filter-feeding zooplankton and other planktonic organisms, can take up plastic particles as they are mistaken for prey (Cole *et al.* 2011). Additionally, plastic particles could adsorb onto biological surfaces. This adsorption could cause effects on the organisms, for example, by affecting algal photosynthesis as it has been reported for plastics in the nano range by Bhattacharya *et al.* (2010). At the same time, it can lead to a transfer of plastic particles through the food chain, for example, if these algae are ingested by zooplankton. According to Cole *et al.* (2011), there is an increasing number of studies that report microplastic ingestion throughout the food chain; microplastics have been found inside the digestive tract of more than 100 different species (Secretariat of the Convention on Biological Diversity and the Scientific and Technical Advisory Panel—GEF 2012). As pointed out by Wright *et al.* (2013a), the availability of different microplastics in the water column will also depend on the plastic particle

density. Low density plastics will likely be more available to organisms inhabiting the upper water column whereas denser particles will be more available to the organisms in the lower parts of the water column or sediment. Other factors include particle size, colour (prey item resemblance) and abundance (Wright *et al.* 2013a). Aside from the concentration and the density of the microplastics to which biota are exposed, the routes of exposure (e.g. ingestion, egestion and translocation) are also determining factors when it comes to the overall extent of adverse impact.

A wealth of evidence of microplastics in wild organisms exists from all over the world as reviewed by Gesamp (2015). Examples of microplastics in wild organisms in the North Sea and Great Belt are shown in Table 4. It is clear from Table 4 that microplastics have been observed in the stomach, gut and tissue of a wide range of different species that live in the North Sea and Great Belt in varying frequency, concentrations and size ranges.

## 2.3.1 Exposure via ingestion

It is well known that, due to their size, microplastics are available for ingestion by a wide range of animals, and ingestion is well documented for marine vertebrate and invertebrate species (Ivar do Sul and Costa 2014). It should be noted that regurgitation (expulsion of material from the pharynx or esophagus) of latex beads has been observed in an estuarine copepod (*Eurytemora affinis*) (Powell 1990), suggesting that some biota is capable of rejecting non-nourishing elements. Moreover, some few specimens of holoplankton (organisms that are planktonic for their entire life cycle) have shown no evidence of ingestion, namely chaetognaths (*Parasagitta sp.*) and siphonophorae (*Cnidaria*) (Cole *et al.* 2013).

Nevertheless, the size of the microplastic particles makes them fall within the same size range of some marine organisms' food, e.g. plankton, and as some of these low trophic marine organisms exert limited selectivity between particles in water and sediment, many of the organisms end up eating anything of appropriate size (Moore 2008).

Ingestion of microplastics by different Baltic Sea zooplankton taxa (mysid shrimps, copepods, cladocerans, rotifers, polychaete larvae and ciliates) was investigated by Setälä  $\it et~al.$  (2014). It was found that 10  $\mu m$  fluorescent polystyrene microspheres were ingested by all taxa studied but with large variations between taxa. Furthermore, the study investigated the potential for microplastic transfer in the food chain and observed a particle transfer to macrozooplankton that were fed with mesozooplankton that had ingested the microplastic particles. The polystyrene microspheres were observed in the intestine of the macrozooplankton (mysid shrimp) after 3 hours' feeding on the macrozooplankton (copepods/Marenzelleria spp. Larvae) (Setälä  $\it et~al.$  2014).

Graham and Thompson (2009) found that the sea cucumbers tested selectively ingested the microand mesoplastics over the sand grains and the other particles that were present in the sediment. In
gyres, the concentrations of plastics are substantial compared to the concentration of plankton, a
likely explanation as to why the contact and thus the probability of plankton ingesting the plastics
are greater in such locations (Moore 2008). Higher trophic planktivores on the other hand could
passively ingest microplastics under normal feeding behaviour or simply by mistaking microplastics
for food (Wright *et al.* 2013b). Fossi *et al.* (2012) concluded that the large filter feeding Mediterranean fin whale *Balaenoptera physalus*, capable of engulfing approximately 70,000 L of water at a
time, had ingested microplastics, both indirectly and directly, from water and plankton. The uptake
by different species depends on the shape, size and density of the microplastic, as this determines
the position in the water column and thus the potential availability (Browne *et al.* 2007).

TABLE 4 EXAMPLES OF MICROPLASTICS IN AQUATIC ORGANISMS IN THE NORTH SEA AND THE GREAT BELT (AS REVIEWED BY GESAMP 2015)

Species	Organism	Area	Individuals with microplastics	Average number of particles per individual	Mean size of particles	Organ	Reference
Harbour seal	Phoca vitulina	Netherlands	11% of 107 stom- achs sampled; 1% of 100 intestines sampled; 0% for scats of 125 scats sampled	n.r.	It is mentioned that the smallest particles are 0.12- 0.3 mm but the focus is on plastics in general	Stomach, intestines, scats	Rebolledo <i>et al</i> . 2013
Herring, Whit- ing	Clupea harengus, Merlangius mer- langus,	Great Belt, Denmark	≈ 30% (≈ 100 individuals sam- pled)	n.r.	0.5 - 4 mm	Stomach	Sørensen et al. 2014
Herring	Clupea harengus	North Sea	8/566	n.r.	n.r.	n.r.	Foekema et al. 2013
Cod	Gadus morhua	North Sea	10/80	n.r.	n.r.	n.r.	Foekema et al. 2013
Whiting	Merlangius mer- langus,	North Sea	6/105	n.r.	n.r.	n.r.	Foekema <i>et al</i> . 2013
Haddock	Melanogrammus Aeglefinus	North Sea	6/97	n.r.	n.r.	n.r.	Foekema <i>et al</i> . 2013
Horse mackerel	Trachurus trachu- rus	North Sea	1/100	n.r.	n.r.	n.r.	Foekema <i>et al</i> . 2013
Grey gurnard	Eutrigla gurnar- dus,	North Sea	1/171	n.r.	n.r.	n.r.	Foekema <i>et al</i> . 2013
Atlantic mackerel	Scomber scombrus	North Sea	1/84	n.r.	n.r.	n.r.	Foekema et al. 2013
Northern fulmar	Fulmarus glacialis	North Sea	95%	0.1 g of plastic exceeded in 58% of individuals and 60% of Dutch birds	All plastic particles >1 mm are included – no focus on microplastics	Stomach	van Franeker et al. 2011
Blue shark	Prionace glauca	North Sea, Channel, Irish Sea, Spitzbergen	4.7%	n.r.	n.r.	Gut	Pinnegar 2014

Species	Organism	Area	Individuals with microplas- tics	Average number of particles per individual	Mean size of particles	Organ	Reference
Cod	Gadus morhua	North Sea, Channel, Irish Sea, Spitzbergen	29%	n.r.	n.r.	Gut	Pinnegar 2014
Grey Gunard	Eutrigla gurnardus	North Sea, Channel, Irish Sea, Spitzbergen	14&	n.r.	n.r.	Gut	Pinnegar 2014
Lesser spotted dogfish	Scyliorhinus canicula	North Sea, Channel, Irish Sea, Spitzbergen	4.8%	n.r.	n.r.	Gut	Pinnegar 2014
Saithe	Pollachius virens	North Sea, Channel, Irish Sea, Spitzbergen	24%	n.r.	n.r.	Gut	Pinnegar 2014
Whiting	Merlangius mer- langus	North Sea, Channel, Irish Sea, Spitzbergen	24%	n.r.	n.r.	Gut	Pinnegar 2014
Blue mussel	Mytilus edulis	Belgium	n.r.	n.r.	200 – 1,500 μm	Tissue	De Witte <i>et al</i> . 2014
Mediterranean mussel	Mytilus gallopro- vincialis	Belgium	n.r.	n.r.	200 – 1,500 μm	Tissue	De Witte <i>et al.</i> 2014
Blue mussel	Mytilus edulis	Germany	n.r.	Average of 0.36 ± 0.07 particles/g	n.r.	Tissue	Van Cauwenberghe & Janssen 2014
Herring	Clupea harengus	Sejerø bay, Denmark	27%	n.r.	1-4 mm	Intestines	Enders et al. (2015)
Whiting	Merlangius mer- langus	Sejerø bay, Denmark	31%	n.r.	1-4 mm	Intestines	Enders <i>et al.</i> (2015)

n.r.: Not reported

Low-density plastic will most likely float and thus be available for filter feeders or planktivores. High-density plastics tend to sink, as mentioned above, and therefore tend to accumulate in sediments where they are available for ingestion by deposit- and detritus-feeding organisms et al. 2013a). In laboratory tests, several marine species have shown ingestion of microplastics: detritivores (amphipods), filter feeders (barnacles), and deposit feeders (lugworms) (mean size 230 µm) (Thompson et al. 2004). More recently, the blue mussel (Mytilus edulis), as well as oysters, were found to have ingested and accumulated nanopolystyrene beads (Wegner et al. 2012). Two effects were observed in the case of nanopolystyrene (30 nm) uptake by mussels (Wegner et al. 2012). First, the production of pseudofaeces (false faeces) was triggered, meaning that the mussel recognizes that the nanopolystyrene is not its natural prey. Secondly, the filtering activity of the mussel was reduced, which may cause the mussel to starve to death. Uptake of microplastics was also investigated in the Shore Crab Carcinus maenas both through aqueous exposure and dietary exposure (pre-exposed common mussel Mytilus edulis). Fluorescently labelled polystyrene microspheres were used as model particles allowing for imaging by Coherent Raman Scattering Microscopy. Results showed retention of microplastics in the foregut after dietary exposure and on the external surface of gills after aqueous exposure (Watts et al. 2014).

## 2.3.2 Egestion or translocation

After ingestion, the microplastic particles can either be egested or translocated within the animal (Browne et~al.~2008; von Moos et~al.~2012). Egestion of microplastics could prompt an avoidance of any potential detrimental effects accompanied by ingestion of nano- and microplastics, depending on how fast it is egested (Wright et~al.~2013a). However, translocation into the tissue of the aquatic biota increases the risk of transfer of nano- and microplastics and any adhered chemicals or imbedded additives in the food web. Experiments confirming egestion of microplastics have been conducted for a wide range of marine biota. Microplastics have been found in faeces from lugworm (Arenicola~marina) (Thompson et~al.~2004). Graham and Thompson (2009) found and enumerated the egested plastic of four different kinds of sea cucumbers (Echinodermata) in order to establish ingestion (Browne et~al.~2008). On the other hand, in a study with mussels (Mytilus~edulis) investigated for ingestion, translocation, and accumulation of microplastics (3.0 or 9.6  $\mu$ m), initial results indicated translocation from the gut into the circulatory system of the mussel. The translocation seemed to have happened within 3 days, and the microplastics persisted in the circulatory system for more than 48 days (Browne et~al.~2008), a timeframe that allows for trophic transfer.

Translocation from the gut cavity to the hemolymph was shown to take place in mussels within three days (Browne, Galloway, and Thompson, unpublished data cited Browne *et al.* 2007). Cole *et al.* (2013) showed that 13 out of 15 tested zooplankton types with varying degrees of indiscrimination ingested microplastics; thereafter, most of them egested it within a couples of hours. The microplastics accumulated on the external parts of the zooplankton, and for the copepod (*Centropages typicus*) a reduced rate of algal ingestion was shown. Therefore, the egestion or/and translocation of nano- and microplastics seem to be dependent on species, but further research is needed.

## 2.3.3 Transfer of microplastics in the food web

A laboratory study (Farrell and Nelson 2013) investigating the trophic transfer between mussels ( $Mytilus\ edulis$ ) that had been exposed to fluorescent nano-sized polystyrene (0.5  $\mu$ m) after which they were fed to crabs ( $Carcinus\ maenas$ ) showed translocation to haemolymph and to the tissues of the crab. The plastics in the nano range were also found inside the stomach, ovary, hepatopancreas, as well as the gills of the crab; however, the amount decreased during the experimental period. The study indicates translocation of plastics in the nano range within the predator animal, and is therefore important in regard to transfer up through the food web. More recently, Setälä  $et\ al.$  (2014) exposed different species of zooplankton (mysid shrimps, cladocerans, copepods, rotifers, and ciliatesand polychaete larvae) to 10  $\mu$ m fluorescent microplastics. All tested zooplankton ingested the plastic particles, and further experiments with mysid shrimps (Neomysis) and copepods ( $Eurytemora\ affinis$ ) showed egestion 12 hours after ingestion. The study is the first to demonstrate

microparticle transfer from one trophic level (meso zooplankton) to a higher trophic level (macrozooplankton). Moreover, the mysid shrimps (macro zooplankton) were exposed both directly and indirectly, implying several possible routes for transfer in the pelagic food web. Another important indication from this study is the fact that the two applied species, mysids and polychaete larvae, both partially live in the pelagial and in benthic realm, respectively, as mysids are nektobenthic animals, and polychaete larvae, when becoming adults, settle to the bottom and live in the sediments. Thus, the two species have the potential to transfer plastic particles from one food web to another. Setälä *et al.* (2014) further conclude that microplastics in high concentrations have the potential to transfer into marine food webs. Trophic transfer has been indicated for both nano- and microplastics; however, the literature is still scarce.

## 2.4 Observed biological effects on biota in the aquatic environment

Observed biological effects of microplastics can be divided into a range of categories of organisms such as zooplankton, benthic organisms, fish and seabirds. The impacts on marine and freshwater organisms of ingesting microplastic particles are largely unknown (Eerkes-Medrano *et al.* 2015) and, as physical impacts of microplastics from field observations are hard to come by, researchers have used laboratory-based experimental facilities instead to investigate particle uptake, retention and effects (GESAMP 2015). According to Browne et al. (2015), less than 1% of all studies have been able to demonstrate conclusive ecological impacts in nature because (i) of difficulties in complex biological systems in demonstrating unambiguously that effects were due to debris, and (ii) many studies inferred impacts from the presence of debris (Browne et al. 2015). A summary of identified results from field and laboratory studies are shown in Table 5.

## 2.4.1 Zooplankton

It is well-known that filter-feeding zooplankton and other planktonic organisms can take up plastic particles that are mistaken for prey (Cole *et al.* 2011). A number of laboratory studies have been published on zooplankton taxa, mainly crustaceans, and it has been reported that there was significantly reduced feeding among copepods in the presence of microplastics (Ivar do Sul and Costa 2014). Additionally, plastic particles could adsorb onto biological surfaces. This adsorption could cause effects on the organisms by, for example, affecting algal photosynthesis as Bhattacharya *et al.* (2010) have reported for plastics in the nano range. At the same time, adsorption can lead to a transfer of plastic particles through the food chain if, for example, these algae are ingested by zooplankton.

TABLE 5 EXAMPLES OF REPORTED EFFECTS FROM MICROPLASTICS EXPOSURE IN THE FIELD AND IN THE LABORATORY

Туре	Organism	Species	Area	Individuals with microplastics	Effects	Reference
Field study	Haddock	Melanogrammus aeglefinus	North Sea	6/97 individuals contained plastic particles	Condition factor significantly lower in individuals that contained plastic than those without plastic. Data deemed insufficient to confirm the hypothesis	Foekema <i>et al</i> . 2013
Field study	White faced storm petrel	Pelagodroma marina	Gough Island, Central South Atlantic Ocean	16/19 birds contained plastic particles in their gizzards	Statistically weak correlation identified between mass of ingested plastic and body mass	Furness 1985
Field study	Wedge-tailed shearwater	Puffinus pacificus	Tropical Pacific	17/85 birds had ingested plastic	Negative relationship identified between plastic ingestion and physical condition (body weight)	Spear <i>et al</i> . 1995
Field study	White-winged petrel	Pterodroma leu- coptera	Tropical Pacific	13/110 birds had ingested plastic	Negative relationship identified between plastic ingestion and physical condition (body weight)	Spear <i>et al</i> . 1995
Field study	Leach's storm petrel	Oceanodroma leucorhoa	Tropical Pacific	70/354 birds had ingested plastic	Negative relationship identified between plastic ingestion and physical condition (body weight)	Spear <i>et al</i> . 1995
Field study	Stejneger's petrel	Pterodroma longi- rostris	Tropical Pacific	34/46 birds had ingested plastic	Negative relationship identified between plastic ingestion and physical condition (body weight)	Spear <i>et al</i> . 1995
Field study	Sooty shearwater	Puffinus griseus	Tropical Pacific	27/36 birds had ingested plastic	Negative relationship identified between plastic ingestion and physical condition (body weight)	Spear et al. 1995
Laboratory study	Phytoplankton	Scenedesmus	-	-	Hindered algal photosynthesis and promotion of algal ROS indicative of oxidative stress	Bhattacharya <i>et al.</i> 2010
Laboratory study	Zooplankton	various species	-	-	7.3 μm microplastics (>4000 mL−1) significantly decreased algal feeding	Cole et al. 2013
Laboratory study	Blue mussel	Mytilus edulis	-	-	Granulocytoma formation (inflammation). Increase in SB haemocytes; decrease in lysosome stability.	von Moos et al. 2012

Туре	Organism	Species	Area	Individuals with microplastics	Effects	Reference
Laboratory study	Blue mussel	Mytilus edulis	-	-	Reduced clearance rate and for 30-nm PS reduced filtering/feeding activity	Wegner et al. 2012
Laboratory study	Lugworm	Arenicola marina	-	-	Statistically significant effects on the organisms' fitness and bioaccumulation, but the magnitude of the effects was not high.	Besseling et al. 2013
Laboratory study	Lugworm	Arenicola marina	-	-	1% of sediment (w/w) reduced total energy reserves by approximately 30%, mainly linked to a reduction in lipid reserves.	Wright et al. 2013b
Laboratory study	Copepod	Tigriopus japoni- cus	-	-	100% survival in 96h tox test. Chronic mortality for 0.05 µm PS >12.5 µg/mL. Reduced fecundity for 0.5 and 6 µm PS.	Lee et al. 2013
Laboratory study	Carp species	Carassius carassius	-	-	Food chain transport of NPs affects behaviour and fat metabolism	Cedervall et al. 2012
Laboratory study	Sea urchin	Lytechinus varie- gatus	-	-	Negative effect on embryonic development	Nobre et al. 2015
Laboratory study	Copepod	Centropages typi- cus	-	-	Significantly decreased copepod feeding on algae	Cole <i>et al.</i> 2013
Laboratory study	Copepod	Calanus helgoland- icus	-	-	Significantly decreased copepod feeding capacity. Prolonged exposure significantly decreased reproductive output (egg hatching success and survival)	Cole <i>et al.</i> 2013
Laboratory study	Marine isopods	Idotea emargina- ta	-	-	No effects on mortality, growth, and intermolt duration after 6 weeks exposure to ~12 microbeads-mg-1 86 food and an estimate ingestion of ~500 plastic particles per day	Hämer et al. 2014
Laboratory study	Japanese Medaka	Oryzias latipes	-	-	Evidence of liver stress and endocrine disruption in Japanese medaka ( <i>Oryzias latipes</i> ) after two-month dietary exposure corresponding to 8 ng of plastic per mL	Rochman et al. (2014c)

## 2.4.2 Benthic organisms

Studies concerning microplastic ingestion by benthic crustaceans are limited and most of the studies related to benthic organism have focused on determining whether microplastic particles have been ingested by benthic organisms and not on the biological effects of this ingestion (Ivar do Sul and Costa 2014). Although microplastics were not observed to accumulate in the digestive tracts of lugworms during a 28-day experiment, Besseling *et al.* (2013) observed a positive relationship between the microplastic concentration in the sediment and the ingestion of plastics on the one hand and the weight loss and reduced feeding activity on the other. Microplastic particles have furthermore been observed to cause an inflammatory response in tissues of blue mussels (*M. edulis*) and reduced membrane stability in cells of the digestive system (Besseling *et al.* 2013; Ivar do Sul and Costa 2014).

Some of the first indications that adhered pollutants comprise a problem for biota were identified by Teuten *et al.* (2007), who modelled the adsorption and desorbing trends for phenanthrene to PE, PP, and PVC in order to the determine whether phenanthrene causes harm to biota, in this case a sediment-dwelling polychaete worm (*Arenicola marina*). The model predicted that a proportion of the sorbed phenanthrene would indeed dissociate to the worm; however, if in competition with clean organic-rich sediment, a significant part of the contaminant would adhere to the soil instead of ending up in the worm. A study by Koelmans *et al.* (2014) suggests that the ingestion of microplastics is not a relevant transfer pathway for nonylphenol and bisphenol A, as the obtained concentrations within the intestinal tract of both a lugworm (*Arenicola marina*) and a North Sea Cod (*Gadus morhua*) did not exceed the lower ends of the global concentration ranges for the two chemicals.

### 2.4.3 Fish

The ingestion of microplastics by fish was discovered many years ago in the wild (Carpenter and Smith 1972; Ivar do Sul and Costa 2014) and in controlled laboratory experiments from 1990; all six different species of fish tested ingested 100-500 mm pellets of microplastics (Hoss and Settle 1990, as cited by Ivar do Sul and Costa 2014). Few studies have reported on the quantitative average number of microplastic particles observed in fish, but Boerger et al. (2010) found synthetic fractions in the gastrointestinal content from 35% (N = 670) of the planktivorous fish in the North Pacific Central Gyre. The average number of plastic pieces ingested (1-2.79 mm) increased with the fish size. The number of studies is limited as regards biological effects observed in fish. However, bioaccumulation and liver stress response and early tumour formation have been reported in the fish Japanese medaka (Oryzias latipes) fed virgin and marine polyethylene fragments of the size <0.5 mm (Rochman et al. 2013b; Eerkes-Medrano et al. 2015). Rochman et al. (2014c) have furthermore found evidence of liver stress and endocrine disruption in Japanese medaka (Oryzias latipes) after two months of dietary exposure to environmentally relevant concentrations of microplastics (<1 mm) and associated chemicals. In a field-collected estuarine Eugerres brasilianus, adults that ingested plastic fragments (<5 mm) were found to have a lower mean total weight of gut contents, which could indicate a reduction in feeding or false satiation (Ramos et al. 2012; Eerkes-Medrano et al. 2015).

## 2.4.4 Seabirds

Seabirds have long been known to ingest microplastic particles and have been used to monitor the quantities and composition of plastic ingestion for decades, but the biological effects of ingested microplastics remain less explored (Ivar do Sul and Costa 2014).

Cole *et al.* (2011) studied the uptake and accumulation of polychlorinated biphenyls (PCBs) in streaked shearwater chicks. Two groups of chicks were served fish and resin pellets, or only fish and the preen gland oil, was analysed weekly for a duration of 42 days. In both groups, PCB concentrations increased over the test period. The contribution from the resin pellets was determined by a congener PCBs analysis that showed that an increase was found to be significantly larger in the chicks eating the plastic pellets. More recent models estimating transfer of adhered pollutants in-

clude conditions found in the gut such as varying pH and temperature as well as the role of gut surfactants. Bakir et al. (2014a) examined the potential for PVC and PE to sorb and desorb for four different carbon 14-labeled persistent organic pollutants (POPs) in both seawater and under simulated gut conditions, in cold and warmed blooded biota respectively. They found desorption rates to be faster under simulated gut conditions and highest when simulating a warm blooded organism. Compared to desorption rates in seawater, these conditions resulted in rates being 30 times greater, suggesting that there is a great risk of transfer of POPs into biota. Furthermore, the combination of POP and plastic type that gave the highest potential for transport to biota was phenanthrene adhered onto polyethylene (Bakir et al. 2014a).

#### Mammals and other large animals in the aquatic environment 2.4.5

As with most other groups of animals, research so far has focussed on studying the ingestion of microplastic particles for marine mammals and little research has been done on studying the biological effects of e.g. ingestion (Ivar do Sul and Costa 2014).

## Microplastics as a carrier of hazardous substances

A number of studies have demonstrated that microplastics found in the environment may contain hazardous substances. The substances may be of two different origins:

- Hazardous substances already present in the plastic particles or items when they were released to the environment. The hazardous substances may either be substances with an intentional function in the plastics (e.g. flame retardants or pigments), they may be traces of unreacted raw materials (e.g. alkylphenols) or they may be traces of auxiliaries used in the production of the plastics (e.g. perfluorooctanoic acid (PFOA)).
- Hazardous substances present in the environment and adsorbed to the surface of the plastics. Over time, the substances may be absorbed into the plastic matrix (i.e. the substances migrate from the surface into the particles).

The following section sets out a description of the levels of hazardous substances found in the environment, followed by subsections describing hazardous substances in manufactured plastic materials. The significance of microplastics for exposure of organisms to hazardous substances and of microplastics as media for long-range transport of pollutants is discussed in two subsequent subsections. Much of the information presented has been extracted from a recently published review for the Norwegian Environment Agency (Nerland et al. 2014) and the assessment from GESAMP (2015).

#### Hazardous substances in microplastics in the environment 2.5.1

Studies of hazardous substances in microplastics in the environment fall into two categories:

- Sampling of microplastics found in the environment
- Experimental exposure of microplastics to hazardous substances in the environment

Mato et al. (2001) first demonstrated that plastic resin pellets, unintentionally released from the plastic industry to the environment, contained measureable concentrations of hazardous substances. The concentrations of PCBs (4-117 ng/g), DDE¹ (0.16-3.1 ng/g), and nonylphenols (NP) (130-16,000 ng/g) varied in polypropylene (PP) resin pellets collected from five Japanese coasts. For the PCBs and DDE, the concentrations were correlated to the concentrations in suspended particles and bottom sediments collected from the same area as the pellets. Field adsorption experiments using PP virgin pellets demonstrated a significant and steady increase in PCBs and DDE concentrations throughout a six-day experiment, indicating that the source of PCBs and DDE was the ambient seawater and that adsorption to pellet surfaces was the mechanism of enrichment.

<sup>&</sup>lt;sup>1</sup> DDE = degradation product of DDT

From the results, it was suggested that beached plastics pellets could be used for monitoring of POPs and other hazardous substances. A similar correlation could not be demonstrated for NP. The major source of NP in the marine PP resin pellets was thought to be plastic additives and/or their degradation products present in the pellets at the time they were released.

## Mechanisms of sorption

The mechanisms of sorption of hazardous substances to plastics in the environment have recently been reviewed by Nerland *et al.* (2014) and reference is made to that study.

In short, the uptake rates are varying and dependent on the physico-chemical properties of the substances and the plastic materials; salinity, temperature and concentration gradient between the plastic material, and the ambient water/sediment.

The rate of uptake and release of POPs from plastics largely depend on the size of the plastics. With increase in the size (thickness) of plastics, sorption and desorption become slower. As an example, to thin polyethylene film with a thickness of 50  $\mu$ m PCBs (congener PCB-52) sorbed in 50 days to reach equilibrium (Adams *et al.* 2007), whereas sorption of PCBs to PE pellets with diameter of 3 mm was slower (Mato *et al.* 2001) and took approximately one year to reach equilibrium (Rochman *et al.* 2013a; 2014a (all, as cited by GESAMP 2015).

Besides the sorption to the plastics themselves, hazardous substances may be accumulated in biofilms formed at the surface of the plastics (GESAMP 2015). Several studies document that marine microplastics are covered with biofilm communities. This organic layer likely acts as a reservoir for hazardous substances such as POPs, although studies demonstrating persistent differences in affinities of POPs among plastic types deployed in marine waters suggest that the biofilms modify rather than control the associations of POPs with aged marine microparticles (GESAMP 2015).

## Use of plastic pellets for monitoring hazardous substances

The International Pellet Watch<sup>2</sup>, IPW, was launched in 2005 as a volunteer-based global monitoring program designed to monitor the pollution status of the oceans. About 80 groups and individuals from approximately 50 countries have been participating in the programme. In the programme, plastic pellets unintentionally released to the environment, both during manufacturing and transport, are collected from beaches all over the world and analysed for the presence of some of the POPs targeted by the Stockholm Convention as well as a few other hazardous substances. Global pollution maps of PCBs, dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexane (HCH) isomers (HCHs), Hopanes and PAHs have been drawn up and maps for several POPs pesticides are in preparation (April 2015).

The global map for PCBs is shown in Figure 6. As illustrated by the map, PCB concentrations in pellets from beaches around the world were found to be spatially different; the coast of the USA had the highest, followed by Japan and Europe. Australia, tropical Asia and southern Africa had much lower concentrations. These findings reflect the PCB usages in the specific countries; for example, as much as 50% of all PCB produced globally was used in the USA. PCBs have not been intentionally used in any country for more than 25 years and the presence of the PCBs in the pellets is clearly the

Microplastics

<sup>&</sup>lt;sup>2</sup> http://www.pelletwatch.org/

uptake of PCBs from the ambient water.

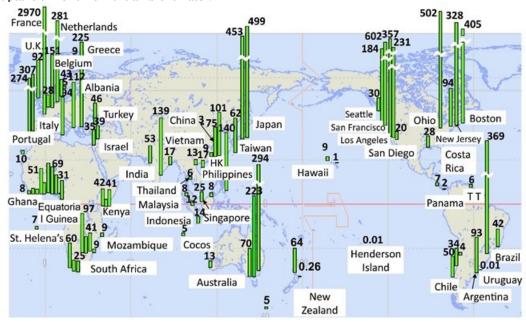


FIGURE 6
CONCENTRATION OF PCBS IN BEACHED PLASTIC RESIN PELLETS IN NG/G PELLET. REPRODUCED WITH PERMISSION FROM THE INTERNATIONAL PELLET WATCH (WWW.PELLETWATCH.ORG)

## Reported concentrations of hazardous substances in collected plastic particles

Reported concentrations of various hazardous substances in plastic particles collected in the marine environment are summarised in Table 6. For each of the substances it is indicated whether the substances are intentionally used in the production of plastic materials.

Substances that have received much attention include the POPs such as PCBs, polycyclic aromatic hydrocarbons (PAHs) and the insecticides DDT/DDE, HCHs, mirex and chlordanes. Apart from the PAH, which might be present as impurities of some types of manufactured plastics, these substances are all are expected to be sorbed from the ambient water. In a majority of cases, these particles have been collected from beaches and the plastic particles were mostly polyethylene and polypropylene, in agreement with high production volumes for these particles, and that these plastic types have a density of below 1 (i.e. they float).

Measurements based on open sea samples and plastics found in the guts of organisms have also been reported (e.g. Colabuono *et al.* 2010, Rios *et al.* 2010). Rios *et al.* (2010) collected plastic debris in the North Pacific Gyre and analysed for 36 individual PCB congeners, 17 organochlorine pesticides, and 16 PAHs. Particles collected included intact plastic items as well as many pieces less than 5 mm in size. Over 50% of the plastic particles/items contained PCBs, 40% contained pesticides, and nearly 80% contained PAHs in measureable concentrations. The concentrations of pollutants found ranged from a few ng/g to thousands of ng/g. The types of PCBs and PAHs found were similar to those found in marine sediments. The plastic particles were mostly polyethylene.

For substances intentionally used as plastic additives, concentrations in plastic particles are likely to be initially much higher than those that could be achieved through sorption from seawater. For example, the observation of the flame retardants decaBDE (BDE-209) concentrations close to 10,000 ng g-1 plastic is most likely the result of its use as an additive flame retardant in the plastics rather than as a result of uptake from seawater.

**TABLE 6**SUBSTANCE CONCENTRATIONS IN PLASTIC PARTICLES COLLECTED IN THE MARINE ENVIRONMENT (EXTENDED FROM NERLAND *ET AL.* 2014)

Substance	Intentionally used in the	Concentrations	Reference
	production of plastic	range ng/g	
	materials*	plastics	
Polycyclic aromatic hydrocarbons	No - may unintentionally be present in some types e.g. in	1-24,364	Ogata <i>et al.</i> 2009; Hirai <i>et al.</i> 2011; Rios <i>et al.</i> 2007; Karapanagioti <i>et al.</i> 2011
(PAHs)	tyres		200/, Karapanagioti et ut. 2011
Polychlorinated biphen- yls, PCBs	Historically used in some sealants, but PCBs in plastic pellets in the environment is more likely sorption from the ambient water	1-5,000 (two samples: 18,600, 18,700)	Ogata <i>et al.</i> 2009; Carpenter and Smith 1972; Hirai <i>et al.</i> 2011; Rios <i>et al.</i> 2007; Karapanagi- oti <i>et al.</i> 2011; Mato <i>et al.</i> 2001; Endo <i>et al.</i> 2005; Heskett <i>et al.</i> 2012; Ryan <i>et al.</i> 2012
DDT and related compounds	No (pesticide)	0.16 - >1,000	Rios <i>et al.</i> 2007; Hirai <i>et al.</i> 2011; Karapanagioti <i>et al.</i> 2011; Mato <i>et al.</i> 2001; Teuten <i>et al.</i> 2009; Ryan <i>et al.</i> 2012
Polybrominated diphenyl ethers (PBDE)	Flame retardant	0.3-9,909	Teuten <i>et al.</i> 2009; Hirai <i>et al.</i> 2011
DecaBDE (BDE-209)	Flame retardant	0.1-9,907	Hirai et al. 2011
Hexachlorocyclohexane isomers (HBCDD)	Flame retardant	<2-36	Ogata et al. 2009
Chlordanes (+ oxychlordanes)	No (pesticide)	4.29-14.4	Colabuono et al. 2010
Cyclodienes	No (pesticide)	2.41-50.9	Colabuono et al. 2010
Mirex	No (pesticide)	6.48-14.6	Colabuono et al. 2010
Hopanes	Natural terpenes	2,000-61,000	Teuten et al. 2009
Aliphatic hydrocarbons	Various	1.1-8,600	Rios et al. 2007
Hexachlorobenzene	No (pesticide)	12.4-17.5	Colabuono et al. 2010
Nonylphenols	May be present as unreacted raw material	0.7-3,936 (one sample: 16,000)	Mato et al. 2001; Hirai et al. 2011; Ryan et al.
		5ample, 10,000)	
Octylphenols	May be present as unreacted raw material	0.1-154	Hirai et al. 2011; Teuten et al. 2009
Bisphenol A (BPA)	May be present as unreacted raw material	0.2-730	Hirai et al. 2011; Teuten et al. 2007
Perfluoroalkyl sub- stances (PFASs)	Used for some coatings but PFCs in particles in the environment most likely is sorption from the ambient water	0.011-0.115	Llorca et al. 2014

<sup>\*</sup> This column has been added as a part of the present review.

## 2.5.2 Hazardous substances in manufactured plastic materials and coatings

Two recent reports for the Norwegian Environment Agency and the Danish Environmental Protection Agency have reviewed the knowledge on hazardous substances in plastic materials (Hansen *et al.* 2013; Hansen *et al.* 2014a).

Hansen *et al.* (2013) describes the use in plastics of 43 hazardous substances adopted on the Norwegian Priority List of hazardous substances or the REACH Candidate list of SVHC-substances.

The report provides information about plastic types in which the substances are used and the main applications of these plastics. Hansen *et al.* (2014a) provides information on 132 hazardous substances used in plastic materials selected from a gross list of 330 substances from the Danish EPA's List of Undesirable Substances (LOUS), the REACH Candidate List, CMR<sup>3</sup>-substances likely to be present in plastic toys and several other lists.

Examples of the main hazardous substances used in plastics are shown Table 7. Many of the same substances are used for similar applications in various coatings (paint, lacquer. varnish, etc.).

## Potential routes of releases to the environment

The potential routes of releases to the environment of the plastic materials containing hazardous substances are:

- Releases of plastic raw materials (pellets and powder) containing the substances
  by transport and manufacturing processes to waste water and directly to surface waters
  (described in section 5.1.2). This release route may be relevant for substances present in
  plastic compounds of thermoplastics e.g. additively used flame retardants such as decaBDE and plasticisers such as the phthalates.
- Wear and tear of plastic materials during use (described in section 5.2.1 and other sections). Examples are plastic dust from flooring containing phthalates or phenylmer-cury compounds and dust from tyres containing residual octylphenol (OP). From coatings, the plastics are released by abrasion and by maintenance of the coatings.
- Losses of small plastic parts by waste handling. Examples are plastic dust from shredders or dust from handling of plastics in landfills. The plastics may contain various hazardous substances.
- **Plastic parts disposed of or lost** to the environment. Examples are plastic foils with heavy metal colourants or plasticisers and fishing tools with heavy metal colourants.

## Main groups of substances

**Plasticizers** - Plasticizers are additives that make the plastics flexible and durable. The plasticizers are most important as softeners for hard plastic material, and over 90% of all additives are used in conjunction with PVC. The phthalates account for about 90% of the consumption of plasticizers. The plasticizers are often used in a combination of a primary plasticizer (e.g. DEHP, DINP or DIDP) and secondary plasticizers (e.g. DBP, BBP or chlorinated paraffins). The main sources of releases of phthalates are abrasive releases from wear and tear of PVC products as has been estimated for the phthalates DEHP, DBP and BBP. As an example, the main releases to the environment of DEHP (historically the main phthalate) are considered to occur from wire and cables left in the environment and abrasion of roofing and flooring materials, coated fabric and shoe soles (COWI *et al.* 2009). In the environment, the plasticizers may be released by diffusion processes and by degradation of the plastics.

Flame retardants - Various brominated flame retardants (often used together with antimony trioxide as the synergist) are the main flame retardants used in plastic materials. The main application areas are electrical and electronic products, building applications and transportation (Lassen *et al.* 2014). For decaBDE and other additive flame retardants with a similar application in e.g. electrical and electronic equipment, the main routes of releases to water are considered to be production processes (including releases of raw materials with the flame retardants) and the use of the substances in textiles (from production processes, washing and textiles disposed of in the environment), but the major release route of the substances is considered to be releases to air. For HBCDD used in EPS insulation materials for buildings, formulation processes and industrial use of the flame retarded raw materials (including releases as microplastics from production processes) are considered to be the main release routes, while releases from EPS in use is estimated to account for

<sup>&</sup>lt;sup>3</sup> CMR: Carcinogenic, mutagenic or reprotoxic

some 10% of the total releases (Lassen *et al.* 2014). Compared to the plasticisers, where released plastic items/dust is a major release pathway, this pathway appears to be of less importance for the flame retardants, but may still be of significance.

**Colourants -** Various pigments based on heavy metals such as lead, cadmium and chromium has traditionally been intensively used in plastic materials and coatings. Release pathways may include manufacture of plastic articles, wear and tear of products in use, plastics disposed of in the environment and maintenance of coatings. Heavy metals have e.g. been used in plastic foils and a significant release pathway may in fact be marine litter. Actual indications of the releases with plastic particles have not been identified. A Danish substance flow analysis for lead estimated that 2-10% of the lead pigments used in the country were released to the environment with paint dust, but did not estimate any releases with other plastic items/particles (Lassen *et al.* 2003).

**Stabilizers** - Stabilizers based on cadmium and lead compounds have traditionally been used for stabilization of PVC used for outdoor applications such as pipes, gutter, windows and door frames. Release pathways may include losses by production and dust generated by abrasion and maintenance of products as well as dust generation by waste treatment. No indication of the significance of releases with plastic items/particles has been identified. A major pathway of release to the environment may be piping left in the ground, but the plastics in the piping most likely would not reach surface water (the cadmium and lead may be released over time from the plastics and mobilised).

**Curing agents** - Phenylmercury compounds have been used for many years as accelerators for curing of PUR elastomers for various applications. The use of the substances are now restricted in the EU, but PUR materials in use may still contain the mercury compounds. An Annex XV restriction dossier for five phenylmercury compounds estimated that the main source of release of the substances to sewage was abrasive releases (dust) from flooring containing the substances as accelerators (Klif, 2010). The lifetime releases factor was estimated to be in the range of 0.5-5% of the content of the flooring. The materials are to some extent used for marine applications, but no releases from wear and tear of the materials were estimated in the Annex XV report.

**Antioxidants** - The EU Risk Assessment report for bisphenol A (BPA) estimated that losses from PVC articles in use account for approximately 50% of the releases to surface waters (ECB 2010). The BPA is used as an antioxidant. The report does not indicate whether the bisphenol A is released by migration from the materials or released in the form of abraded materials (microplastics).

**Unreacted raw materials** - Dust from abrasion of tyres (where the OP may be bound in the abraded rubber particles) is considered as likely being the major source of OP releases to surface water) and sewage treatment (Lassen *et al.* 2015). Some uncertainty regarding the content of OP in the particles abraded from the tyres does, however, exist because the OP is present in the core of the tyres whereas the major part of the abrasion would be from the outer part of the tyres.

Bisphenol A is present as residual monomer in polycarbonate, epoxy resins and unsaturated polyester resins. The EU Risk Assessment Report does not indicate any losses to the aquatic environments or sewage from final materials (ECB, 2010).

 $\begin{array}{l} \textbf{TABLE 7} \\ \textbf{EXAMPLES OF HAZARDOUS SUBSTANCES USED IN PLASTICS (BASED ON HANSEN \textit{ET AL}. 2013; 2014a IF NOTHING ELSE IS INDICATED)} \end{array}$ 

Substance group	Examples of hazardous substances	Application in	Typical concentration in material,					
Substance group	Examples of nazardous substances	plastics	percent					
Organic compounds								
Alkylphenols	Nonylphenol (NP), octylphenol (OP), 4-tert-butylphenol (4-t-BP)	Unreacted raw materials; Catalyst (nonylphenol); Heat stabilizers (barium and calcium salts of NP)	Some 3-4% of the AP is present as unreacted AP in phenolic resins, the concentration in final plastics is approximately 0.2-2% residual NP (Lassen <i>et al.</i> 2015)					
Bisphenol A	Bisphenol A	Unreacted raw materials; Antiox- idant	Up to 0.0003-0.1% as unreacted monomer in polycarbonate 0.2% as antioxidant in PVC					
Brominated flame retardants	Decabrominated diphenylether (DecaB-DE), hexabromocyclododecane (HBCDD), tetrabromo bisphenol A (TBBPA), decabromodiphenyl ethane (DBDPE), ethylene (bistetrabromophthalimide) (EBTEBPI)	Flame retardants	2-28% (various applications) 0.7% in EPS, 1-3% in XPS (HBCDD)					
Chlorinated paraffins	Medium-chain chlorinated paraffins (MCCP) Short-chain chlorinated paraffins (SCCP) (mainly historic use)	Plasticizers; Flame retardants	MCCP: 9-13% SCCP: 10-15% (in sealants)					
Chlorinated phosphates	Tris(2-chloroethyl)phosphate (TCEP), Tris(2-chlor-1-methylethyl)phosphate (TCPP)	Flame retardants; Plasticizers	TCEP: 1-10% in PMMA and PA, 0-6% in PUR, 0-20% in unsaturated polyester (UPE)					
Phenylmercury compounds	Phenylmercury acetate, phenylmercury propionate, phenylmercury 2-ethylhexanoate, phenylmercuric octanoate, phenylmercury neodecanoate (mainly historic)	Accelerators, curing agents	0.1-0.3% mercury in PUR					
Phthalates	Bis (2-ethylhexyl)phthalate (DEHP), dibutyl phthalate (DBP), benzyl butyl phthalate (BBP), diisononyl phthalate (DINP), diisobutyl phthalate (DIBP)	Plasticizers	10-40% (total phthalates)					
Inorganic compou	inds							
Boric com- pounds	Boric acid	Flame retardant	Up to 8%					
Antimony triox- ide	Antimony trioxide (together with brominated flame retardants	Flame retardant	4-10% (Lassen <i>et al</i> . 2014)					
Lead com- pounds	Lead chromate molybdate sulphate red, lead sulfochromate yellow, lead stearate	Stabilizers; Colourants	o- 5% (colourants) ~2% (Stabilisers)					
Cadmium com- pounds	Cadmium chloride, cadmium oxide	Stabilizers, colourants	~0.01-1% (colourants) ~0.1% (stabilizer)					
Cobalt(II) compounds	Cobalt(II) diacetate	Catalyst, pigments	<1% (pigments) no information regarding use as catalyst					

## 2.5.3 Significance of microplastics for exposure of lower-trophic-level organisms to hazardous substances

Substances used intentionally in the plastics as well as contaminants sorbed during exposure of microplastic particles to the substances in the environment can ultimately be transferred into organisms. If a contaminant concentration gradient between the organism and the ingested plastic exists, gut fluids have the potential to facilitate the transport of chemicals from the plastics to the organism. While laboratory studies have demonstrated that transfer of chemicals from ingested plastic particles to the organism is possible, it is still uncertain to which extent this process significantly contributes to the exposure of the organisms in the environment to hazardous substances. In this respect, it is essential to distinguish between substances present in the plastic particles when released (e.g. flame retardants) where the concentrations can be relatively high and the plastic particles serve as primary sources of contamination, and plastic particles where the substances have been sorbed from the ambient water or sediment to which the organism are exposed as well.

### Substances sorbed from ambient water and sediment

The available studies of plastic-sorbed contaminant transfer into organisms shows contradictory results.

Gouin *et al.* (2011) investigated the environmental risks associated with microplastics, with an emphasis on assessing the interaction of PBT (persistent, bioaccumulative and toxic) substances with microplastic material in the environment and its subsequent fate within a food-web model. In a two model system representing a coastal marine system and an open ocean environment, respectively, partitioning to polyethylene was <0.1% for all of the assessed substances. In a system where the concentrations of polyethylene was increased by three orders of magnitude (1000 times) and the concentration of natural organic matter was low, results suggest that chemicals with  $\log K_{\rm OW}>5$  have the potential to partition at >1% to polyethylene. Food-web model results suggest that reductions in body burden concentrations for nonpolar organic chemicals are likely to occur for chemicals with  $\log K_{\rm OW}$  between 5.5 and 6.5 when microplastics are present in the system. Thus, according to the authors, the relative importance of microplastics as a vector of PBT substances to biological organisms is likely of limited importance, relative to other exposure pathways. Nevertheless, a number of data gaps were identified, largely associated with the understanding of the physical fate of microplastics in the environment.

Koelmans *et al.* (2013) developed and analysed a conceptual model that simulates the effects of plastic on bioaccumulation of POPs. The model accounts for dilution of exposure concentration by sorption of the POPs to plastic particles (POP "dilution"), increased bioaccumulation by ingestion of plastics containing POPs ("carrier"), and decreased bioaccumulation by ingestion of clean plastic ("cleaning"). The model was applied for the lugworm *Arenicola marina* and evaluated against recently published bioaccumulation data for this species from laboratory bioassays with polystyrene microplastic. Further scenarios included polyethylene microplastic, nanosized plastic, and open marine systems. Model analysis showed that plastic with low affinity for POPs such as polystyrene would have a marginal decreasing effect on bioaccumulation, governed by dilution. For stronger sorbents such as polyethylene, the dilution, carrier, and cleaning mechanism are more substantial. In closed laboratory bioassay systems, dilution and cleaning dominate, leading to decreased bioaccumulation. As well, in open marine systems, a decrease is predicted due to a cleaning mechanism that counteracts biomagnification. However, according to the authors, the differences are considered too small to be relevant from a risk assessment perspective.

Chua *et al.* (2014) evaluated the effect of micro-size PE particles obtained from exfoliating face scrub soap on the assimilation of PBDEs in the marine amphipod *Allorchestes compressa*. In one experiment, polybrominated diphenyl ethers (PBDEs) were added to seawater containing microplastics and the amphipod. In this system, the microplastic particles acted as an additional sorption phase in the system, resulting in an overall reduction of bioaccumulated PBDEs when compared

with a seawater-only treatment. The presence of microplastic particles influenced which PBDE congeners were incorporated into the amphipods. When microplastic particles were present, higher-brominated PBDEs (e.g., BDEs-153, -154) were preferentially taken up and bioaccumulated compared with lower-brominated PBDEs. In another experiment, the PBDEs were sorbed to the PE particle prior to amphipod exposure to these particles, and the amphipod was exposed solely to PBDEs sorbed to plastic particles. The amount of PBDEs added to the system in the two experiments was the same. The proportion of PBDEs added to the system that was assimilated by the amphipod was lower than when the contaminants were added directly to the water compared to the experiment where the PBDEs were sorbed to the plastic particles. Even the proportion taken up was lower in the second experiment. The study demonstrated that substances present in the microplastics particles may be taken up by the organism.

Rochman *et al.* (2014b) sampled and modelled microplastic contamination along a transect in the Southern Atlantic Ocean in relation to contaminant concentrations measured in mesopelagic lanternfish. Lanternfish sampled from each station and analysed for bisphenol A, alkylphenols, alkylphenol ethoxylates, PCBs and PBDEs, exhibited variability in contaminant levels, but this variability was not related to plastic debris density for the targeted compounds with the exception of PBDEs. The data indicate that apart from the PBDEs (which may have been added to the plastics as flame retardants), plastic-mediated transport of the substances is not a significant source of exposure of the fish. It was found that fish sampled at stations with greater plastic densities did have significantly higher concentrations of the PBDE congeners BDE-183 to BDE-209 in their tissues. The results indicate that plastics containing the higher brominated congeners of PBDEs, added as flame-retardants, are a source of contamination of the marine environment with the PBDEs.

Rochman *et al.* (2014c) conducted a chronic two-month dietary exposure study in the laboratory using Japanese medaka (*Oryzias latipes*) and environmentally relevant concentrations of microplastics (<1 mm) and associated chemicals. The fish were exposed to three treatments: a no-plastic (i.e. negative control), virgin-plastic (i.e. virgin polyethylene pre-production pellets) and marine-plastic treatment (i.e. polyethylene pellets deployed in San Diego Bay, CA for 3 months). The marine plastics contained measurable concentrations of PAHs, PCBs and PBDEs. Altered gene expression was observed in male fish exposed to the marine-plastic treatment, whereas altered gene expression was observed in female fish exposed to both the marine- and virgin-plastic treatment. The authors concluded that their study suggests that the ingestion of plastic debris at environmentally relevant concentrations may alter the endocrine system function in adult fish and warrants further research.

In a previous study, Rochman et al. (2013b) showed that fish exposed to a mixture of polyethylene with chemical pollutants sorbed from the marine environment bioaccumulate these chemical pollutants and suffer liver toxicity and pathology. The plastics concentration in the experiment was 8 ng of plastic per mL of water (particle concentration not reported). According to the authors, maximum concentrations reported in the North Pacific Subtropical Gyre are 300 ng/mL; therefore, the concentrations of plastic used in the experiment may be considered environmentally relevant. Fish fed virgin polyethylene fragments also show signs of stress, although less severe than fish fed marine polyethylene fragments. The study did not indicate how much of the exposure to the hazardous substances in the environment could be attributed to the plastics, but demonstrated that pollutants in the plastics in themselves may result in effects. The authors conclude that polyethylene ingestion is a vector for the bioaccumulation of PBTs in fish, and that toxicity resulting from plastic ingestion is a consequence of both the sorbed contaminants and plastic material. Thus, hazards related to plastic debris are not one-sided - supporting the idea that the mixture of plastic and sorbed pollutants associated with plastic debris should be acknowledged in aquatic habitats. The authors suggest that future studies should examine the hypothesis that plastics comprise a multiple stressor in aquatic habitats, shifting the focus to health effects from the combination of the type, size and shape of the material, the chemical ingredients and the concentration of chemicals that sorbs to the material from the environment.

According to GESAMP (2015), many organisms are able to translocate assimilated microplastic particles within their tissues, storing the foreign body within intracellular structures. It is possible that this defence mechanism would deliver microplastic-associated POPs and additive chemicals to different tissue types and locations than those resulting from uptake from food and water. The relatively low frequency of occurrence and density of tissue-encapsulated microplastics in field-collected marine specimens suggests that this mechanism is likely not an important vector in overall chemical delivery to marine species (GESAMP 2015). However, it is possible that this unstudied mechanism may provide a unique process to deliver chemicals to specific organs, especially for very small plastic particles that can cross membranes.

## Substances present in the plastics when released to the environment

For substances intentionally used as additives in plastic materials or present in plastic materials due to their use in the manufacture of the materials (described in section 2.5.2), the plastic items/particles are not only a vector of pollutants transfer, but a primary source. Examples of substances demonstrated in microplastics in the environment are PAHs, PBDEs, HBCDD and alkylphenols (e.g. nonylphenol and octylphenol), though it is not known how much of the measured concentrations can be attributed to sorption within the environment. The bioavailability of the substances embedded in the matrix (and present in an even concentration all through the particles) may likely be different from the availability of substances sorbed to the particles in the environment.

Koelmans et al. (2014) assesses the potential of leaching of nonylphenol and bisphenol A in the intestinal tracts of *Arenicola marina* (lugworm) and *Gadus morhua* (North Sea cod). The authors used a biodynamic model that allows calculations of the relative contribution of plastic ingestion to total exposure of aquatic species to chemicals residing in the ingested plastic. The scenario studies covered BPA and NP concentrations measured in marine plastics *in situ* of 25-2,660 mg/kg NP and 5-284 mg/kg BPA. The conservative analysis showed that plastic ingestion by the lugworm yields NP and BPA concentrations in the tissue that stay below the lower ends of global NP and BPA concentration ranges in lugworms, and therefore are not likely to constitute a relevant exposure pathway. For cod, plastic ingestion appeared to be a negligible pathway for exposure to NP and BPA. The authors note that the anticipated limited relevance of chemical leaching after ingestion by fish does not imply that leaching from marine plastics as such is irrelevant for aquatic species, as additives are known to be leached directly into fresh and marine waters due to the natural breakdown of plastic in the environment.

Investigations under the EU ECsafeSEAFOOD (2015) programme have aimed at studying the significance of plastic-associated contaminants for the total body burden of marine biota. On the basis of the results, the authors conclude that the analysis of plastic associated contaminants (or additives) in the tissue of these studies organisms seem to suggest that the levels of plastic-specific contaminants (i.e. additives such as PBDEs and other brominated flame retardants and bisphenol A) are not directly related to the microplastics body burdens of these animals. The interpretation of the results - plastics acting as vectors for plastic-associated contaminants, such as PAHs adsorbed to the plastic from the environment - was further complicated by the fact that these contaminants are also present at high concentrations in the surrounding environment (ECsafeSEAFOOD 2015).

## 2.5.4 Significance of microplastics for exposure of higher-trophic-level organisms to hazardous substances

There have been several field studies to examine the transfer of POPs from ingested plastics to marine organisms with a focus on seabirds (GESAMP 2015). Ryan (1988, as cited by GESAMP 2015) measured amounts of plastics in the digestive tracts of Great shearwaters (*Puffinus gravis*) and concentrations of PCBs in the fat tissue of the bird and examined the correlation between amounts of plastics and PCB concentrations. A positive correlation was observed between the mass of ingested plastic and the PCB concentrations in the fat tissue of birds, suggesting the transfer of PCBs in plastics to the organisms. However, correlation was weak because marine organisms, especially higher-trophic animals, are exposed to PCBs through natural prey in addition to ingested plastics.

Tanaka *et al.* (2013; as cited by GESAMP 2015) examined the transfer of PBDEs from the ingested plastics to the tissue of the seabirds with focus on higher brominated congeners not found in prey items of the seabirds. PBDEs in abdominal adipose tissue of oceanic seabirds (short-tailed shearwaters, *Puffinus tenuirostris*) collected in northern North Pacific Ocean were analysed. In three of twelve birds, higher-brominated congeners (viz., BDE-209 and BDE-183) which were not present in the natural prey (pelagic fish) of the birds were detected. The same compounds were present in plastic found in the stomachs of the three birds. These data and their follow-up observations of the same species of seabirds indicated the transfer of plastic-derived chemicals from ingested plastics to the tissues of marine-based organisms. However, the mechanism of the transfer of the chemicals from the plastics to the biological tissue was not revealed. Because the ingested plastics were relatively large (mm to cm-size) and BDE-209 and BDE-183 are highly hydrophobic, slow release and low bioavailability of the chemicals have been suggested (GESAMP 2015).

## 2.5.5 Microplastics as media for long-range transport of pollutants

Microplastics and larger plastic debris have been suggested as a potential media for long-range transport of hazardous substances (Mato *et al.* 2001; Zarfl and Matthies 2010) and several studies have attempted to assess the significance of this transport mechanism.

Zarfl and Matthies (2010) assessed the risk of plastic particles being a significant mode of transport of contaminants to the Arctic. The authors estimated mass fluxes of plastic particle-bound PCBs, PBDEs and PFOA to the Arctic via the main ocean currents and compared them to the transport of the substances in the dissolved state and in air. Contaminant concentrations in microplastics were estimated from literature data for contaminant concentrations in sub-polar seawater and polymerwater partition coefficients. According to the estimates, substance fluxes by atmospheric long-range transport or sea water currents accounted for several tons per year, whereas those mediated by plastics were four to six orders of magnitude smaller. For the estimate, a plastic flux to the Arctic Ocean of 62,000 to 105,000 tons per year was used, assuming the maximum volume transport of ocean water. This calculation was based on average quantities of plastic floating on the world's ocean surface, reported by the United Nations Environment Program. The authors note that plasticmediated transport may be of greater importance for substances that are not transported in air or by ocean currents, e.g. due to rapid degradation or sorption to sediment particles. Highly hydrophobic organic chemicals (for instance with a log Kow>6.5), which typically partition strongly to sediment, may have enhanced mobility if adsorbed to buoyant microplastic material. The authors do not point at specific substances for which plastic-bound transport may be of higher significance.

On a more local/regional level, the transport of persistent organic pollutants by microplastics from riverine to brackish and marine waters and the influence of the salinity gradient on the sorption/desorption of the hazardous substances has been investigated (Bakir *et al.* 2014b). The proposed transport model suggested that estuaries could represent an important source for contaminated microplastics under conditions of pulse release to the marine environment through natural (e.g. flushing) and anthropogenic processes (e.g. dredging).

## 2.6 Microplastics as a carrier for organism transport

The surface of any object exposed to seawater rapidly becomes coated with a variety of inorganic and biological coatings. The rapid increase in floating plastics that do not disintegrate in transit has the potential to bring about a rapid increase in the importance of this vector (GESAMP 2015). Colonization of plastic objects by larger sessile organisms is observed frequently. There is emerging concern that microplastics may also act as a carrier for microorganisms, including pathogenic species of bacteria, resulting in an increase in the occurrence of non-indigenous species and GESAMP (2015) suggests evaluating the potential significance of plastics and microplastics as a carrier for organisms in future assessments.

## 2.7 Macroplastics as a source of microplastics in the aquatic environment

Macroplastics are the most conspicuous kind of marine litter and are precursors to microplastics in the marine environment. Macroscopic plastic litter makes its way to the sea from both land based and sea based sources. It may be transported over long distances or deposited close to where it was released, and its presence in the environment may be due to deliberate or involuntary actions. The many potential sources are reflected in the diverse kinds of plastic litter that are found in the sea.

Marine litter concentrations, including macroplastics, are highest in the Northern Hemisphere, and in particular close to urban regions, in enclosed sea areas and along water fronts (Barnes *et al.* 2009). Litter is present in large quantities on beaches, it floats around on the water surface and in the water column and it is found deposited on the sea floor. The abundances in these different compartments largely depend on the density of the plastic material and the degree of adhered material, such as fouling mussels or barnacles, which increases the density of the litter. The highest abundances are found in heavy populated areas, but no aquatic environment on earth is today free of plastic litter (Barnes *et al.* 2009). Litter on the sea floor tends to reach the highest concentrations on accumulation bottoms (Galgani *et al.* 2000), whereas floating litter reaches the highest abundances in the fronts between different water masses (Law *et al.* 2010).

There is a large general interest in society in marine litter and in particular in marine plastic litter. In contrast to this, the number of field studies of the actual abundances of plastic litter in the sea, are surprisingly few, both in Danish coastal water and elsewhere around the world.

## 2.7.1 Occurrence of macroplastics in the aquatic environment Macroplastics in water and sediment

There is to our knowledge no available data on macrolitter abundances in Danish coastal waters. However, results from visual surveys from ships indicate that macroplastic concentrations are larger close to the coast than in offshore waters. In the German Bight >50 litter items were detected along one kilometre long transects in the waters around Helgoland and East Frisia, and more than 70% of this was made up of plastic (Thiel *et al.* 2011). During a cruise from the high Arctic, across the North Atlantic and along the South American east coast to the Antarctic Peninsula densities of floating litter in the area between the equator and 50°N generally were found to range between 0 and 20 litter items/km (Barnes and Milner 2005). The highest concentrations by far, 10 to >100 litter items/km, were found in the English Channel. Abundances declined further north, and in West Spitzbergen they ranged between 0 and 3 items/km.

The distribution and abundance of marine benthic litter in European coastal waters were reported by Galgani *et al.* (2000). The data was obtained from 27 cruises between 1992 and 1997 and the sampling was done by trawling. The plastic densities ranged between 0 and 101,000 pieces/km² depending on currents and bottom morphology. Accumulation bottoms had higher densities than transport bottoms. The mean concentration in the North Sea was 1.56±0.37 items/ha but a regional hotspot of litter was found 200 km west of Denmark with a density of >6 litter particles /ha (see section 2.9.3 for further discussion). Mean concentrations in the West Baltic Sea were 1.26±0.82

items/ha. Several types of litter were enumerated such as plastic, plastic and glass bottles, metallic objects and fishing gear. Plastic material accounted for more than 70% of all litter at most sampling stations.

The abundance, spatial distribution and characteristics of marine litter on the sea floor were investigated in five areas of the Eastern Mediterranean and Black Seas (Ioakeimidis *et al.* 2014). The abundance varied between 1 211 items/km² in the most polluted area (Saronikos Gulf in Greece) and 24 items/km² in the least polluted (Limassol, Cyprus). Up to 95% of the litter was found to be made up of plastic. Most of the litter was in the range of 10-20 x 10-20 cm. There was also an elevated abundance of plastic items <5 x 5 cm.

Very high abundances of marine litter were found on the sea floor of the Arctic Sea. Analyses of images from the eastern Fram Strait west of Svalbard indicate an increasing trend between 2002 and 2011; from 3,635 to 7,710 items/km² (Bergmann and Klages 2012). More than half of the Arctic litter (59%) consisted of plastics.

## Monitoring of plastic litter on beaches

About 15% of all marine litter is believed to reach the coastline at some point (Save the North Sea, year not indicated). Monitoring of beach litter has been carried out within both the OSPAR and HELCOM region. The OSPAR beach monitoring programme started in 2001 and is still running (OSPAR Commission 2010). All Parties are included, although some Parties have a more ambitious approach than others. A similar initiative initiated by HELCOM was limited to the central Baltic Sea area and was run between 2011 and 2013 (MARLIN 2013). Furthermore, as a consequence of the Marine Strategy Framework Directive (Directive 2008/56/EC), many EU Member States including Denmark, have initiated marine litter monitoring programmes. In addition to these monitoring programmes, NGOs like Keep Denmark Tidy organize national beach cleaning campaigns.

The OSPAR beach monitoring programme includes a number of beaches where litter items along 100 m transects are counted and categorized according to a standard survey sheet. Each beach should be surveyed four times a year. In the North Sea region, data is available from the following countries and number of beaches: Belgium 3 beaches, Netherlands 4 beaches, Germany 5 beaches. Denmark 3 beaches, UK 6 beaches, Sweden 9 beaches and Norway 2 beaches. The first surveys were done in 2001 and the latest available data is from 2012. Three Danish beaches are part of the monitoring programme, two in Jutland, Nymindegab (2011-2012) and Skagen (surveyed 2003-2006) and Suggan on Streymoy on the Faroe Islands (surveyed 2002, 2005 and 2006). The time series are far from complete; with several gap years with no monitoring for most beaches, a great deal of interesting information could still be obtained from the acquired data.

In the present report, the OSPAR data was used to compare the amount of plastic litter on the Danish OSPAR beaches with other beaches in the North Sea region. Abundances were calculated as the sum of all plastic litter items collected at each beach at all sampling episodes in a year. The first year in a suite of sampling years was always omitted since it is uncertain how long those plastic items had been lying on the beach.

Skagen beach and Nymindegab beach (monitored by KIMO Denmark) and two beaches on Sealand (monitored by Aarhus University) are reference beaches and monitored as part of the Danish environmental surveillance programme.

Time trend analyses of a selection of beaches representing different parts of the North Sea did not reveal any obvious changes between the years 2002 and 2012 (Figure 7). The beaches included in the comparison were, besides the three Danish ones, Balmedie in northern Scotland, Saltö at the Swedish Skagerrak coast, close to the Norwegian border, and Sylt in the northern part of Germany.

When comparing the average plastic abundance over the total survey period (2002-2012), the load was generally lower on the beaches in the southern North Sea than on those in the northern North Sea (Figure 7). The abundances at the two Jutland beaches were high compared to the southern North Sea beaches, but moderate compared to e.g. several of the beaches on the Swedish Skagerrak coast.

Three of the four Arctic beaches, including Streymoy on the Faroe Islands, had relatively low abundances of plastic. The exception was Rekvika, close to Tromsö, where levels were very high (>20,000 plastic items per 100 m in a year).

During the beach surveys, all plastic litter objects were identified and the most common objects (based on number) on the Danish beaches were connected to fishing and aquaculture activities (e.g. nets, ropes and plastic sheets used in culturing of oysters and mussels, fishing nets, lobster pots), various kinds of storage containers (e.g. bottles, hard bottle caps, various food packages), plastic bags and consumer products (e.g. combs, shoes, toys). The single most common type of plastic item was cigarette lighters.

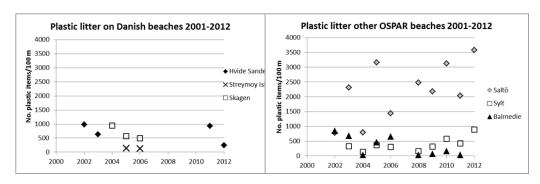


FIGURE 7
TEMPORAL TRENDS IN THE NUMBER OF PLASTIC ITEMS ON BEACHES IN THE OSPAR REGION. TO THE LEFT IS SHOWN THE THREE DANISH BEACHES INCLUDED IN THE MONITORING PROGRAM AND TO THE RIGHT ONE BEACH FROM THE SWEDISH SKAGERRAK COAST (SALTÖ), ONE FROM THE GERMAN NORTH SEA COAST (SYLT) AND ONE FROM THE SCOTTISH EAST COAST (BALMEDE). THE LITTER LOADS ARE EXPRESSED AS THE SUM OF PLASTIC LITTER ITEMS PER YEAR ALONG A 100 m TRANSECT. THE NUMBER OF SURVEYS PER YEAR VARIED BETWEEN ONE AND FOUR AT THE DIFFERENT BEACHES.

Between the years 2011 and 2013, HELCOM also arranged a beach monitoring program called MARLIN. This was limited to the Central Baltic region, and included surveys of beaches in Sweden, Finland, Estonia and Latvia (MARLIN 2013). Denmark has not been part of any beach monitoring in the Baltic Sea.

The MARLIN protocol was based on UNEP/IOC guidelines adapted to Baltic Sea conditions. Similar to the OSPAR surveys, all litter items along 100 m transects were characterized and counted. Twenty-three reference beaches in the four countries were selected, representing both urban and rural areas. Monitoring was conducted three times per year and there were in total 138 assessments carried out within the program (MARLIN 2013). The amount of plastic litter was found to range between 40 items per 100 m on the rural beaches and 160 items/100 m on the urban beaches. The plastics made up around 60-65% of the total amount of litter items found along the transects.

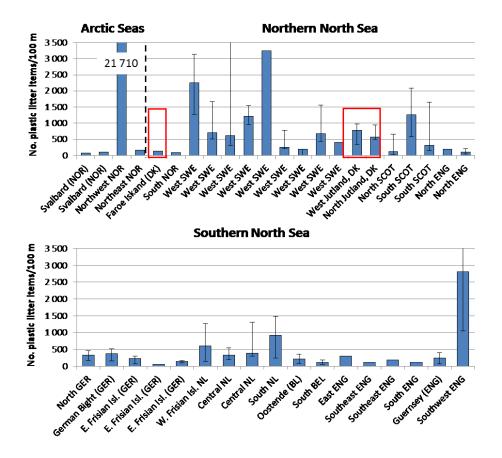


FIGURE 8

NUMBER OF PLASTIC ITEMS WITH RESPECT TO OSPAR DEFINED CATEGORIES ON A 100 m LONG TRANSECT AT BEACHES IN THE ARCTIC SEAS AND THE NORTH SEA. DANISH BEACHES ARE MARKED WITH RED SQUARES. DATA IS PRESENTED AS MEDIAN VALUES (75% CENTRAL RANGE) OVER THE SURVEY PERIOD 2001-2012±SE. NUMBER OF SAMPLED YEARS INCLUDED FROM THE DIFFERENT BEACHES VARIES BETWEEN 1 AND 10. ABBREVATIONS: NOR=NORWAY, DK=DENMARK, SWE=SWEDEN, SCOT=SCOTLAND, ENG=ENGLAND, GER=GERMANY, NL=THE NETHERLANDS, BL=BELGIUM, E=EAST, W=WEST

## 2.7.2 Sources of macroplastics to the aquatic environment

It is frequently stated that 80% of the marine litter comes from land-based sources, but this figure is not confirmed by field data. How much of the litter produced on land that will end up in the sea varies considerably between countries, largely depending on waste management infrastructure (Jambeck *et al.* 2015). Jambeck *et al.* (2015) have calculated that 270 million tons of plastic waste was generated in 192 coastal countries in 2010 and, of this, 4.8 -12.7 million tons entered the oceans. The mismanaged plastic waste derives from badly kept disposal sites and landfills, storm water, etc. If no improvements are made in this area the amount of plastic waste from land to the oceans could be expected to increase by an order of magnitude by 2025.

Plastic litter also derives from activities at sea. Fishing gear lost at sea makes up an important portion of marine plastic litter. In the summer of 2012, 4,980 m of ghost nets (lost fishing nets) were collected in a campaign arranged by KIMO Baltic and Keep Sweden Tidy (KIMO Baltic Sea and Håll Sverige Rent 2012). The collection was carried out by professional fishermen at four locations along the Swedish south coast and around Gotland. A particular issue involves the so-called "dolly ropes", mainly used by bottom trawlers, which consist of dozens of smaller twisted pieces of rope manufactured in such a way that small pieces of thread easily loosen when the rope is used (DollyRopeFree, 2015).

The composition of the macrolitter may give an indication of the sources of the litter. The OSPAR Pilot Project on Monitoring Marine Beach Littering investigated marine litter on beaches of nine OSPAR Contracting Parties (Belgium, Denmark, France, Germany, The Netherlands, Portugal, Spain, Sweden and the United Kingdom) (OSPAR 2009). The project's final report is based on a statistical analysis of marine litter from 609 surveys, using a common, standardized survey protocol on 100 metre stretches of 51 regular reference beaches monitored during the pilot project period (2001–2006), supplemented by 335 surveys of 1 kilometre stretches on 31 regular reference beaches during the same period. The overall results of composition and numbers of marine litter items found on reference beaches is shown in Figure 9.

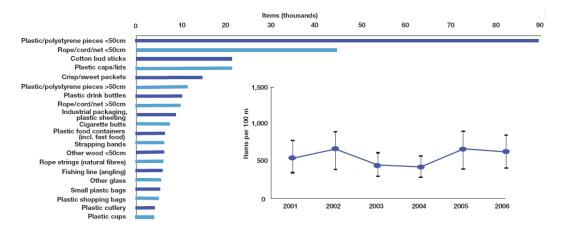


FIGURE 9
COMPOSITION AND NUMBERS OF MARINE LITTER ITEMS FOUND ON REFERENCE BEACHES. THE BAR CHART SHOWS THE TOTAL NUMBERS OF THE MOST COMMON ITEMS. THE POINTS IN THE LINE DIAGRAM REPRESENT THE AVERAGE OF ANNUAL AVERAGES FOR INDIVIDUAL BEACHES (ERROR BARS = 95PER CENT CONFIDENCE INTERVAL) (OSPAR 2009)

In a review for the European Commission, Arcadis (2014) summarises information on composition and possible sources of marine litter in the North Sea (based on same data as shown in Figure 9) and the Baltic Sea (based on similar data from OSPAR). The main conclusions regarding the North Sea Region are (Arcadis 2014):

- "Based on the analysis, plastic/polystyrene pieces of both small (18%) and medium (14%) size were found to be the dominant fraction, followed by string and cord items (12%) and plastic caps/lids (7%).
- The dominant fraction is mainly made of plastic (80%), followed by sanitary items (6%), clothes (5%) and items made of paper/cardboard (2%), processed wood (2%), rubber (2%) and metal (2%).
- A considerable fraction of the beach litter items (30%) is potentially transported over a longer distance, 35% of the items is transported over a short distance from the site, with 35% produced/discarded on site;
- The main contributing sectors are coastal/beach tourism (18%), professional fishing (13%) and the shipping sector (9%). Other important sectors are port activities (8%), households (7%), and other marine industries (8%). This is also reflected in the numbers showing that professional activities tend to have a larger contribution (40%) to marine/beach litter than consumers (33%), resulting in a higher share (51%) of long lasting use goods than in other regional seas.
- The same proportion (32%) of beach marine litter items is likely to be generated during the consumption phase of the life-cycle of the item as during the production phase.
- Direct littering accounts for 43% of beach litter items, whereas 23% comes via inland waterways, 11% from sewerage sources and 24% from other diffuse sources."

Regarding the Baltic Sea Region, the main conclusions are (Arcadis 2014):

- "Based on numbers, plastic pieces with sizes between 2.5 cm and 50 are the dominant fraction (24%) having an average frequency of 34 items per 100 m of coast line, followed by cigarette butts (10%) and other items such as plastic bottle caps/lids (5%), foam sponges (5%), ceramic/pottery items (5%) and plastic (shopping) bags (4%).
- Plastic items are the dominant fraction in the Baltic Sea (58%), followed by items made of paper/cardboard (17%), metal (7%) and ceramic (6%).
- The most important sectors contributing to marine/beach litter are the coast beach tourism sector (the probability of beach litter items being related to this sector is 24%) and the recreational fishing sector (14%).
- Individual consumers (48%) tend to make a larger contribution to marine/beach litter than professionals (17%). 35% remains unknown."

A recent study on marine littering and sources in Nordic waters (Blidberg *et al.* 2015) has undertaken some pilot studies based on pick analyses of litter items collected during clean-up campaigns or in connection with beach litter monitoring. The results confirm that the most common types of litter found on beaches in all Nordic countries are made of polystyrene and other plastics. Short-life items and packaging were the most common product types, strongly linking littering to individual consumers, although the authors note that the litter can originate far beyond the borders of the Nordic countries. At Amager Strandpark in Copenhagen, the Danish beach studied, 80% of the beach litter could be associated with individual consumers, whereas 19% could be related to industrial or professional sources. Litter from the beach studied in Norway differed from the others as it had a higher proportion of industrial packaging from, for instance, the fishing and agricultural sectors, as well as packaging related to the transport of goods.

Many publications state that it has been estimated that 20,000 tonnes of marine litter is dumped into the North Sea annually. According to a report from KIMO from 2000, the estimate originates from a summary record of the October 1995 meeting in Stockholm of the OSPAR Working Group on Impacts on the Marine Environment (Hall, 2000).

The "Save the North Sea" project (Save the North Sea, year not indicated) has estimated that 70% of the litter sinks to the bottom, 15% floats on the surface and 15% is washed up on the coastlines (time perspective is not indicated).

# 2.7.3 Long range transport of macroplastics

In addition to local land and sea-based sources, long-range transportation of marine macroplastics also takes place, both with wind and currents from distant sea areas and via rivers from land-based sources away from the ocean. River estuaries have been identified as hot spots for both macro- and microplastics, and examples of this are findings of large quantities of debris in both the Danube River and in the area around the Danube delta in the Black Sea (Lechner *et al.* 2014; Suaria *et al.* 2015).

On a global scale, the long range wind and current-driven transportation tend to gather floating plastic debris in accumulation zones, where the most important ones are found in the subtropical area of the northern hemisphere. However, modelling based on introduction of plastics from sea and land based sources on a global scale and over a time period of 30 years, revealed that smaller sea areas in densely populated regions like the North Sea and the Baltic Sea have their own gyres of floating litter (Lebreton *et al.* 2012). Most of the plastic debris released here will not be transported away, but rather stay in the area. The model showed that 98% of the land-based litter in the North Sea stayed in the East Atlantic area, and in the Baltic Sea, 100% of the released litter released remained there.

Within the North Sea and the Baltic Sea, the spatial distribution of macroplastics is affected by hydrodynamic conditions, proximity to anthropogenic activities and proximity to river effluents. In surveys of litter concentrations on the sea floor of European waters conducted between 1992 and 1998 (Galgani et~al.~2000), the sediments were trawled with 20 mm nets. In the North Sea two areas with distinctly elevated concentrations of litter compared to the surroundings were detected, and one of them was 200 km west of Esbjerg. The concentrations here were >6 litter items/ha compared to an average concentration in the North Sea of 1.56 litter items/ha. The exact ratio of plastic objects in relation to the total litter content was not stated for this particular area, but the average proportion at all North Sea stations was ~50%. However, when sampling was carried out at the same location several times per year the seasonal variations were found to be substantial.

Information on long-range transport of plastic debris not only reveals what is brought to an area from other countries. It also gives information on how plastic litter from one's own country is transported to other nations. Norwegian data shows that some of the litter on Norwegian beaches originates from Denmark, Sweden, UK and Germany.

# 2.7.4 Formation of microplastics from macroplastics in the environment

The mechanisms and rates of degradation of microplastics have been described in section 2.2.1, whereas this section describes the overall degradation rates and the formation of microplastics from fragmentation of macroplastics.

Abrasion and decomposition of marine litter exposed to environmental conditions will at some point result in cracks to the surface of the plastics causing a weakening of the plastic, followed by embrittlement and micro-fractionation (Andrady 2011, GESAMP 2015) and resulting in the generation of powdery plastic fragments in various sizes. This process is often termed fragmentation or disintegration (Lambert *et al.* 2014). This process occurs in oceans as well as on beaches; however, the potentially higher temperatures achieved in the surface of beach sand, relative to the lower temperature in water, accelerates the light-initiated oxidative degradation, and is the reason why degradation occurs much faster on beaches than in the oceans (Arthur *et al.* 2009; Andrady 2011, GESAMP 2015).

In the oceans, the formation of microplastics is known to be influenced by a combination of environmental factors and the properties of the polymer, but there is a general lack of research information on weathering and fragmentation of plastics in the marine environment and how the combined effects of photo-oxidation, fragmentation, mechanical abrasion and additive chemicals affect the formation of microplastics (GESAMP 2015).

The mechanisms behind the generation of microplastics from macroplastics have been reviewed by Andrady (2011), which the following descriptions, if no other references mentioned, are based on. The likely mechanism for generation of microplastics is the *in situ* weathering of macroplastics. This occurs on the beaches, in the surface water and in the deep-water environments.

When the plastic polymers in the marine environment are exposed to UV-B radiation in sunlight, photo-oxidative degradation is initiated. Once this is initiated, the degradation process can also precede thermooxidatively (i.e. a slow, oxidative breakdown, which occurs at moderate temperatures) for some time, without the need for further exposure to UV radiation. These reactions can continue as long as there is oxygen available to the system. Light-induced oxidation is the fastest degradation process, compared to other types of degradation (e.g. biodegradation and hydrolysis) which are orders of magnitudes slower. Hydrolysis is also usually not a significant mechanism in seawater. Biodegradation of the plastic polymers does occur in the marine environment, but the rate of this process, even in the benthic environment, is still several orders slower than the light-induced oxidative degradation. The initial photo-oxidative degradation, however, renders the plastic more susceptible to further microbial action such as biodegradation.

The breakdown of the macroplastics by weathering occurs at very different rates, depending on where the breakdown occurs. Degradation initiated by solar UV radiation is a very efficient mechanism in plastics exposed to air or lying on a beach surface. However, when the same plastic material is exposed to sunlight at the same location, but while floating in seawater, degradation is severely retarded. This phenomenon is mainly due to the relatively lower temperatures and oxygen levels in the water environments. When exposed in the water environments, the plastics may also be subject to surface fouling, where the surface of the plastic debris is covered with a biofilm, followed by an algal mat and a colony of invertebrates (Muthukumar et al. 2011, as reviewed by Andrady 2011). The fouling increases the density of the plastic debris, causing the plastic to sink down in the water column. Due to the lower temperatures, oxygen levels and fouling effects, plastics that are directly discharged to the water or plastic debris washed in the water prior to any significant weathering degradation are unlikely to yield microplastics via weathering breakdown. The same is the case for plastics debris that sink in the water column. The lack of UV-B (which is rapidly attenuated in the water column) to initiate the process, the low temperatures and the lower oxygen concentration relative to that in air, makes extensive degradation far less likely than for the floating plastics debris. Therefore, the most likely site for generation of microplastics in the marine environment is the beach (Andrady 2011).

Data on the time perspective on degradation of different types of marine debris is generally scarce, and the data that do exist are not very well substantiated and the original references are in some cases no longer possible to locate. References to data from different institutions such as South Carolina Sea Grant Consortium (referred by Ocean Conservancy, no date), Mote Marine Laboratory and the Italian Legambiente have been made in several reports, including reports from UNEP (e.g. Ten Brink et al. 2009; Cheshire et al. 2009), which again are referred to in other reports (e.g. Mouat et al. (2012)). Thus, it is difficult to backtrack the references and the original data sources Table 8 shows summarised degradation rates of different types of marine litter in the sea reported in the literature. For some litter types, the rates are based on actual measurements, whereas for other the rates are based on "expert estimates". For comparison, data for litter items other than plastics are included. For some of the materials, great inconsistency between degradation rates can be seen – for plastic bags, for example, the South California Sea Grant Consortium suggests that they will persist for 1-20 years, which is less than what is suggested by Mote Marine laboratory (10-20 years) and especially Legambiente (1000 years) (as referred by Ecowarriors n.d.; Ocean Conservancy n.d. and The Center for Microbial Oceanography: Research and Education (C-MORE) n.d., respectively). This discrepancy might be due to inconsistency in the understanding of degradation between the references, which is not specified (Cheshire et al. 2009). It should be noted that it is uncertain whether the rates are related to degradation in the sense of fragmentation or mineralisation or both.

It should also be noted that the estimated degradation rates vary substantially depending on environmental conditions such as sun exposure, particularly UV levels, temperature, oxygen level, wave energy and the presence of abrasive factors (sand, gravel or rock) (Cheshire *et al.* 2009).

 TABLE 8

 ESTIMATES OF DEGRADATION RATES OF SELECTED TYPES OF LITTER IN THE OCEAN

Material	Degradation rate	Reference
Plastic items		
Plastic beverage holder	400 years	1; 3
Plastic bags	1 up to 1000 years	3; 1; 2
Disposable diapers	50 - 450 years	1; 2; 3
Plastic bottle	100-1000 years	1; 3: 2
Plastic lighter	100-1000 years	2
Synthetic fabric	500 years	2

Foamed plastic cup	50 years	3
Monofilament fishing line	600 years	1; 3
Polystyrene case	100-1000 years	2
Telephone and top-up cards	1000 years	2
Other items		
Paper towel	2-4 weeks or 3 months	1; 3; 2
Newspaper	6 weeks – 2 months	1; 3; 2
Cardboard box	2 months	1
Waxed milk carton	3 months	1; 3
Apple core	2-6 months	1; 3; 2
Cotton gloves	1-5 months	1
Wool gloves	1 year	1
Cotton or wool garments	8-10 months	2
Wool socks	1-5 years	3
Plywood	1-3 years	1; 3
Painted wooden sticks	13 years	1
Photo-degradable beverage holder	6 months	1; 3
Glass bottle and jars	Undetermined or 1000 years	1; 2
Tin can	50 years	1; 3
Aluminium can	200 -500 years	1; 3; 2
Chewing gum	5 years	2
Cigarette	4-6 years	2
Matches	6 months	2
A banana skin	2 years	2
Cotton rope	1-5 months	3
36 ( 36 '	0 1 6 16 11 10 1	( ) x 1' ·

<sup>1:</sup> Mote Marine Laboratory 1993 as cited by The Center for Microbial Oceanography (2015); 2: Legambiente no date as referred by Ecowarriors (no date); 3: South Carolina Sea Grant Consortium, as referred by Ocean Conservancy (no date)

There is currently a lack of studies where the rates at which macroplastics are broken down into microplastic particles have been investigated. Sundt *et al.* (2014) have summarised the results from the few relevant studies, but all of these studies were terminated before any large defragmentation took place (see Table 9).

TABLE 9
EXAMPLES ON RESULTS FROM STUDIES ON PLASTIC LITTER BREAK DOWN AS REVIEWED BY SUNDT *ET AL.* (2014)

Plastic item type	Defragmentation rate	Comments on study conditions	Reference
Polyurethane foam piece (mattress e.g.)	Brittle and partial disintegration already after 4 weeks exposed	Full sunlight conditions in summer	Hale <i>et al.</i> 2002 as reviewed by Sundt <i>et al.</i> (2014) (reference no. 228)
Extruded polystyrene blocks or buoys (EPS)	Depending on the shape of the element: more than 10% per year	Within a year the outer surface (5 cm) might be completely defragmented	Davidson 2012 as reviewed by Sundt et al. (2014) (reference no. 229)
Plastic films (bags)  Polyethylene  Polypropylene	Brittle within a year, annual mass loss estimated from 1%, up to around 5% when exposed to sun and air. Fouling interrupts the UV degradation largely after short time in sea. Estimated 300 years for total degradation in soil.	Consistent results from several studies. Exposed to air, sun or shallow water	E.g. Andrady 2011; O'brine et al. 2010 and Ohtake et al. 1998 as reviewed by Sundt et al. (2014) (reference no. 230)
Polyurethane flexible foam	40 years: total loss of coherence resulting in powdering.	Suitcase padding, archived in a muse- um	Lattuati-Derieuxa <i>et al.</i> 2013, as reviewed by Sundt <i>et al.</i> (2014) (reference no. 231)
Polypropylene ropes and polyamide (nylon) fish farming nets	Strength loss of 50% in 180 days found in India, more in Oman. Linear relationship: breakdown by time.	Exposed to Oman or Indian sunlight. In Northern Europe sun radiation is lower	Al-Oufi et al. 2004 and Thomas and Hridayanathan 2006, as reviewed by Sundt et al. (2014) (reference no. 232 and 233)

Model estimates on the basis of actual observations of plastics in the ocean indicate that the majority of the plastics in terms of weight in the ocean's surface water occur as larger fractions. Eriksen et al. (2014) estimated the total number of plastic particles and their weight floating in the world's oceans from 24 expeditions (2007-2013) across all five sub-tropical gyres, coastal Australia, Bay of Bengal and the Mediterranean Sea, conducting surface net tows (n = 680) and visual survey transects of large plastic debris (n = 891). Using an oceanographic model of floating debris dispersal calibrated by the available data, and correcting for wind-driven vertical mixing, they estimate a minimum of 5.25 trillion particles in the World's oceans weighing 268,940 tons. When comparing between four size classes, two microplastic <4.75 mm and meso- and macroplastic >4.75 mm, a tremendous loss of microplastics is observed from the sea surface compared to expected rates of fragmentation, suggesting there are mechanisms at play that remove <4.75 mm plastic particles from the ocean surface (see Figure 10). Please note that the smallest fraction is 0.33 mm, and thus still quite large particles of microplastics. The authors note that the observations that there is much less microplastics at the sea surface than might be expected suggests that removal processes are at play. These include UV degradation, biodegradation, and ingestion by organisms, decreased buoyancy due to fouling organisms, entrainment in settling detritus, and beaching. The authors further suggest that fragmentation rates of already brittle microplastics may be very high, rapidly breaking small microplastics further down into ever smaller particles, making them unattainable for the applied nets (0.33 mm mesh opening). Bacterial degradation and ingestion of smaller plastic particles by organisms may facilitate their export from the sea surface. In this manner, incorporation of smaller plastics into marine food chains may not only generate impacts on the health of the involved organisms, but also contribute to the removal of small microplastics from the sea surface (Eriksen et al. 2014).

The data shown are, as mentioned, weight densities. In terms of numbers of particles/km², the smaller fractions (0.33-1.00 mm) are present in abundancies up to 100,000-1,000,000 pieces/km², while the larger fragments (>200 mm) are generally below 1,000 pieces/km² (Eriksen *et al.* 2014).

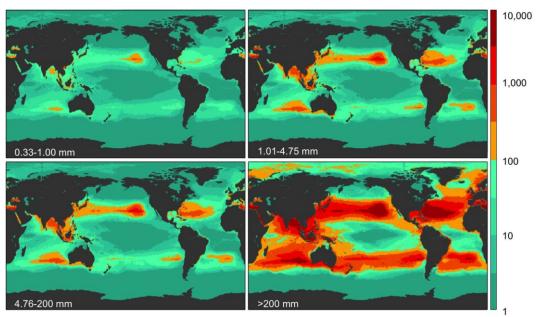


FIGURE 10
MODEL RESULTS FOR GLOBAL WEIGHT DENSITY IN FOUR SIZE CLASSES. MODEL PREDICTION OF GLOBAL WEIGHT DENSITY (g/km; SEE COLOURBAR) FOR EACH OF FOUR SIZE CLASSES (0.33–1.00 mm, 1.01–4.75 mm, 4.76–200 mm, and >200 mm). (Eriksen et. al 2014, open source)

The results demonstrate that the mechanisms of degradation and transport of the microplastics between different environmental compartments are still not fully understood. A better understanding and mass flow models are necessary in order to assess the possible long-term effects of the pollution of the oceans with macro- and microplastics.

# 2.7.5 Significance of macroplastics as source of microplastics in the marine environment

Only one attempt to assess the significance of the degradation of macroplastics in the environment into microplastics as compared with other sources of microplastics has been identified.

Sundt et al. (2014) establish a "best guess" Norwegian emission scenario for secondary microplastics. In the absence of actual data on the quantities of macroplastics reaching the seas around Norway from local sources and by long range transport, the authors make the assumption that of 20,000 tonnes (4% of total plastic annual consumption of plastic products) of plastic littering in Norway, 60% originates from land and 40% from the sea. A further assumption is made that 50% of the land-based litter ends up in the soil on land, while the rest ends up in the ocean as macroplastics. Six thousand tonnes end up in the ocean, and together with the 8,000 tonnes from ocean based sources, the authors reach a total of 12,000 tonnes to the macroplastic litter to the sea. The macroplastics will partly sink and partly float around, while the rest will end up on beaches and there, to a certain extent, degrade to microplastics. Assuming that the macroplastics released in the last 20 years are still present in the environment, and that 1-5% is annually degraded into microplastics, the total formation from the macroplastics in the environment is estimated at 360-1,800 t/y. For comparison, the best estimate for the total releases from other sources was 4,000 t/y (without an indicated range). The authors conclude that "from a pure mass flow perspective" that the annual Norwegian contribution of macroplastics to the sea (and hence also fragmented microplastics from this) can hardly be very much larger than the direct primary emissions of microplastics. By stating that the conclusion concerns "a pure mass flow perspective", the authors emphasise that from e.g. a perspective of possible impacts on the marine organisms it would be necessary to know more about the actual transport, distribution, bioavailability, effects etc. of the microplastics from the various sources.

# 2.7.6 Macroplastics and biota

Besides the possible effects of microplastics formed from the macroplastics, the macroplastics have in themselves become an increasing problem for aquatic biota all over the globe, even at the most remote places. Sea-living species may be entangled in plastic litter and strangled or starved to death. A large number of marine species are also known to ingest plastics, probably mistaking them for food, and plastics are frequently detected in the guts of stranded marine mammals and sea birds. In a study from the North West Atlantic, plastic particles were found in the gut of 21 out of 38 species (Moser and Lee 1992).

Within the OSPAR North Sea countries, the abundance of plastic in stomachs of northern fulmars (*Fulmaris glacialis*) is being used as a biological indicator, Ecological Quality Objective (EcoQO), for marine litter. An ecologically acceptable EcoQO has been defined as a situation where a maximum of 10% of the analysed North Sea fulmars have more than 0.1 g of plastic in the stomach. Analyses of 1,295 North Sea fulmars between 2003 and 2007 revealed that 95% of the birds had plastics in their stomachs, and that the EcoQO was exceeded in ~58% of them (van Franeker *et al.* 2011). Between 2007 and 2011 there was a declining trend from the North Sea going north, where the percent of birds with >0.1 g of plastic went from 62% in the North Sea, to 40% on the Faroe Islands, 28% on Iceland and 14% in Arctic Canada (Kühn and van Franeker 2012; van Franeker and Law 2015).

Plastics are also used as nest material by several species of birds (Hartwig *et al.* 2007). In 1992, 39% of 466 nests of the kittiwake colony at Bulbjerg at the Jammerbugt in Northwest Denmark contained plastic garbage, and in 2005 plastic was found in 57% of 311 nests. The increased share of plastics in the nests was presumed to reflect an increased amount of plastic waste in the environment around the nesting area. It was observed that plastic material resembling natural structure had been preferred and that the share of a particular category of plastic debris corresponded to the amount found on the beach.

In a study of marine debris in nests at gannet colonies in Newfoundland a positive correlation was found between the proportion of nests containing fishing gear and the number of gillness set in the sea around the breeding grounds (Bond *et al.* 2012).

One of the few reports on plastic litter in marine mammals from the North Sea is from a study made on harbour seals (*Phoca vitulina*), stranded at the Dutch island Texel during an outbreak of Phocine Distemper Virus (PDV) (Rebolledo *et al.* 2013). The analyses showed that 11% of 107 animals had plastic in their stomach. The prevalence was highest in animals up to 3 years of age.

# 2.8 Microplastics in the soil environment

Microplastics may end up in soil from the application of sewage sludge to agricultural soils or by direct releases of secondary microplastics from abrasion or maintenance of outdoor plastic items and coated surfaces.

Very limited information on the fate and effects of microplastics in soil has been identified and it would appear that this field has simply not (yet) been studied (Rillig 2012, Lambert *et al.* 2014). The only study of relevance identified is the one by Zubris and Richards (2005) who examined soil and the spreading of synthetic fibre via sewage sludge. Zubris and Richards (2005) found that the fibres were detectable in soil columns more than 5 years after application and they were detectable in field site soils up to 15 years after application. No information was identified, but it has been argued by Rillig (2012) that it is likely that microplastics would have potential adverse effects on soil living organisms, as many soil biota are essentially aquatic and thrive in a thin film of water covering soil surfaces. As with the aquatic environment, soil also has filter feeders that are active on the water films on soil surfaces (ciliates and rotifers) as well as micro- and mesofauna (mites, collembola, or

enchytraeids that could ingest microplastics, thus leading to accumulation in the soil detrital food web<sup>4</sup> (Rillig 2012).

# 2.9 Human exposure to microplastics via the environment

The general population may be exposed to microplastics from different sources, among others:

- Primary microplastics in cosmetics; in particular toothpaste.
- Food items contaminated via the environment e.g. blue mussels and oysters.
- Food items contaminated by manufacturing.
- Indoor dust containing e.g. fibres of synthetic textiles and flakes of paints.
- Dust generated by cleaning and maintenance of plastic items and coated/painted surfaces.

In the working environment, occupational exposure to primary microplastics may take place by use of primary microplastics in manufacturing processes and exposure to secondary microplastics may occur by various processes where plastic items or painted surfaces are cut, ground, crushed, etc.

Aside from possible particle toxicity effects, hazardous substances within the microplastics or adsorbed to the surface may contribute to the dietary exposure of humans to various hazardous substances such as some phthalates or PCBs. The significance of indoor dust (of which some may be plastics) for exposure of particular children has been demonstrated for several hazardous substances and, to some extent, similar mechanisms may be in play with regard to exposure to microplastics associated with hazardous substances from other sources.

No summaries on human exposure to microplastics and the possible health effects have been identified, and it has been beyond the scope of this survey to establish an overview of possible human exposure and health effects.

As described in section 2.3, microplastics may accumulate in marine animals, some of which are used for human consumption. In some animals, the microplastics may be translocated from the gastrointestinal tract and stored in other parts of the animals.

Researchers van Cauwenberghe and Janssen (2014) investigated the presence of microplastics in two species of bivalves commercially grown in Belgium, the blue mussel  $Mytilus\ edulis$  and the oyster  $Crassostrea\ gigas$ . Microplastics were recovered from the soft tissues of both species after a three-day depuration period, which reduced the microplastics content by about one third. After the depuration period, corresponding to the time of human consumption, the blue mussel contained on average 0.36  $\pm$  0.07 particles/g (wet weight), while the microplastics content of the oyster was of 0.47  $\pm$  0.16 particles/g w.w. (detection limit >5  $\mu$ m). The authors estimate that when consuming an average portion of mussels (250 g wet weight) one consumes around 90 particles; the annual dietary exposure for European shellfish consumers can amount up to 11,000 microplastics particles per year.

Similar concentrations were found in another Belgian study, which analysed microplastic contamination in farmed mussels and wild type mussels (mainly *Mytilus edulis*), collected at Belgian department stores and Belgian breakwaters and quaysides, respectively (De Witte *et al.* 2014). The number of total microplastics varied from 0.3 to 0.5 g fibres/g mussel.

A maximum concentration of 105 particles/g dry weight (d.w.) (1-5,000 µm) was reported in wild mussel (*Mytilus edulis*) from the Dutch coast (Leslie *et al.* 2013); recalculated by GESAMP (2015)

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 $<sup>^4</sup>$  In detrital food webs, bacteria and fungi consume detritus from primary producers or consumers.

into a wet weight concentration of 13.2 particles/g (w.w.), more than ten times the levels reported in the Belgian studies.

Microplastics have also been reported in other food items such as beer (Liebezeit and Liebezeit 2014) and honey (DR 2015, Liebezeit and Liebezeit 2013), but it is still unknown whether these food items are contaminated during the manufacturing process (e.g. from plastics filters) or whether the microplastics are present as contaminants in the natural raw materials.

As reviewed by van Cauwenberghe and Janssen (2014), once inside the human digestive tract, intestinal uptake of the ingested particles may occur. Translocation of various types of microparticulates across the mammalian gut has been demonstrated in multiple studies involving rodents (particle size 0.03-40  $\mu m$ ), rabbits (particle size 0.1-10  $\mu m$ ), dogs (particle size 3-100  $\mu m$ ) and humans (particle size 0.16-150  $\mu m$ ). However, contrasting reports exist on (i) the upper size limit of particles capable of being translocated and (ii) the magnitude of this type of transport (Hussain  $\it et~al.~2001$  as cited by van Cauwenberghe and Janssen 2014). According to van Cauwenberghe and Janssen (2014), in current literature there are no data (either from  $\it in~vivo~$  or  $\it in~vitro~$  studies) on the toxicity of translocated microplastics in humans.

The German Federal Institute for Risk Assessment (BfR) recently concluded that it currently does not have any reliable data on the chemical composition, particle size or concentration of microplastic particles in food (BfR 2015). Due to a lack of robust data, a health risk assessment of the consumption of food contaminated with microplastic particles is presently not possible. The BfR has requested the European Food Safety Authority (EFSA) for a scientific opinion on the occurrence of microplastics and particles in the nano range in food, especially in seafood.

The health risk resulting from the use of skin cleansing and dental care products with polyethylene-containing microplastic particles has been evaluated by the BfR in 2014 (BfR 2014). The institute concludes that microplastic particles used in peelings and shower gels are larger than one micrometre and given this particle size, it is not to be expected that foreseeable use of the products lead to absorption via healthy and undamaged skin. Microplastic particles from toothpaste can inadvertently be swallowed and thus ingested orally. Due to their size, it is not to be assumed that the particles are absorbed via the gastrointestinal tract. Instead, they are much more likely to be excreted with the faeces. It is unlikely that during their passage through the gastrointestinal tract, toxicologically relevant amounts of ethylene are released from the microplastic particles. Based on the current state of knowledge, the BfR therefore concludes that the use of cosmetic products containing PE microplastic particles does not pose a health risk to consumers (BfR 2014).

GESAMP (2015) concludes in their review on microplastics: "Although it is evident that humans are exposed to microplastics through their diet and the presence of microplastics in seafood could pose a threat to food safety (Van Cauwenberghe and Janssen 2014), our understanding of the fate and toxicity of microplastics in humans constitutes a major knowledge gap that deserves special attention. Therefore, an analysis and assessment of the potential health risk of microplastics for humans should comprise dietary exposure from a range of foods across the total diet in order to assess the contributing risk of contaminated marine food items."

# 3. Review on use of primary microplastics and sources of microplastics to the environment

This chapter includes a review of the current knowledge on uses of microplastics and sources of microplastics to the environment. The information serves as background for the assessment of the use of primary microplastics in Denmark and the estimates of releases of microplastics from various sources in Denmark in Chapter 5. For some applications of primary microplastics and some sources of secondary microplastics, limited information has been identified in the literature, and for these applications, the description is supplemented by information collected as part of this study.

# 3.1 Use of primary microplastics

# 3.1.1 Introduction

An overview of uses of primary microplastics identified in the literature and pathways of releases of primary microplastics to the environment are shown in Table 10.

**TABLE 10**OVERVIEW OF SOURCES AND PATHWAYS FOR RELEASES OF PRIMARY MICROPLASTICS TO THE ENVIRONMENT (PARTLY BASED ON SUNDT ETAL. 2014; ESSEL ETAL. 2015)

Source group	Description	Potential environmental release pathways
Raw materials for production of plastic items (plastic pellets)	Losses of plastic pellets by transport, reloading, processing, etc.	Surface water Municipal sewage Industrial sewage Urban run-off
Plastic particles used for cosmetics	Exfoliating beads in e.g. face and body wash. Released by washing	Municipal sewage
	Microbeads for colour or glitter effect e.g. in toothpaste	Municipal sewage
Plastic particles in abrasive media	Plastic blasting grit for <u>outdoor</u> applications e.g. "plastic blasting" at shipyards, offshore maintenance, bridges, etc. Released by drift (via air) or by cleaning of surfaces	Surface water Municipal sewage Industrial sewage Urban run off
	Plastic blasting grit used <u>indoors</u> at different production facilities, e.g. plastic production. Released by cleaning of surfaces	Municipal sewage
Plastic particles used in paints	Plastic particles in water-based paint. Released by washing of brushes and other tools.	Municipal sewage

Source group	Description	Potential environmental release pathways
	Plastic particles in organic solvent-based paints. Under certain circumstances released by washing of brushes and other tools	Municipal sewage
	Plastic particles to be mixed with paints (e.g. antiskid powder). May be released by washing of brushes and other tools	Municipal sewage
Rubber granules and powder from recycling of tyres	Rubber granules used for artificial turfs, running lanes, playgrounds, etc.	Soil Sewage
EPS beads used for other applications than plastics produc- tion	EPS beads used in furniture, pillows, hollow wall insulation, etc. Released during transport, reloading, application, demolition of houses, tear of furniture, etc.	Soil Municipal sewage Urban run-off
Plastic particles used in medical products and in research	Vectors for active pharmaceutical ingredients. Widely used for a range og medical and biological applications	Not considered a signifi- cant source of environ- mental releases
Plastic particles used in other products	Plastic beads used in professional dish washing machines Various plastic beads and ironing beads used by children Printer toner	Sewage

# 3.1.2 Primary microplastics in personal care and cosmetics

# **Application**

Primary microplastics in personal care and cosmetic products is a minor source of releases of microplastics to the environment, but as use in cosmetic products has attracted much attention in recent years, the use is described here in relatively high detail. Very recently, and after the drafting of this section, UNEP published a report on microplastics in cosmetics (2015). To the extent possible, the UNEP report's new information has been added to this section.

Polymers have been applied as ingredients in personal care and cosmetic products for several decades with early patents dating from the 1960s (as cited by Leslie 2014). Though patented for use in cleansers already in 1972, for decades, plastic microbead abrasives were rarely used in consumer products and were considered only a minor source of plastic pollution (Zitko and Hanlon 1991). Starting in the 1990s, manufacturers of consumer products began to replace natural materials such as ground almonds, oatmeal and sea salt in personal care products with plastic microbeads more extensively (Zitko and Hanlon 1991)

The polymer materials applied as ingredients in personal care and cosmetic product formulations include both thermoplastics (e.g. polyethylene, polypropylene, polystyrene and polytetrafluoroethylene) and thermosets (e.g. polyester and polyurethanes).

As reviewed by Leslie (2014), polymers are made up of mixtures of macromolecules of different chain lengths and thus different molecular weights. The physical form of the polymers depends on the chain length and the branching of the chains. As an example, PE molecules up to about 700 carbons chain length are waxy solids, and alkane chains with less than 20 carbons are liquids or gases (Peacock 2000 as cited by Leslie 2014).

Polymer waxes (e.g. polyethylene waxes), which are macromolecules that result from shorter chain lengths, are popular gellants for personal care and cosmetic products and are available as powders, flakes or granules (Cosmetic Ingredient Review 2007 as cited by Leslie 2014). Polyethylene waxes are nondegradable, water insoluble, solid materials with melting points well above maximum sea temperatures and fall under the definition of microplastics as defined by Leslie (2014). As discussed elsewhere, the waxes are not included as microplastics in the current report and waxes in cosmetics are not included in the quantities of the consumption of microplastics in cosmetics products and the releases from the use of these products.

Longer polymer chains produce materials that are more rigid, e.g. poly(ethylene terephthalate) glitters or styrene/acrylates copolymer colour spheres. Increasing the chain length of ethylene oxide polymers to 20,000 carbons results in solid materials. Cross-linking tends to decrease water solubility of polymers, e.g. 'water-dispersible' polymers known as 'microgels', which have been described as 'soft, clear, round, water-logged microscopic sponges' (Gruber 1999, as cited by Leslie 2014). Plastic particles (i.e. with a distinguished form) in personal care and cosmetic products are designated throughout this report as 'plastic microbeads' to distinguish them from other uses of polymers in the products and microbeads of natural origin.

Polymers in personal care and cosmetics products are named in accordance with the International Nomenclature of Cosmetic Ingredients (INCI) and indicated in the ingredients list of the marketed products. INCI names and functions of the polymers in personal care and cosmetics products formulations are further described in Appendix 1 - Content and size distribution.

Relatively few actual analyses of plastic microbeads in cosmetics have been reported. Results for plastic beads in toothpaste, scrubs, hand cleaning and shaving foam are shown in Table 11. The reported levels of PE microbeads in scrubs range from 0.4 to 10.6% with a size range from 40 to 1000 µm. These results are generally consistent with a U.S. patent on the use of micro-plastic beads as scrubbers in cosmetic products, which suggests that beads  $<60 \mu m$  are generally ineffective as scrubbers, with the ideal size being in the region of 420 µm (U.S patents cited by Gouin et al. 2015). Varying results have been obtained for toothpaste. In a Danish study, the analysed toothpastes contained plastic microbeads in the size range 40-800 µm (median diameters of 200-300 µm), similar to the sizes found in scrubs, whereas a Dutch study found the particles to be in the range 2-5 μm (Verschoor et al. 2014a). In the two Dutch toothpastes, 91% and 97% of the microbeads, respectively, were below 10 µm in diameter. The reported concentration in the Dutch study of 1.9-3.9 was ten times higher than reported in the Danish study. It should be noted that smaller particles may be present in the Danish study because test sieves of 38 µm were used (Strand 2014). Small-sized microbeads in the range of 5-15 µm have also been mentioned in US patents for shaving foam (as cited by Sundt et al. (2014)). The small particle sizes found in the Dutch study of toothpastes are lower than the sieve sizes used in studies of the fate of microplastics in sewage treatment plants (as discussed in section 4.1) and the retention efficiency for these small particles in sewage treatment plants is still poorly understood.

Cosmetic products with plastic microbeads may contain several hundred thousands of plastic particles. A study from the USA estimated that a facial scrub tube contained approximately 350,000 plastic particles (5-Gyres 2013) while a Dutch study found on average 1,700 and 6,400 plastic particles per g toothpaste in two toothpastes, respectively (Verschoor *et al.* 2014a).

In a Danish study of nine products, the dominant colours were blue and white (in many products both were present), whereas one product contained white and green plastic particles (Strand 2014). In the investigated products in a German study the beads were blue and white spherical particles (Hintersteiner *et al.* 2015).

As an example of a commercial product, DupontTM Gotalene® Exfoliating Micropowders for personal care are recommended in concentrations of 3-10% (Dupont 2009). The technical pamphlet indicates that 3% is the minimum percentage for an efficient scrubbing effect. The microbeads are available in different grades for mild and medium abrasive properties. Particles sizes of the different grades are indicated as <200  $\mu m$ , <315  $\mu m$ , <400  $\mu m$ , <500  $\mu m$  and <630  $\mu m$  (Dupont 2009).

TABLE 11
CONTENT OF PLASTIC MICROBEADS IN ANALYSED PERSONAL CARE PRODUCTS

Product type (as indicated by reference)	Weight % plastic mi- crobeads	Size of particles, µm	Plastic type	Number of samples	Country	Reference
Toothpaste	0.1-0.4	40-600*	PE	3	Denmark	Strand 2014
Toothpaste	1.4-5	2-5	PE	2 (with mi- crobeads)	The Netherlands	Verschoor <i>et al.</i> 2014a
Toothpaste	0.3	>100 (blue spherical) >10 (white spherical)	PE	1	Germany	Hintersteiner <i>et</i>
Facial scrubs	1.6-3.0	100-200	PE	3	New Zealand	Gregory 1996
Facial scrubs	0.9-4.2	not reported	PE	3	USA	5Gyres 2013
Facial cleaner	not report- ed	4-1240 (in three of the products the mean was <100)	PE	4	New Zealand	Fendall and Sewell 2009
Scrubs	0.4-10.5	40-800*	PE	6	Denmark	Strand 2014
Exfoliating scrub	10.6	100-1000	PE	1	The Nether-lands	Leslie 2012
Anti-callus scrub	0.6	>200 (blue spherical) >100 (white spherical)	PE	1	Germany	Hintersteiner <i>et</i>
Hand cleaning	0.2-6.9	100-1000	PE	3	New Zealand	Gregory 1996
Shaving foam	0.1-2.0	5-15	PFTE	not reported	USA	US patents referred to in Sundt et al. 2014
Peeling	2.1-7.5 **	>200	PE	3	Germany	Hintersteiner <i>et</i> al. 2015
Shower gel	0.5-3.0	>300 (blue elongated) >100 (white spherical)	PE	2	Germany	Hintersteiner <i>et</i> al. 2015

<sup>\*</sup> Smaller particles may be present but not recorded because of the use of 38-µm test sieves (Strand 2014).

# Consumption reported in the literature

According to a recent survey by the European Cosmetic Industry Association (Cosmetics Europe) and major market actors, plastic microbeads used in cosmetic products in the European market are largely dominated by their use in niche consumer products, particularly in liquid soaps that make a claim of an exfoliate function (Gouin *et al.* 2015). In estimating the usage of plastic microbeads used in cosmetic products, the survey utilized two complementary approaches. The first approach was based on feedback to a user questionnaire received from companies that are members of Cosmetics Europe. The information obtained from the survey provided no geographic or company specific details, but rather provided an estimate of the relative use of plastic microbeads used in cosmetic

<sup>\*\*</sup> Results obtained by weighing the dried residues after twofold density separation. Lower values were obtained by GPC measurements.

products within the European market. The second approach attempted to assess usage spatially, utilizing data reported by Euromonitor International, which is a consumer products database that reports market data of various product types.

The Cosmetics Europe survey was focused on products marketed in the European Union, Norway, and Switzerland. Members were requested to provide the INCI Name of the plastic microbeads used, the particle size, and particle shape (i.e. sphere, fragment, fibre) for the year 2012. For the purposes of the survey, participants were asked to provide information on petroleum-based microplastic beads in cosmetic products of any size <5 mm, and used for any purpose in the cosmetic formulation (Gouin *et al.* 2015). PE plastic microbeads were reported to be the dominant type of plastic material used as an exfoliating agent in skin cleansing products, with a total reported amount of 4,073 tons used in European countries, including Norway and Switzerland. The total amount of plastic microbeads other than PE was 287 tons. Typical inclusion levels of PE microplastic beads reported in the various products ranged between 0.05% and 12%, consistent with the results shown in Table 11. The survey results also found that approximately 70% of micro-plastic beads used was >450 µm, based on the size distribution of PE plastic microbeads for which size data were reported (660 tonnes) in the Cosmetics Europe survey. Given the documented evidence that the efficacy of plastic microbeads as an exfoliating agent is significantly reduced with decreasing size, the authors expect limited use of plastic microbeads <450 µm.

Using the Euromonitor data, estimates of the volumes of plastic microbeads contained in shower gels, face wash and liquid hand soap were derived (Gouin *et al.* 2015). Based on the authors' analysis of the data, it was estimated that 6% of all liquid soaps contained plastic microbeads in 2012. The 6% is indicated as a "conservative" estimate (i.e. likely to be too high). Assuming a maximum inclusion level of 10% polyethylene beads, it was estimated that plastic microbeads accounted for 0.6% of the total volume of skin cleansing products sold. The total estimated content all for countries within the European Union plus Norway and Switzerland was 4,130 t/year plastic microbeads. This figure is consistent with the data reported in the Cosmetics Europe survey of 4,360 t/year. The reported total consumption in Europe is also consistent with an estimate of 4,000 t/year (the year not indicated) reported by the trade association Plastics Europe, which represents the plastics manufacturing industry in Western Europe, to the Norwegian assessment of releases of microplastics (Sundt *et al.* 2014).

The *per capita* consumption based on Euromonitor data varied by country by a factor of three with an average for EU Member States plus Norway and Switzerland of 17.5 mg/day, corresponding to 6.4 g/year. Based on the Euromonitor data, the consumption in Denmark is estimated at 29 tonnes in 2012 (Gouin *et al.* 2015), which is further discussed in section 1.1.1.

A recent German assessment (Essel *et al.* 2015) estimated the total *per capita* consumption at 6.2 g/year distributed across shower gels and liquid soaps (1.9 g/year), cleansers for body care (2.2 g/year), skin-care and sun protection products (0.5 g/year), dental hygiene products (1.2 g/year) and other body-care articles (0.4 g/year). This estimate was not based on Gouin *et al.* (2015) but rather on independent estimates derived from German figures.

In the USA, Gouin *et al.* (2011) estimated the *per capita* consumption of PE microbeads in cosmetic products of approximately 2.4 mg/d corresponding to 1 g/year. This estimate was based on general sales volumes for hand wash in the USA and some general assumptions on market share of the microbead products and their content.

**Ecolabels** - The Nordic Ecolabelling board decided on 11 December 2013 that microplastics should be added to the list of prohibited substances in cosmetics products awarded the Nordic Swan ecolabel. The microplastics is defined as "...undissolvable plastic particles of less than 1 mm size and not biodegradable according to OEC 301 A-F" (Swan, no date).

The European ecolabel (the flower) ecological criteria for "rinse-off cosmetic products" of December 2014 defines that microplastics are excluded from products awarded the ecolabel (COM 2014). The microplastics are not further defined in the criteria.

# Trend in consumption

The European industry association Cosmetics Europe decided in 2012 not to support the use of micro plastic beads in cosmetics and urged their member companies to take this into consideration in their CSR policy. In recent years, several of the major European manufacturers of cosmetics have phased out the use of microplastics and many companies have announced that they are in the process of phasing out the microbeads, as also illustrated in Table 35 in Appendix 1. The trend in consumption is downward.

# 3.1.3 Primary microplastics as raw materials for plastics production

Lost plastics pellets for production of plastic items have been reported from the marine environment all over the world and are systematically used for monitoring of persistent organic pollutants as described in section 2.5. In some studies, their presence in the environment has been assigned to a single plastics manufacturing point source nearby (e.g. Fabbri *et al.* 2000; Norén 2007; 2010; Lechner and Ramler 2015). A recent study of microplastics in the river Danube has demonstrated that industrial raw material (pellets, flakes and spherules) accounted for 79.4% of the plastic debris in the water.

# Application

Plastics can be divided into two types: thermoplastics and thermosets.

<u>Thermoplastics</u> do not undergo chemical change in their composition when heated. During conversion of thermoplastics compounds into finished items, the materials are shaped by heat and pressure using moulding, extrusion and other processes. Examples of thermoplastics include polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and polytetrafluoroethylene (PTFE). The thermoplastic raw materials are typically pellets of a size of 1-5  $\mu$ m or powder of smaller sizes (here collectively designated as preproduction plastic pellets).

<u>Thermosets</u> can melt and take shape once; after they have solidified, they stay solid. In the thermosetting process, a chemical reaction with the establishment of a tight crosslink between the plastic molecules occurs that is irreversible. Examples of thermosets include polyurethanes (PUR), unsaturated polyesters, phenolics and epoxy resins. The raw materials for thermosets are typically liquid.

The smaller sized raw materials are used for specific conversion processes; in particular rotational moulding and sintering of PTFE powder. The typical size of particles for rotational moulding is 150-500  $\mu$ m and PE represents about 85-90% of the plastics used for rotational moulding (ARMA 2015). Batches of pellets might also contain finer plastic dust as contamination from raw material production or created during transport and transfer (Dhodapkar *et al.* 2009).

# Consumption reported in the literature

The total consumption of plastic raw materials in the EU in 2013 was 57 million tonnes (PlasticsEurope 2015). The thermoplastics PE (different types), PP, PS, PVC and PET accounted for the major part of the total consumption as illustrated in Figure 11. About 40% of the demand was for packaging while building and construction accounted for 20% of the total demand. Besides the potential losses of plastic raw materials to the environment, the use of plastics leads to releases of macroplastics to the environment (in particular packaging materials) and to abrasive releases of microplastics from the plastic items (in particular, building materials as discussed in Section 5.2.6).

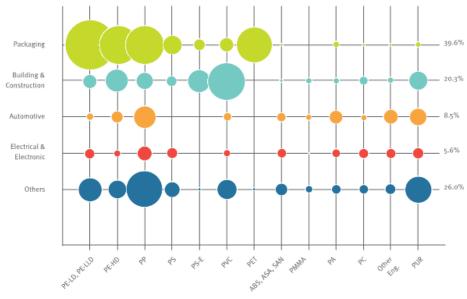
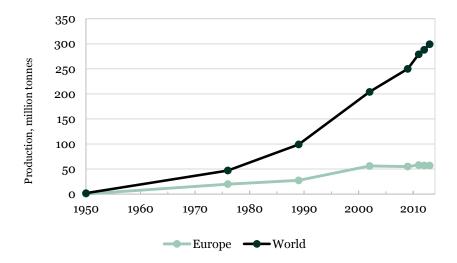


FIGURE 11 EUROPEAN PLASTICS DEMAND BY SEGMENT AND POLYMER TYPE 2013 (PLASTICS EUROPE 2015). REPRODUCED WITH PERMISSION FROM PLASTICS EUROPE.

# **Trend in consumption**

While the plastics production in Europe has been stable during the period 2002 to 2013, the global production of plastics has increased from 204 million tonnes in 2002 to 299 in 2013 ( Figure 12). The increasing trend in the global production and use of plastics is of concern as it potentially may lead to increasing releases of microplastics preproduction pellets and macroplastic items to the global environment. As the increase in production and use is occurring in countries without the same level of waste management as in Europe, the trend in pollution of the marine environment with plastics may even be steeper than the increase in the production.



 $\begin{array}{l} \textbf{FIGURE 12} \\ \textbf{TREND IN PLASTICS PRODUCTION IN EUROPE AND THE WORLD (BASED ON DATA FROM PLASTICSEUROPE 2013)} \end{array}$ 

# Release pathways and reported releases

The release pathways of preproduction pellets are described in the Plastic Pellet Loss Prevention Guidance Manual of the Operation Clean Sweep, which is an international programme designed to prevent resin pellet loss and help keep pellets out of the marine environment (Operation Clean Sweep 2010; 2015). According to the manual, there are many steps involved in the movement of

plastic pellets from the resin production facility, through the distribution network, to the processor. Spills and pellet loss to the environment can occur at any step.

The manual provides guidance for loss prevention from pellet transport/packaging, unloading hopper railcars and lorries, bagging and boxing, palletizing, "other transport vehicle concerns" and marine transport. The manual does not indicate the level of losses from the different steps and to what extent these losses can be prevented by the proposed procedures. To date, however, no figures have been published on the success of Operation Clean Sweep and the quantities of microplastics that had been prevented from entering the environment because of the project. Some information on the release levels which can be obtained is provided in section 5.1.2 based on information collected by the Danish Plastics Federation from companies which have signed commitments to the Operation Clean Sweep.

Norway - An assessment of releases of microplastics in Norway (Sundt et al. 2014) makes reference to a worst case emission factor of 0.5% for transport losses of solid powders from the US EPA/OPPT Dust Emissions from Transferring Solids Model used by the OECD emission scenario document on adhesive formulation (OECD 2009a). The model concerns "solid powder", not further specified, but likely of smaller particle size than the larger pellets mainly used in the manufacture of plastics items. The Norwegian report notes that the emission factor applies to transport inside or outside the production line. In addition, there would certainly be spills from the production line itself through the effluent system and even past best available control and treatment measures. The authors have extracted an emission factor for this problem from real data from a Norwegian polystyrene plant during the last decade where the spill was 0.4 grams per kg produced (=0.04%). Using these worst case emission factors without any control measures, the releases from the transport of plastics in Norway would be about 2,500 t/y. The authors note that there is no exact information on the effect of spill control measures on the transferring processes, but expect these to be in place in a large proportion of the industrial sites, hence they are adjusting the estimate down to 250 t/y; of this, 22 t/y is assumed to be released to the sea. The losses from the Norwegian production sites are estimated at 200 t/y using an emission factor of 0.04%; of this, 180 t/y is expected to end up in the sea and 20 t/y released to sewage. Spill from accidents is indicated as "not assessed".

**Germany** - A recent German assessment assumes that the pellet loss account for 0.1 to 1.0% of total plastics production and estimates the loss of microplastics from the German plastics industry at 21,000-210,000 t/year (Essel et al. 2015). It is not indicated to what extent the loss is to waste water or the aquatic environment.

**Significance of the source** - Both the Norwegian and the German assessments identify pellet loss to be the major source of releases of primary microplastics, far beyond the releases from microplastics in cosmetics.

A recent study of microplastics in the river Danube has demonstrated that industrial raw material (pellets, flakes and spherules) accounted for 79.4% of the plastic debris in the water (Lechner *et al.* 2014). The plastic input via the Danube into the Black Sea was estimated to 4.2 t per day. If 79.4% of this originated from plastic raw materials, the total quantity transported by the river would be some 1,200 t/year. Lechner *et al.* (2015) report that one manufacturer of plastics raw materials, according to the company's own reporting, discharged 0.2 kg/day to the Danube River under normal operating conditions but 50-200 kg was lost during a heavy rainfall event. The releases were far below the applicable legal limits and the company may legally discharge up to 94.5 t/year of plastics (Lechner *et al.* 2015).

## Release of hazardous substances

The lost pellets may contain environmentally hazardous additives such as some flame retardants or plasticizers and thus serve as a source of environmental pollution. Sometimes, additives are already added to preproduction pellets (and may be lost to the environment with the pellets), whereas other

additives may be added in the conversion step, when the plastic is being processed into the final item (and therefore not lost with the pellets). It is beyond the limits of this survey to establish an overview of which additives may be present in the preproduction pellets and lost to the environment with the pellets.

# **3.1.4** Primary microplastics for use in paint Application

Primary microplastics may be added to paint and wood preservatives to provide surface effects (such as matting effects) or as a colour enhancer. Apart from the use of waxes described by Essel *et al.* (2015), limited information has been identified on this application in the literature on microplastics. Information on the use of microplastics and waxes in paint and wood preservatives has been obtained from the Danish Coating and Adhesives Association (DFL) and from product literature from manufacturers of the polymer particles for paints. Details on quantities used in Denmark are provided in section 0, whereas this section includes a general description of the applications.

The following main applications of microplastics in paint and wood preservatives have been identified:

- To decrease the density of the paints
- Pigments extenders/spacers (titanium oxide spacers)
- To improve the hardness and provide greater durability and scratch resistance
- To apply structure (mainly waxes are used for this application)
- To provide "pop-up" effects
- To provide glitter effects (similar applications in cosmetics and inks for textiles)

In order to exemplify the applications, information from manufacturers' websites of products used in the production of paint in Denmark are provided below.

Expancel® expanded microspheres are lightweight fillers of very low density (Akzo Nobel 2015). The microspheres consist of a thermoplastic polymer shell encapsulating a gas (polymer type not indicated). When heated, the internal pressure from the gas increases and the thermoplastic shell softens, resulting in a dramatic increase of the volume of the microspheres. It is used as filler for paint, inks and various plastics applications. The microspheres are available in different sizes between 20 and 120  $\mu$ m after expansion (Boud Minerals 2015). In paint, the microspheres are used to decrease the density of the paint, improve the applicability, and give higher water vapour permeability, matting effect, and low emission of VOC (Akzo Nobel 2015).

ROPAQUE<sup>TM</sup> Opaque Polymer products are water-based styrene/acrylic copolymer pigment formulations used to replace a portion of  $TiO_2$  in paint (DOW 2015). The average particle size is 0.4  $\mu$ m, and the majority of the particles are just below 1  $\mu$ m and thus outside the  $\mu$ m-range. Orgawhite 2000, with the same function, has similar average particle sizes (OrganikKimya, 2015).

An example of the various applications of polymer particles used for structured paint is the application range of Coathylene® Polymer Powders (Axalta, 2013). The powders are provided in different grades with maximum grain size ranging from 35 to 400 µm and made from PP, HDPE, LDPE and polylactic acid (PLA). The typical applications of the powders are as anti-slip agents, for structural effects and satin and matte finishes. The application areas for the paints are road marking; out-door/indoor structured paint; roofs and underbodies for cars; wood & metal furniture; metal casings; electrical boxes; marine paint, ship decks; swimming pools and surroundings; garage floors and other heavy duty industrial flooring. The low specific gravity of the particles allows them to migrate to the surface of the painted area. Therefore, in general, the addition level is between 2 and 10% based on the dry content of the paint or varnish. It is indicated that the particles are added to the paint by simply stirring into the liquid paint and that the particles have no modification or reaction with paint formulations.

Geoshine® glitter is an example of particles used for glitter effects cut from 12 and 25  $\mu$ m brilliant metallized polyester foils (Geotech, 2015). The Geoshine® range is available in 36 different colours. Each colour is available in 12 different particle sizes from 100  $\mu$ m up to 3 mm. The glitter offers high brilliance and reflection. The particles are used in a wide variety of applications: cosmetics, automotive coatings, inks, flooring, textiles and decoration. The products are light fast, water and solvent resistant and heat-stable up to a temperature of 180 degrees Celsius. Geoglit® is a special range of polyester glitter used for all kinds of decorations such as crafts and Christmas decorations.

# Consumption reported in the literature

No specific data on the consumption of microplastics in paints and wood preservatives have been identified in the literature. The application is mentioned in the Norwegian and German assessment reports. Paints, lacquers and dyes rank high in a Dutch "Quick scan and Prioritization of Microplastic Sources and Emissions" as consumer use of these products is often accompanied by a relatively large emission of polymers, but this concerns rather the releases of secondary microplastics from maintenance of painted products (Verschoor *et al.* 2014b).

# Trends in consumption

No data on the trend in consumption of microplastics in paint have been obtained.

# 3.1.5 Blasting abrasives

### **Application**

Primary microplastics may be used as a blasting abrasive for cleansing of surfaces. Microplastics may be used as an alternative to other blasting agents, such as sand, corundum and steel grit, when a more gentle procedure is needed in order not to damage the surface, e.g. for removing paint from aluminium components of aircrafts (Miles *et al.* 2002).

The application of plastics blasting abrasives have been mentioned in published assessment reports from Norway and the Netherlands (Sundt *et al.* 2014; Essel *et al.* 2015; Verschoor *et al.* 2014b), but the products and applications have not been described in detail. The following is mainly based on information from suppliers of plastics blasting abrasives in Denmark.

Plastic blast media may comprise different types of plastics such as urea, melamine, acrylic, polyester, polyamide, polycarbonate and polyurethane. Commercial plastic granulates for sandblasting are available in sizes from 0.012 – 2.03 mm based on products available on the Danish market. The plastic granulates may be re-used between 10 and 50 times.

The applications for the plastic blast media are removal of paint from aircrafts and their components, for deburring of plastic and light metal parts, for car maintenance (e.g. cleansing of rims), cleansing of tanks used in the offshore industry and in the marine industry, cleansing aboard ships and cleansing of turbine blades at power plants. Plastic blasting media have also been used for building sanitation and for graffiti removal.

There are different commercial products that use microplastics for sandblasting, available on the Danish market. One of these is "Sponge Jet". Sponge jet consists of small sponges made of polyure-thane reinforced with different types of materials, depending on the application. These materials are steel grit, aluminium oxide, and plastic chips. The polyurethane sponges may also be used without further reinforcement. The size of the material is  $<12-125~\mu m$ . After cleansing of the collected material, the sponges can be reused approximately 10-15 times.

Another commercial product is Dry Strip®, which is a granulated amino resin that comes in two different qualities: one approved for use in the air transport industry and one for other industrial uses. The product is available in four different types (polyester resin, urea resin, melamine resin

and acrylic), and the size of the particles is in the range of 0.6-1.2 mm. According to the importer, Dry Strip® is only used in the air transportation industry.

Other commercial products imported to Denmark are JETplast®, which are granulates made of urea combined with melamine and phenol (1.0–1.6 mm) and granulates made of urea (0.6-0.8 mm), as well as products from the German company Eisenwerk Würth GmbH (made of either melamine resin, urea resin, polyamide or polycarbonate) in sizes from 0.15 – 2.03 mm.

Another Danish importer of synthetic granulates for use as blast media specifies that they import three types of granulates: urea, melamine and acrylic. The size of the granulates is >0.3 mm, and the material can be reused up to 50 times, as long as the granulates are larger than 0.3 mm.

# Consumption reported in the literature

No information on use volumes was given in either the Norwegian or the German assessments. In this report, the use of microplastics in blasting activities in Denmark has been investigated further, and the results are further elaborated in section 5.1.4.

# 3.1.6 Rubber powder and granules for artificial turfs and other applications

The diameter of rubber granules from recycling of tyres varies between 0.7 and 3 mm (Genan, 2015), which thereby classifies the rubber granules as primary microplastics as defined in this report. The applications have not been described in detail in existing surveys of microplastics.

The granules are used as infill for artificial turfs for football, rugby, tennis and golf fields. The application is described in further detail in section 5.1.5. Combined with a binder, the granules are also used for playgrounds, athletic tracks and similar applications. Rubber granulate is mixed with polyurethane, and the playground or the track is built on the spot (Genan, 2015). Rubber granules are furthermore used an elastomer bitumen and asphalt modifier in concentrations of about 10% (Genan, 2015). Rubber granules are added to roads because it makes the roads less inclined to rutting and cracking.

Wear of artificial turf fields and other areas with rubber coating will release rubber granules used in the fields and, furthermore, secondary microplastics will be formed and released from synthetic grass fibres in the turfs. Sundt et al. (2014) suppose that the rubber granules are collected if spread outside the field and do not include the rubber granules in the inventory of releases of microplastics in Norway. Another conclusion is reached for Denmark, as further described in section 5.1.5.

# 3.1.7 Primary microplastics in other applications

Primary microplastics may also be used in other areas of application, such as for cleaning and maintenance products, active pharmaceutical ingredients, professional dishwashing machines, and as ingredients in drilling fluids used in the oil and gas industry. Several uses in technical applications as micronized synthetic waxes have also been identified by Essel *et al.* (2015). As part of this survey, websites of manufacturers of plastics applied in microsize in mixtures and articles have been reviewed in order to identify applications beyond those described in the literature.

These applications are further elaborated in the following sections.

# Microbeads of expanded polystyrene (EPS) in furniture and for insulation

The majority of plastic pellets of expanded polystyrene (EPS) is formed into blocks or shape moulded for use in construction, packaging and few other applications. A small portion of the pellets is used directly without being formed into blocks. The pellets are typically 1-5 mm in diameter and thus considered microplastics in this context. The pellets are used for furniture (beanbags), pillows and as hollow wall insulation. For these applications, both primary pellets and particles of shredded, recycled EPS are used.

For insulation purposes, the pellets are to some extent used by professionals for hollow wall insulation of old houses and bags with EPS bead are sold in DIY (do it yourself) stores for DIY applications.

If released from articles or by application as an insulation material, it is assumed that the majority is collected as waste either manually or by use of a vacuum cleaner. A small part may be released to sewage by cleaning with water or, due to the low density, released to the surroundings and spread by the wind. Small secondary microplastics particles generated by cutting of EPS block may be released by the same pathways. No data on total consumption or possible emission factors have been identified.

# Cleaning and maintenance products

It seems likely that microplastics to some extent may be used as abrasive materials in some cleaning and maintenance products with a similar function as they have in cosmetics. According to the German survey (Essel *et al.* 2015), in the German cosmetics industry association, the member companies do not use microplastics in these products. The German assessment concludes that it cannot be ruled out that other manufacturers of detergents, cleaning and maintenance products use plastic abrasives. The Danish Association of Cosmetics, Toiletries, Soap and Detergent Industries has been requested for information on microplastics in cleaning and maintenance products. The only product indicated was hand cleaners used by professional and consumers for removing grease, oil, paint, etc. from hands.

# Plastic beads and ironing beads

This use is not described in the literature. Some types of small ironing beads used by children to form pictures and patterns, and some plastic peals used by children and adults may fall within the definition of microplastics. The size of ironing beads is typically about 5 mm, whereas plastic peals may be slightly smaller. Ironing beads sold in Denmark in 2002 were made of PE and ethylenevinylacetate copolymers (EVA) (Pors and Fuglendorf, 2002). As the size is just at the limit of the size range and releases to sewage are not considered a significant disposal route (although it cannot be ruled out that it might happen by floor washing), these applications have not been investigated further.

# **Medicine and research**

According to Essel *et al.* (2014), microplastics are used in the medical sector as well. Hollow particles may be used as a carrier for active pharmaceutical ingredients, where the active substances are placed in the hollow interior and slowly diffuse through the body. Microplastics are also being considered to treat reflux – the backflow of gastric acids into the oesophagus due to weak sphincter muscles. Today, preparations containing aluminium hydroxide and magnesium hydroxide gels, or mixtures of calcium and magnesium carbonate or aluminium magnesium silicate hydrate are being used in the treatment. Hydroxides, carbonates and silicate hydrates help to buffer the gastric acids, thus alleviating their acidic effects. Microplastics may be used as an alternative to aluminium-containing products, due to the potential toxic impact of the aluminium compounds, and the application of microplastics in medicine may increase in the future.

Polymeric microbeads are widely used in medical and biological applications as carriers, such as in immunoassays and cell separation, in site-specific drug delivery systems, in nuclear medicine for diagnostic imaging, in studying the phagocytic process, in affinity separation of biological entities, etc. (Piskin *et al.* 1994).

However, there are no detailed studies available on either the use quantities or the possible entry pathways into the environment of microplastics used in pharmaceuticals. Further assessment of this application area, which is assumed to be rather small and mainly dealing with processes with limited releases to the environment, has not been undertaken.

# Specialty chemicals in oil and gas industry

According to Sundt *et al.* (2014), drilling fluids based on microbeads have been used in oil and gas exploration for decades, and new products such as PTFE strengthened particles have been patented and marketed heavily for drilling purposes internationally. Depending on the classification of these types of chemicals, releases directly to the oceans are possible (i.e. if they are regarded as green chemicals). Recovery and waste collection of the muds and drilling fluids does happen; however, the treatment processes have not been designed for handling plastic particles. The use of microbeads in specialty chemicals used in the oil and gas industry in Norway has not yet been documented (Sundt *et al.* 2014).

# **Printer toner**

According to Sundt et al. (2014), laser printer toner to a large extent consists of microscopic thermoplastic powder. The polymer diameter of the plastic particles is about 2- 10  $\mu$ m and thus falls within the definition of microplastics as applied here. Usually the powder is a styrene acrylate copolymer. The powder is melted onto the paper when printing. Spill of toner products will therefore add microplastic particles to the indoor environment which may eventually be discharged to the sewer. No data on the amounts used or releases have been identified in previous surveys.

# **Expandable microspheres**

As mentioned under paints in section 3.1.4, Expancel® expandable microspheres are used for various applications (Akzo Nobel, 2015). Product data sheets indicated the following examples of applications: Shoe soles, wine stoppers (plastic and agglomerated), wood plastic composite, wallpaper, silicone rubber, underbody coatings, leather, polyester putty, and model marking board. It is assumed that in these applications, the microspheres are embedded in a cured matrix and not released from products as free micro particles. As described for other plastic raw materials, some releases may take place by production, loading and transport, but the total releases of the speciality microspheres are expected to be small compared to the total releases of plastic raw materials.

# Polymer powders used in construction industry

Dispersible polymer powders are powdery products widely used in the construction industry e.g. in cementitious dry mortars. As described by one of the main suppliers (Wacker 2015), the powders are produced from dispersions which usually have a particle size of 0.1 to 5  $\mu$ m by spray drying which separates the dispersion particles (~1 μm) in the powder particles (~100 μm) from one another and prevents the polymer particles from irreversibly forming a film during drying and storage. The spray-dried powder is treated with antiblocking agent, which prevents the powder particles sticking together during storage. As soon as the dispersible polymer powder comes into contact with water, the dispersible polymer powder grains disintegrate and release the individual dispersion particles again. The dispersed polymer powder has the same properties as the original dispersion, creating the right binders for better building. In the hardened cement, the dispersion particles form a bridging polymer film and so act as an additional binder. The term used is "improved cohesion". At the same time, they form bridges to the substrate, thus improving the adhesion of the mixed mortar to the substrate (Wacker 2015). Even the polymer powder particles may be considered microplastics when they are applied; they disintegrate when coming in contact with water and are not considered a significant source of releases of microplastics, and consequently not considered further.

# Plastic beads for commercial potwashing

According to Sundt *et al.* (2014), plastic beads for commercial dishwashing ("Power granules") are supposed to be used on ships. An example of these granules are those from GRANULDISK (2015), optimised to be used in GRANULDISK potwashers, some of these for marine applications. According to the manufacturer, the granules are made from polyoximetylen, a copolymer with a diameter of 3 mm and reused for up to around 2,500 - 3,500 cycles. The manufacturer indicates that it may

be necessary to top up with fresh granules in order to always have the right volume of granules in the machine, indicating some loss of granules during use. Twenty litres of granules are indicated to be able to wash up to 20,000 pans.

# Polyacrylamide used in waste water treatment

Polymers based on polyacrylamide are used for waste water purification, as well as for thickening and dewatering of municipal and industrial sewage sludge (centrifuges, filter belt presses, and chamber filter presses). The polymer may be delivered as granular particles, and thus considered microplastics as defined here, but by the application, the granules are dissolved in water (not suspended). One example of products marketed for use in Danish sewage treatment plants is the Praestol® polymer products range from Ashland. Praestol® cationic polymers are high molecular weight, water-soluble flocculating agents made by the copolymerization of acrylamide with various cationic monomers (Demolscorp, 2015). These polymers are available in both liquid emulsion and granular dry solid grades and cover the full spectrum of cationic charges. Praestol® dry polymers cannot be fed into an application without pre-diluting in water. The recommended concentration range is 0.1-0.5 percent with 0.25 percent being optimum. As the polyacrylamide granules are dissolved in water, the polyacrylamide will not be present as particles in the water; therefore, polyacrylamide used for this purpose is not considered a source of microplastics in the environment as defined in this survey. Polyacrylamide is used for various other applications such as enhanced oil recovery or in fracking fluid for shale gas extraction, but the polyacrylamide is used in dissolved form as well for these applications.

# Micronized synthetic waxes in technical applications

In the German assessment (Essel *et al.* 2015), micronized synthetic waxes are also considered as primary microplastics. The synthetic waxes do not fall under the definition of microplastics used in this report, but the application is briefly described below. As described in section 1.1, the wax particles are on the borderline of the definition and it may be relevant to further assess to what extent they should be subject to further studies. The wax may be present in the products as particles and released as particles.

These synthetic waxes are homopolymeric waxes of fine powders with particle sizes in the  $\mu m$  to lower mm range. The authors of the report note, however, that in some applications, the microparticles of synthetic waxes may lose their micro-character by being bound in a matrix, and the physical state of the synthetic waxes in applications mentioned below is therefore uncertain.

The main applications of synthetic waxes in technical applications are presented in Table 12 below.

TABLE 12
MAIN APPLICATIONS OF MICRONIZED SYNTHETIC WAXES IN TECHNICAL APPLICATIONS (ESSEL ET AL. 2015)

Application of synthetic waxes	Function
Pigments/Masterbatch	Binding material and carriers for pigment concentrates for dyeing and additive finishing of plastics
Plastics processing	Lubricant or abherent for moulding plastics, such as PVC
Adhesion promoters, hotmelts	Additives for controlling viscosity
Care products	- Additive for wear-resisting films in floor care - Protection against water marks and dirt in vehicle care - Protection of surfaces in leather and furniture maintenance
Inks	Additive to enhance rubbing fastness
Paints	Additive to protect surfaces or to create matting effects
Food coating	Additive to protect fruits

As an example, synthetic waxes may be added to polyvinylchloride, where the waxes act as a lubricant to prevent the polymer from sticking to the hot surface of the machine during processing. In care products for shoes, furniture, car paint and floors, synthetic waxes are used in order to enhance shine or improve safety through better slip resistance. When synthetic waxes are used in floor maintenance products, for example, the product is applied to the floor and the particles set in a polyacrylate matrix as the product dries. Subsequent cleaning removes these particles again, embedded in larger matrix sections. It is uncertain whether sewage treatment can filter out these particles from the sewage once they have entered the sewerage system (Essel *et al.* 2015).

Other examples cover dispersing synthetic waxes in water used for textiles processing, which may provide the textiles with smoother surfaces, thus making them easier to handle (e.g. for sewing) as well as protecting against linting. Coating the paper of glossy magazines with wax prevents the ink from coming off on the hands of the reader and a thin layer of polyethylene wax on the peel of citrus fruits protects them from drying out or bruising (Essel *et al.* 2015).

# 3.2 Formation and releases of secondary microplastics

Secondary microplastics result from the fragmentation and weathering of larger plastic items and from abrasion and maintenance of coatings. This can happen during the use phase of products, by waste disposal or within the environment from larger plastics items, which have been released into the environment.

The formation of microplastics from macroplastics in the environment is further described in section 2.7.

A Norwegian assessment of sources of microplastics has provided a conceptual model of mechanisms and different sources of microplastic pollution (Sundt *et al.* 2014). The list of sources presented in Table 13 originates in the Norwegian model, but has been reconsidered and organised slightly differently.

Apart from the Norwegian assessment, very limited information is available from the literature and the detailed description of the sources are included in the description of sources of secondary microplastics in Denmark in section 5.2.

TABLE 13 OVERVIEW OF SOURCES OF SECONDARY MICROPLASTICS FORMATION AND RELEASE (DEVELOPED BASED ON SUNDT ETAL. 2014).

Main source group	Product group / source subgroup	Description	Release pathways
Particles re- leased from handling of plas- tic products during industri- al and profes- sional use	Various plastic items	Dust from cutting, polishing and moulding plastic items e.g. at boat repair and shipyards, car repair shops, building and construction.	A small part to municipal sewage and surface water
Particles re- leased from plastic items	Various plastic products used indoors	Plastic dust from the abrasion or wear of floor coverings, furniture, plastic toys, etc.	A part to municipal sewage by cleaning of surfaces
during use		Kitchen utensils, scouring pads and similar	The main part to municipal sewage

Main source group	Product group / source subgroup	Description	Release pathways
	Various plastic parts used outdoors	Plastic dust from the abrasion or weathering of footwear, roof cover- ings, piping, tarpaulins, garden plastic furniture, etc.	Soil Surface water Urban run-off (with separate sewer system) Sewage (not separate sewer system)
	Textiles (incl. carpets)	Textile fibres ripped loose from cloth- ing, carpets, furniture and other household textiles during use	A small part in dust to sewage by cleaning of surfaces
		Textile fibres ripped loose in laundry machines (private and professional) and textile fibres from cloth used in the kitchen	Sewage
	Tyres	Dust from abrasion of tyres	Soil Surface water Urban run-off (paved areas with separate sewer system) Sewage (paved areas without separate sewer system)
	Polymer modified bitumen	Dust from abrasion of polymer modified bitumen	Same as above
	Artificial turfs	Dust from the abrasion of the turfs	Urban run-off (paved areas with separate sewer system) Sewage (paved areas without separate sewer system)
	Plastic film used in agriculture	Weathering and abrasion of the films	Soil Surface water
	Plastic items in fishing tools and	Effluent from aquaculture and fishing net cleaning facilities	Surface water
	aquaculture: nets, ropes, etc.	Abrasion and weathering of plastic in fishing tools during use	Surface water
		Abrasion and weathering of plastic ropes and surfaces in harbours	Surface water
Particles re- leased from painted surfaces	Paint for outdoor applications	Dust from paint application, abrasion and maintenance work; other outdoor applications of paint	Soil Surface water Urban run-off (with separate sewer system) Sewage (not separate sewer system)
	Paint for indoor applications	Dust from paint application, abrasion and maintenance work; indoor application	Sewage from cleaning of dust on surfaces
	Road paint	Abrasion of road paint	Soil Surface water Urban run-off (paved areas with Sew- age (paved areas without separate sewer system)
Waste handling	Plastic particles from shredding and fragmenting plastic	Composting plastic contaminated organic waste, and runoff from reuse of this	Soil Surface water
	waste and waste contaminated with	Shredding of plastic parts of vehicles, electrical and electronic equipment, etc.	Soil (via air) Surface water (via air)

Main source group	Product group / Description source subgroup		Release pathways
	plastics	Food waste shredders on ships	Surface water
		Food waste shredders in institutions  Shipbreaking/decommissioning of ships and offshore installations	Sewage Surface water Industrial sewage
	Illegal waste burning	By illegal burning of waste, particles of incomplete burned plastics may be formed	Soil Surface water (via air)
	Landfills and waste dumps	Particles formed by fragmentation of larger plastics parts in landfills and waste dumps	Soil (via air) Surface water (via air) Sewage (via leachate)
Fires	Fires	Particles of incomplete burned plastics may be formed	Soil Surface water (via air)
Fragmentation of plastics waste in the environ- ment (by biolog- ical and non- biological pro-	Macro plastic debris from illegal, un- wanted or unregu- lated <u>terrestrial</u> waste handling	Sources: Littering in public spaces; drift from landfills, waste dumps, construction sites, agriculture, etc.; dumping of industrial and construc- tion waste.	Soil Surface water (streams and lakes) Urban run-off (with separate sewer system) Sewage (not separate sewer system
cesses)	handling  Macro plastic debris from illegal, unwanted or unregulated maritime waste handling  Macro plastic debris from illegal, unwanted or unregulated side leisure activity and recubating; weathering and do ing of wrecks and abandon storm loss of floaters and of items		Surface water (sea)

# 3.3 Examples of overall releases of microplastics

Two surveys of releases of microplastics have been carried out recently in Norway and Germany.

## **Norway**

A recent assessment of sources of microplastics pollution in Norway has estimated the total annual microplastic emissions from Norwegian land-based sources and ship paint at more than 8,000 tonnes annually (Table 14). The assessment covered plastics in a broad sense including rubbers, synthetic textile fibres and paints. The report applies the term "primary sources" for all sources except the formation of microplastic from macrolitter in the environment.

Tyre dust was by far the most important single source, followed by abrasion and particle shedding from polymer based paint and textiles. Release of microplastics from cosmetics for consumer applications was estimated at 40 t/y, corresponding to 0.5% of the total releases of microplastics.

The 8,000 tonnes annually exclude secondary microplastics formed from macrolitter in the environment, as the figures for macrolitter were not complete. Some secondary sources are indicated by the annual generation of lost or discarded macroplastic items, and the authors provide a "best guess" of annual Norwegian macrolittering of the sea would of about 10,000 tonnes, but according to the authors, there is not enough knowledge about rates and amount of these plastics ending up as microplastics. Further studies are therefore necessary before the different sources in Norway can be compared.

 $\begin{tabular}{l} \textbf{TABLE 14} \\ \textbf{SUMMARY OF EMISSION ESTIMATES FOR NORWEGIAN SOURCES TO MICROPLASTIC POLLUTION (SUNDT $ET$ $AL$.} \\ \textbf{2014}). \\ \end{tabular}$ 

Mechanism	Source group		Tonnes **	% of total	Possible path- way	Dominant plastic
Designed ***	Consumer products, all		40	0.5	Drain, sludge	PE, PMMA, PTFE
	Commercial products, all		100	1.2	Drain, soil, air, sea	
Production spill	Transport spil	I	250	3.0	Sea, soil	PS, PE, PET, PVC
***	Production dis	charge	200	2.4	Drain, sea, air	
	Accidents		n.a.		Sea, runoff, air	
Abrasion by commercial	Ship paint		330	3.9	Sea, seaside	Epoxy, PU, A****, S****
maintenance	Marinas		400	4.8	Sea, seaside	Epoxy
	Building repai	r	270	3.2	Sewer, soil	
	Laundries		100	1.2	Drain	
Wear and tear	Household	Laundry	600	7.1	Drain	PS, PA, A****, PU
during use		Dust	450	5.4	Drain, waste, air	
	City dust outdoor	Road paint	320	3.8	Sewer, soil, air	SIS****, EVA, PA
		House paint	130	1.5	Sewer, soil, air	PVA, A****, PS, SBC****
		Tyre dust	4500	53.6	Sewer, soil, air	SBR
	Indoor city	Dust	130	2.4	Drain, air	
Waste shred- ding	Plastics recycli	ng	n.a.		Drain, sludge, air	PS, PA
	Illegal dumpin	g, paint	100	1.2	Soil, sea	
	Landfills		n.a.		Air, water	
	Biowaste		336	2.4	Soil, water	
	Paper recycle		60	1.2	Water	Latex, PE, S****
	WEEE and EL	V	10	0.1	Air, water	ABS and more
Total primary sou	rces *		8,396		Upstream	
Macrolitter	Macrolitter Tonnes as macrolitter					
	Fishery		>1000	n.a.	Dumped, lost	PA, EPS, PP
	Sewage		460	n.a.	Drain	various
	Plastic bags		60	n.a.	River, sea	PE, LDPE, PET
	Other			n.a.		
Total secondary s	ources *		n.a.			

<sup>\*</sup> The distinction between primary and secondary sources is different from the distinction between primary and secondary microplastics in the present review.

<sup>\*\*</sup> Emission by all pathways incl. sewage.

 $<sup>\</sup>ensuremath{^{***}}$  Applications considered primary microplastics in this report.

<sup>\*\*\*\*</sup> As abbreviated in Sundt *et al.* (2014). No further explanation of these abbreviations is given in Sundt *et al.* (2014); these abbreviations are not familiar to the authors of this project.

TABLE 15 "BEST GUESS" ON PROBABLE SHARES TO SEA FROM DIFFERENT NORWEGIAN MICROPLASTICS SOURCES (SUNDT ET AL. 2014)

Source grou	p	Up- stream t/y	Pathway to sea	Possible share to sea*	Fraction to sea, tonnes
Consumer produ	icts, all	40	Drain, past STP	Small	4
Commercial pro	ducts, all	100	Drain, sea	Medium	50
Transport spill		250	To sea	Large	225
Production disch	narge	200	To drain or sea	Large	180
Ship paint		330	Sea, seaside	Large	297
Marinas		400	Sea, seaside	Large	360
Building repair		270	Sewer, dump	Medium	135
Laundries		100	Drain	Medium	50
Household	Laundry	600	Drain, past STP	Small	60
	Dust	450	Drain, air	Small	45
City dust out-	Road paint	320	Sewer, air	Medium	160
door	Exterior paint	130	Sewer, air	Small	13
	Tyre dust	4500	Sewer, air	Medium	2250
Indoor city	Dust	200	Sewer, air	Small	20
Illegal dumping,	paint	100	Soil, sea	Large	90
Biowaste		336	Soil, water	Small	34
Paper recycle		60	Water	Large	54
WEE and ELW		10	Air, water	Medium	5
Total		8,396			4,323

<sup>\*</sup>small = 10%, medium = 50%, large = 90%

# Germany

In a recent German study, the use of microplastics in cosmetic products as well as other applications on the German and European markets was investigated and the uses of primary and secondary microplastics were quantified, based on data from the literature and interviews with companies (Essel *et al.* 2015).

The authors estimate a total use of primary microplastics in Germany of approximately 100,700 t/year, which is a rough estimate. In the report, microplastics are defined as all plastics particles with a diameter of 1-5000  $\mu$ m. In the German study, micronized synthetic waxes are also considered as primary microplastics. Even though synthetic waxes differ from 'real' plastics as the molar mass of the used polymer is much lower (2,000 – 20,000 g/mol) compared to actual plastic (>100,000 g/mol), according to the authors, scientific studies have still identified the synthetic waxes as a source of microplastic, and they were therefore included in the study. The use quantities of micronized synthetic waxes are estimated to be 100,000 t/year, and thus by far the most important use of microplastics in products according to the German figures.

Primary microplastics in cosmetic products account for 500 t/year (data only available for polyethylene), and use in detergents, cleaning and maintenance products for industry and as blasting abrasive account for less than 100 t/year each. No figures were available for the quantities of microplastics used in cleaning agents for private households or in medicine.

The identified sources of secondary microplastics were fragmentation of plastic debris, synthetic fibres from clothing and other textiles, pellets loss during production and processing of plastics, as well as tyre abrasion. The overall results are shown in Table 16. Please note that the release pathway is not considered; i.e. the estimated releases do not represent releases to the aquatic environment. Fragmentation of plastic debris was found to be the most significant source of microplastics even though no exact figures on releases were given for Germany. For Europe, the number is estimated to be between 3.4 and 5.7 million t/year. Pellet loss and tyre abrasion release between 21,000 – 210,000 and 60,000 – 111,000 t/year, respectively, while the quantity of fibres shed from synthetic textiles are assumed to be 80-400 t/year.

 $\begin{array}{l} \textbf{TABLE 16} \\ \textbf{SOURCES OF PRIMARY AND SECONDARY MICROPLASTICS IN GERMANY (ESSEL \textit{ET AL.} 2015)} \end{array}$ 

Primary microplastics	Consumption, t/year
Cosmetic products	500
Detergents, cleaning and maintenance products for commercial and industrial use	<100
Blasting abrasives for deburring surfaces	<100
Micronized synthetic waxes in technical applications	100,000
Secondary microparticles	Releases, t/year
Fragmentation of plastic debris	unknown
Synthetic fibres from clothing and other textiles	80 - 400
Pellet loss during manufacture and processing of plastics *	21,000 - 210,000
Tyre abrasion	60,000 - 111,000

<sup>\*</sup> The pellet loss is considered a loss of secondary microplastics by the authors because the pellets are unintentionally used in products (different use of the terms than in the present study).

# 4. Fate of microplastics by sewage treatment

# 4.1 Introduction

Sewage treatment plants (STPs) in northern Europe often receive sewage from a mixture of sources like households, industries, business related units and storm water. Microplastics enter sewage from all these entities. The quantities and characteristics of the plastic particles depend on the activities carried out by all users of the STPs. The recognition of private homes as sources of microlitter to the environment is fairly recent. Microplastics (plastic fragments and plastic fibres) are intentionally or unintentionally released into the sewage from various household activities. The origin could be e.g. synthetic textile fibres from washing of clothes and wet cleaning of floors and dusty surfaces, plastic pellets from personal care products, and various objects deliberately thrown into sinks or toilets (Gregory 1996; Browne *et al.* 2011; Hintersteiner *et al.* 2015). Although there are only a few studies available on the subject, there is no doubt that STP effluents have become entrance routes for microplastics to the aquatic environment and possibly also to the terrestrial environment through spreading of sewage sludge on farmland (Habib *et al.* 1998; Zubris and Richards 2005). There is, however, still not enough data to determine the magnitude of the importance of STP effluents relative to other sources.

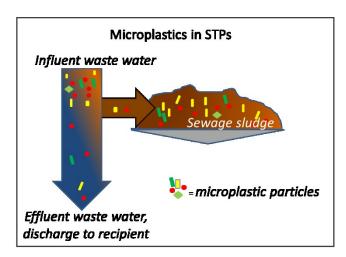


FIGURE 13
THE VAST MAJORITY OF MICROPLASTIC PARTICLES REACHING THE STPS VIA INFLUENT WATER ARE RETAINED IN THE SEWAGE SLUDGE. THE NUMBER OF PLASTIC PARTICLES IN EFFLUENT WATER REMAINS SUBSTANTIAL

# 4.2 Retention efficiency and microplastics in the effluents

Published results on microplastics in sewage to and from STPs are available from three Swedish, three Norwegian, and ten German STPs (Magnusson 2014b; Magnusson and Norén 2014; Magnusson and Wahlberg 2014; Mintenig *et al.* 2014). Analyses in the Swedish and Norwegian plants were done on influent and effluent water, and from the German plants only on effluent water. From the German STPs and the Swedish Långevik analyses were also done on sewage sludge. Henriksdal in Stockholm and Ryaverket in Gothenburg are the largest STPs in Sweden, whereas Långevik in Lysekil is representative of small municipal STPs. The Norwegian plants were located in the south-

ern part of the country and included the largest plant, VEAS in Oslo, and also Tönsberg and Fuglevik. The personal equivalents (PEs) for these STPs are shown in Table 18 and 20. The German plants were all situated in Lower Saxony, a region in north-western Germany bordering the North Sea. Eleven of them were in the size range of 7,000 and 56,000 PE, and one had 210,000 PE. Like most STPs in northern European countries, all investigated STPs were equipped with mechanical, chemical and biological treatment of the sewage.

Microplastics in the influent water to sewage treatment plants - Microplastics and non-synthetic fibres in the influent sewage to three Swedish STPs are shown in Table 17. Notably the number of non-synthetic anthropogenic fibres (e.g. fibres of cotton or viscose) per  $m^3$  is approximately 10 times the number of plastic fibres. The number of plastic fibres ranges from 5,000 to 35,000 per  $m^3$  and accounts for the majority of microplastics  $\geq 300 \ \mu m$ .

TABLE 17
MICROPLASTIC CONCENTRATION IN INFLOW OF SWEDISH STPS BY SIZE AND PARTICLE TYPE (AVERAGE VAUES FROM THREE SAMPLING OCCASIONS) (MAGNUSSON AND WAHLBERG 2014)

	Plastic fibres no/m³	Plastic fragments no/m³	Plastic flakes no/m³	Total mi- croplastics no/m³	Non- synthetic fibres no/m³
≥300 µm					
Henriksdal; 750,000 PE	14,250	1,500	800	9,400	76,800
Ryaverket; 740,000 PE	5,000	1,150	850	7,000	84,600
Långevik; 12,000 PE	8,300	1,350	600	10,250	56,200
≥20 µm					
Henriksdal; 750,000 PE	17,000	13,700	22,100	58,800	284,000
Ryaverket; 740,000 PE	8,900	6,850	6,800	22,500	132,800
Långevik; 12,000 PE	7,200	7,200	6,250	27,700	146,600

Retention efficiency - In the Swedish STPs, two size fractions of microplastics, \$\ge 300\$ and \$\ge 20\$ \$\mu m\$, were analysed in the sewage. It was found that \$\ge 99\%\$ of the microplastics \$\ge 300\$ \$\mu\$ m that reached the STPs via incoming water were retained in the sewage sludge and thereby prevented from being discharged to the recipient (Magnusson and Norén 2014; Magnusson and Wahlberg 2014). The data from Magnusson and Wahlberg (2014) are shown in Table 18. The most efficient retention was found in Ryaverket, which is equipped with a 15-\$\mu\$m disc filter as an additional treatment step before the water is released to the recipient. Also, when including particles down to 20 \$\mu\$m the retention in Ryaverket was the most efficient - but only if the concentrations were expressed as weight of particles (data not shown). However, when sticking to the common practice of expressing the microplastic concentration in numbers of particles, Ryaverket appeared to have the least efficient retention. This is because the effluents from Ryaverket contained more lightweight particles, mainly small fibres and paint flakes, whereas effluents from the other two STPs contained fewer but larger particles.

No significant differences in the retention efficiency depending on the morphology (fibre, fragments or flakes) were demonstrated, but the retention of non-synthetic anthropogenic fibres was generally more efficiently retained.

TABLE 18
RETENTION EFFICIENCY OF MICROPLASTIC PARTICLES AND NON-SYNTHETIC ANTHROPOGENIC FIBRES IN THREE SWEDISH STPS BY PARTICLE SIZE AND TYPE (MAGNUSSON AND WAHLBERG 2014)

	Plastic fibre %	Plastic fragments %	Plastic flakes %	Total mi- croplastics %	Non- synthetic fibres, %
≥300 µm					
Henriksdal; 750,000 PE	99.4	95.2	99.9	98.9	100.0
Ryaverket; 740,000 PE	100.0	98.5	100.0	99.9	100.0
Långevik; 12,000 PE	99.8	98.5	99.8	99.5	99.9
≥20 µm					
Henriksdal; 750,000 PE	89.4	74.0	99.2	90.0	96.6
Ryaverket; 740,000 PE	64.4	88.6	69.9	69.9	77.8
Långevik; 12,000 PE	86.4	89.3	82.9	87.0	93.0

Similar retention efficiencies as in the Swedish STPs were found in three Norwegian plants, over 96% for particles >300  $\mu$ m and over 87% for microparticles >20  $\mu$ m, as shown in Table 19 (Magnusson 2014B). However, whereas data from the Swedish STPs are average values from three separate sampling occasions, data from the Norwegian plants are based on just one single sampling.

TABLE 19
RETENTION EFFICIENCY OF MICROPLASTIC PARTICLES AND NON-SYNTHETIC ANTHROPOGENIC FIBRES IN THREE NORWEGIAN SEWAGE TREATMENT PLANTS BY SIZE AND PARTICLE TYPE (MAGNUSSON 2014b)

	Microplastics %	Non-synthetic fibres %
≥300 µm		
VEAS, Oslo; 700,000 PE	99.9	100.0
Tönsberg; 185,000 PE	97.4	96.2
Fuglevik; 85,000 PE	99.4	98.9
≥20 µm		
VEAS, Oslo; 700,000 PE	97.0	92.1
Tönsberg; 185,000 PE	87.6	87.0
Fuglevik; 85,000 PE	90.7	93.2

**Microplastics in effluents** - Although most microplastics in incoming sewage were found to be retained in the STPs, the amount in effluent water was still substantial. The concentrations in effluent water varied considerably depending on both the concentrations in incoming water and on how the sewage was being treated in the plant. In effluents from the German STPs, the concentrations of microplastics  $\geq$ 10  $\mu$ m varied between 260 and 1,900 particles/m³ (an extreme concentration of 13,700 particles/m³ was found in one plant) (Mintenig *et al.* 2014). Concentrations of microplastics  $\geq$ 20  $\mu$ m in the Swedish STP effluents were found to be higher, between 2,100 and 5,500 particles/m³ (Magnusson and Wahlberg 2014). The cut-off size for the collected particles was lower in the German study than the smallest particles collected in the Swedish study, 10  $\mu$ m compared to 20  $\mu$ m. Still, particles between 50 and 100  $\mu$ m were found to be dominant in the German samples, which should make the results from the two studies comparable.

However, the microplastic concentrations are in themselves mostly of interest when calculating the retention efficiency. In order to estimate the load of microplastics that actually reaches the recipient waters from the STPs, the flow rate of the effluent water also has to be taken in account. Estimated data on yearly release of microplastics from the German and Swedish plants are presented in Table 20 (data from the German STPs was not available from individual plants). Comparing the German data, which are based on particles  $\geq 10~\mu m$ , with Swedish data on particles  $\geq 20~\mu m$ , reveals that the highest number of particles released from German STPs per year was in the same range as the number released from Långevik, the smallest of the Swedish plants.

The plants included in the Swedish study are of very different sizes (12,000 – 750,000 PE). By normalizing data on the amount of microplastics reaching the STP recipients to the PE of the plants, the number of users is compensated for (Table 20). The discharge via effluent water was then found to be between 1,350 and 10,000 particles  $\geq$ 300  $\mu$ m per year and PE. When including particles down to 20  $\mu$ m, the contribution was 350,000-850,000 plastic particles/year/PE. The Swedish STPs generally had similar or slightly better retention efficiency than the Norwegian ones, however, still the number of microplastics per unit time and PE was higher (Table 20). This means that fewer microplastics were entering the Norwegian than the Swedish STPs.

TABLE 20 ESTIMATED YEARLY RELEASE OF MICROPLASTIC PARTICLES FROM STPS IN GERMANY (12 PLANTS) AND SWEDEN (3 PLANTS) TO RECIPIENT WATERS (MAGNUSSON AND WAHLBERG 2014; MINTENIG  $ET\,AL.$  2014). THE PRESENTED DATA SHOWS THE NUMBER OF PLASTIC PARTICLES IN EFFLUENT WATER PER YEAR, AND ALSO THE NUMBER OF PARTICLES PER YEAR DIVIDED BY THE POPULATION EQUIVALENT (PE) OF THE PLANTS (ONLY DATA FROM THE SWEDISH PLANTS).

	Microplastics in effluent water (number particles/year)	Microplastics in effluent water (number particles/year/PE)
German STPs (particles ≥10 μm)	0.093·109 - 8.2·109 (12 STPs: 7,000 – 210,000 PE)	(not available)
Swedish STPs (particles ≥300 μm)	0.12·10 <sup>9</sup> (Långevik, 12,000 PE) 1.0·10 <sup>9</sup> (Ryaverket 740,000 PE) 4.5·10 <sup>9</sup> (Henriksdal 750,000 PE)	10,000 (Långevik) 1,350 (Ryaverket) 5,900 (Henriksdal)
Swedish STPs (particles ≥20 µm)	0.1·10 <sup>11</sup> (Långevik, 12,000 PE) 6.5·10 <sup>11</sup> (Ryaverket 740,000 PE) 2.5·10 <sup>11</sup> (Henriksdal 750,000 PE)	850,000 (Långevik) 880,000 (Ryaverket)* 350,000 (Henriksdal)*
Norwegian STPs (particles ≥300 μm)	0.22·10 <sup>9</sup> (Fuglevik, 85,000 PE) 2.8·10 <sup>9</sup> (Tönsberg, 185,000 PE) 3.2·10 <sup>9</sup> (VEAS, 700 000 PE)	2,640 (Fuglevik) 15,350 (Tönsberg) 4,700 (VEAS)
Norwegian STPs (particles ≥20 μm)	25.5·10 <sup>9</sup> (Fuglevik, 85,000 PE) 30.7·10 <sup>9</sup> (Tönsberg, 185,000 PE) 309·10 <sup>9</sup> (VEAS, 700 000 PE)	300,000 (Fuglevik) 166,000 (Tönsberg) 438,000 (VEAS)

When the microplastic content was expressed as *weight* instead of *number* of particles the load in effluent water from Ryaverket was significantly lower than in effluents from Henriksdal (see further in text).

# 4.3 Microplastics in sewage sludge

The studies of STPs clearly show that the vast majority of microplastic particles in incoming water are retained in the sewage sludge. Since the sludge is often used as fertilizer of farmland, there is a definite risk that this has become a pathway for microplastics to enter the terrestrial environment.

Data on sludge is available from the German STPs and from the Swedish STP Långevik (Magnusson and Norén 2014; Mintenig *et al.* 2014). Sludge from German STPs was found to contain 1,000-24,000 microplastics particles ≥10 µm per kg dry weight. From Långevik, only data on particles ≥300 µm was available, and the concentration was ~17,000 microplastic particles per kg dry weight

of sludge. All values on sludge were based on small sampling volumes and should be regarded as indicative rather than as true values.

The further fate of the microplastics in the soil is briefly discussed in section 2.8.

None of the studies has estimated the full mass balance of the microplastics in the sewage treatment plants and consequently no data are available on the possible degradation of microplastics by the various process steps in the treatment process. It is, however, expected that the mineralization of microplastics by the treatment process will be insignificant.

# 4.4 Polymer composition of microplastics in sewage

The polymer composition, colour and morphology of the microplastics in the sewage may provide some indications of the sources of the microplastics in the sewage.

The polymer composition of microplastic fragments and fibres collected in the German and Swedish STPs were analysed with Fourier transform infrared spectroscopy (FTIR). A wide spectra of plastic materials were identified, but e.g. PE, PP, polyester (PEST) and PA were found to be particularly common (Magnusson and Norén 2014; Mintenig *et al.* 2014).

The detailed polymer composition of the microplastics ( $<500 \mu m$ ) in the effluent water of 12 German STPs are shown in Figure 14. In most plants the dominant polymers were PE, PS, poly(vinyl alcohol) (PVAL in figure, English PVA), PA, and PEST. All the polymers are used for textile fibres, while PE is the main polymer used for microplastics in cosmetics. Notably, paint-like particles ("lackartig" in the figure) accounted for only 5-10% of the particles but it cannot be ruled out that some of the other polymer particles (e.g. PUR or polyester) may be paint particles.

For microplastics >500  $\mu$ m, separate datasets are reported for textiles and particles, respectively. Fibres varied from 33 to 9923 fibres/m³ in the effluent, with polyester as the dominant polymer accounting for more than 50% of the total, while PA (nylon) was the second most abundant, followed by PP. One plant receiving sewage from a textile factory did not differ significantly from the average figure. The particles >500  $\mu$ m ranged from 1 to 52 particles/m³ and were totally dominated by PE particles (more than 75% in most plants) followed by PP and PVC. Particles indicated as paint were insignificant apart from one plant where the paint accounted for less than 10%.

The authors note that since the subsamples were relatively small, the amounts have to be seen as indicative values (Mintenig *et al.* 2014). The data indicates that more data on the polymer composition of influent and effluent from sewage treatment plants may be valuable for a further assessment of the sources.

Styrene-butadiene rubber (SBR), used in tyres, is not among the polymers tested for.

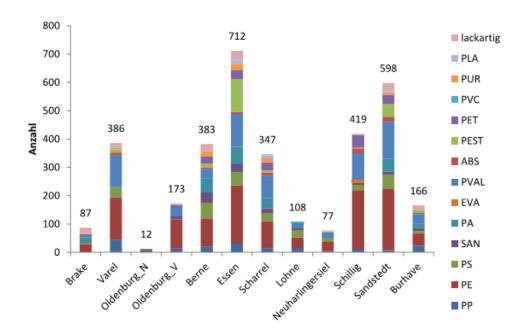


FIGURE 14 POLYMER COMPOSTION OF MICROPLASTICS ) <500  $\mu$ m; IN EFFLUENT FROM GERMAN SEWAGE TREATMENT PLANTS (MINTENIG *ET AL.* 2014). NUMBER PER m³. LACKARTIG = PAINT LIKE, ANZAHL= NUMBER, PEST = POLYESTER, PVAL = PVOH, (MINTENIG *ET AL.* 2014) (REPRODUCED WITH PERMISSION FROM THE MAIN AUTHOR)

# 5. Releases of microplastics and occurrence in the water environment in Denmark

A general description of uses of primary microplastics and sources of releases of primary microplastics to the environment is provided in Chapter 3.1

This chapter focuses on the use of primary microplastics in Denmark and the contribution of releases of primary microplastics to the general pollution by microplastics in the environment in Denmark and the surrounding waters. In order to assess to what extent primary microplastics contribute to the microplastics pollution, the section also addresses sources of secondary microplastics and the fate of microplastics entering the sewerage systems in Denmark.

The comparison is made based on the estimated mass flows in terms of tonnes per year. Due to differences in shape, size, density and composition, the transport of the microplastics within the environment, and the possible effects on organisms and ecosystems may be very different between the different types of microplastics. One tonne of small, buoyant particles with a high concentration of a brominated flame retardant may potentially have much higher environmental impact than one tonne of larger, pure polymer particles with a high density which is buried in the sediment close to the outlet. The estimated releases should consequently be compared with caution, but the estimates may be used for the prioritization of further research and initiation of measures for reduction of the releases.

A major release pathway for releases from processes and products is releases to sewage. As a majority of the microplastics is retained in the sludge at the STP, the resulting releases to the aquatic environment from processes and products is considerably lower than the initial releases to sewage. In order to allocate releases from sewage treatment to the initial sources, a summary table is prepared where both initial releases and resulting releases by source category are estimated.

# 5.1 Use and releases of primary microplastics

# 5.1.1 Personal care products

# **Application and consumption in Denmark**

A general description of the use of microplastics in personal care products is provided in section 3.1.2 and this section focuses on the Danish situation. The majority of personal care products marketed in Denmark are produced by large international companies, and the type of applications and products are not supposed to be different from the applications described in section 3.1.2.

Data on microplastics in personal care products as well as in cleaning and maintenance products in Denmark have been collected by a survey undertaken by the Association of Danish Cosmetics, Toiletries, Soap and Detergent Industries (SPT). The answers summarised in the table below are highly variable and do not allow the estimation of the total content of microplastics in personal care products sold in Denmark in 2014. The Cosmetics Europe survey for 2012 described in section 3.1.2

(Gouin *et al.* 2015) will therefore be used as a source for an estimation of the likely consumption in 2014.

The Cosmetics Europe survey provides an estimate of the use of microplastics beads in personal care products sold in Denmark in 2012. Based on the Euromonitor data on consumption of shower gel, face wash and liquid soaps, the Cosmetics Europe survey estimates the total content of microbeads in personal care products sold in Denmark in 2012 at 29 tonnes (Gouin *et al.* 2015).

The Euromonitor data indicates the following market volume in Denmark of relevant personal care products:

- Shower gel: 2.9 million litres
- Face wash (premium): 0.06 million litres
- Face wash (mass prod): 0.2 million litres
- Liquid hand soap: 1.7 million litres

The largest volumes are accounted for by shower gel and the liquid hand soap.

The 29 tonne estimate is based on the assumption that the microbeads account for 0.6% of the total weight of the sold products. The 0.6% was reached assuming that 6% of the sold products contained plastic microbeads in 2012 with a concentration of 10% polyethylene microbeads. The percentage of products and the concentration is the same for all products and all countries and the 6% is indicated as a conservative estimate. Similarly, the 10% is indicated as a maximum concentration.

Results from the SPT survey are shown in Table 21. The table is based on answers from 9 companies, who answered that they still use microbeads in some products or have used them in 2012. The members of SPT represent about 90% of the total Danish market. No data have been requested from suppliers that are not members of the trade association. Besides the 9 companies, one company answered that microplastics would be phased out in all products by 2017, but this company did not provide quantitative data. The answers do not indicate the market share of the individual companies and there is no basis for weighting of the answers. Due to the low number of answers, the data do not allow detailed statistical analyses.

As mentioned, the Cosmetics Europe survey estimated an average content of 10% in these products containing microplastics. The actual average concentrations as reported in Table 22 are rather around 5%, which is accordance with the concentrations in analysed personal care products summarised in Table 11. A maximum of 11% was reported for a few products.

The highest uncertainty is related to the estimate of the share of the total marketed products that contain microbeads. The indicated percentages vary considerably among the companies. For face wash as an example, one company indicated that 30% of the products contained microplastics in 2012, while several companies indicated the percentage to be 0.1%.

Except for hand cleaners (Danish: håndrens), the overall impression is that the 2014 level is about half of the 2012 level, reflecting the ongoing phase-out of microplastics in many products. All uses of microplastics are indicated to be phased out by 2017 with the exception of 0.1% of the products from one supplier. For hand cleaners used by professionals and consumers for cleaning hands contaminated with oil, grease, paint etc., a decrease from 23-25% of all products in 2012 to 15-19% of all products in 2017 is indicated. It is not clear how much of the liquid hand soap indicated in the Cosmetics Europe survey is accounted for by these hand cleaners or if they are included at all. According to information from SPT, manufacturers of hand cleaners are also working on developing products with substitutes for microplastics.

The percentages of face wash/scrub products containing microplastics indicated in the SPT survey and the Cosmetics Europe survey is relatively low compared with the results of surveys of products marketed in the Netherlands shown in Table 35, where it is indicated that 34% of cleansing products and 40% of scrubs contained microplastics. If the Cosmetics Europe survey estimates are recalculated assuming that 40% of the face wash products contained microplastics and assuming an average content of 5% microplastics for all products (instead of the 10% used), the total for Denmark, however, would be lower than the 29 tonnes estimated in the Cosmetics Europe survey (because the face wash represent a small part of the total volume).

The Cosmetics Europe survey does not include microplastics in toothpaste which in a German assessment was estimated to account for 20% of the total use of microplastics in cosmetics in Germany (Essel *et al.* 2015), and data from the Netherlands indicate that about 10% (in terms of number of different products) of the toothpastes contained microbeads (Appendix 1). None of the companies answering the SPT survey has answered regarding toothpaste but it is not clear if the reason is that they do not market toothpastes. According to SPT, none of the toothpastes supplied by its member companies contains microplastics today.

If the estimated German per capita consumption of 6.2 g/year (Essel *et al.* 2015) was used for Denmark, the total would be 35 tonnes per year.

All in all, the available data indicate that the Cosmetics Europe survey results of 29 tonnes in 2012 may realistically reflect the maximum consumption in Denmark in 2012 and that the consumption in 2014 likely is considerably lower.

Some applications of microbeads e.g. for colouring toothpaste and microplastics used for glitter effects of some cosmetics are not included in the estimate, but likely relatively small as compared with the consumption of microplastics in the addressed personal care products.

Based on the available data, the total content of microplastics of personal care products and other cosmetics in 2014 is estimated to be in the range of 10-30 tonnes.

TABLE 21
MICROBEADS IN PERSONAL PROTECTION PRODUCTS SOLD ON THE DANISH MARKET. RESULT OF SURVEY OF MEMBERS OF SPT

Product group	Concentration, individual answers where the concentration is above 0%, % of product weight	Percentage of the company's products containing microbeads (individual answers)  2012 2014 2017				
Shower gel	5; 1-2; 5-10	0.1; <0.5; <1	0.1; 0: 50% of 2012 level; <0.05	0.1; 0; 0; 0		
Face wash	5; 0.05- 1; 2.5; 5-10; 2.8	0.1; <0.1; 3; <1; 30	0.1; 0; 40% of 2012 level; 0; 10	0.1; 0; 0; 0; 0		
Liquid hand soap	9	0; 0	0; 0.1	0; 0		
Toothpaste	-	-	-	-		
Hand cleaners	1-5	25; 23	23; 19	19; 15		

**Ecolabels** - A large portion of cosmetic products produced and sold in Denmark are labelled by the Nordic Swan ecolabel. Microplastics are, as previously mentioned, prohibited in products with the Swan ecolabel and with the European flower label.

#### Release pathways and total releases

The major release pathway of microplastics in cosmetics is direct discharge to the municipal sewerage system. The microplastics are used in scrubs, soaps and toothpaste, which are intended to be washed off immediately after application. The further fate of the plastic particles in the sewerage system and the municipal sewage treatment plants is described in section 4.1. It is assumed that some 90-95% of the microplastics in the products are released to sewage. A small percentage, left in the packaging or left in cleaning tissue, would be disposed of with the municipal solid waste. Unlike sun creams and other cosmetics left on the skin, direct releases to the aquatic environment e.g. by bathing are considered insignificant.

Based on the estimated consumption (based on consumption figures from Gouin *et al.* 2015) and the emission factors, the total release of microplastics in cosmetics to sewage is estimated at 9-29 t/y. Using the distribution factors for microplastics in sewage as described in section 5.3, the total releases to aquatic environment from the use of microplastics in cosmetics is estimated at 0.5-2.9 t/y while 5-14 t/y is applied to agricultural soils.

#### 5.1.2 Raw materials for plastics production in Denmark

As described in section 3.1.3, nearly all plastics raw materials are used in form or plastics pellets or powder of plastic resins (smaller particles) which fall under the definition of microplastics as used in this report.

The pellets and powders are converted by heating into the final plastic items, which do not themselves contain microplastics. The potential releases would this occur from the manufacture of the plastics raw materials (both manufacture of the resins and compounds), during transport and by the conversion. Plastic raw materials are not manufactured in Denmark and all raw materials for Danish plastics converters are imported.

The distribution of plastics raw materials in Denmark by plastics type is quite similar to the total European plastics production described in section 3.1.3 with polyethylene (PE) and polypropylene (PP) and ethylene copolymers representing 55% of the total of 583,000 tonnes in 2014. Some of the raw materials are not traded in pellet or dust form, but rather as liquid resins (e.g. the epoxy resins and some of the polyesters) and the total use of solid plastics raw materials is likely in the range of 500,000-550,000 t/y. PE, PE copolymers and PP account for about 60% of the solid raw materials and are thus the most likely to be found in the environment due to pellet loss. The majority of the solid plastic materials are used as pellets while a minor part is used as powder for rotational casting, sintering of PTFE powder (for Teflon® coating) and possibly other applications.

The plastic raw materials are used by approximately 250 plastic converters (Danish Plastics Federation 2015a); of these, many companies for which plastic conversion is not the main activity (main activity: wind turbines, district heating pipes, etc.).

TABLE 22

NET IMPORT OF PLASTICS GROUPED INTO MAIN TYPES OF PLASTICS \* (SOURCE: STATISTICS DENMARK, EXTERNAL TRADE STATISTICS)

	Abbreviations (examples) **	Net import, tonnes	% of total
Polyethylene	PE, LDPE	120,129	21%
Ethylene, copolymers	HDPE, EVA	34,214	6%
Polypropylene and copolymers	PP	162,346	28%
Polystyrene, expandable	EPS, XPS	24,545	4%

Polystyrene, other + copolymers	PS, HIPS, ABS, SAN	37,169	6%
Polyvinyl chloride, plasticised	PVC	6,117	1%
Polyvinyl chloride, other + copolymers	PVC	31,140	5%
Acrylic polymers and copolymers		40,999	7%
Epoxy resins		19,139	3%
Polycarbonate	PC	9,530	2%
Alkyd resins		4,191	1%
Polyethylene terephthalate	PET	22,219	4%
Polyesters	UP	11,826	2%
Polyamides	PA, nylon	13,209	2%
Polyether alcohols (for PUR)	PUR	21,219	4%
Polyurethanes	PUR	2,528	0%
Phenolic plastics		6,181	1%
Aminoplastics		3,016	1%
Polymers and copolymers of vinyl acetate (including dispersions)		10,332	2%
Other		3,209	1%
Total		583,259	

<sup>\*</sup> The commodity groups from the statistics have been grouped into main type of plastics by the authors of this report, based on the description of each commodity group in the statistics.

# Release pathways and total releases

The major releases are expected to take place by transport, loading and reloading of trucks and similar operations, whereas the releases from the conversion of the pellets are considered small or insignificant.

Plastic raw materials are not manufactured in Denmark and only some of the steps involved in the movement of plastic pellets from the resin production to the processor would take place in Denmark. Steps in the movement of plastic pellets from the resin production facility, through the distribution network, to the processor where spills may take place according to the Operation Clean Sweep guidelines are shown in Figure 15. Raw materials used in Denmark are typically transported by trucks from resin production facilities in Germany or Sweden. Only the steps on the right side of the figure take place in Denmark.

<sup>\*\*</sup> Some subgroups within the indicated groups of plastics may be designated using other abbreviations.

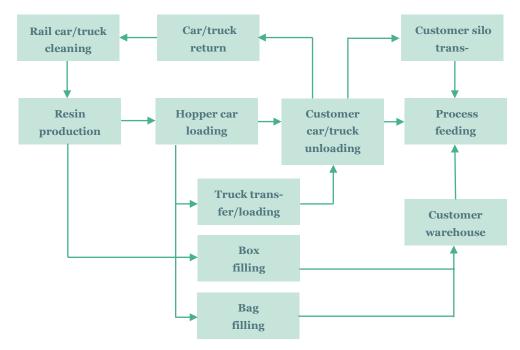


FIGURE 15
STEPS INVOLVED IN THE MOVEMENT OF PLASTIC PELLETS FROM THE RESIN PRODUCTION FACILITY, THROUGH THE DISTRIBUTION NETWORK, TO THE PROCESSOR. SPILLS AND PELLET LOSS TO THE ENVIRONMENT CAN OCCUR AT ANY STEP (BASED ON OPERATION CLEAN SWEEP 2015)

In order to obtain more specific information on the possible levels of spill and the fate of the spill, as part of this study, the Danish Plastics Federation has contacted nine member companies with a questionnaire. Eight of the companies have answered the questionnaire. The results of the survey are show in Table 23. The results concern spills within the property of the companies.

In total, the eight companies use about 40,000 t raw materials per year corresponding to approximately 7% of the plastics raw materials consumption in Denmark. All of the eight companies, except one, have committed themselves to the Operation Clean Sweep and have a particular focus on reducing spills and the discharges of spills to sewage and the environment.

The reported spill varies among the companies and is on average 0.04%. Only a minor part of this spill is discharged to the drain and no companies report on direct releases to the environment.

Two of the companies (No 2 and 3) report some discharges directly to the drain (one of the companies is not committed to Operation Clean Sweep). For both companies, the estimated discharges to the drain are in the order the raw material consumption. One company reports that some releases to the drain take place but provide no quantitative estimate, and one company reports that it has installed a filter in the outdoor drain, but does not have any estimates of the quantity of spill or the fate of the spill.

If the highest emission factor of 0.0013%, indicated for one company, is used for the entire plastics conversion in Denmark, the total releases to the drain (and thereby to the public sewerage system) would be some 5 t/year.

The data demonstrates that it is possible to reduce the releases and that the measures are relatively easy, low-tech solutions. A challenge, as also described in the Operation Clean Sweep manual, is the commitment of the workers and the leaders of the company.

SPILL OF PLASTICS RAW MATERIALS FROM PLASTIC CONVERTERS IN DENMARK, ALL BUT ONE HAVE COMMITTED THEMSELVES TO OPERATION CLEAN SWEEP, SURVEY UNDERTAKEN BY THE DANISH PLASTICS FEDERATION

Company No	Consumption t/year	Spill t/year *	Spill % *	Any risk for spill being discharged to sewage or directly to the environment (as indicated by the companies)
1	65	0.600	0.92%	No. The production hall has no drain. Handling of pellets take place indoors
2	15,000	15.000	0.10%	Yes. Typically to the drain - estimated at a few hundred kilograms per year
3	1,700	0.550	0.03%	Yes, <10 kg. No drain in the production hall and the drain cover outdoors is closed
4	1,100	0.130	0.01%	No, it is not possible. Gratings on all drains that may come into contact with pellets.  Developed plug for raw material container to avoid spillage during fill up
5	400	0.052	0.01%	No. We sweep the refuse
6	6,100	?	?	Installed filter in rainwater drains
7	15,000	0.036	0.0002%	Yes. No drains indoor. Experiments with filters in the outdoor drain.
8	1,000	0.260	0.03%	No. The refuse is vacuum cleaned

Includes solid and liquid waste

The answering companies, however, represent nearly all companies in Denmark who have committed themselves to the Operation Clean Sweep (June 2015), and the releases from these companies are likely lower than the average releases from plastics converters in Denmark.

The plastic raw materials are either transported in closed containers (where no spill from transport is expected) or as bags on pallets. The pallets may be transport by trucks, trains or ships and some spill may occur by transferring from one mean of transport to another. No data are available, but some leaks from bags may happen.

The Statutory Order of Environmental Permitting includes standard requirements for plastics converters (D 208). The requirements do not identify releases of plastic pellets as a significant risk of contamination of the environment or sewage and do not indicate any specific requirement regarding spills of solid raw materials; they only address spill of liquid raw materials and auxiliaries (BEK no 682 of 18/06/20145).

The OECD emission scenario document (ESD) for plastics additives (OECD 2004) provides some default worst case emission factors for various plastics additives, but not for the preproduction pellets. For "polymeric impact modifiers" which are polymeric powders, the ESD suggests for raw materials handling an emission factor of 0.6% to solid waste/sewage for powders of particle size <40 µm and 0.2% for powders of particle size >40 µm. The emissions by raw material handling may be split between the compounding step and the conversion step (which may take place in different countries).

For compounding, the emission factors are 0.05% for powders of particle size <40 µm and 0.01% for powders of particle size >40 µm. The emission factor for the conversion process is o (zero) for all pathways. For powder of >40 µm, the total emission factor is thus 0.25% (2.5 gram released/kg

<sup>&</sup>lt;sup>5</sup> https://www.retsinformation.dk/pdfPrint.aspx?id=162486 (in Danish)

handled) which may end up in the sewage. The total emission factor of 0.25% covers all processes including production and compounding of the raw materials and the shipment, which for the most part will not take place in Denmark. Please note that the default worst case emission factors represent worst case for a single company, not a worst case average for the sector.

In the absence of actual data from companies which have not committed themselves to Operation Clean Sweep, the authors estimate that the average emission factor for the sector would be within a range of 0.0005% (half of the highest reported factor) and 0.01% (ten times the highest reported). This estimate also includes any leaks outside the premises of the companies.

**Total estimated releases** - Based on the estimated consumption and the used emission factors, the total release of preproduction pellets to sewage is estimated at 3-56 t/y. Using the distribution factors for microplastics in sewage as described in section 5.3, the total releases to the aquatic environment from the use of preproduction pellets is estimated at 0.2-5.6 t/y while 2-28 t/y is applied to agricultural soils along with sewage sludge (the remaining part of the sludge is incinerated).

#### 5.1.3 Primary microplastics for use in paint

The application of primary microplastics in paint is described in section 3.1.4, which is largely based on information collected as part of this study from manufacturers of paints in Denmark via the Danish Coatings and Adhesives Association (DFL).

#### **Application and consumption in Denmark**

The total content of microplastic particles in paints sold in Denmark has been estimated by DFL based on answers from member companies. Not all members have answered the request and DFL has extrapolated the obtained answer to cover the entire market.

The main application area of microplastic particles in paint in Denmark is in building paint incl. lacquers for floors. In wall paint, the microplastic particles are in particular used as white pigment extender and to lower the density of the paint, while in the lacquers for floors, the pigments are used to increase the hardness, provide greater durability and scratch resistance. The total sale of building paints by DFL Member Companies in 2014 is estimated at 64,000 tonnes. By extrapolation of obtained answers, the total amount of microplastic particles is estimated at 254 tonnes, corresponding to 0.4% of the weight of the paint. The members of DFL represent approximately 90-95% of the total Danish market of building paints. Considering the uncertainties regarding the results, the total content of microplastics in building paint sold in Denmark is estimated at 200-350 tonnes. As described in section 3.1.4, some of the particles used as white pigment extenders are in fact just below the  $\mu$ m-range, and do not fall under the definition of microplastics as applied here. However, no attempt has been made to estimate how much of the 200-350 tonnes is represented by particles with a diameter of just below 1  $\mu$ m.

In some niche products, microplastics particles are used for some pop-up colours (e.g. with Expancel microspheres) and in glittering colours (e.g. with polyester glitter). The microplastics content of these products is according to DFL typically 10%. The total content of microplastics in sold niche paint products is estimated by DFL to be <0.5 tonnes (note that these types of particles may be used for other applications as well).

#### Release pathways and total releases

Microplastics used in the production of paints may be lost during transport in the same way as the losses of preproduction pellets described in the previous section. Furthermore, the particles may be lost in the application of the paints before the binder is cured (hardened). In the cured paint, the plastics particles will be bound in the polymer matrix and are assumed to be released as part of the cured paint (i.e. not as unbound particles) e.g. by abrasion and maintenance (see section 5.2.3).

Emission factors for the manufacture of paint are available from The European Council of Paint, Printing Ink and Artists 'Colours in the form of Specific Release Categories (SPERCs) emission factors (CEPE 2013). For the standardized supply chain communication of environmental assessments under REACH, a number of industry sector groups and trade associations have developed SPERCs, which describe typical operational conditions that are relevant with regard to the emissions of substances to the environment (CEFIC 2012). The SPERCs generally represent "good practice" where risk management options are considered (CEFIC 2012). For emission from diffuse sources, such as releases from the application of paints, the emission factors from SPERCs and are generally considered "realistic/reasonable".

For solids used in the formulation of paints, the applied emission factor for emission to waste water is 0.0097% (CEPE 2013). In Denmark, industrial waste water (as further described in 5.3 will usually be pre-treated by a flocculation process before it is sent to a municipal STP, and it is estimated that the microplastics particles reaching the municipal STP are insignificant.

The main release pathway is estimated to be releases to sewage from the use of water-based paints and wood preservatives by cleaning of equipment and to air by application by spray (if relevant for the types of paint). CEPE SPERCs use different emissions factors for paint used by consumers, professionals and industry, and apply the following emission factors: Consumer use, all applications and professional application by brush and roller: 1% to sewage for all applications and 0.5% to soil for applications outdoors. Professional application by spray: 2.2% to air, 2% to sewage, soil indicated as "to be advised".

Poulsen *et al.* (2002) assessed the waste generation (including discharges to waste water) and environmental impact of the application of paints by consumers. The study found that the actual waste generation (spillage, remaining paint in the tools, etc.) from paint jobs typically accounted for 8 to 30% of the purchased amount of paint. In most of the waste scenarios, the waste generated was disposed of as follows: between 65 and 97% of the generated paint waste ended up as solid waste, while 3 to 35% of the total waste (corresponding to an average of 4% of the purchased paint) ended up in the sewer system. Regarding outdoor paint jobs without any covering material, the spillage to the ground was estimated at approximately 1% of the total consumption of paint. The assessment demonstrated that the releases from consumers' painting were greater than from professional painting, because the professional painters generally apply waste reducing routines.

Based on this, an average emission factor for all types of releases to sewage of 1-2% is applied. The releases outdoors are assumed to be spill where the spilled paint cures on the ground and the release of free microparticles is considered to be small and that this release will ultimately add to the releases of secondary microplastics from paint.

The total release of microplastics in paint to sewage is estimated at 1.0-5.3 t/y. Using the distribution factors for microplastics in sewage (taking into account that some of the sewage is not treated) as described in section 5.3, the total releases of primary microplastics to the aquatic environment from the use of paints is estimated at 0.1-0.5 t/y while 0.5-2.6 t/y is applied to agricultural soils.

# 5.1.4 Blasting abrasives

A general description of the use of plastic particles as blasting abrasives is provided in section o which is largely based on information collected as part of this study from Danish suppliers of blasting abrasives.

#### **Application and consumption in Denmark**

Several Danish companies offer sandblasting solutions using microplastics as abrasive material, described in section o. The application areas in Denmark, according to the interviewed companies are building sanitation (e.g. removal of PCB), removal of paint from air planes, cleansing of airplane rims, cleansing of moulds used in the manufacture of plastics or rubber, cleansing of tanks used in

the off-shore industry and in the marine industry, cleansing aboard ships, cleansing of turbine blades at power plants, trimming of Bakelite materials and removal of graffiti from walls. Microplastic blast media is mostly used in cases where a mild blast material is necessary.

All of the contacted companies state that the consumption of plastics for sandblasting is very low compared to other materials, such as corundum, stainless steel beads or glass beads. This situation is mainly due to the relatively high cost of using the plastic blast media. Based on the information from the contacted companies, it is estimated that the consumption of microplastics for use in sandblasting in Denmark is in the range of 5-25 tons per year.

#### **Potential releases**

As previously mentioned, in Denmark, blasting activities usually occur in closed or semi-closed units (e.g. cabins), where the microplastic blasting media are collected by vacuuming or ventilation systems. If the blasting occurs outside, the area must be covered with tarpaulins or similar. Emissions to the environment are assumed to be low, but may occur when the plastic media is used outdoors, e.g. when transferring airplane to and from the hangars where the sandblasting occurs, or due to ineffective or sloppy handling and collecting routines. Use for cleansing aboard ships and cleansing of turbine blades at power plants may result in some releases to the aquatic environment.

No specific emission factors for blasting abrasives have been identified, and with uncertainties on the distribution of the consumption between different applications, the total releases can only be estimated with very high uncertainties. For uses with the possibilities of direct releases e.g. cleansing aboard ships, some percent of the total may be released to the aquatic environment, but such applications only account for a minor part of the total consumption. In the absence of actual data it is estimated that 0.5-5% of the total use is released to sewage and the aquatic environment, respectively, corresponding to a total of 0.05-2.5 t/year.

Using the distribution factors for microplastics in sewage for the half of the releases (taking into account that some of the sewage is not treated) as described in section 5.3, and assuming 100% is released directly to the aquatic environment, the total releases to aquatic environment from the use of blasting abrasives are estimated at 0.03-1.3 t/y while 0.03-1.2 t/y is applied to agricultural soils with sewage sludge.

# 5.1.5 Rubber granules for artificial turfs and other applications

As described in section 3.1.6, the diameter of rubber granules from shredded tyres varies between 0.7 and 3 mm (Genan 2015), which thereby classifies the rubber as primary microplastics as defined in this report. The granules are used as infill for artificial turfs for football, rugby, tennis and golf fields. This application is assessed to lead to the most significant release and described in further detail below. Combined with a binder, the granules are also used for playgrounds, athletic tracks and similar applications. Rubber granulate is mixed with polyurethane, and the playground or the track is built on the spot (Genan, 2015). Rubber granules may furthermore be used as elastomer bitumen and asphalt modifiers in concentrations about 10% (Genan, 2015), but according to major suppliers, this application area is very small in Denmark.

The total consumption of rubber granules and powder in Denmark is estimated at 10,000-15,000 t/year, which is nearly 100% used for artificial turfs, playgrounds, athletic tracks and similar applications whereas a very small part is used in the rubber industry. The production of rubber granules is higher, but a significant part of the produced granules is exported.

Artificial turfs are used for football, rugby, tennis and golf fields, as well as for smaller areas such as roadsides and playgrounds (DHI 2013) and the use of artificial turfs is increasing, for example for use in football fields, where the number of artificial turfs has increased from 45 in 2007 to 191 in 2013 (DHI 2013). According to the website of the Danish Ballgame Union (Dansk Boldspil-Union,

DBU), there are currently 254 registered artificial football fields in Denmark<sup>6</sup>. However, as artificial turfs are also used in other applications, it is expected that the number of artificial turfs in Denmark is considerably higher.

Most artificial turfs used in Denmark are so-called 3<sup>rd</sup> generation fields, which contain an infill of a mixture of sand and resilient rubber granules. The 3<sup>rd</sup> generation fields are built with a drainage system in the bottom. The synthetic grass mostly consists of plastic fibres attached to a perforated polypropylene or polyester fabric. A latex-based glue is applied to the fabric, which is then cured. Infill is used between the fibres, in order to stabilise the fibres as well as to achieve the desired functional characteristics.

Most infill granules used in Denmark are made of recycled rubber from various tyres (SBR, styrene-butadiene-rubber), but other types of rubber are also being used, such as granulated tyres coated with polyethylene, residues from industrial rubber production, ethylene propylene diene monomer (EPDM) and thermoplastic elastomer (TPE). Infill based on natural fibres, such as cork or coconut, is also used in some cases (DHI 2013; Nilsson *et al.* 2008). The lifetime of an artificial turf used in a regular football field is approximately 10 years.

Besides the infill material itself, which is a source of releases of primary microplastics, secondary microplastics may be formed from wear and tear of these granulates as well. Formation of microplastics from wear and tear of the artificial grass fibres may also occur.

No studies on the release of microplastics from artificial turfs have been identified. However, several studies have investigated the release of hazardous compounds from the artificial fibres and infill (e.g. DHI 2013; Nilsson *et al.* 2008).

Rubber granulates are also used for running lanes, shock absorbing mats for playfields and for rubber asphalt used for playgrounds. The running lanes are made of a layer consisting of rubber granulate made from either used tyres or new rubber, combined with polyurethane as a binder. This is coated with a layer of EPDM with polyurethane as a binder material. The shock absorbing mats and the rubber asphalt consist of the same material as the running lanes, usually with a coating of EPDM (Borgersen and Åkesson 2012).

#### Release pathways and estimated releases

Wear and tear of the artificial grass fibres as well as spreading of the infill material may result in the release of microplastics to the environment. The following release pathways are considered for infill granulate and abrasions from artificial grass fibres:

- Release to surrounding soil area.
- Release to paved areas surrounding the field, and subsequently release to sewerage system via grates (includes releases from shoes and clothing).
- Release of infill particles to the indoor environment, as the particles get stuck in sportsbags, shoes and clothing where they 1) are removed by vacuum cleaning or 2) are released to sewerage system via discharges from washing machines.
- Release to drainage via drainage water. The fate of the drainage water is: 1) downward seepage; 2) release to sewerage system or 3) release to nearby streams due to heavy rainfall.

<sup>&</sup>lt;sup>6</sup>http://www.dbu.dk/klubservice/Kampe og baner/kunstgraes fodboldbaner/Find en kunstgra esbane

Around 100-120 tonnes of rubber infill granulates are used for a regular football field. Parts of the infill granulates will disappear from the field to the surrounding area and must therefore be continuously replaced, while replacement sometimes is necessary due to compression of the infill granulates on the field. It is estimated that the consumption of infill granulates is 3-5 tonnes per year for a standard football field (DHI 2013). It is assumed that the release is equal to half of the consumption of infill granulate i.e. 1.5-2.5 t/year. This equals a total release of infill granulates of 380-640 t/year from all 254 artificial football fields in Denmark.

Furthermore, microplastics will be released from the artificial grass fibres due to wear and tear. It is estimated that 5-10% of the grass fibres are abraded and released per year. According to a report from the Norwegian Institute for Water Research (NIVA) (Källquist 2005), the amount of grass fibres is equal to  $0.8 \text{ kg/m}^2$ . A standard football field is  $7,140 \text{ m}^2$ ; therefore, the amount of fibres is 5,712 kg/field. The release of microplastic particles from the grass fibres from all 254 artificial football fields in Denmark is therefore approximately equal to 70-150 tonnes.

The total release of microplastics (infill granulate and abraded fragments from artificial grassfibres) is therefore 450-790 t/year. However, rubber granulates are furthermore being used in other applications, such as running lanes, rubber asphalt/shock absorbing mats used for playgrounds etc., and it is therefore estimated that the total release from these applications, together with the use in artificial turfs for other applications than football fields, is approximately a factor 2 greater than the upper estimated release from the artificial football fields. Therefore, it is estimated that 450-1,580 t/year of microplastics is released from such applications annually.

No data have been identified to quantify the share of microplastics released via the above-mentioned pathways. However, direct releases to drainage water are considered negligible, except in cases where the fields are surrounded by grates leading to the drainage water. Releases to the surrounding soil are considered the main route for the infill granulates, but no further information on the fate of the granulates on the surrounding soil has been identified. Releases to sewerage systems is also considered possible in the following ways: 1) via grates in the surrounding paved areas, in cases where the users of the fields are carrying the granulates in their shoes and clothes, and 2) during laundering of clothes. It is assumed that 5-20% of the released material ends up in sewage. For the part that reaches the sewer, it is estimated that 3-6% ends up in surface water, since the granulates account for >80% of the total amount released and the sizes of the granulates are >300  $\mu$ m (see section 5.3.1), giving a total release to surface water of 1-20 t/year. Consequently, 94-97% ends up in sludge, i.e. 20-310 t/year. Assuming that 55% of the sludge goes to agricultural soil, 10-170 tonnes of microplastics from artificial turfs is estimated to end up here annually.

It should be noted that the releases from the use of rubber granulates are many times less than the releases from the tyres before they are shredded.

# 5.1.6 Other applications of primary microplastics

Other applications of microplastics are described in section 3.1.7. Within the limits of the project it has not been possible to provide estimates on all uses and potential releases in Denmark.

# Microbeads of expanded polystyrene (EPS) in furniture and for insulation

EPS pellets may be used for furniture (beanbags), pillows and for hollow wall insulation. Direct releases to sewage or surface water are considered to be small, as the EPS pellets in most cases only are released from articles e.g. in the case of leakage from a damaged beanbag or spill during DYI application of the pellets. In these cases, it is assumed that the vast majority of the pellets are collected using a vacuum cleaner due to the low density of the pellets and the difficulties in handling. When used as insulation material by professionals, it is also assumed that the pellets only are released in cases of spill, and that the majority is collected as waste, without reaching any water bod-

ies. Thus, releases from these applications have not been considered further in this report due to the lack of significant direct release pathways to either sewage or surface water.

#### Plastic beads for commercial potwashing

As mentioned in section 3.1.7, plastic granules are used in some commercial potwashers. Potwashers using this technology are marketed in Denmark. The plastic beads are sold in 15 litre containers, indicating that the total quantities used are limited and that the use has not been further investigated. A supplier to the Danish market has been requested for information on the fate of the plastic particles used in the washing machines and the total volume of particles for the Danish market. The total consumption is less than 0.1 t/year and according to the supplier, the majority of the beads are disposed of as solid waste when the beads are replaced in the machines.

#### Cleaning and maintenance products

The enquiry was sent to Members of SPTs manufacturing and/or marketing washing and cleaning agents. All answered that they do not use microplastics in these products except for in some professional hand cleaning products included in the estimates in section 1.1.1.

#### Medicine and research

The releases to the environment from se in medicine and research are considered insignificant and will not be further investigated.

#### Oil and gas exploration

Possible use of microplastics for oil and gas exploration in the Danish part of the North Sea has been investigated by the Danish EPA which monitors the releases of chemicals from the oil and gas sector. No data on the use of microplastics particles for oil exploration are reported from the sector.

#### Laser printer toner

Printer toners for laser printers are usually based on thermoplastic polymers, which are heated and adhere to the paper. According to Sundt et al. (2014), the powder consists of polymer particles of a diameter of about 2- 10  $\mu$ m. Toner for laser printers are not covered by specific commodity codes in the trade statistics and no statistical data on the total use of printer toner in Denmark has been identified, but likely the total used volume is in the order of magnitude of thousands of tonnes. Spill of toner products and dust generated from the printing process may add microplastic particles to the indoor environment. The majority of the dust is expected to be vacuum cleaned, but a part of the dust may be released to the sewer by wet wash. The quantities could potentially be significant.

# 5.1.7 Summary on consumption and releases of primary microplastics in Denmark

Data on the quantified consumption and releases of primary microplastics by application area in Denmark in 2014 are summarised in Table 24. For some applications, it has not been possible within the limits of the project to establish an estimate of consumption and releases. Applications for which releases to waste water may be significant, but likely not exceeding the quantified releases, are use of microbeads of expanded polystyrene (EPS) in furniture and for insulation, plastic beads for commercial potwashing, oil and gas exploration and laser printer toner.

TABLE 24
SUMMARY OF COSUMPTION AND RELEASES OF PRIMARY MICROPLASTICS IN DENMARK

Product group	Total consumption (content of sold products),	Total releases, total lifecycle, t/y *	Main pathways	Resulting release to the aquatic environment, t/y **
Personal care prod- ucts	10-30	9-29	Sewage	0.5-4.4
Raw materials for plastics production	540,000-560,000	3-56	Sewage	0.1-4.5
Paints and wood preservatives	200-350	2-7	Sewage	0.3-1.8
Blasting abrasives	ng abrasives 5-25		Sewage	0.03-1.3
Rubber granules	ibber granules 10,000-15,000		Soil	1-19
Other applications	?	?	?	?
Total quantified 550,000-575,000 (rounded)		460-1,670	Soil, sewage	2-31

<sup>\*</sup> Total releases to sewage and the environment; does not include releases to solid waste.

#### Primary microplastics in industrial sewage with direct discharges to the environment

None of the Danish manufacturers of plastic items, paints and wood preservatives or cosmetics has direct discharges to the aquatic environment in Denmark. According to the Danish inventories of releases from point sources, point sources with direct discharges to surface water in Denmark mainly include industries in the food and feed sector, airports, sewage treatment plants, shipyards, land-fills/deposits and power plants (Danish Nature Agency 2015a). The available information on the use of plastics blasting abrasives indicate that use in shipyards is not a significant application area but it cannot be ruled out that some releases directly to the aquatic environment or in sewage from shipyards takes place.

Based on the above, direct releases of primary microplastics from industrial users of primary microplastics are considered to be small and the releases are not further quantified. Contrary to this, several industrial sources of secondary microplastics exists as further described in section XX

# **5.2** Formation and releases of secondary microplastics

# 5.2.1 Formation from tyres

During their life, tyres can lose small particles due to abrasion and these particles can be classified as microplastics under the definition used in this study. The polymer parts of the tyres typically consist of a complex mixture of synthetic rubber (elastomers) and natural rubber with different filler materials. Evans and Evans (2006) states in a publication on the composition of tyres that a common-sized all season passenger tyre made by Goodyear contains: 30 kinds of synthetic rubber (elastomers), 8 kinds of natural rubber, 8 kinds of carbon black, steel cord for belts, polyester and nylon fibre, steel bead wire, 40 different chemicals, waxes, oils, pigments, silicas and clays. Elastomers are synthetic polymers, which are turned into an elastomer through the linking of polymer chains by sulphur bridges.

<sup>\*\*</sup> For the releases to the aquatic environment it is estimated that a part of the microplastics discharged to the sewer system are ultimately released to the aquatic environment (indicated as resulting releases). The percentage depends on the size of the particles; reference is made to the sections describing the various uses.

The content is slightly different between passenger car tyres, lorry tyres and "off the road" (OTR) tyres, with a total carbon based material of 74% and 76% in the passenger car and OTR tyres, respectively, and 67% carbon based material in lorry tyres (Evans and Evans 2006). The typical composition of a passenger car tyre is as follows (Evans and Evans 2006):

Rubber/elastomers: ~47%

• Carbon black: ~21.5%

Additives: ~7.5%

Metal cord: ~16.5%

Textile: ~5.5%Zinc oxide: ~1%Sulphur: ~1%

• Total carbon-based material: ~74%

The rubber/elastomers are members of the family of styrene-butadiene rubbers (SBR).

Tyre wear particles are mainly generated by shear forces between the tyre thread and the road surface and are predominantly coarse particles in the size range of 2.5-10  $\mu$ m (PM<sub>2.5-10</sub>) but may also be generated by volatilization (as reviewed by Pant and Harrison 2013). The particles are formed from the outer parts of the tyres and consist of a matrix of rubber/elastomers with carbon black and other additives embedded in the matrix. Other sources indicated that the particles might be larger. Adachi *et al.* (2004) studied tyre dust from street dust samples sieved though a 149- $\mu$ m screen. The length of the selected particles ranged from 220 to 1,230  $\mu$ m. The embedded particles of black carbon and heavy metals in the tyre dust were typically below 2  $\mu$ m. The released particles are here considered microplastic particles with additives comparable with other plastics with high content of additives (e.g. PVC) and the total particle releases are considered microplastics (in contrast to Sundt *et al.*, 2014 which considers only the polymer part as the plastics).

#### Release pathways and estimated releases

The total releases of microplastics from tyres (tyre dust) may be estimated using two approaches:

- Estimate based on vehicle-type-specific emission factors per vehicle-km, and the total road transport in Denmark
- Estimate based on consumption of tyres in Denmark and percentage of the abrasive loss (in percentage) during the service life of the tyre.

Using emission factors from a Russian study, provided as an informal document for The United Nations Economic Commission for Europe (UNECE) (GRPE, 2013) and Danish transport intensity data, the total emission of tyre dust from vehicle transport is estimated at 1,915 tonne/y (see Table 25). The emission factor for passenger cars from the Russian study is lower than reported in a UK study (Pant and Harrision 2013), where an emission factor of 0.1 g/vehicle km is used (based on a review of several studies using various emission factors). If the emission factor of 0.1 g/vehicle km is used for the passenger cars, the emission from the passenger cars can be estimated at 3,580 tonnes.

TABLE 25
EMISSION OF TYRE DUST IN DENMARK ESTIMATED ON THE BASIS OF EMISSION PER VEHICLE-km

Vehicle type	Emission factor g/km *	Transport intensi- ty in 2014, billion km/year **	Total emission, tonne/year
Passenger car	0.033	35.8	1,181
Light commercial	0.051	7.4	377
Commercial	0.178	2	356
Total			1,915

- \* GRPE 2013.
- \*\* Trafikstyrelsen 2015.

An alternative estimate is based on the following information on the consumption of tyres in Denmark. Tyres are not produced in Denmark and are either imported together with the vehicles or as separate tyres. According to data from the organisation Dækbranchens Miljøfond (2015) the total sale of separate tyres in Denmark in 2014 was about 45,000 t and around 38,000 t was collected for treatment. The quantity of new tyres is estimated from the tax revenue for tyres combined with estimated average weights of the new tyres. If assuming a steady-state situation, the quantity of the used tyres collected is equal to the number of new tyres entering the country (a couple of years back), i.e. the annual consumption of tyres. The annual consumption of tyres is therefore approximately equal to 38,000 t + 10-15% (the weight of a new tyre compared to a used one, see below), i.e. 41,800-43,700 tonnes/y. It should be noted that this estimation does not take, for instance, an increase in the fleet of cars in Denmark into account. Slightly different estimates on the percentage of the tyres released during its entire service life are available:

- By comparing the average weight of new tyres with the weight of used tyres, based on data from Dækbranchens Miljøfond it can be estimated that on average 17% has been removed by abrasion of the tyre before it is sent for recycling.
- Environment Agency (2005) assumes that as a worst case that 15% of the rubber of the tyre is lost during service life.
- Pant and Harrison estimates that up to 10% of the tyre will be lost during service life.
- For re-threaded tyres in Norway, each has an average life of 2-4 years and the weight when collected is about 10-15% less than new tyres, or re-threaded tyres. (Sundt *et al.*, 2014).

The losses from the vehicles' last set of tyres before it is disposed of would on average not be more than half of tyres used during the vehicle's service life. Based on the available data it is estimated that 10-15% of the tyre is abraded during its service life and the total emission of tyre dust (loss of tyre) in Denmark in 2014 is thus estimated at 4,200-6,600 tonnes. This is quite well in accordance with estimates based on the UK emission factors quoted above and considered reliable.

The tyre dust will either be spread to the surroundings by the wind or be washed off the pavement by rain.

The total area covered by paved roads in Denmark was in 985 km² in 2001 (Hvidberg and Studnitz 2001) and is currently just below 1,000 km² (Den Store Danske Encyclopædi, 2015). According to the Danish Nature Agency (2012), the paved areas with sewer systems was approximately 770 km²; of this, 350 km² have a combined sewer system and the remaining 415 km² have separate storm water systems. Approximately 16% of the paved area in Denmark is consequently not connected to a sewerage or a storm water system. The significant part of dust from the tyres will be generated outside areas with sewer systems; here, the dust will mainly be released to the surrounding soil while a minor part is released to surface water e.g. when driving on bridges.

Some of the dust on paved areas with sewer system is discharged to the sewer system during rainfall but a part may be spread to adjacent soil by the wind. The percentage spread to the soil depends on the particle size and is e.g. reported to be higher from brake lining dust (with smaller particle size) than for tyre dust (Sörme and Lagerkvist 2002). Sörme and Lagerkvist (2002) estimates for Stockholm that 40% of the tyre dust ends up in run-off water, while the remaining is released to soil. This distribution will here be considered realistic for the part released in areas with sewerage system.

For the separate sewer, 34% are connected to a sedimentation basin, where it is estimated that 40% of the emitted tyre dust would settle. It is therefore assumed that 80-90% of the tyre dust ending up in the separate sewer is released to surface water, while 10-20% is retained in the sedimentation basin.

The total emission is estimated applying the following emission factors:

TABLE 26
EMISSION FACTORS USED FOR EMISSION ESTIMATE FOR TYRE DUST

Sewerage type	Area, % of km² total area			Emission factor for dust emission, %			Emission fac part reachin system	
			Soil	Surface water	Sewage	Surface water	Sludge*	
Drain to combined sewer	350	38%	50-70%		30-50%	15-25%	75-85%	
Drain to separate sewer	415	45%	50-70%		30-50%	80-90%	10-20%***	
Without drain to sewer **	150	16%	95-98%	2-5%				

- \* 55% of the sludge is applied on agricultural soil
- \*\* Calculated by subtraction of total road areas with the area with sewer, the area without drainage can be estimated at 230 km<sup>2</sup>. The figure used here is applied from a recent report on discharges from paved areas (Petersen *et al.* 2013).
- \*\*\* In this case sludge refers to the sediment in the sedimentation basin.

Under these assumptions, the total emissions to surface water is estimated at 500-1700 t/year, to roadside soil 2,400-5,000 t/year, while 220-770 t/year is released to agricultural soil from application of sewage sludge.

The particles are reported predominantly to be in the range of 2.5-10  $\mu$ m. Particles of this size would not be included in the available investigations of microplastics in sewage treatment plants and the environment, which may explain why black rubber particles are not more abundant in the samples.

Tyre dust has been demonstrated to be the main source of carbon black releases to sewage treatment plants and the environment in Denmark (Gottshalk *et al.* 2015) and the main source of zinc to sewage treatment plants in Stockholm, Sweden (Sörme and Lagerkvist 2002). Furthermore, tyre dust may contain residual octylphenol and heavy metals.

#### **5.2.2** Formation from textiles

Textiles made of synthetic polymers represent one source of microplastics. Microplastic fibres are shed from synthetic textiles, such as clothing, curtains, furniture, carpets and other household textiles (e.g. synthetic cleaning cloths) during everyday use. The microplastics particles are formed and

released from the textiles due to wear and tear of the products during normal use and during washing, where textile fibres are ripped loose in laundry machines.

As described below, the main sources of microplastics fibres discharged with sewage are considered to be textiles and non-woven cloths used for cleaning purposes, and the description will focus on these two groups.

About 89,000 tonnes, corresponding to 16 kg per capita of new clothing and household textiles, are put on the Danish market each year for consumption (Palm *et al.* 2014). The distribution of the total consumption between different textile fibres shows that textiles made of synthetic fabric (viscose, polyester, acrylic, polyamide, polyurethane and polypropylene) cover 45% of the total consumption, based on figures from a British study from 2009 (WRAP 2012). These values are assumed to be relatively similar to Nordic consumption (Nielsen and Schmidt 2014). A German investigation puts the market share of synthetic fibres in 2010 at 59% (IVC 2012 as cited in Essel *et al.* 2015), whereas a Norwegian study estimates that more than half of the textiles used today are plastic polymer based (Sundt *et al.* 2014). For the further calculations, it is assumed that synthetic fibres account for 50% of the clothing.

Synthetic cloths used in kitchens are included in section 5.2.8 along with other articles used in kitchen.

#### Release pathways and estimated releases

The following pathways for releases of synthetic fibres are considered:

- Fibres discharged by laundering of clothing and linens, in households and industrial laundries: to sewerage system
- Fibres released from the use of synthetic cloth used for wet cleaning in households and institutions: to sewerage system
- Fibres releases from synthetic clothing in use, carpets, carpet tiles, furniture, household textiles, etc.: 1) removed by vacuum cleaning, 2) discharged to sewerage system by wet wash of floors and furniture surfaces, or 3) released to the outdoor environment
- Fibres released outdoors from clothing and outdoor textiles.

A small portion of the fibres released from synthetic clothing in use, carpets, carpet tiles, furniture, household textiles, etc. will be discharged to sewerage system by wet wash of floors and furniture surfaces. No data have been identified to quantify the share of the fibres released by this pathway or the total amount of fibres released. The majority of fibres in the house dust is considered to be removed by vacuum cleaning. Even when floors are wet cleaned it is common to vacuum clean before the wet clean. Sundt *et al.* (2014) estimates based on an assumed dust deposition rate of 2 grams microplastics/m²/year that the total quantity of microplastics settling on floors and surfaces in Norwegian households could be above 400 tonnes per year (0.08 kg per capita per year). In addition, some tens of tonnes of microplastics in dust would be emitted to the outdoors through doors and windows. As only a small percentage of the settled dust will be released to sewage, compared to the quantities removed by laundering (estimated below), these quantities are assumed to be relatively small, and therefore not investigated further. Microplastics in household waste are not included in the present survey.

Only a few relatively data are available on the release of plastic fibres by laundering of textile. In order to assess the representativeness of these studies, the results of the studies are compared to data on fibres in the inflow to sewage treatment plants described in section 4.2. The available data only allow very rough estimations of the total releases. Below, total releases are estimated using different approaches in order to see if a result of the same order of magnitude is reached.

For the estimates, the following assumptions regarding the intensity of household textile washing are applied based on a review of electricity and water consumption for laundry washing by washing machines worldwide (Pakula and Stamminger 2010): 75 wash cycles/capita/year and 4 kg laundry per standard wash.

Browne et al. (2011) investigated the amount of synthetic fibres in washing machine effluent after washing blankets, fleeces and shirts, all made out of polyester. They found the following mean total releases for the three types per wash (40°C, 600 rpm): fleece (>1,900 plastic fibres), shirt (approx. 1,160 fibres), blanket (approx. 900 fibres) (recalculated from data in fibres per litre). The weight of the garments is not reported. If it is assumed that the weight is 0.2 kg, the total amount of fibres in a wash of 4 kg laundry of synthetic garment would be in the range of 18,000 to 38,000 synthetic fibres per wash.

Fibres >300 µm in inflow to three Swedish sewage treatment plants (Table 13) ranged from 5,000 to 14,250 plastic fibres/m<sup>3</sup>. Recalculation of these figures to estimate the fibres per wash has been done using the following assumptions:

- Total quantity of sewage in Denmark: 644 million m³ (Danish Nature Agency 2015a)
- Total number of laundries per capita: 75
- Of these, garment of plastic fibres: 50%
- Population in Denmark 5.6 million

In the total amounts of synthetic fibres (644 million m<sup>3</sup>) is divided by the total number of washes of synthetic garments (196 million), the total releases per wash can be estimated at 31,000 to 87,000 plastic fibres per wash. The data indicates that the measured concentration represents the actual situation well and even may underestimate the releases.

Sundt et al. (2014) recalculates the measured 1,900 fibres into a quantity of 280 mg, assuming an average fibre length of 5 mm and a weight of a typical textile fibre of  $300~\mathrm{g}$  per  $10{,}000~\mathrm{m}$ . The  $280~\mathrm{m}$ mg from the wash of 0.2 kg can be recalculated into 5.7 g per standard wash. Using these figures, the total annual releases from all washes of synthetic garments can be estimated at 1,100 t/year. As the average length of the fibres in the micrometre range (0.001-5 mm) is close to 2.5 mm, the total weight of the microplastics fibres are half of the 1,100 t/year. On the other hand, data based on measurement of fibres in inflow to sewage treatment plants indicated that this estimate would be on the low end.

Dubaish & Liebezeit (2013) refer to experiments which show that between 220 and 260 mg fibres were released from a single 660 g polyester garment/washing. If it is assumed that the garment is washed 19 times during its lifetime the total release would correspond to 0.74% of the weight. The 19 times represents the average a garment is washed, calculated on the basis of the total amount of garment washed per year (75 wash cycles/capita \* 4 kg/wash cycled \* 5.6 million capita), and the total amount of new textiles of 89,000 tonnes. If it is assumed that 0.74% of the 89,000 tonnes is released to sewage during the lifecycle, the total can be estimated at around 330 t/year, or of the same order of magnitude as the estimate above.

These estimates concern laundry in private households. A study on the laundry sector in Finland showed that about 10% of the annual total of textiles washed is performed in commercial or public laundries as compared to private households (as quoted in Sundt et al. 2014). Specific data for Denmark have not been obtained.

Based on the above estimations, it is estimated that, likely, the total quantity of microplastics fibres from laundering of textiles is in the range of 200-1000 t/year. Considering the large range, the contribution from other fibre sources is considered to be so small that it would not change the range significantly.

Microplastics fibres have been demonstrated in the inflow to sewage treatment plants as further described in section 0. The number of fibres in the range of 20-30  $\mu$ m was about twice the number of particles >300  $\mu$ m. Due to the higher weight of the longer fibres, on a weight basis, the majority of the fibres would be in the >300  $\mu$ m and an aggregate emission factor of 3-6% to the aquatic environment is applied. The resulting releases to the aquatic environment are estimated at 6-60 t/year while some 100-530 t/y is released to agricultural soil by application of sewage sludge.

German analysis of fibres in outflow of sewage treatment plants shows that the dominant fibre accounting for more than 50% of the total is polyester, followed by polyamide (nylon) and PP, while other polymers were not detected (Mintenig, 2014). Polyester fibres are among other applications used for fleece.

#### 5.2.3 Formation from paints other than marine

Microparticles from non-marine paints are formed from painted surfaces mainly by three mechanisms:

- Upon UV irradiation, the binder in the paint may degrade and small particles may be released from the surface.
- The paint may flake off e.g. due to weathering and changes in the underlying layer (e.g. rust on metal surfaces or expansion of wood surfaces).
- The paint may be sanded or scratched off by maintenance (re-painting) of the painted surface.

While flakes may be in the mm-size, dust generated from sanding of paint will typically be of a size below 10  $\mu m$  and would not be included in current investigations of microplastics in sewage and the environment. Koponen *et al.* (2009) studied sand dust generated from a sanding machine. The dust was in the range of 50 nm to about 5  $\mu m$  with modes (peaks) around 1 and 2  $\mu m$ . The same article cites a study of Choe *et al.* (2000) where size distribution modes around (top points) 3  $\mu m$  and around 400 nm were observed.

The paint typically consists of a polymer binder, pigments, fillers and other additives. When cured, the paint is comparable to a plastic material. The particle size of pigments and fillers is typically sub-micrometre, and it is estimated that released particles would consist of the cured binder with the pigments and filler embedded in the material and resemble particles of plastic materials.

The paint released as particles today may have been applied up to a couple of decades ago; using average consumption figures for a period of time would be the most relevant for estimating the releases today.

The consumption of paints for all applications in Denmark was, based on statistics from Eurostat, approximately 60,000 t in 2013 (as summarised by Sørensen *et al.* 2014). The dataset was extracted from the Eurostat database combining production, import and export by the PRODCOM (production statistics) nomenclature.

The Danish Coatings and Adhesives Association (DFL) keeps some statistics on the market for building paint building paints (decorative paints).

For this survey, DFL has undertaken a consultation of key actors in the market. Based on data from key suppliers (provided in litres), the market for building paint (decorative paint = indoor and outdoor paints for walls, ceilings, woodwork, concrete and metal on buildings), the total supply in Denmark in 2014 from members of DFL is estimated at 60,000 tonnes. To this total should be added 5-10% sold by companies that are not members of DFL. This figure also includes other chemical products for buildings such as some oils, adhesives, etc. It is estimated that 36% of the tonnage is

applied outdoors. Considering that the total consumption, according to Statistics Denmark, is approximately 60,000 tonnes and decorative coatings only account for a part of this amount, the total consumption of the coatings is estimated here at 45,000-50,000 t. Of this total, approximately 16,000-18,000 t was for outdoor applications.

For other applications than building paint, the consumption of paint in Denmark would not reflect the actual quantities of paint on materials and articles in Denmark. As an example, manufacture of vehicles in 2001 accounted for 5% of the total EU market of paint of 5.5 million tonnes (OECD, 2009), but none of this was used as paint in Denmark, whereas the paint on the imported cars may be released during use in Denmark. The OECD emission scenario document (ESD) for the coating industry provides some information on the EU market for paint in 2001 (OECD, 2009). If it is assumed that materials and articles used in Denmark account for 1% of the EU market in 2001, the total amount of non-decorative paints would be 18,790 t, split out into automotive use and vehicle finishing (3,850 t), various industrial products (6,000 t), wood paints (3,300 t), powder coating (2,750 t), protective coating (1,650 t), and coil coating (1,100 t). From the description of the applications, it is not possible to estimate the share of painted articles/materials for outdoor use, but at least the protective coatings and automotive use and vehicle finishing are expected to be for outdoor use.

The ESC provides specific environmental emission factors from the service life phase for automotive, marine and "decorative paints". The "decorative paints" include (OECD 2009):

- Interior wall and ceiling paints for plaster, etc.
- Exterior wall paints for masonry, brick, etc.
- Interior/exterior wood/metal primers, undercoats and finish paints.
- Interior/exterior stains and varnishes for wood.

The "decorative paints" also include the protective coatings.

To the estimate for building paints above, some 2,000-5,000 tonnes protective paints and other types of paint used outdoors are therefore added and the total for outdoor paints is estimated at 18,000-23,000 tonnes. The average dry matter content of building paint is usually about 45-60% by weight. Furthermore, a portion of the applied paint is disposed of as waste by the application of the paint. The ESD assumes for decorative paints used by consumers that 26% is disposed of during the application phase (left in paint cans and spill), while the percentage for professionals is assumed to be 4%. It will be assumed in this study that the average for all applications is 10-15%. On this basis, the weight of the final coatings can be estimated at 7,000-12,000 t/y.

#### Releases and emission factors, indoor applications

Releases due to UV degradation and other environmental factors are assumed to be much lower for paint for indoor applications than for outdoor application. This applies both to decorative building paint and paint on painted articles. An exception may be releases from varnished flooring where the varnish is abraded by use. The released particles would largely be removed by vacuum cleaning or sweeping while a small part is released to the sewer by wet cleaning. Compared to the releases from paints applied outdoors, the releases from indoor applications are estimated to be small and are not further considered.

# Releases and emission factors, outdoor applications

Outdoor application of paint consists mainly of decorative building paints (for walls and wood/metal parts), paints on vehicles and machines and protective coatings on construction.

**Decorative paints** - For "decorative paints" (including building paint and protective paint), the emission scenario document (ESD) from the OECD assumes that approximately 3% of the coating will be lost to industrial soil by flaking/chipping during the useful life of the product (for profes-

sional applications this corresponds to 3% of the initial applied quantities, for the general public it corresponds to 2%). The ESD uses the term "flaking/shipping" for the release mechanism, and it is not clear if the emission factor also includes sanding for maintenance of the paint. The ESD assumes that by the end-of-life of the coated products and the coating is disposed of as solid waste. The life-cycle flow chart does not indicate any re-painting step. Danish substance flow analyses have usually assumed that some 3-4% of paint applied outdoors is lost to the environment by maintenance (re-painting) and abrasion during the lifetime of the paint; of this approximately 25% has been assumed to be lost to sewage e.g. from paved areas while the rest ended up in the soil (e.g. Lassen *et al.* 2003).

In addition to the paint released during the service life, spill by the application of the paint, which cures on the ground, may also contribute to the formation of secondary microplastics. In a study on spill from consumer paint applications, Poulsen *et al.* (2002) estimates that for outdoor paint jobs without any covering material, the spillage to the ground was approximately 1% of the total consumption of paint.

Here, a total consumption of 7,000-12,000 t/year of paint for outdoor applications on buildings and constructions (dry weight) and an emission factor of 2-6% will be applied (incl. spill by application). Of this, it is assumed that 10-30% of the releases is discharged to the sewerage system while 2-10% is released to surface water, either by direct emissions from buildings and constructions close to surface waters or indirectly by storm water. As the particles are relatively small, it is assumed that 15-25% of the quantities reaching the sewerage system is ultimately released to the aquatic environment. Based on these assumptions, the total releases to sewage are estimated at 140-720 t/year, while the total ultimate releases to surface water (direct and via sewage treatment plants) are estimated at 5-126 t/year.

**Automotive paint** - The ESD (OECD 2009) assumes that about 10% of automotive coating is lost to industrial soil during the vehicles' useful life due to flaking and chipping. Whereas this may have been the situation 15 years ago (the ESD builds on an older ESD on automotive painting), it is estimated that today a much smaller portion of automotive coatings will be lost during the service life of the vehicles. Assuming some that 1-3% of 1,000-3,000 t/y paint on imported and repainted vehicles is lost and the releases are distributed similarly to the releases from tyres, the total releases can be estimated at 10-90 t/y, of which 1-23 tonnes is ultimately released to surface water.

**Total releases** - The total releases from paint other than marine paint can be estimated at 150-810 tonnes on the basis of the abovementioned, while the ultimate releases to surface water is estimated at 6-150 t/year.

#### **5.2.4** Formation from marine paints

Microplastics may be formed from marine paint by the same mechanisms as described for other types of paints in the previous section. In addition to these mechanisms, some microparticles may be released from self-polishing antifouling paints by a self-polishing mechanism.

According to the OECD emission scenario document for the coating industry, there are six key areas of shipboard paints (OECD, 2009):

- Underwater (hull bottom).
- Waterline.
- Topside superstructures.
- Internal spaces and tanks.
- Weather decks.
- Loose equipment.

According to the emission scenario document, anticorrosive paints include vinyl, lacquer, urethane, and epoxy-based coating systems while typical finishes include one and two-pack polyurethanes. Varnishes used on boats are often also polyurethane based (OECD, 2009). For antifouling paints, several self-polishing copolymers are often used. Sundt et al. (2012) report that there is a movement away from polyurethane paints over to epoxy paints and even newer paint formulations with a range of various polymers. However, industry contacts for this survey inform that at the same time the trend for topcoat is towards polyurethane-based paints. Furthermore a move away from one-component acrylic and alkyd paints has been reported because on-board maintenance is becoming less common. Instead longer lasting two-component paints systems are used.

Like the situation for other paints, particles released from the coatings may be considered secondary microplastics.

No statistical data on the market for marine paints in Denmark are available and marine paints are not specifically registered in the import/export statistics. The paints are not produced in Denmark today. The marine paint market in the EU in 2001 (excluding antifouling paints), was more than 110,000 tonnes (OECD 2009). The market may be divided into a market for manufacture and maintenance of recreational boats and a market for manufacture and maintenance of commercial vessels. Based on information from suppliers of marine paints, the total market for marine paints in Denmark is estimated as follows:

- Recreational boats, hull bottom paints: 50-100 t/year
- Recreational boats, other paints: 50-100 t/year
- Vessels, hull bottom paints: 800-900 t/year
- Vessels, other paints: 3,000-3,500 t/year.

For the paints for large vessels, paint sold in Denmark is not necessarily used in Denmark, and the paint used in Denmark may be purchased abroad and brought to Denmark on the vessel. However, the quantities of marine paints sold in Denmark are used in this study as the best indicator of the actual consumption in Denmark.

# Releases and emission factors - recreational boats

The application of antifouling paints (part of hull bottom paints) on recreational boats in Denmark and the resulting releases of antifouling agents while the vessels/boats are in the water or out of water for maintenance are described in some detail in the literature. The focus has been on the releases of heavy metals and other antifouling agents. For other types of paints for recreational boats, no literature has been identified.

According to an investigation from 2009, the number of recreational boats in Danish marinas was 57,000; of these, 57% was sailing yachts while 43% was motorboats (small rowboats, canoes, kayaks, etc. are not included in the figure).

As indicated above, the market for marine paints for recreational boats is estimated at a total of 100-200 t/year equally split even between hull bottom paints and other paints. If it is assumed that 10% ends up as waste by the application (assuming that the paint is mainly applied by brush) and that the dry matter content is about 55% by weight (OECD 2009 indicated about 55% solids), the total amount of dried paint applied is 50-100 t/year.

The majority is used for maintenance work. Every 10-20 years all paint from the surface is removed (Højenvang, 2003) and the total removed from the boats is assumed to correspond to the consumption. During the life of the boats, the paint will be totally removed several times. Some of the bottom paints are self-polishing, and a part of the paint is consequently hydrolysed during use.

Procedures for maintenance of recreational boats and the resulting releases have been studied by Højenvang (2002, 2003) with the aim of developing methods and guidelines for better maintenance practices. As part of the study, the maintenance practices and the quantities of paint removed from the maintenance of the hull bottom of a typical recreational boat of 30 feet were estimated. The data were used to estimate total releases of paint from a marina by multiplying by the number of boats. Using a similar approach for all 57,000 recreational boats in Denmark, the total releases can be estimated at 78-141 t/year. This range is higher than the estimated consumption of paints for bottom hull paints in Denmark but the estimate is sensitive to the assumption that a 30-foot boat can be considered an average boat. The results, however, indicate that the total consumption is a good representation of the total releases; even a part of the antifouling paint is released when the boat is in the water.

TABLE 27
MAINTENANCE PATTERN AND PAINT REMOVED FROM A TYPICAL RECREATIONAL 30-FEET BOAT (BASED ON HØJENVANG. 2002)

	Percentages of the boats every year	Removed by the maintenance, kg/boat	Total removed from 57,000 boats, t/year *
Total removal (fibreglass repair):	1-5%	24 kg	13 - 66
Coarse cleaning	10%	10 kg	55
Fine grinding	5-10%	3.4 kg	10 - 20
Total			78 - 141

<sup>\*</sup> Estimated as part of this study

The release by the different pathways is highly dependent on the measures taken to prevent that the paint is released to the environment. The Danish Sailing Association has developed guidelines for maintenance of bottom paints and recommends that all paint dust and scrapings are collected, preferably with a vacuum cleaner. Furthermore, the standard rules for marinas and small fishing harbours stipulate that removal of biocide-containing bottom paints is only allowed in designated areas and that waste from maintenance activities (any) shall be collected and disposed of in accordance with the applicable regulations. As the focus has mainly been on the bottom paint, the procedures used for maintenance of other parts of the boat are less strict and a larger portion of the dust may be released to the surroundings.

No studies of the actual practices used today by boat owners have been identified. Sundt et al. (2014) report that in Norway, based on recent findings, less than 10% of boat owners use any paint dust collection or control system when removing paints other than antifouling paints; therefore, the expected spill of paints over time is likely to be >90% of all paints used.

The paint which is not collected will mainly be released to the ground below the maintenance site and may over time be released to the sea; the releases will here be considered releases to surface water. Under the assumption that most boat owners collect paint dust and scraping where maintaining bottom paints, but more often do not collect dust and scrapings of other paints, it is roughly estimated that 10-50% of the applied paint over time is released to surface water, corresponding to 5-50 t/year.

# Releases and emission factors - professional vessels

Marine paints for the professional market in Denmark are used by a number of small and medium sized shipyards and boatyards. The shipyards and boatyards offer new building, rebuilding of vessels and repair and maintenance. The Danish producers of maritime equipment and ships are or-

ganised in the industrial association Danish Maritime which organises 5-7 shipyards while about 20 boatyards are organised by the "Skibs- og Bådebyggeriernes arbejdsgiverforening".

No data are available for estimating the releases from vessels during maintenance activities, and in the absence of actual data, the potential releases are estimated based on the quantities of paint sold in Denmark. As mentioned above, paints sold in Denmark may not be the same as the paints used in Denmark, but the amounts sold will be used as representative of the consumption in Denmark. Some of the paints may be used for building of new vessels but the majority is assumed to be used for maintenance activities; i.e. where some of the existing paint on the vessel is removed prior to the application. Furthermore, the quantities applied are used as a proxy for the quantities removed. As for the recreational boats, not all paint is removed every time, but during the lifetime of the vessel, at least the bottom paint may be totally removed several times. In an analysis of the releases of tributyltin in antifouling paints as part of a substance flow analysis for tributyltin, Lassen et al. (1997) report that approximately in 1/5 of all maintenance cycles, the antifouling paint and the underlying primer was totally removed by sand blasting, whereas for the remaining 4/5 maintenance cycles, the surface was cleaned by high-pressure cleaning. The OECD emission scenario does not mention high-pressure cleaning, but assumes for the non-antifouling paint that at the end-oflife stage of the coating, the coating is removed by blasting. It is assumed that 90% of the coating is captured while the remaining 10% is released to the environment (half to soil and half to surface water). Total removal of paint above the waterline by maintenance seems to be more uncommon than removal below the waterline, and based on the available data it is not possible to state that coatings in general will be totally removed several times during the life of the vessel. Alternatively, the coating would simply be thicker during the life of the vessel and the coating will be removed when the vessel is scrapped/recycled.

According to the OECD emission scenario document, the paint is mainly applied by spray, and for non-antifouling coatings (the coatings covered by the emission scenario document) it is assumed that only 65% of the paint ends up on the vessel. If the same is assumed for the antifouling paints, and assuming a dry matter content of 55% for both paint types, the quantities of solid paint ending up on the vessel corresponds to 400-450 and 1,490-1,730 t/year, respectively. Of the selfpolishing antifouling paints, some 60-70% is released while the vessel is in the water (Lassen et al. 1997, quoting major suppliers of marine paints) and consequently only a quantity corresponding to 120-180 t/year remains on the vessel.

Lassen et al. (1997) report that the waste water from the docks of the larger shipyards is collected and treated by sand traps, sand filters, oil separators or other waste water treatment procedures, and that a very small part of the organotin (antifouling paint biocide) passes the filters. That is likely the situation as well as concerns the removed paint. The study reaches the conclusion that the major pathway for releases from the maintenance at shipyards is dust releases from the sand blasting. Even though some protection is used to avoid drift of the dust, it is assumed in the study that 2-15% of the total removed paint is released to the surroundings (half to soil and half to surface water). Today, it is commonly required to use wet sand blasting to prevent drift when the sand blasting is applied above the edge of the dock. This applies to the larger shipyards with dry docks whereas at smaller boatyards with a slipway releases by drift may be higher, but data has not been available.

Sundt et al. (2014, quoting a paint industry source) include airborne droplets from spray application in the estimate of microplastics releases in the survey for Norway, under the assumption that the droplets dry/cure while they are in the air and consequently end up in the environment as microplastics particles.

The OECD emission scenario document assumes as a worst case that ultimately 5% of the paint on the vessel (3.2% of the quantity originally applied) is released to soil and water by maintenance, respectively, which is in the middle of the range indicated above for Denmark. In the emission scenario

nario document it is furthermore assumed that 1.8% of the applied amount is lost to both the water and soil from the spray application of the paint.

Considering the large uncertainties and in the absence of actual measured releases, a range of 2-20% is applied as best estimate (half to soil and half to surface water). Releases for boatyards are expected to be higher than the releases from shipyards. On this basis, the total releases from professional uses of marine paints are estimated at 16-150 t/y to surface water and soil, respectively. From some yards, some untreated waste water may be released to sewage but in the absence of data this release pathway has not been considered as the resulting releases are expected to be small compared to direct releases to the environment.

#### Releases from self-polishing antifouling coating

The self-polishing behaviour of antifouling paints is achieved through an erosion process of the polymeric binder, which prevents fouling of the surface and enable release of incorporated biocides (if any) at a constant rate. When in contact with seawater, the polymer films exhibit a thin surface erosion zone controlled by hydrolysis/erosion that leads to a constantly polished surface. A question has been raised by industry as to whether the self-polishing behaviour may lead to releases of small paint particles to the water. No studies have been identified on the subject but further studies could be relevant.

#### 5.2.5 Formation from road marking materials

Formation of microplastics may be a result of weathering and abrasion of material used for road marking. Thermoplastics is the most commonly used material for road markings in Denmark, and according to a Danish manufacturer/supplier, it is estimated that approximately 98-99% of all markings on Danish roads is based on thermoplastic materials. The thickness of thermoplastic road marking is usually 2-3 mm. Paints may be used for temporary road marking and for parking lots, and the road markings in airports are usually based on paints in order to avoid fragmentation of the thermoplastics. In certain cases, marking tape may also be used for temporary road marking, usually in cases where a gentle treatment of the covering is necessary.

The composition of a typical thermoplastic material for road marking according to a Danish manufacturer is given in Table 28. Table 29 shows the composition of a water-based road marking paint according to safety data sheets from a Danish company<sup>8</sup>.

TABLE 28
COMPOSTITION OF A TYPICAL ROAD MARKING THERMOPLASTIC MATERIAL ACCORDING TO A DANISH MANUFACTURER

Component material	Content
Plastic polymer (light density PE, EVA)	~ 0.5-2%
Resin (wood-based or based on crude-oil)	~ 10-15%
Fillers (e.g. sand, dolomite, calcinated flint)	~ 50%
Glass beads (approx. 100 μm – 1.5 mm)	~ 30%
Titanium dioxide	~ 5-10%

 $<sup>^{7}\,\</sup>underline{\text{http://www.lkf.dk/lkftraffic/trafikmarkering/markering-med-maling.html}}\,\text{(in Danish)}$ 

<sup>8</sup> http://www.trafikprodukter.dk/produktkataloget/afmaerkning/vejstribemaling/ (in Danish)

TABLE 29
COMPOSTITION OF TYPICAL ROAD MARKING WATER BASED PAINT (OF THE BRAND MERCALIN ®) ACCORDING TO THEIR SAFETY DATA SHEET

Component material	Content	
Fillers and pigment	40-60%	
Acrylic polymer	15-40%	
Titanium dioxide	5-10%	
Water	10-30%	
Ammoniac	0.15-0.2%	

The fillers and glass beads appear to be significant compared to the generated microplastics particles and the dust emitted from weathering. Abrasion of the thermoplastic road marking is assumed to consist of fragments of the glass beads, filler fragments and microplastic particles consisting of the plastic polymer, the cured resin and the titanium dioxide.

#### Release pathways and estimated releases

The degradation and removal of road markings depends on several different factors e.g. the composition of the marking, the location of the marking (e.g. whether the marking is located on the edge or in the middle of the lane), the load on the marking, the weather conditions and so on. The use of snowploughs in the wintertime will also remove some of the road marking, making new applications necessary.

There are no exact data available on the applied road marking which, due to weathering and abrasion, may lead to releases of particles to the environment.

Sundt *et al.* (2014) have estimated the emission of microplastic based on an assumption that the annual consumption reflects the annual abrasion, even though it is known that some markings are ultimately removed. This is, however, assumed not to represent the Danish situation, as there are several practises that differ between the Norwegian and Danish situation. Norwegian cars are, for example, required to be equipped with studded tyres in the winter season, which has a huge abrasive effect on the road marking. Secondly, the estimated emission factor of 100% from Sundt *et al.*, (2014) inevitably will lead to an overestimation, as they did not take into account the fractions being removed by renovation of the road, whereby the material does not end up in the environment.

For the Danish situation, it is estimated that 15-25% of the annual consumed thermoplastic material is used for reapplication of existing road marking, thus indicating the level of degraded and released material (i.e. the abrasion factor is 15-25%). The remaining 75-85% of the applied road marking is used on new roads or rehabilitation of existing roads and it is assumed that 30-50% of the existing road marking is worn off due to wear and tear and weathering before rehabilitation, giving a emission factor of 23-43%. When roads are rehabilitated, the existing pavement including the road marking is ground or broken up into small pieces, and the road marking is built into new asphalt or disposed of.

The annual consumption of thermoplastic materials for road marking in Denmark is assumed to be 5,000-6,000 tonnes according to information from several of the contacted companies and manufacturers. Based on the above-mentioned composition of the thermoplastic material it is assumed that 15-27% of released particles are plastic particles (plastic polymer, cured resin and titanium dioxide), thus the estimated consumption of microplastic material in thermoplastic road marking is 750-1620 t/year. Based on the abovementioned assumptions, the abrasion factor is 15-43%, thus the estimated release of microplastic particles from thermoplastic road marking material is 110-690 tonnes per year.

It is assumed that the release pathways of the microplastics released from the road markings are similar to the release pathways of the tyre wear particles (see section 5.2.1). The same emission factors are therefore used as those presented in Table 26. Under these assumptions, the total emission to surface water is estimated at 10-18 t/year, to roadside soil 60-510 t/year, while 6-80 t/year is released to agricultural soil from application of sewage sludge.

As mentioned, paint is also being used as a road marking material but accounts for 1-2% only and is used for particular purposes. The release of microplastics from road marking paint is therefore not estimated in present study.

#### 5.2.6 Formation from building materials and furniture

Microplastic particles may be formed from abrasion and wear of plastic building materials in use

The main sources of microplastics particles from plastics items (apart from items specifically addressed in other sections) are considered to be:

- Abrasion and wear of articles and building materials used indoors (for most applications the main route is solid waste from vacuum cleaners' sweeping):
  - Flooring of PVC, PVC tiles, and other plastic materials
  - Wall covering and wall paper of PVC and other plastic materials
  - Table tops of melamine, Corian (PMMA), polyester composite and other plastics (for kitchen and bathroom table tops, release to sewage will be the main route).
- Abrasion and wear of articles and building materials used outdoors (main routes are to soil and separate and common sewer systems):
  - Roofing membranes of PVC and PFO (flexible polyolefin)
  - Plastic coatings on metal roofs (coil coated PVC)
  - Various outdoor building materials of plastics such as gutter, sheets, window and door frames.

For the possible emissions from indoor and outdoor applications of plastics, the following two approaches have been tried in order to establish some first rough estimates of the formation and releases:

- Information on plastic particles in indoor dust. Such data may be used to obtain an aggregate estimate of the formation of microplastics from various plastic items in the indoor environment. An exemption is plastic particles formed from surfaces and utensils in kitchens and bathrooms, where the formed microplastics may be cleaned off without being part of the general indoor dust.
- Available information on abrasive releases of PVC. Abrasive releases of PVC have been studied in some detail in order to estimate the releases of phthalates from the use of plasticised PVC and releases of heavy metals from the use of rigid PVC in building and construction. PVC accounts for a significant part of plastics use in building and construction and consequently the knowledge on the PVC can be used in the estimation of total releases from plastic materials. As shown in Figure 11, PVC accounts for about 50% of all plastics used in building and construction followed by EPS and PUR, both used for insulation, and HDPE used for various applications. Of the outdoor application, where the plastics may be exposed to the weather, and indoor application with high level of wear, PVC account for the major part of the applied plastics.

In addition to this, the possible emission of microplastics from kitchen and bathroom table surfaces, scouring pads and kitchen utensils of plastics is considered.

Furthermore, the generation of particles from handling of plastics materials and articles during industrial and professional use is discussed.

#### Indoor dust

A wealth of information exists on various hazardous substances in household dust and vacuum cleaner dust, but no detailed studies of the typical plastics content of the dust in households, institutions or offices have been identified. As the hazardous substances in the dust often are evaporated from surfaces and adhere to the dust particles via the air, the concentrations of these substances cannot be used to estimate the rate of abrasive releases of plastics.

Sundt et al. (2014) have tried the same approach for Norway, and note that there are some indications on the relative amounts of plastics in the indoor dust. For example, synthetic fibres were found to constitute 1-5% of household dust, and building material debris/acrylic plastic flakes 15-40% by volume in a single Boston home (acrylic plastic flakes probably mainly paint particles) (Webster et al. 2009). Similarly, high amounts of organic microparticles believed to originate from paint and textiles have been documented in several studies of indoor air, this fraction often constituting several tens of percent of total indoor dust as reviewed by Sundt et al. 2014). The results mainly document that plastic fibres (addressed in section 5.2.2) and from paint (addressed in section 5.2.3) are present in the household dust, whereas no documentation for plastic particles from plastic items/surface exist. Based on reported deposition rates of 1-8 g/m<sup>2</sup>/year, a deposition rate of 1-2 g/m<sup>2</sup>/year of microplastics, of this mainly textile fibres, is assumed for Norwegian households. Based on this deposition rate of microplastics of 2 g/m<sup>2</sup>/year, a total of 400 t/y of microplastics generated as dust in Norwegian households and in addition some tens of tonnes would be emitted to the outdoors via air. Considering the available data, the 400 t/y is a worst case, and only a small part of this would be from plastic items. The major part of the dust in households are vacuum cleaned or wiped and disposed of as municipal solid waste and only a small part would be released to the sewer system by wet cleaning.

In contract to the situation in the households, flooring in institutions and offices are often regularly wet cleaned. Due to the large surface and high abrasion rates, PVC and other plastic flooring could obviously be an important source, where a significant part of the generated dust is removed by wet cleaning. The potential abrasive releases from flooring is estimated below on the basis of information on emission factors for releases from PVC flooring

#### **Articles of PVC**

Abrasive releases of phthalates in plasticised PVC - Abrasive releases of phthalates are releases where the phthalate is released as a part of small or larger part of the material, in contrast to leaching where the phthalates are slowly released from the PVC material. The EU Risk Assessment for the general-purpose phthalate DEHP (ECB 2008) identified "waste remaining in the environment" i.e. particles and items of PVC released to the environment, as the major route of releases of DEHP. On the basis of emission factors from the EU Risk Assessment and updated data on the use of DEHP, COWI et al. (2009), in a study for ECHA, estimated the total abrasive releases of DEHP in the EU to the environment in 2007 at 4,700 t/y (approximately 25% to sewage and 75% to soil). With an average content of 30% DEHP, this corresponds to approximately 4,000 tonnes DEHP-containing PVC per year. As DEHP, at that time, accounted for about 30% of the phthalate consumption for plasticised PVC, and same emission factors are expected for PVC for the same purposes with other phthalates (mainly DINP and DIDP), the total abrasive releases of plasticised PVC can be roughly estimated at around 52,000 t/year. If Denmark represents 1% of this (approximate percentage of EU population), the total releases in Denmark would be 520 t/y. This indicates that plasticised PVC may be a significant source of microplastics in Denmark. At the EU level, the main indoor sources of emission to sewage were abrasive releases from PVC from flooring (vinyl flooring). The main sources of outdoor emissions were abrasive releases from roofing materials, soles of footwear and PVC-coated fabric.

**Flooring** - The lifetime emission factors for abrasive releases from PVC flooring is estimated at 6% based on an emission factor of 0.3% per year to sewage for 20 years with a distribution of 50% released to sewage while 50% is vacuum cleaned. PVC flooring is typically used in institutions and offices and is regularly wet cleaned. The releases today will reflect the use for flooring the last 20 years. The PVC use for flooring was in 1995 2,300 tonnes (and 3,000 tonnes to wall covering) (Plastindustrien 1996); in 2001 the PVC used for flooring and wall covering was estimated at 1,018 tonnes (Skårup and Skytte 2003). The average for 2002-2004 can (Brandt and Hansen 2009 who note that the consumption may be underestimated) be recalculated from the phthalate consumption into 960 tonnes. If it is assumed that the average consumption of PVC flooring for the last 20 years is 600-1000 tonnes and the lifetime emission factors for releases to sewage is 2-4%, total releases to sewage can be estimated at 12-40 tonnes to sewage while a similar amount is vacuum cleaned.

**Roofing** - The emission factor for lifetime emissions from calendared roofing material (relatively thick membranes, similar to flooring) is estimated at 5% while the emission factor for coil coated roofing sheets (thin PVC coating) is estimated at 50% distributed with 25% to sewage and 75% to surrounding soil (COWI *et al.* 2009). At EU level, the coil coated material accounted for approximately 80% of the PVC consumption for roofing. The PVC consumption with roofing membranes and roofing sheets (type not indicated) in Denmark in 2001 was 722 tonnes (Skårup and Skytte 2003) while the average for 2002-2004 (Brandt and Hansen 2009) was estimated at 900 t (Brandt and Hansen 2009, who notes that not all of the 900 tonnes may be roofing). If it is assumed that the average PVC consumption for roofing is about 500-800 tonnes and the aggregate emission factor is 10-30%, the resulting emission can be estimated at 50-240 t/year, of which 13-60 t/year is released to sewage and 38-180 t/year to soil. Also, plastic membranes of other materials are used for roofing e.g. of PFO (flexible polyolefins) and the total releases of plastic particles from roofing will be higher than the estimates based on PVC roofing. Data on the market of plastics roofing materials have not been sought.

#### Other PVC building materials

The consumption of other PVC building materials that may be subjects to weathering and abrasion in 2001 was: gutters and downspouts ( $\sim$ 3,000 t), roofing plates and skylights of rigid PVC ( $\sim$ 6,000 t). No data on emission factors for these materials have been identified, which are probably lower than the factors for the flexible PVC. If the lifetime emission factors for these materials are just 1%, the resulting releases would be nearly 100 t/y, indicating that the releases could be significant. The major releases are expected to occur from the mm-thin roofing sheets. Total releases are roughly estimated at 20-200 t/y distributed as 25% to sewage and 75% to soil.

Comparison PVC emission with data from sewage treatment plants - Notably, a German study of plastics composition of microplastics of 10-500  $\mu m$  in effluent from German sewage treatment plants did not find PVC in any significant amount in any of the plants (Figure 14). In this fraction, the dominant polymers were PE and PVA (polyvinyl alcohol) accounting for more than 50%, followed by polystyrene, polyester, PP and polyamide. PVC was also insignificant in the fractions >500  $\mu m$  (Mintenig et al. 2014) where PE and PP accounted for more than 90% in all plants. A possible explanation for the absence of PVC in the samples may be that the particles are below the size of particles analysed in the effluent of the sewage treatment plants. Particles generated by sanding of paint or from abrasion of tyres are reported to be in the size range <10  $\mu m$  and particles from abrasion of flooring, shoes, roofing, etc. may likely be in the same size range and would not be included be in the available investigations of polymer composition of microplastics in sewage treatment plants and the environment.

#### Kitchen and bathroom table surfaces

Microplastic particles from kitchen/bathroom table surfaces would mainly be released with sewage and would not end up in the indoor dust. No data are available for estimating the possible releases from table tops.

#### Total releases from building materials

Based on the available data, the total releases to sewage from building materials are estimated at approximately 30-150 t/y to sewage and 50-330 t/y to soil. Of the part released to sewage, the emission factor of 15-25% for relatively small particles is applied and the resulting releases to surface water is 5-38 t/y.

#### 5.2.7 Shoe soles

Shoe soles - At EU level abrasive releases of DEHP from shoe soles in 2007 were estimated at 1,940 t/year (25% to sewage, 75% to soil). Using the same approach as described above for recalculation into total PVC quantities, the total PVC releases from soles can be estimated at approximately 21,000 t/y PVC (with all types of phthalates). The estimate is based on the assumption that 10% of the soles are abraded during the life of the footwear. A major manufacturer of shoes has been contacted but the manufacturer did not hold any data on average abrasive releases from shoes. If it is assumed that the releases in Denmark account for 1% of the EU total, it corresponds to 210 tonnes. Shoe soles are typically made from PVC, polyurethane or synthetic rubber (elastomers). Data on the share of different plastic types is not available, but the total releases would be higher than the releases from the PVC soles. Considering the uncertainties on the total consumption of shoe soles and the emission factor (10% seems to be quite high), the total releases from shoe soles are roughly estimated at 100-1000 t/year. The main part of the releases is assumed to be outdoors from paved areas with sewerage systems, and the same emission factors as those presented in Table 26 for tyres are therefore used. Under these assumptions, the total emission to surface water is estimated at 10-260 t/year, to roadside soil 60-750 t/year, while 10-120 t/year is released to agricultural soil from application of sewage sludge.

#### 5.2.8 Formation from plastic kitchen utensils, scouring pads and cloths

Microplastic particles may be formed during use of various plastics cooking utensils of PE, PP, PTFE and other polymers, scouring pads of polyurethane foam/polyester/nylon or use and washing of synthetic cloths. For some of the products such as scouring pads or dish brushes (with plastic brushes), significant abrasion of the articles are visible to the naked eye and releases from these products are inevitable. Loss of material during washing of synthetic cloth also represents a common scenario. For many other articles, releases are visible as scratches in the surface (e.g. plastic chopping boards).

# Release pathways and estimated releases

Release via sewage is the main route for microplastics formed from the abovementioned products. Some of the released microplastics will be transferred to other products, e.g. plates when using the scouring pad for dishes, and some microplastic particles may be transferred to food items from the chopping boards, but these releases are assumed to be minor and not considered in the following scenario. A rate of 90-100% release to sewage is therefore considered.

Within the limits of this survey, it has not been possible to estimate the release from all of the abovementioned articles, but in order to assess whether these releases may be a significant source of microplastics to sewage, the potential releases from scouring pads and synthetic cloths are estimated, as these are assumed to account for the major part of the released microplastics in this group of products.

Scouring pads consist of a foam layer usually made of polyurethane with a scouring layer of polyester and nylon (special scouring pads with different abrasive materials are not considered in this

case). The weight of a typical scouring pad article is 4.2 g, and the contribution of foam and scouring material is 43 and 57%, respectively. The estimated consumption of scouring pads, based on sales figures from the sector of trade/industry is 30-90 million scouring pads per year, corresponding to approximately 130-380 t/year. An abrasion factor of 5-20% and 10-30% is being assumed for the sponge layer and scouring layer, respectively, giving a total estimated release of 10-100 t/year.

The most commonly used synthetic cloths in Denmark either consist of 15-20% polypropylene and 80-85% viscose (everyday use cloths) or 70-80% polyester and 20-30% polyamide (so-called microfibre cloths). Release from these either occurs during use or during laundering of dirty cloths in washing machines. The estimated consumption of synthetic cloths, based on sales figures from the sector of trade/industry, is 20.5 million–45 million for everyday use cloths and 1.7 million – 4.8 million for microfibre cloths. The weight of the cloths are approximately 23 and 40 g/piece, respectively, giving a total consumption of 470-1000 and 70-190 t/year for everyday use cloths and microfibre cloths, respectively. Combining these figures with the content of plastic material (15-20% and 100%, for everyday use cloths and microfibre cloths, respectively) and an estimated release of 10-20% and 5-10% from everyday use cloths and microfibre cloths, respectively, a total release of 10-60 t/year is assumed.

It should be noted that in this survey, semi-synthetic viscose made from cellulose is not considered as a plastic material.

Combining these estimates with an uncertainty regarding the releases from other articles in use (approximately 20 tonnes), an estimated release of 20-180 t/year from plastic kitchen utilities, scouring pads, synthetic cloths and similar products is assumed. As mentioned in the beginning of the section, it is assumed that 90-100% is released to sewage. For the part that reaches the sewer, it is estimated that 3-25% ends up in surface water, as the size of the released microplastics is not known, resulting in a total release to surface water of 1-50 t/year. Consequently, 75-97% ends up in sludge, i.e. 10-170 t/year, respectively, from which 10-90 t/year ends up in agricultural soil.

# **5.2.9** Other sources of secondary microplastics

A number of other sources of secondary microplastics exist. The following sources have been included in a survey of microplastics sources in Norway (Sundt *et al.*, 2014):

- Handling of plastics materials and articles during industrial and professional use:
  - Dust from cutting and polishing plastic items in industry
  - Dust from cutting plastic items in building and construction (e.g. cutting EPS sheets or PVC gutter)
  - Dust from cutting plastic items in boat repair- as well as shipyards and car repair shops
- Nets and other fishing tools
- PE foils used in agriculture
- Polymer modified bitumen
- Waste handling
  - Biowaste
  - Paper recycling
  - Shredders
  - Food waste shredders
  - Other waste handling
  - Plastics recycling facilities.

# Handling of plastics materials and articles during industrial and professional use

By cutting, turning, polishing, etc. plastic items in industry, building and construction as well as ship repair, small plastic particles may be formed. Sundt et al. (2014) mention the source, but do not include it in the estimated generation of secondary microplastics. Dust generated e.g. outdoors on paved areas may be discharged to the sewer or to the stormwater sewerage system, while activities in ports and other areas close to the sea, lakes or streams may be released directly to surface water. No data are readily available for estimating the total quantities generated and within the limits of the survey, it has not been attempted to establish an estimate.

#### Nets and other fishing tools

Microplastics fibres and particles may be released from nets and other fishing tools in use as well as from equipment for aquaculture. Compared to the formation of microplastics from fishing tools and larger parts of rope, cords and nets, lost to the sea, the releases from the equipment during use is probably lower for most application. The same conclusion has been reached for Norway by Sundt *et al.* (2014). It has been estimated that professional fishing contributes 13% of the total macrolitter in the North Sea and rope/cord/nets <50 cm was the second most abundant type of marine litter items found on reference beaches (section 2.7.2). The significance of this as a source of microplastics in the aquatic environment is further discussed in section 5.5.

A particular issue is the so-called "dolly ropes", mainly used by bottom trawlers, which consist of dozens of smaller twisted pieces of rope manufactured in such a way that the small pieces of thread easily loosen when the rope is used (DollyRopeFree, 2015). The releases from these ropes are significant during use, but no data on the total use of these types of ropes in Denmark are available.

Within the limits of the project, it has not been attempted to establish an estimate of the releases of microplastics from this source, but likely releases from the nets in use is a significant source of releases to the marine environment.

# Polyethylene foils used in agriculture

Polyethylene foils are widely used in agriculture. Sundt et al. (2014) mention the source, but do not include it in the estimated generation of secondary microplastics. Even some small plastic particles may be generated from the films during use and lost to the soil environment or spread as dust with the wind. The most likely pathway of releases of plastics from this application to the environment is, however, loss of larger pieces of plastics, which may later be fragmented in the environment. The loss of larger plastics parts is beyond the scope of this report.

#### Polymer modified bitumen

According to Sundt et al. (2014), in order to improve the properties (viscosity) of asphalt, polymers are added to some bitumen. The materials used are SBR (styrene butadiene rubber) and SEBS (styrene ethylene/butylene styrene copolymer/ "SEBS Rubber"). In brief, the polymers make the asphalt stiffer on warm summer days and more flexible on cold winter days (Sundt et al. 2014). According to an article in Dansk Vejtidsskrit [Danish Road Journal] from 2000, the application in Denmark was increasing though no volumes were provided (Jensen 2000). According to product data from a major producer of polymer modified bitumen, the polymer content of the products varies from about 3 to 7%. It may be questioned as to whether dust from the mixtures of rubber and bitumen may fall under the definition of microplastics applied here. Within the limits of the project, it has not been attempted to establish an estimate of the releases of microplastics from this source.

#### Waste handling

Sundt et al. (2014) establishes some estimates for the formation of secondary microplastics from various waste management operations. The major source of generation of secondary microplastics is compost and biogas sludge. No Danish data on plastic particles in compost and biogas sludge are available.

**Biowaste** - A European review shows that impurities (defined as: above 4 mm size, visible impurities) in compost and biogas digestate from European biowaste is typically in the range from almost zero to about 0.3% dry weight (Saveyn and Eder, 2014). Declarations for compost in Denmark typically indicate a content of plastic particles (<2 mm) of <0.1% dry weight. Summarised data on the actual plastics content of compost and biogas digestate in Denmark below 2 mm and in the 2-5 mm range have not been identified, although some information on specific waste products may exist.

Sundt et al. (2014) establish a worst case estimate for plastics in biowaste in Norway, using the 0.3% dry weight from the European review as a worst case for particles >4 mm, a similar content of impurities of <4 mm and assuming that 50% of the impurities are plastics. A total microplastic factor for biowaste is estimated at about 0.12% wet weight. On this basis, they estimate a worst case content of microplastics in biowaste at 336 t/year; of this, they assume that 90% is released to soil while the remaining 10% is released to surface water (by surface run-off after application.

Even the 336 t/year is a worst case, and the actual value may be significantly lower; the estimate indicates that biowaste may be a significant source of microplastics to soil and further data on microplastics in biowaste would be relevant is order to evaluate this source.

Paper recycling - As described by Sundt et al. (2014), paper recycling factories receive large amounts of printed paper (some printing techniques apply thermoplastic to the paper surface), glossy paper (which may have a plastic film), grease- or waterproof food packing paper (which may be plastic laminated) and other paper that might also have been polymer modified in some way. Often paper, cardboard and corrugated board are coated by plastics. According to the Reference Document on Best Available Techniques (BREF document) in the Pulp and Paper Industry, plastics in paper sent for recycling can account for up to several percent of the waste fraction and several OECD reports on emissions from de-inking facilities and factories for recycled paper production show that discharges of polymer particles are possible (as cited by Sundt et al. 2014). Furthermore, measurements by the outlet of a Dutch paper recycling plant demonstrated high concentrations just by the outlet (Dubaish and Liebezeit 2013 as cited by Sundt et al. 2014). Sundt et al. estimates for Norway, based on Dutch effluent measurements, a total release of microplastics of 60 t/year in 700,000 m<sup>3</sup> waste water. The largest Danish manufacturer of paper from recycled paper produces approximately half this amount of waste water (Danish Nature Agency, 2012) and has a separate industrial waste water treatment plants. It has not been attempted to estimate the potential releases of microplastics for recycling of paper within the limits of the project.

**Shredders** - Shredding of cars, white goods and other articles generates large quantities of lightweight waste, "fluff" which largely consists of plastic materials from the articles. By the shredding process, some dust is generated from coatings, plastic foams and plastic articles within this waste. Based on data on dust emission from shredder plants, Sundt et al. (2014) estimated the total microplastic emission from shredder plants in Norway at 10 t/year. It has not been attempted to estimate the potential releases microplastics from shredder plants within the limits of the project but the Norwegian estimates indicate that this is likely not a major source of secondary microplastics emission.

**Landfills** - Plastics within landfills may disintegrate and form microplastics particles, which may be released to the surroundings by windborne dust or discharged to sewage suspended in leachate from the landfill. In Denmark, plastic waste which is not recycled is incinerated and furthermore, the waste in the landfills is covered. Although emissions may occur, it is estimated that releases from landfills are not a significant source in Denmark.

**Food waste shredders** - According to Sundt et al. (2014), food waste shredders on ships and institutions are also a possible pollution source of secondary microplastics. Aboard fishing boats and probably other vessels in Nordic waters, these shredders are common. As an example, according to GreenPort (2013), the Swedish waste management system provider, Uson Marine (UMS), has

launched a food waste shredder with integrated macerator designed for use on board ships and offshore platforms. After passing through the shredder head, the shredded material is collected into a second hopper connected to the macerator. The ground up waste can then be discharged overboard or stored in a holding tank when the vessel is operating within an area where discharge is prohibited. According to Sundt et al. (2014), there may therefore be a risk that plastic film and food wrapping follows the food waste through this maceration, and then goes directly overboard as macro or microplastics. No data which could be used for an estimate are available.

**Plastics recycling facilities** - According to Sundt et al. (2014), plastic recyclers often have a system for sink/float sorting of plastic waste according to the specific gravity of different plastic types. The waste water obviously contains contamination, possibly also some plastic items or particles from the rough washing and/or agglomeration processes. About 25% of the plastics waste, corresponding to approximately 80,000 t/year, is collected for recycling in Denmark (Danish Plastics Federation, 2015). It has not been attempted to estimate the potential releases of microplastics for recycling of paper within the limits of the project.

#### Summary on other sources

With a view to the estimates for some of these applications in Norway, it is estimated that the total generation of microplastics from other sources likely is in the range of 100-1000 t/y. A reservation is made for polymer modified bitumen which, depending to what extent the particles fall under the definition and what types of microplastics are formed, may be a more substantial source. Considering that one of the main sources is biowaste (releases to soil) and releases from nets and fishing tools in use occur directly to the water environment it is roughly assumed that 20-50% is released to sewage (of this 15-25% is released to surface water), 20-50% to soil and 5-25% directly to surface water (note the ranges are interrelated).

# 5.2.10 Summary of emission estimates for Danish sources to microplastic pollution

The estimated releases of primary and secondary microplastics in Denmark are summarised in the table overleaf.

TABLE 30 SUMMARY OF RELEASES OF PRIMARU AND SECONDARY MICROPLASTICS IN DENMARK (EXCL. FORMATION FROM MACROPLASTICS IN THE ENVIRONMENT)

	Total emis- sion t/year	% of total (average)	Emission to sewage treat- ment plants (STP) t/year	Ultimate emission to the aquatic environment *	% of total ultimate emission to the aquatic environment (average)				
Primary microplastic	Primary microplastics								
Personal care products	9-29	0.2	10-22	0.5-4.4	0.1				
Raw materials for plastics production	3-56	0.3	3-56	0.1-4.5	0.1				
Paints	2-7	0.1	2-7	0.3-1.8	0.1				
Blasting abrasives	0.05-2.5	0.01	0.03-1.3	0.03-1.4	0.04				
Rubber granules	450-1,580	10.5	20-330	1-20	0.6				
Other applications	?	?	?	?	?				
Total, quantified primary microplastics	460-1,670	11	35-416	2-31	0.9				
Secondary microplas	stics								
Tires	4,200-6,600	56	1,600-2,500	500-1,700	60				
Textiles	200-1,000	6.2	200-1,000	6-60	1.8				
Paints (excl. ship paints)	150-810	4.9	14-220	6-149	4.2				
Ship paints	40-430	2.4	0-0	21-240	7.1				
Road markings	110-690	4.1	40-260	10-180	5.1				
Building materials of plastics	80-480	2.9	30-150	5-38	1.1				
Footwear	100-1,000	5.7	40-380	10-260	7.3				
Cooking utensils, scouring sponges and cloths	20-180	1.0	20-180	1-50	1.4				
Other uses	100-1,000	5.7	20-500	8-375	10				
Total, secondary microplastics (rounded)	5,000- 12,200	89	2,000-5,200	600-3,100	98				
Total quantified microplastics (rounded)	5,500-13,900		2,000-5,600	600-3,100	99				

 $<sup>\</sup>ensuremath{^{*}}$  Indicates the resulting emissions after sewage treatment.

#### 5.3 Fate of microplastics in sewerage systems in Denmark

#### 5.3.1 Municipal sewage

Microplastics entering the municipal sewerage systems may be released to the aquatic environment via the sewage treatment plants (STP) or directly to the receiving waters by sewage overflows.

#### **Overflows**

A part of the sewage is released directly to the recipients by STP overflows due to heavy rain and flooding. The percentage varies significantly between Danish municipalities. The publication "Water in figures, 2009" shows data for a number of municipalities with overflow percentages ranging from less than 1% to more than 10%, with an average of approximately 4% of the sewage directly discharged to the recipient via overflows (DANVA 2009b). The percentage is probably, as a result of investment in overflow retention basins, decreasing, but newer data have not been identified and 4% is used as the best available estimate.

#### **Releases from STPs**

No actual data on the fate of microplastics in Danish STPs have been identified, but it is for the estimations assumed that the fate of microplastics in Danish STPs is probably similar to the results reported from Swedish and Norwegian STPs reviewed in section o. As some variance between plants has been observed, the retention efficiencies are represented by ranges in the estimations.

According to the review in chapter o, microplastics reaching the STPs will mainly end up in the sewage sludge. The percentage withheld in the sludge differs on the size of the microplastics whereas more or less similar percentages are found for fibres, flakes and particles.

For microplastics >300  $\mu$ m, approximately 99% of the microplastics ends up in the sludge whereas the percentage for microplastics >20  $\mu$ m (including microplastics >300  $\mu$ m) is lower and varies from approximately 70% to 90% among the studied STPs. No data are available for microplastics smaller than 20  $\mu$ m.

The data from other countries indicates that microplastics  $>300 \mu m$  is dominated by fibres (most likely textile fibres), whereas flakes and particles dominate the smaller particles.

The combined emission factor for the large particles >300  $\mu m$  will be totally dominated by the percentage discharged directly without treatment.

Based on data the following combined emission factors for emissions to aquatic environment from the waste water system:

Particles >300 μm: 3-6%
 Particles 20-300 μm: 15-25%

For applications where particle sizes do not fall within one of these ranges a combined emission factor is roughly estimated.

#### Disposal of sludge

The majority of the microplastics in sewage end up in sewage sludge. In Denmark, the sludge is either applied on agricultural soils or it is incinerated. The figures for the distribution between the two pathways differ slightly from year to year and between information sources. Lassen *et al.* (2015) estimates average figures of 55% to agricultural soils and 45% incinerated on the basis of various sources (DANVA 2009a; Kirkeby *et al.* 2005; DEPA 2009). Approximately 3% of the agricultural area in Denmark receives sludge.

No data are available on microplastics in sewage sludge and the resulting concentrations of microplastics in the agricultural soils in Denmark.

Ten years ago, Zubris and Richards (2005) suggested synthetic fibres as an indicator of land application of sludge. The context was not possible effects of microplastics. Their study demonstrated that fibres (isolated by water extraction and examined using polarized light microscopy) were detectable in sludge products and in soil columns over 5 years after application, retaining characteristics observed in the applied sludge. Fibres were also detectable in field site soils up to 15 years after application, again retaining the characteristics seen in sludge products. The data indicates that the microplastics will remain in the soil for a long time after the application.

As described in section 2.8, very limited data on possible effects of microplastics on soil organisms are available and more data on the possible long-term effects of microplastics in sludge applied to agricultural soils are warranted.

#### 5.3.2 Urban runoff

About 5% of the total area of Denmark is paved and equipped with systems for drainage of stormwater (Danish Nature Agency 2015b). There are two types of sewer systems:

- Combined systems where sewage and stormwater is collected in one sewer and discharged to a STP.
- Separate systems where sewage and stormwater is discharged in separate sewers to STPs and recipients, respectively.

According to the Danish Nature Agency (2012) the paved area with sewerage system was approximately 770 km $^2$ ; of this 350 km $^2$  has common sewerage system and the remaining 415 km $^2$  has separate storm water systems.

No data on microplastics in stormwater in Denmark and the fate of microplastics within the separate sewer system is available. The stormwater consists of road runoff (from roads and squares) and water from roofs. The sources of microplastics in the road runoff are mainly dust from tyres (section 5.2.1), road marking materials (section 5.2.5) and decorative coatings used outdoors (section 5.2.3). The sources of microplastics in runoff from roofs is mainly abrasive releases from roof membranes and PVC gutters (Section 5.2.6)

As mentioned, microplastics in approximately half of the stormwater are discharged directly to the aquatic environment without treatment in sewage treatment plants. In about 1/3 of the separate systems, the water is directed through lagoons where some of the particles in the stormwater settle (Danish Nature Agency 2015b). Microplastics with a density above one (e.g. dust from tyres) may likely to some extent settle in the lagoons, but it is unknown to what extent they are re-suspended during heavy rainfalls.

Studies specifically addressing the retention efficiency of the sedimentation lagoons for microplastics have not been identified, but some assessments of the retention efficiencies of the lagoons in general exist. Vollertsen et al. (2012) reviewed a number of Nordic and US studies of the fate of suspended matter, nutrients and heavy metals i sedimentation lagoons. Furthermore, an assessment of possible control of EU priority substances in Danish Waters (Kjølholt 2007) and a study of discharges of storm water (Petersen et al. 2013) include some information on retention efficiencies. Even the variation is high, in general the retention efficiency for suspended matter is high (80-90%). As the density of some of the microplastics is lower than the density of water, the retention efficiency for microplastics on average may be lower and here assumed to be 40-50%. Combined with the information that about 1/3 of the separate systems have lagoons, the combined emission factor for all stormwater in separate systems is roughly estimated at 80-90%.

#### 5.3.3 Industrial waste water with direct discharges to the environment

No data are available on microplastics in industrial waste water with direct discharges to the environment. None of the enterprises within the plastics, cosmetics or paint industries, using primary

microplastics, has direct discharges to the environment (Danish Nature Agency 2015a). Secondary microplastics may be present in waste water discharged (after treatment in treatment facilities) from e.g. shipyards and recycling facilities. Some secondary microplastics may in principle be present in waste water from most activities, but none of the industrial discharges are considered as major sources at national level, but it cannot be rules out that the discharges may lead to elevated levels of microplastics in the vicinity of the outlets of some facilities.

#### 5.4 Sources of microplastics in the sea around Denmark

Levels of microplastics in the Danish waters are described in section 2.1 whereas this section is a discussion about the interpretations that can be drawn from the available data.

As the sources of microplastics can only be estimated with high uncertainty and it is furthermore very uncertain how the different types of microplastics are transported in the environment, an attempt is done in this section to analyse the data on the geographic distribution of microplastics in the environment by type, in order to assess to what extent is it possible to indicate significance of the different sources for the occurrence of microplastics in the environment.

A limitation in such an analysis it that investigations of microplastics in the environment have analysed particles of a size down to about 10  $\mu$ m, while the particle size of particles generated by many of the sources of secondary microplastics is below this size.

#### 5.4.1 Occurrence by type of microplastics and by distance to sources

Levels of microplastics in the Danish waters are described in section 2.1 whereas this section focuses on the information on sources that can be drawn from the available data.

There is no available data on microplastics in Danish waters that could be directly related to sources. However, studies on microplastics concentrations made in neighbouring water areas can be used with the assumption that similar littering processes rule both Danish, Swedish and North German waters (in particular the Eastern Danish areas and the Western Swedish areas). The authors also use data from surveys of microplastics made close to Danish water borders.

In a study of litter in surface water in German/Danish North Sea and Baltic waters there was one offshore location west of Jutland (Station 12 in Figure 3 in Section 2.1), at the level of Limfjorden, with both an elevated concentration and a different composition of plastic particles compared to other sampling stations (Mintenig 2014). The particles detected here were dominated by PE particles of a character that suggested that they were industrial pellets made up of recycled plastics (see description in Section 2.1). The same kind of particles were found at this location in both 2012 and 2013, but whereas the particle surfaces were clean in 2012 they were covered with a biofilm in 2013. Although it may be presumed that these particles derive from a particular source it is not possible to draw any conclusions about what source that could be. In the same study, microplastics concentrations even higher than at station 12 were found at Station 15, northeast of Skagen. However, unlike Station 12, the composition of particles at Station 15 did not differ from the other sampling stations in the survey. This implies that the elevated concentration at station 15 was the result of hydrological conditions rather than of local sources.

In a survey of microplastics in marine sediments, higher concentrations were found in the Belt Sea area than at the North Sea coast and Kattegat stations (Strand *et al.* 2013). Whether this reflects regional differences or differences related to local sources is not possible to say.

The correlation between distance from urbanized areas and concentration of microplastics was analysed by Norén *et al.* (2014). The concentrations of microplastics in urban water areas were higher compared to rural water areas. The concentration of microplastic particles (>10µm, fibres and potential antifouling paint flakes excluded) was 21,000±1,000/m³ in the Gothenburg area (mean value of three stations in the estuary area outside Gothenburg) compared to a mean of

7,000±6,000/m³ for eleven stations at the Swedish west coast north of Gothenburg. The same pattern was also visible for textile fibres (both plastic and non-plastic fibres) and potential antifouling paint-flakes. An area with higher concentrations of microplastic was found outside a plastic production plant in the city of Stenungssund on the Swedish west coast (see Norén 2007 and Norén et al 2014). In Sweden, large quantities of PE and PVC are produced in the area and transparent PE particles in particular have been found in higher abundance in the surface waters. It can be mentioned that the PE industry recently has installed fine-meshed filters on the outgoing storm and process water as a result of permit regulations from the county administration.

Landbecker (2012) studied the microplastic concentrations in Lake Mälaren west of Stockholm (1  $\sim$ 1.4 million inhab.) and the results showed the highest concentration of microplastic closest to the urban centre and lowest concentrations at the most remote stations. Textile fibres were the most abundant type of microplastics found.

Sewage treatment plants (STPs) as entrance points for microplastic particles to the marine environment appear to be important and were evaluated by Magnusson and Norén (2014). The microplastic concentration in the recipient was elevated compared to an area presumed not to be directly affected by the effluent; 1.1 - 1.8 plastic particles per  $m^3$  were found in the effluent plume compared to  $0.45 \ m^{-3}$  in the reference area. Higher particle concentrations were found close to the mouth of the tube compared to  $200 \ m$  downstream. Only plastic textile fibres, and no other types of plastic particles, were found in the recipient. The study demonstrates that the supply of microplastics from STP effluents to the marine environment may be substantial.

# 5.4.2 Occurrence in the open sea and potential transport by sea currents and sediment transport

Due to hydrographic conditions there are areas on the sea floor where sediment particles are only settling temporarily (transportation bottoms), and others where they settle more or less permanently (accumulation bottoms). The accumulation bottoms are characterized by high concentrations of very small organic particles. In their study of marine sediments in Danish coastal waters, Strand *et al.* (2013) found a general positive correlation between the percent total organic carbon (TOC) content in the sediments and the concentration of microplastic particles. This indicates that microplastics with a density higher than sea water, which therefore settle on the sea floor, are transported with sediment particles and end up on accumulation bottoms.

An accumulation zone for benthic marine litter >20 mm far from any point sources and which was presumed to be the result of hydrodynamic conditions, was detected in the North Sea  $\sim$ 200 km west of Esbjerg ( $55^{\circ}55N-05^{\circ}20E$ ) (Galgani *et al.* 2000). Unlike the case in other accumulation zones for marine litter in e.g. in the Mediterranean or south of Brittany, this area did not coincide with fishing activities, but was supposed to be a result of displacement of the litter by sea currents.

In a study of pelagic microlitter particles >10  $\mu m$  along a transect between south Norway and Hirtshals the highest concentrations of microlitter was found close to the Danish coast (Norén and Naustvoll 2010). The water in this area had a high salinity, which indicates that it was of North Sea origin and that the particles were not from local sources. Most of the collected anthropogenic material consisted of red and blue particles that were suspected to derive from boat paint.

The general concentration of the plastics in the North Sea has decreased over the past decades. This is as described in Section 2.1 was supported by analyses of both sea water and of gut content of North Sea fulmars (Morét-Ferguson *et al.* 2010; van Franeker and Law 2015).

Efforts to model drift patterns of litter particles in the southern North Sea showed that that variability of currents was so strong that it was impossible to use hydrological factors to backtrack the source of detected particles (Neumann *et al.* 2014). As shown by other models (Lebreton *et al.* 2012) it is however likely that litter found in the North Sea and the Baltic Sea derive from the North

Sea/Baltic Sea region. On a global scale, there are a number of important gyres in the open oceans where pelagic litter accumulates, but there are also smaller gyres in local semi enclosed sea areas like the North Sea and the Baltic Sea.

#### Assessments of the origin of microplastics in the Danish waters 5.4.3

There is at present very limited information on the quantities of microplastics in Danish coastal waters, and even less is known about the composition of the microplastic material. In most studies where the microplastic composition have been analysed, only a subset of the collected material has been selected and it has not been possible to use the analyses to determine the origin of the particles.

One of the most comprehensive studies, thus far, in determining the composition of marine microlitter was done by Mintenig 2014 (Figure 5) In this study, efforts were made to carry out FTIR analyses of all microplastic particles collected from water samples from a number of stations in the German/Danish North Sea and Baltic coasts. The analyses were successful for particles >500 µm but for the fraction <500 µm not all particles could be analysed. The most common kind of microplastic particles  $>500 \mu m$  at the sampling stations along the Jutland west coast was PUR fragments (51%). In addition, fragments of PE (29%) and a fragment and a fibre of PP (17%) were found. PE and PP are the plastics used in the highest volume, have a density below one and are widely used in packaging and films so it is not surprising that these polymer types are abundant. Polyurethane has a density above one, unless it is used as a foam, and it is not obvious which applications could result in the high concentrations. Polyurethane is used in some ship paints, but is not the dominant polymer for this application. Rigid polyurethane foam is widely used for building insulation while flexible polyurethane foam is used for mattresses and upholstered furniture. It is not clear how the microplastic particles are generated from these applications and end up in the sea. The data from sewage treatment plants in Figure 14 indicate that PUR accounts for only a few percent of the particles in the effluent from sewage treatment plants. For particles <100 µm, the dominant polymer, accounting for more than 75%, was polypropylene (PP). PP is widely used in packaging. PP is to some extent used in fishing nets, but nets of nylon (polyamide) are more common, and fragmentation of lines from fishing nets is consequently not the most obvious source of the high number of PP particles.

Exceptionally high concentrations of plastic particles were found at a sampling station located in the open sea outside the mouth of Limfjorden (station 12 in Figure X). Here a high number of industrial pellets, spheres and fragments of PE and PP were found, with the PE particles being most frequent.

Strand et al (2014) applied FTIR spectroscopy to identify specific polymers in some selected flakes and granules from sediments in Danish waters (fibres could not be analysed). Particles of the polymers polypropylene, acrylate, and polyester/alkyd were identified, but data on abundances are not reported. Both the acrylate and polyester/alkyd may originate from paint.

#### Formation from macroplastics in the sea as compared with other 5.5

Microplastics are formed by fragmentation of larger plastic pieces which are dispersed in the environment. There exists considerable knowledge about types of plastic found on beaches, and hence much knowledge about the sources of the macroplastics in the environment.

It is estimated that in the 1990s about 20,000 tons of waste were dumped in the North Sea, of which a large part was plastic waste (see section 2.7.2). Of this total, approximately 15% ended up on the beaches while the rest was expected to float around partly fragmented before it ultimately sunk to the bottom. There are no specific inventories for plastics and no estimations of the rate and to what extent the macroplastics floating or at the bottom were fragmented into microplastics. The timeframe for a complete degradation (mineralization) of plastics may be many hundreds of years.

Available data for fragmentation indicate that many types of macroplastics will be fragmented to a certain degree within a timeframe of years or decades.

A substantial portion of the plastic pieces in the marine environment will therefore fragment before they are removed from the beaches or before they are covered by sediment. There are no model calculations estimating the formation of microplastics based on the occurrence of macroplastics in the environment. A Norwegian survey provides what the authors of the survey would term a "qualified guess" with respect to formation of microplastics formed through the fragmentation of macroplastics in the aquatic environment of Norway. The estimates show a total amount of 360 to 1,800 t/year from the main sources, but the actual uncertainty of the estimate is likely to be greater than the range suggests. Nonetheless, the estimate suggests that the fragmentation of macroplastics in the environment is likely to contribute significantly more than the emissions of primary microplastics, whereas it is more uncertain as to whether the quantities formed in the environment are of the same magnitude as the releases of secondary microplastics. Since there are no data allowing better estimates than this "qualified guess", no attempt has been made to prepare another "qualified guess", and the present study refers to the Norwegian estimate.

#### 5.6 Occurrence of microplastics in streams and lakes

No data on the occurrence of microplastics in streams and lakes in Denmark have been identified.

# 6. Ongoing initiatives regarding microplastics and marine litter

#### 6.1 Initiatives of Danish authorities

#### National authorities

The Ministry of Environment and Food, The Danish Environmental Protection Agency, have initiated this project.

As a consequence of the Marine Strategy Framework Directive, a Danish Marine Strategy ("Danmarks Havstrategi") was published in 2012. The base analysis provided an overview of the environmental status of the Danish seas and outlined existing biological, chemical and physical conditions, pressures and impacts on the North Sea and the Baltic Sea with the aim of establishing a good environmental status in these areas by 2020. A monitoring programme under the Danish Marine Strategy was published in 2014. The monitoring programme includes monitoring of marine litter at four reference beaches, marine litter on the sea floor by bottom trawl investigations, and analyses of microplastic in sediments, as well as analyses of macro and microplastics in stomachs of two fish species. The monitoring programme was complemented with an action programme in 2015.

# **6.2** Initiatives of the European Commission and EU Member States The European Commission

The European Commission has engaged in a number of activities related to plastic waste and microplastics. For instance, a Green Paper on a European Strategy on Plastic Waste in the Environment was published by the EC in March 2013<sup>10</sup> with the purpose of initiating a broader discussion on plastic waste leading up to a review of the European waste legislation in 2014. After having acknowledged that plastic waste is a horizontal waste stream that cuts across waste types and sectors, the Commission mentioned microplastics as an area of particular concern – both from primary (virgin) and secondary sources. According to the Commission, plastic waste is not covered by one separate piece of EU legislation at present but through several different pieces of legislation, thereby posing policy challenges. Following the publication of the green paper, a public consultation was carried out by the BIO Intelligent Service for the European Commission<sup>11</sup>. Responses were received from NGOs, trade organisations, industry federations, public administrations, private companies and citizens. One question in this consultation, taken from the Green Paper, was: "How can challenges arising from the use of microplastics in products or industrial processes and of nanoparticles in plastics be best addressed?" The general response to this question was that the use of microplastics should be prohibited or limited or that precautionary principles should be enforced and more detailed testing carried out before pacing microplastics on the market.

<sup>&</sup>lt;sup>9</sup> Naturstyrelsen, Danmarks Havstrategi

 $http://naturstyrelsen.dk/media/nst/12323931/samlet\_overv\_gningsprogram\_for\_hsd.pdf$ 

 $<sup>^{10}\,</sup>http://www.ab.gov.tr/files/ardb/evt/greenpaper\_march\_2013.pdf$ 

<sup>11</sup> http://ec.europa.eu/environment/waste/studies/pdf/green\_paper\_plastic.pdf

Another example of current activities related to the Marine Strategy Framework Directive (MSFD). This directive was adopted in 2008 and provides the overall framework to protect the European marine environment and for Member States to achieve Good Environmental Status (GES) by 2020. One of the criteria listed as a qualitative descriptor "for determining good environmental status" in this directive is that "Properties and quantities of marine litter do not cause harm to the coastal and marine environment" (Descriptor 10)<sup>12</sup>. To support the implementation of the directive, a document with more detailed information on criteria and methodological standards was published in 2010. For descriptor 10 on marine litter it is further detailed that microplastics is a particular topic of interest with regards to biological effects as well as "the amount, distribution and, where possible, composition of micro-particles (in particular microplastics)"<sup>13</sup>.

A final example of European Commission activities is the funding of Seventh Framework Programme (FP7) projects and Marie Curie actions. Several ongoing FP7 and FP8 projects deal with microplastics, for instance:

- The project **Marlisco<sup>14</sup>** (with Danish participation through KIMO Denmark) ran from 1 June 2012 31 May 2015 and had the overall aim of reducing marine litter through public awareness and dialogue. Microplastics were part of their awareness-raising campaign. In addition, information specific to microplastics was included in the project deliverables on quantities and distribution of marine litter, monitoring methods and policy.
- **ECsafeSEAFOOD**<sup>15</sup> is another ongoing FP7 project (February 2013 January 2017) with the aim of addressing issues related to food safety and contaminants present in seafood. WP2 of the project ("Monitoring of environmental contaminants in seafood products with unknown information") will monitor levels of micro-plastics in selected seafood as well as optimize detection and quantification methods in water, sediment, particulate matter and seafood. Blue mussel (*Mytilus edulis*) and European flounder (*Platichthys flesus*) are used as case studies. Deliverable D2.3<sup>16</sup> of the project is specifically focussed on microplastics and is entitled: "Microplastics and the associated contaminants in various environmental compartments and biota".
- The **CLEANSEA**<sup>17</sup> project is as an example of an ongoing FP7 project (January 2013 December 2015) focussed on the management and potential impacts of marine and costal litter. It aims to provide estimates of the quantities of marine litter and to describe its composition and distribution, including the rates of fragmentation of micro-sized particles. As part of this project work has been carried out on biological effects of microplastics and microplastic-mediated uptake of PAHs by European shore crab.
- MARMICROTOX (June 2014-June 2016) is a FP7 Marie Curie action with the full title "Marine microplastics toxicity: investigating microplastics and their co-contaminants in marine organisms" with the goal to qualitatively and quantitatively assess microplastics found in wild mussels in Scotland. Furthermore, the project will investigate accumulation, effects, trophic transfer and microplastics as carriers for co-contaminants.
- The COMMON SENSE<sup>18</sup> project (November 2013 February 2017) is a Marie Curie fellow-ship that aims to contribute with design and develop new generation sensors for detection of microplastics in support of the implementation of the Marine Strategy Framework Directive (MSFD) and other EU policies.
  - The Marie Curie Action FreshwaterMPs<sup>19</sup> has been funded under H2020 and has the full title "The environmental fate and effects of microplastics in freshwater ecosystems".

15 http://www.ecsafeseafood.eu/

16 http://www.ecsafeseafood.eu/images/ECsafeSEAFOOD/Project/WPs/WP2.pdf

<sup>12 (</sup>http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32008L0056&from=EN)

<sup>13</sup> http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32010D0477(01)&from=EN

<sup>14</sup> http://www.marlisco.eu/)

<sup>17</sup> http://www.cleansea-project.eu/drupal/index.php

<sup>18</sup> http://cordis.europa.eu/project/rcn/110790\_en.html

<sup>19</sup> http://cordis.europa.eu/project/rcn/195924\_en.html

The project is ongoing (January 2015 – April 2017) with the aim of building "towards a sophisticated state-of-the-art mesocosm study that will evaluate both MP fate and impacts in model ecosystems" and thereby establish a novel framework for the environmental risk assessment of MPs".

#### **European Environmental Agency**

The European Environment Agency (EEA) has developed **Marine LitterWatch** mobile app to strengthen Europe's knowledge base and thus provide support to European policy making<sup>20</sup>. The EEA is working together with many existing communities working on marine litter from all regional seas in Europe. Marine LitterWatch also aims to inspire new communities to form. These can range from NGOs, business and industry, coastal communities (e.g. local sports club, scouts), schools and universities, expert communities (i.e. science and research) to public authorities and other. The current community list can be found here.

#### **EU Member States initiatives at EU level**

In December 2014, member countries of the European Union (Belgium, the Netherlands, Austria, and Sweden with support from Luxembourg) - issued a joint information note entitled "Elimination of micro-plastics in products - an urgent need" calling for a European ban of microplastics in cosmetics and detergents in order to prevent pollution of the aquatic environment. Also, the statement calls for an effort to close scientific gaps as well as a clarification of the role the European Environment Agency and the European Chemicals Agency with regard to addressing the issue. As a follow-up on this activity, a stakeholder conference was organised and hosted by the Permanent Representation of the Netherlands in Brussels in May 2015<sup>21</sup>. It was highlighted by Commissioner Vella (the European Commissioner for Maritime Affairs and Fisheries) that tackling the problem of microplastics is an urgent need that requires local, European and international efforts and collaboration<sup>22</sup>. With regard to actions to eliminate microplastic pollution, the main conclusions<sup>23</sup> from this conference included urgent needs for: increased public awareness and education, material substitution and improved product design, improved plastic recycling and resource efficiency as well as eco-design and marine litter targets as part of the ECs Circular Economy Strategy<sup>24</sup>. Definitions of microplastics and harmonised quantification and monitoring methods were highlighted as challenges. It was further decided that the EPA Network is an appropriate forum to continue this work and will be discussed at the EPA Network plenary meeting in September 2015 in Iceland. Next steps on a European level are to be discussed between the EPA Network members and Commissioner Vella and his cabinet. It is suggested that microplastics is to be included in the next European State of the Environment Report (SOER) (to be published in 2020).

# 6.3 Initiatives of international organisations UNEP

Global Programme of Action for the Protection from the Marine Environment from Land-based Activities (GPA) - United Nations Environment Assembly of the United Nations Environment Programme (UNEP) at its first session on 27 June 2014 adopted a resolution on marine plastic debris and microplastics, noting with concern the serious impact which marine litter, including plastics stemming from land and sea-based sources, can have on the marine environment, marine ecosystem services, marine natural resources, fisheries, tourism and the economy, as well as the potential risks to human health.

<sup>&</sup>lt;sup>20</sup> http://www.eea.europa.eu/themes/coast\_sea/marine-litterwatch/at-a-glance/marine-litterwatch-in-a-nutshell

<sup>21</sup> http://epanet.pbe.eea.europa.eu/ad-hoc-meetings/workshop-plastics-environment-11-12-may-2015

<sup>&</sup>lt;sup>22</sup> https://ec.europa.eu/commission/2014-2019/vella/announcements/eliminating-plastic-and-microplastic-pollution-urgent-need\_en

<sup>&</sup>lt;sup>23</sup> http://epanet.pbe.eea.europa.eu/ad-hoc-meetings/workshop-plastics-environment-11-12-may-2015/conclusions-workshop

<sup>&</sup>lt;sup>24</sup> http://ec.europa.eu/environment/circular-economy/index\_en.htm

The Global Programme of Action for the Protection from the Marine Environment from Land-based Activities (GPA)<sup>25</sup> is a global intergovernmental mechanism directly addressing the connectivity between terrestrial, freshwater, coastal and marine ecosystems. UNEP hosts the GPA Coordinating Unit and coordinates some activities in support of the programme. Intergovernmental Review Meetings are organized every 5 years to review the progress made by countries in the implementation of the GPA through their respective National Action Plans.

Global Partnership on Marine Litter - Under the GPA, land-based marine litter has been highlighted in the Manila Declaration as a priority source category for 2012-2016. To prevent the influx of marine litter in the environment, the Global Partnership on Marine Litter (GPML)<sup>26</sup> was launched in June 2012 at Rio + 20. The GPML, besides being supportive of the Global Partnership on Waste Management, seeks to protect human health and the global environment by reducing and managing marine litter. The GPML is a global partnership gathering international agencies, Governments, NGOs, academia, private sector, civil society and individuals. Participants contribute to the development and implementation of GPML activities. Contributions may be in the form of financial support, in-kind contributions and/or technical expertise.

A specific objective of the GPML is to enhance international cooperation and coordination through promotion and implementation of the Honolulu Strategy. The Honolulu Strategy is a framework for a comprehensive and global effort to reduce the ecological, human health and economic impacts of marine debris. http://www.unep.org/esm/Portals/50159/Honolulu%20Strategy%20Final.pdf

A number of regional initiatives is described at the UNEP marine litter website at: http://www.unep.org/regionalseas/marinelitter/initiatives/unepregions/default.asp

**Global Marine Litter Gateway** - The Global Marine Litter Gateway is in information gateway with information relevant to the issue of marine litter. The Gateway is a co-operative effort of the UNEP GPA Coordination Office and the UN International Maritime Organization Available from: http://marine-litter.gpa.unep.org/about/about.htm

#### Other initiatives and support

Besides the GPML, UNEP supports a number of initiatives by NGOs among these the Plastics Disclosure Project, Beat the Microbead and Plasticity Forum (see further description in section 6.5 and 6.6).

#### Other UN bodies

**GESAMP** - The Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) is a multidisciplinary body of independent experts nominated by a number of sponsoring UN organizations (IMO, FAO, UNESCO-IOC, WMO, WHO, IAEA, UN, UNEP). Its mission is to provide advice to the Sponsoring Organizations, at their request, on pollution and other problems that face marine and coastal environments. The GESAMP Working Group 40, on Sources, fate & effects of microplastics in the marine environment has recently finished its global assessment report (GESAMP 2015).

**IMO** - The International Maritime Organization (IMO) is an agency of the United Nations that deals with safety and security of shipping and the prevention of marine pollution by ships. Dumping of waste material at sea is regulated by the London Convention and the International Convention for the Prevention of Pollution from Ships (MARPOL), both adopted by IMO. IMO participates in a number of activities addressing marine litter and plastic pollution such as GESAMP and the Global Marine Litter Information Gateway.

<sup>26</sup> http://unep.org/gpa/gpml/gpml.asp

<sup>25</sup> http://unep.org/gpa/default.asp

### 6.4 Regional initiatives by OSPAR, HELCOM and Nordic Council of Ministers

#### **OSPAR**

The Oslo and Paris Commission (OSPAR) is the administrator of the OSPAR Convention protecting the marine environment of the North East Atlantic (incl. Kattegat) involving Denmark and other 14 contracting parties.

The OSPAR Regional Action Plan for prevention and management of Marine Litter in the North-East Atlantic was adopted by OSPAR Contracting Parties in 2014 (OSPAR 2014). The Regional Action Plan is designed as a flexible tool providing a set of actions to address marine litter. It contains actions requiring collective activity within the framework of the OSPAR Commission through OSPAR measures (i.e. Decisions or Recommendations) and/or other agreements such as guidelines. The Action Plan sets out a number of actions within the following themes and sub-themes (each indicated in the plan with Lead Party and implementation year, see Appendix 2 for details):

- Theme A: Actions to combat sea-based sources:
  - Harmonised system for port reception facilities
  - o Incentives for responsible behaviour/disincentives for littering
  - Develop best practice in relation to fishing industry
  - o Fines for littering at Sea
- Theme B: Actions to combat land-based sources:
  - o Improved waste prevention and management
  - o Reduction of sewage and storm water related waste
  - o Incentives for responsible behaviour/disincentives for littering
  - o Elimination, change or adaptation of the products for environmental benefits
  - Development of sustainable packaging
  - Zero pellet loss
- Theme C: Removal Actions:
  - Application of Fishing for Litter activities
  - Cleaning environmental compartments and keeping them clean
- Theme D: Education and outreach:
  - Education
  - Outreach

For each action, lead Party/Parties and assisting Parties are listed. Denmark is not lead or assisting Party for any of the actions.

Furthermore, the OSPAR Litter Expert Group has a leading role in developing regionally coordinated SMART reduction/operational targets linked to relevant actions as contained in the implementation plan, starting from 2015.

#### HELCOM

The Helsinki Commission (HELCOM) is the administrator of the Helsinki Convention protecting the marine environment of the Baltic Sea area (incl. Kattegat) involving Denmark and nine other contracting parties.

The HELCOM Recommendation 36/1, Adopted 4 March 2015, includes a regional action plan on marine litter (RAP ML) in the Baltic Sea area (HELCOM 2015). The RAP ML lists a range of adopted regional actions (see Appendix 2 for details):

- Regional actions addressing land-based sources of marine litter:
- General improved waste prevention and management
- Measures to tackle top items:

- Micro particles
- Sewage related litter including sanitary waste
- Expended Polystyrene (Polystyrene Foam)
- Plastic bags
- Bottles and containers
- Actions addressing third parties
- Remediation and removal actions
- Regional actions addressing sea-based sources of marine litter:
- · Actions addressing shipping related waste
- Actions addressing waste delivery in ports/marinas
- Actions addressing waste related to fishing and aquaculture
- Remediation and removal measures
- Regional actions addressing education and outreach on marine litter
- General improved waste prevention and management

Furthermore, the RAP ML lists a number of proposed actions for the Contracting Parties to the Helsinki Convention for voluntary implementation. These actions aim at information exchange and coordination but are primarily of national concern and in the responsibility of the Contracting Parties.

The action plan does not indicate lead and assisting Parties and mentions a detailed implementation schedule, which is expected to be, finalized late 2015.

#### **ICES**

The International Council for the Exploration of the Sea (ICES) with secretariat in Copenhagen is a global organization that develops science and advice to support the sustainable use of the oceans. ICES is a network of more than 4000 scientists from over 350 marine institutes in 20 member countries and beyond. ICES has since 2011 been involved in the development of technical recommendations for the monitoring of marine litter and has recently on request from OSPAR developed common monitoring protocol for plastic particles in fish stomachs and selected shellfish (ICES 2015).

#### **Nordic Council of Ministers**

Marine litter has since 2013 been among the priorities or the Marine Group (HAV) under the Nordic Council of Ministers and the Nordic Council of Ministers. The council has in recent years supported several activities:

- Study on Marine litter in Nordic waters (Strand et al. 2015).
- Study on marine litter and its sources in Nordic waters (Blidberg et al. 2015)
- Icelandic conference on plastics. (referenced in Strand et al. 2015)
- The importance of sewage treatment plants as sources of marine microlitter in Finland, Sweden and Iceland (referenced in Strand *et al.* 2015)
- Study on plastic loading in Northern Fulmars, to gather new knowledge and to assess
  pollution levels of fulmars from Faroe Islands, Iceland and Svalbard (referenced in
  Strand et al. 2015)

For 2016, the Marine Group has indicated the following areas of relevance for microplastics and marine litter as prioritised for support:

- Car and road-related microparticles (such as rubber tire fragments, road dust and asphalt particles) as a source of pollution of the marine environment in the Nordic seas.
- Microparticles from paint residues, etc. as a source of pollution of the marine environment in the Nordic seas.

- Micro pollution from textile fibres (such as plastics and synthetic fibres, and/or cotton
  and other natural fibres) as a source of pollution of the marine environment in the Nordic seas.
- Based on the Islands recycling system for fishing gear, assess whether similar systems can be used advantageously in other Nordic countries.

#### **KIMO**

The Local Authorities International Environmental Organisation (KIMO International) representing local communities in the North Sea and Baltic Sea area have over the years coordinated a number of activities on marine litter (KIMO 2015). KIMO has adopted four resolutions on microplastics and marine litter, most recently a resolution on identifying and improving legislation to reduce marine litter in 2011 (Resolution 3/11). According to the organisation's website, ongoing activities include a project called "Fishing for Litter" and a proposal for scientific study on the environmental effects of microplastics. KIMO Denmark is currently involved in monitoring of beach litter for the Danish Nature Agency and has recently finalised an information dissemination project under the Marlisco project including a video competition, a travelling exhibition and various teaching materials.

# 6.5 Initiatives of environmental and consumer organisations Plastic Change

Plastic Change is an international organization based in Denmark, which aims a creating awareness about the consequences of the increasing plastic pollution in the oceans and in the environment. Specifically, Plastic Change works with documenting plastic pollution and educating the public and politicians. This work includes sampling expeditions in the Caribbean and the launch of a series of campaigns trying to engage citizens in cleaning and removing of plastic litter from the environment and exploring why the use of plastics leads to it ending in the environment.

Along with the Danish Plastics Federation and the Danish Ecological Council, Plastic Change is running the project "Projekt Plastfrit Hav" (Project Plastics-free Sea)<sup>27</sup>. The project includes two components; i) analyse the plastic litter washed ashore along the west coast and ii) identifying and analysing the microplastics that ends up in sewage from Danish households and from industrial sources and identify possible solutions<sup>28</sup>.

#### The Danish Ecological Council

As mentioned the Danish Ecological Council, Plastic Change and Danish Plastics Federation are running "Projekt Plastfrit Hav" (Project Plastics-free Sea)<sup>29</sup>. Besides that, the Danish Ecological Council has been heavily involved in discussions about how to reduce the use of plastics in general and plastic waste specifically. For instance, the Danish Ecological Council provided a series of recommendations in response to a public hearing regarding the Green book prepared by the European Commission. This included the call for the application of the Precautionary Principle when it comes to the use of microplastics as microplastics do not have any positive consumer effects and are known to have adverse environmental effects<sup>30</sup>.

28

http://www.plasticchange.org/dk/om-plastic-change/vision-og-mission/

http://www.plasticchange.org/dk/nyheder/

http://www.plasticchange.org/dk/kampagner/hvorfor-ender-plastik-i-naturen/

http://www.plasticchange.org/dk/kampagner/strandrensning/

<sup>&</sup>lt;sup>27</sup> http://www.plast.dk/aktuelt/nyhed/Helle-Fabiansen-Utraditionelt-samarbejde-skaerper-kampen-imod-plastaffald-i-havet-

<sup>&</sup>lt;sup>29</sup> http://www.plast.dk/aktuelt/nyhed/Helle-Fabiansen-Utraditionelt-samarbejde-skaerper-kampen-imod-plastaffald-i-havet-

 $<sup>{\</sup>color{red}^{30}}\ http://www.ecocouncil.dk/en/udgivelser/horingsvar/2013/2150-europaeisk-strategi-for-plast affald-i-miljoet {\color{red}^{30}}\ http://www.ecocouncil.dk/en/udgivelser/horingsvar$ 

#### **Danmark Naturfredningsforening**

Danmark Naturfredningsforening (DN) is the largest nature and environmental NGO in Denmark and works on a number of issues related to waste and plastics. When it comes specifically to microplastics, Danmark Naturfredningsforening have called for the Danish Government and the Danish Parliament to set the long-term and ambitious objectives of no new releases of waste into the marine environment by 2035 and the amount of marine waste found in the marine environment shows a strong downward trend. DN has also called for adequate monitoring of the amount and type of marine waste as well as the exploration of ways to collect waste from the sea and the removal of waste facilities and vessels at sea<sup>31</sup>.

#### **Beat the Microbead**

In 2012, two Dutch NGOs - the North Sea Foundation and the Plastic Soup Foundation launched a smartphone app under the campaign known as "Beat the Microbead" that allows consumers in the Netherlands to scan personal care products to check for the presence of plastic microbeads. By scanning the bar code of a given product with a smartphone, the App will indicate whether the product contains microbeads, whether manufacturers have indicated that they will replace microbeads from the product or whether the products in free from microbeads. In 2013, UNEP and UK based NGO Fauna & Flora International has partnered with these two Dutch foundations to further develop the App for international use. More than 70 NGOs from 33 countries support the campaign including Plastic Change from Denmark32.

#### Seas At Risk

Seas At Risk is a coalition of 28 national and international environmental NGOs from fifteen countries including Danmarks Naturfredningsforening and Levende Hav. In 2014, Seas At Risk published a joint NGO paper on priorities for the Marine Strategy Framework Directive (MSFD) programme of measures, which included detailed calls for a EU wide ban of plastic microbeads in personal care products and more funding for research to close the knowledge gaps related to marine litter: including sources, pathways, and effects on ecosystems and organisms<sup>33</sup>.

#### Greenpeace Austria, BUND and Forby Mikroplast

Greenpeace Austria and other NGOs such as Forby Mikroplast in Norway and Bund für Umwelt and Naturschutz Deutschland (BUND) have launched public online campaigns to sign a petition that calls for various kinds of action to be taken. For instance, the petition of Greenpeace Austria calls for the environmental ministers of Austria to make a binding commitment to, among others, reduce the use of plastic bags by 80% within 4 years in Austria and implement a ban of free plastic bags and of microplastics in cosmetics34.

#### Clean Coasts and Project Blue Sea

A number of NGOs are involved in activities related to engaging volunteers in removing marine litter from beaches and coasts all over Europe. For instance, in Ireland Clan Coasts is organizing the "Clean Coasts Big Beach weekend" in 2015 and according the Clean Coasts, 560,000 volunteers removed more than 7,000 tonnes of litter in more than 90 different countries35.

https://secured.greenpeace.org/austria/de/aktivwerden/proteste/konsum/mikroplastik/

<sup>31</sup> http://www.dn.dk/Default.aspx?ID=17

<sup>32</sup> http://www.beatthemicrobead.org/en/in-short

<sup>33</sup> Seas At Risk member organisationshttp://www.seas-at-risk.org/about-us/members.html;

http://www.seas-at-risk.org/images/pdf/archive/2014/NGO\_priorities\_for\_PoM\_with\_additional\_chapters\_-\_FINAL\_17\_October\_2014.pdf

<sup>;</sup> http://www.bund.net/index.php?id=19340; https://www.bund.net/index.php?id=22479

<sup>35</sup> http://www.projectbluesea.de/muell-im-meer.html

# 6.6 Initiatives of industry and business organisations Operation Clean Sweep®

Operation Clean Sweep® (OCS) is an international programme designed to prevent resin pellet loss and help keep pellets out of the marine environment <sup>36</sup>. The programme was launched in 1992 by the Society of the Plastics Industry (SPI) which is the plastics industry trade association representing the third largest manufacturing industry in the United States. The program is administrated in Europe by the Association of Plastic Manufacturers in Europe, PlasticsEurope. In Denmark, the program is administrated by the Danish Plastics Federation and has today eight participating companies<sup>37</sup>. As part of the programme, guidelines on good practices to prevent pellets loss have been developed. A large number of companies worldwide have taken the OCS pledge and have become OCS program partners to help control pellet loss worldwide. Operation Clean Sweep® provides support to configure the industrial sites to prevent pellet loss and assist companies to implement the company's prevention plan, including containment and cleaning procedures to be developed in a five-step training programme.

Operation clean sweep is part of the worldwide project "Marine litter solutions".

#### Projekt Plastfrit Hav (Project Plastics-Free Sea)

The Danish Plastics Federation co-operates with the two environmental NGOs Danish Ecological Council and Plastic Change on "Projekt Plastfrit Hav" (Project Plastics-free Sea)<sup>38</sup>. The project includes two components; i) analyse the plastic litter washed ashore along the west coast and ii) identifying and analysing the microplastics that ends up in sewage from Danish households and from industrial sources and identifying possible solutions.

#### **Marine Litter Solutions**

In March 2011, representatives of plastics organizations from around the globe released a "Declaration for Solutions on Marine Litter. Forty-seven world plastic organizations in 29 countries signed the pledge, which describes steps that the industries will take, and suggests approaches and platforms for global cooperation and future partnerships. Organisations around the world run many initiatives under the umbrella "Marine litter solutions"<sup>39</sup>.

In Europe, Plastics Europe has initiated or participates in a number of initiatives within the framework of Marine Litter Solutions (see details at the website):  $^{40}$ 

- Information campaigns and public participation: Cool Seas Bottle Champion, Cuaderno de bitácora (Spain), Goletta Verde (Italy), Love where you live, MARLISCO, Recycling Rejs 2015 – Baltic Sea, Spiagge e Fondali Puliti (Italy), Vacances Propres.
- Scientific studies and conferences: Identiplast Conference, Mosa Pura (research project on the presence of floating litter in the river Maas), Mussels and lugworm study
- Management: Operation Clean Sweep (see above), Zero plastics to landfill, Seeking solutions for the north Sea

#### **Waste Free Oceans (WFO)**

Waste Free Oceans (WFO) is a public-private Foundation aimed at mobilizing and uniting the fisheries sector, public authorities and the international plastics industry in combatting the growing issue of floating litter on the coastlines, at the rivers and in the seas <sup>41</sup>. The partners are mainly

<sup>36</sup> http://www.opcleansweep.org/; http://www.opcleansweep.eu/;

<sup>37</sup> http://www.plast.dk/Miljoe/Operation-Clean-Sweep/

<sup>38</sup> http://www.plast.dk/aktuelt/nyhed/Helle-Fabiansen-Utraditionelt-samarbejde-skaerper-kampen-imod-plastaffald-i-havet-

<sup>39</sup> http://www.marinelittersolutions.com/

<sup>40</sup> http://www.marinelittersolutions.eu/

<sup>41</sup> http://www.wastefreeoceans.eu/

private companies in the plastics sector, ports, universities and other research institutions. The website of the organisation does not include a description of actual projects (July 2015).

#### **Plastics Disclosure Project (PDP)**

The Plastics Disclosure Project<sup>42</sup> is an international initiative with the objective of reducing plastic waste in the environment, encouraging sustainable business practices vis-a-vis plastic and inspiring improved design & innovative solutions. The work of the PDP is overseen by a global Steering Committee, drawn from investors and other stakeholders who support PDP's mission. PDP's is supported by UNEP and several other organisations.

The PDP requests annual reporting regarding the production, use, handling and management of plastic and plastic waste by organisations. By measuring the amount of plastic that flows through an organisation, efficiencies can be gained in cost and waste reduction, new design, new materials, and better recycling. By reviewing how the material is managed, organisations can recognise risks and seize opportunities. All types of companies can participate in PDP, whether publically or privately held, since almost all of us use plastic in one way or another for different parts of our businesses or organizations. The main focus of the PDP is on companies in the consumer goods and technology sectors, as well as other service industries such as airlines and hotels that may use or sell plasticintensive products on a relatively large scale. Annual reports are still not available, but the organisation has co-funded a recent publication on measuring, managing and disclosing plastic use in the consumer goods industry (UNEP 2014b).

<sup>42</sup> http://www.plasticdisclosure.org

# 7. Possible new studies and initiatives

#### 7.1 Possible new studies

#### 7.1.1 Main data gaps identified

Recommendations from GESAMP (2015) for further research activities are listed in Appendix 3. They represent the research activities considered of most relevance by the international scientific community. The GESAMP report does not include a list of data gaps, but the research activities listed in Appendix 3 are aimed at closing identified data gaps, and the list thus indicates the main data gaps of relevance for the international scientific community.

The main data gaps identified in this report are summarised in Table 31. Besides the general data gaps of importance to the international community, the table include data gaps more specific to the Danish situation. It is furthermore indicated if the data gaps are linked to the catalogue of possible new studies potentially to be initiated by Danish authorities listed in next section.

TABLE 31 MAIN DATA GAPS IDENTIFIED

Area	Data gaps	Linked to catalogue of possible new studies
Use, consumption and releases of primary microplastics	Several applications of primary microplastics are still not assessed	Partly - study on micro- plastics in households Complementary study of sources not covered in this survey.
	Data on the use of synthetic wax is limited and it is not known whether synthetic waxes may lead to environmental problems of the same kind as microplastics	Synthetic wax - a source of emissions of persistent particles?
	The total releases from the use of plastics raw materials in Denmark are still quite uncertain	
Releases of secondary microplastics	The formation of secondary microplastics from paint (in particular marine) and the resulting releases to the aquatic environment is very uncertain.	
	The significance of the formation of microplastics from self-polishing antifouling paints is not known.	
	The significance of releases from the use of shoes, plastic utensils, scouring pads, synthetic fibre cloths, etc. in households are very uncertain.	Study on formation on microplastics in households.
	The formation of secondary microplastics from a number of expectedly minor sources in a Danish context is not described	Complementary study of sources not covered in this survey.

Area	Data gaps	Linked to catalogue of possible new studies
Sources of microplas- tics in the environ- ment	Much data on sources of macroplastics in the sea are available but modelling data of the formation of microplastics from the macroplastics in the Danish waters is lacking.	
	Data on the polymer composition of microplastics in the environment are scarce and would be very useful for the understanding of the sources. Detailed data on fibres, to be able to distinguish between clothing, other textiles and fishing tools are missing.	Modelling of formation from marine litter and fate of microplastics in the environment.
Occurrence and fate in the sewage treatment plants	No data are available on the fate of microplastics in Danish STPs and no data are available on microplastics in inflow to the plants (morphology, size, quantities, and polymer content).	Study on the fate of microplastics in Danish sewage treatment plants.
	The fate of particles <20 $\mu m$ in sewage treatment plants is unknown - a large fraction of secondary microplastic particles is <20 $\mu m$ .	
Occurrence in the environment	Data on microplastics in the freshwater and estuarine environment in Denmark are missing.	
	More data on microplastics in sediments and the water column are needed in order to model the fate of microplastics in the environment.	Ongoing monitoring activities.
Effects, exposure, risks in the environment	Limited data are available on the effect on freshwater organisms.	Effects of freshwater organisms and especially species used for regulatory testing purposes.
	Hardly any data are available on the fate of microplastics in the soil environment, the exposure of soil organisms and effects on soil organisms	Study on the fate of microplastics in soil and the possible effects of on soil organisms.
Formation and fate of microplastics in the environment	More data on decomposition/fragmentation rates of plastics in the micro and nano range in the environment in order to determine the long-term fate of plastics in the environment is needed.	Degradation rates of dif- ferent kinds of plastics and controlled uptake and bioaccumulation studies.
	Of particular importance is to study the persistence of secondary microplastic particles as compared with primary microplastics.	Fate of microplastics from freshwater to the marine environment.
	More detailed modelling of long-range transport of macro-plastics and microplastics is needed in order to assess the influence of microplastics on the global transport of hazardous substances.	
	Data and modelling of vertical transport of microplastics in the ocean and the further fate of the microplastics in the deep oceans is needed in order to understand potential long-term effects plastics in the ocean.	
	Hardly any data on occurrence, fate and effects of plastics in the nano-range are available.	Fate of plastics in the nano range on the aquatic environment and effects of such plastics on organisms.

Area	Data gaps	Linked to catalogue of possible new studies
Human exposure, health effects and risks	Data on human exposure to microplastics from all sources and data on possible effects are need- ed in order to assess possible adverse effects of microplastics in food, the indoor environment and the outdoor environment	Monitoring of microplastics in food in Denmark.
Sampling and analysis	Development of standardised methods for sampling and analysis in the environment and sewage treatment plants	

#### 7.1.2 Catalogue of possible new studies to be initiated by Danish authorities

The following catalogue includes studies that are considered of highest relevance for the further development of policies and strategies in Denmark, and focuses on possible new studies to be initiated by Danish authorities.

Studies already initiated or planned as part of the Danish Sea Strategy (Danmarks Havstrategi) as described in section 6.1 are not included in the catalogue.

CATALOGUE OF POSSIBLE NEW STUDIES INITIATED BY DANISH COMPETENT AUTHORITIES

Study	Objectives	Initiator	Rationale	Schedule	
Application and releases of microplastics					
Microplastics in the households in Denmark.	Improved knowledge on possible use of primary microplastics in households and better understanding of the releases from everyday products such as shoes, textiles, kitchen utensils, etc. and the potential for reducing the releases from households. The survey may also provide better data on microplastics in household dust.	Danish EPA	The survey indicates that many everyday products may be significant sources, but the estimates are still based on a limited database and it might be appropriate to obtain better understanding on release rates in order to suggest measures for reduction of formation and releases.  Denmark has a strong tradition for surveys of chemicals in consumer products and the possible exposure of consumers.	2016	
Further information on applications of primary microplas- tics and sources of secondary micro- plastics not covered by this study.	To complement the assessment of the sources covered by this assessment.	Danish EPA	Within the limits of this survey, a detailed assessment of all sources has not been possible. In order to have a comprehensive view of all sources, it might be useful to cover minor sources as well.	2015-2016	
Synthetic wax - a source of emissions of persistent particles?	Better understanding of the significance of polymer waxes as sources of particles to the environment.	Danish EPA	Some uncertainties exist on whether polymer waxes should be considered microplastics with the potential for effects similar to microplastics.	2016	

Study	Objectives	Initiator	Rationale	Schedule
Occurrence and fate				
Occurrence and fate on microplastics in Danish sewage treatment plants (STPs).	Improved knowledge on the sources of microplastics to the Danish STPs and on the releases from the STPs to the environment.	Danish EPA	With respect to further measures on reducing the environmental load with microplastics nationally and internationally, it is appropriate to have national data on sources of microplastics to the sewage treatment plants and releases from the plant. It is suggested to include analysis of polymer composition of microplastics in inflow to the STP, which would be useful in the identification of the main sources. For the fibres, it is e.g. relevant to have more knowledge on the significance of various textiles.  Furthermore, it is proposed to include mass balances.	2015-2016
Microplastics in food in Denmark.	Survey of microplastics in food in Denmark.	Danish Veterinary and Food Admin- istration	Very limited information on microplastics in food is available and data across the EU would be valuable for an assessment of the possible risks of microplastics in food. It is suggested to include analysis of polymer composition of microplastics and measure particle number and composition, particle mass and - if possible - presence of hazardous substances. Particle mass could be useful for the first assessment of the potential for the microplastics as carrier of hazardous substances.	2016
Modelling of formation from marine litter and fate of microplastics in the environment.	Better understanding of the importance of marine litter for the formation of microplas- tics in the environment in the waters around Denmark.	Danish Nature Agency	A wealth of information on sources of macroplastics and plastics on the beaches are available, but it is poorly understood to what extent this contributes to microplastics in the environment.	2016-2017
Fate of secondary microplastics in sedimentation basins.	Better understanding of sedimentation and removal of microplastics from sedimentation basins.	Danish Nature Agency	Knowledge on capacity of sedimentation basis is important for an assessment of the potential and the need to establish more sedimentation basins.  The efficiency of sedimentation basins in reducing the releases of heavy metals and hazardous substances has been evaluated in recent year and the evaluation could be supplemented with an evaluation of microplastics.	2016

Study	Objectives	Initiator	Rationale	Schedule
Polymer composition of microplastics in the environment in Denmark.	Improved understanding of the sources of microplastics in the environment.	Danish Nature Agency	The available data on polymer composition of microplastics in the environment indicates that such data may be very useful in understanding the sources of microplastics in the environment, but more data are needed for a better understanding.	2016
Degradation rates of different types of microplastics and controlled uptake and bioaccumulation studies.	Improved understanding of how the degradation rates are impacted by physical and biological parameters and vary between different types of plastics and how this affects uptake and bioaccumulation in the food chain.	Danish EPA	Quantification of the degradation rates of different types and sizes of plastics is key when it comes to understanding the fate and behaviour of microplastics as well as when it comes to investigating uptake and bioaccumulation in the food chain. It would be highly relevant to investigate the persistency of the secondary microplastics, e.g. from paint and tyres, in comparison with the primary microplastics.	2016-2017
Fate of microplastics from freshwater to the marine environments.	Improved knowledge about the fate and behaviour of microplastics and how microplastics are dispersed once released into the environment and into the food web.	Danish EPA	Most microplastics are released into the marine environment via fresh- water pathways; understanding the fate and behaviour of microplastics is important when it comes to mini- mizing environmental impacts most effectively.	2015-2017
Effects of microplastic	es			
Effects on freshwa- ter organisms and especially species used for regulatory testing purposes.	Improved knowledge about the adverse effects that microplas- tics might have on freshwater organisms e.g. algae, crustaceans and fish.	Danish EPA	Limited research has been conducted on the effect of microplastics on freshwater organism despite the fact that most microplastics are released to the marine environment via freshwater pathways.  For regulatory actions on the use of microplastics, data on effects on standard organisms may be desirable.	2015-2017
Fate of microplastics in soil and the possible effects on soil organisms.	Improved understanding of the possible effects of using sewage sludge with microplastics for agricultural purposes.	Danish EPA	It is of importance for the considerations about microplastics in waste water to evaluate whether microplastics in soil constitute a problem. If the evaluation indicates a potential significant risk to soil organisms, the consequences may be not to apply sludge on agricultural soil and/or further reduce the sources of microplastics to sewage.	2015-2017

Study	Objectives	Initiator	Rationale	Schedule
Fate of plastics in	Better understanding of	Danish EPA	It is assumed that microplastics	2016-2018
the nano range in	degradation rates for		ultimately are fragmented into	
the aquatic envi-	various kind of plastics		plastics in the nano range, and it is	
ronment and effects	in the nanorange, and		important to know if plastics in the	
of plastics in the	possible effects on		nano range are persistent and accu-	
nano range on or-	organisms.		mulate in the environment or are	
ganisms.			relatively quickly decomposed.	
			Furthermore, a better understanding	
			of the possible effects of plastics in	
			the nano range can indicate the	
			possible long-term consequences of	
			the pollution with plastics.	
			During recent years, detailed as-	
			sessments of the possible environ-	
			mental and health impacts of nano-	
			materials have been undertaken by	
			the Danish EPA, and there a strong	
			tradition is in Denmark for assess-	
			ments of materials in the nano-	
			range.	

#### 7.2 Possible other new initiatives

This section includes a list of possible new initiatives beyond those already taken or planned as part of the Danish Sea Strategy (Danmarks Havstrategi).

The list focuses on initiatives directly linked to the use of primary microplastics and the releases of microplastics and macrolitter to the environment. Some initiatives for improved waste management, resource management and recycling of plastics may indirectly also reduce the potential for releases of the plastics to the environment, but are considered beyond the scope of this report.

The OSPAR Regional Marine Litter Action Plan sets a number of actions for the period 2014 to 2018 (see Appendix 2). The initiatives are mainly of a preparatory character (establishment of database and exchange platform and development of maps, identify hot spot areas, promote initiatives, evaluate all products and processes). At this stage, the initiatives do not commit the Parties to specific actions, but various follow up actions are expected on the basis of the preparatory work. As part of the Intersessional Correspondence Group on Marine Litter (ICG-ML) Denmark is involved in initiatives including: i) Develop and agree on regionally coordinated SMART reduction/operational targets linked to relevant actions as contained in this implementation plan, starting from 2015, including those linked to sources. ii) Identify best practice in relation to inspections for MARPOL Annex V ship generated waste, including better management of reporting data, taking into consideration the Paris Memorandum of Understanding on Port State Control. iii) Reduce the consumption of single use plastic bags and their presence in the marine environment, supported by the development of quantifiable (sub) regional targets, where appropriate, and assist in the development of relevant EU initiatives. iii) Develop marine litter assessment sheets to assist Contracting Parties in developing material for education programmes, including those for professional seafarers and fishermen.

The HELCOM Regional Marine Litter Action Plan (see Appendix 2) proposes a number of actions at the regional level. Lead countries for the various actions are still not appointed.

#### 7.2.1 Catalogue of potential other new initiatives

Possible initiatives of Danish Authorities (other than studies mentioned in previous sections) are listed in Table 33.

 $\begin{array}{c} \textbf{TABLE 33} \\ \textbf{CATALOGUE OF POSSIBLE OTHER INITIATIVES TO BE INITATED BY DANISH AUTHORITIES} \end{array}$ 

Initiative	Objectives	Partners	Rationale/background	Time perspective
Development of national action plan on microplastics and marine litter.	Establish an overview and coordinate actions on microplastics and marine litter.	Danish EPA, Danish Nature Agency, Danish Veterinary and Food Administration, Danish Health and Medicines Authority	Today, no overview of actions taken by the various national authorities exist.  It has, furthermore, been suggested by Danish Chamber of Commerce to extend the scope of the action plan to include actions by other stakeholders.	2016
Working for a ban of microplastics in cosmetics and cleaning agents in the EU.	To eliminate the use of primary microplastics in cosmetics and cleaning agents and the releases to the environment due to this use.	The Danish EPA in cooperation with other Member States encourage the European Commission to propose a ban on microplastics in cosmetics.	A ban is considered the only effective measure for eliminating the use of microplastics in cosmetics. Alternatives are available and many companies have already phased out the microplastics.  According to the cosmetics industry, a voluntary agreement on the phase out would not be possible due to the legal issues.	2016-2017
Include in the standard requirements for plastics converters (under Order of Environmental Permitting) requirement concerning losses of plastic pellets.	To reduce and possibly eliminate the losses of plastic pellets to the environment from transport, unloading as use of the pellets.	Danish EPA	Loss of plastic pellets can be effectively reduced by simple management measures. By including requirements in environmental permits, it can be ensured that such measures are taken.	2015-2016
Distribution of information on options to reduce losses of plastic pellets to municipalities and companies in the plastics sector.	Same as above, but targeting companies not addressed by the environmental per- mitting require- ments.	Danish EPA	Same as above	2015-2016
Extend existing guide- lines for management of bottom paint to include other types of marine paints.	Provide information to owners of pleasure boats and shipyards about microplastics in order to reduce the releases from maintenance activi- ties.	Danish EPA	During the last decade, much awareness has been raised regarding preventing releases of antifouling paints, and procedures for reducing the releases have been implemented. The procedures is to a lesser extent applied when other marine paints are maintained.  May be implemented in cooperation with Danish Sailing Association and Maritime Denmark.	2016

Initiative	Objectives	Partners	Rationale/background	Time perspective
Participation in the implementation of the OSPAR and HELCOM Regional Marine Litter Action Plans.	Work for an efficient implementation of the regional marine litter action plans.	Danish Nature Agency	The regional marine action plan include a number of activities. In addition to the regional actions, which Denmark is already obliged to implement, the Helcom action plan suggests voluntary national actions, which should be considered.	2015-2018
Ask the European Food Safety Authority (EFSA) for a scientific opinion on the occurrence of microplastic particles and plastics in the nano range in food, especially in seafood.	More information on human exposure to microplastics and the possible health effects.	Danish Veterinary and Food Administration	More information on the possible health effects of microplastics is needed. The activity is linked to a survey of microplastics in food in Denmark.	2015

# 8. Abbreviations and acronyms

ABS Acrylonitrile butadiene styrene

BBP Benzyl butyl phthalate

BFr Bundesinstitut für Risikobewertung (German Federal Institute for Risk Assesment)

BPA Bisphenol A

BUND Bund für Umwelt and Naturschutz Deutschland CMR Carcinogenic, mutagenic or toxic to reproduction

DBDPE Decabromodiphenyl ethane

DBP Dibutyl phthalate

DDE Dichlorodiphenyldichloroethylene degradation product of DDT

DDT Dichlorodiphenyltrichloroethane

DecaBDE Decabrominated diphenylether, same as BDE-209

DEHP Bis(2-ethylhexyl)phthalate DEHT Dioctyl terephthalate

DFL Danish Coating and Adhesives Association

DIBP Diisobutyl phthalate
DINP Diisononyl phthalate
DIY Do It Yourself

DN Danmark Naturfredningsforening/Danish Nature Conservancy Association

EBTEBPI Ethylene bistetrabromophthalimide

EcoQO Ecological Quality Objective
EFSA European Food Safety Authority
EPA Environmental Protection Agency
EPDM Ethylene propylene diene monomer

EPS Expanded polystyrene
ESD Emission Scenario Document
EVA Ethylene-vinylacetate copolymers

FTIR Fourier transform infrared spectroscopy

GESAMP Joint Group of Experts on the Scientific Aspects of Marine Environmental Protec-

tion

GPA The Global Programme of Action for the Protection from the Marine Environment

from Land-based Activities

GPML Global Partnership on Marine Litter

HAV Marine Group under the Nordic Council of Ministers

HBCDD Hexabromocyclododecane
HCH Hexachlorocyclohexane isomer
HDPE High density polyethylene
HELCOM The Helsinki Commission
HIPS High-impact polystyrene

ICES The International Council for the Exploration of the Sea ICG-ML Intersessional Correspondence Group on Marine Litter

ICIS database Integrated Compliance Information System IMO International Maritime Organization

INCI International Nomenclature of Cosmetic Ingredients
IOC Intergovernmental Oceanographic Commission

KIMO The Local Authorities International Environmental Organisation

LDPE Low density polyethylene

LOUS List of Undesirable Substances (of the Danish EPA)

MarLIN The Marine Life Information Network
MCCP Medium-chain chlorinated paraffins
NIVA Norwegian Institute for Water Research

NP Nonylphenols

OCS Operation Clean Sweep®

OECD Organisation for Economic Co-operation and Development

OP Octylphenols

OSPAR The Oslo and Paris Commission

OTR Off the Road PA Polyamides (nylon)

PAH Polycyclic aromatic hydrocarbons
PBDE Polybrominated diphenyl ethers
PBT Persistent, Bioaccumulative and Toxic

PC Polycarbonate

PCB Polychlorinated biphenyl
PDP Plastics Disclosure Project
PDV Phocine Distemper Virus

PE Polyethylene PE Polyethylene

PE Population equivalent = person equivalent

PEST Polyester

PET Polyethylene terephthalate (polyester)

PFO Flexible polyolefin
PFOA Perfluorooctanoic acid

PLA Polylactic acid

PM<sub>2.5-10</sub> Particulate matter in the size from 2.5 to 10  $\mu$ m

PMMA Polymethyl methacrylate POPs Persistent organic pollutants

PP Polypropylene
PS Polystyrene
PS Polystyrene

PTFE Polytetrafluoroethylene

PU Polyurethane
PUR/PU Polyurethane
PVA Polyvinylacetate
PVC Polyvinyl chloride

REACH Registration, Evaluation, Authorisation and Restriction of Chemical substances

(Regulation EC 1907/2006).)

ROS Reactive Oxygen Species
SAN Styrene Acrylonitrile
SBR Styrene-butadiene rubber

SEBS Styrene ethylene/butylene styrene copolymer

SPERCs Specific Release Categories SPI Society of the Plastics Industry

SPT Association of Danish Cosmetics, Toiletries, Soap and Detergent Industries

STP Sewage (municipal waste water) treatment plant

SVHC Substances of Very High Concern

 $\begin{array}{ll} {\it TBBPA} & {\it Tetrabromo\ bisphenol\ A} \\ {\it TCEP} & {\it Tris2-chloroethylphosphate} \end{array}$ 

TCPP Tris2-chlor-1-methylethylphosphate

TOC Total Organic Carbon
TPE Thermoplastic elastomer

UNECE The United Nations Economic Commission for Europe

UNEP United Nations Environment Programme

UPE Unsaturated Polyester
UV Ultraviolet (light)
UV-B Ultraviolet B (light)
WFO Waste Free Oceans
XPS Extruded polystyrene

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### Appendix 1 Complementary information on microplastics in cosmetics

Polymers in personal care and cosmetics products are named in accordance with the International Nomenclature of Cosmetic Ingredients (INCI) and indicated in the ingredients list of the marketed products. INCI names and function of the polymers in personal care and cosmetics products formulations are summarised in the table below on the basis of information from the EU Cosmetic Ingredient 'CosIng' Database. The fact that the polymeric ingredients have an INCI name and are included in the CosIng database do not imply that they are actually in current use.

The INCI name indicates the type of polymer but is not sufficient to determine whether the polymer is present as a liquid, a gel or a solid. To determine if a polymeric ingredient is a solid particle or not, additional chemical information on top of the INCI name would be required; it is consequently not possible from the ingredient list of cosmetic products to determine if the products contain plastics microbeads. For some product types such as exfoliating scrubs, the polymers are typically present as plastic microbeads and the information on the ingredient list gives a good indication of the possible presence of plastic microbeads.

For many of the polymers, the indicated function in Appendix 1 is 'viscosity controlling', 'film forming', 'emulsion stabilising' and other functions where the polymer most likely is not present as plastic microbeads, but rather in a solution or gel. Functions that indicate the presence of microbeads are 'abrasive' (PE), 'coloured microspheres' (styrene acrylates copolymer) and 'aesthetic agent' (polyethylene terephthalate (PET)).

Polymer ingredients are part of the formulation for a variety of personal care and cosmetic products such as toothpaste, shower gel, shampoo, creams, eye shadow, deodorant, blush powders, make-up foundation, skin creams, hairspray, nail polish, liquid makeup, eye colour, mascara, shaving cream, baby products, facial cleansers, bubble bath, lotions, hair colouring, nail polish, insect repellents and sunscreen (Leslie 2014). Polymer ingredients are present in different products at different percentages, ranging from a fraction of a percent to more than 90% in some cases. In an unpublished survey from the USA in 2013, PE had a highest maximum use concentration of 67.6%, which was reported in skin cleansing preparations (Cosmetics Ingredient Review 2015). Hydrogenated polyisobutene had a highest maximum use concentration of 95%, which was reported in lipstick (Cosmetics Ingredient Review 2015). Concentrations of plastic microbeads are further described below.

Besides the carbon-based polymers, inorganic-organic polymers with a silicon-oxygen backbone, based on siloxanes or polysiloxanes are used. Most of the silicon-based polymers are water soluble or water-dispersible, and therefore are not solid materials. However, some cosmetics (e.g. foundation makeup) contain solid silicone resins for their film-forming properties, or because they are able to add structure to products, such as lipstick (Leslie 2014).

TABLE 34
SELECTED EXAMPLES OF POLYMER INGREDIENTS CURRENTLY APPLIED IN PERSONAL CARE AND COSMETICS PRODUCTS ACCORDING TO THE COSING DATABASE (LESLIE 2014)

INCI name of polymer or co-polymer	Function in personal care and cosmetics products formulations *
Nylon-12 (polyamide-12)	Bulking, viscosity controlling, opacifying (e.g. wrinkle creams)
Nylon-6	Bulking agent, viscosity controlling
Poly(butylene terephthalate)	Film formation, viscosity controlling
Poly(ethylene isoterephthalate)	Bulking agent

INCI name of polymer or co-polymer	Function in personal care and cosmetics products formulations *
Poly(ethylene terephthalate)	Adhesive, film formation, hair fixative, viscosity controlling, aesthetic agent, (e.g. glitters in bubble bath, makeup)
Poly(methyl methylacrylate)	Sorbent for delivery of active ingredients
Poly(pentaerythrityl terephthalate)	Film formation
Poly(propylene terephthalate)	Emulsion stabilising, skin conditioning
Polyethylene	Abrasive, film forming, viscosity controlling, binder for powders
Polypropylene	Bulking agent, viscosity increasing agent
Polystyrene	Film formation
Polytetrafluoroethylene (Teflon)	Bulking agent, slip modifier, binding agent, skin conditioner
Polyurethane	Film formation (e.g. facial masks, sunscreen, mascara)
Polyacrylate	Viscosity controlling
Acrylates copolymer	Binder, hair fixative, film formation, suspending agent
Allyl stearate/vinyl acetate copolymers	Film formation, hair fixative
Ethylene/propylene/styrene copolymer	Viscosity controlling
Ethylene/methylacrylate copolymer	Film formation
Ethylene/acrylate copolymer	Film formation in waterproof sunscreen, gellant (e.g. lipstick, stick products, hand creams)
Butylene/ethylene/styrene copolymer	Viscosity controlling
Styrene acrylates copolymer	Aesthetic, coloured microspheres (e.g. makeup)
Trimethylsiloxysilicate (silicone resin)	Film formation (e.g. colour cosmetics, skin care, suncare)

<sup>\*</sup> Some polymers may be available in various forms, as dispersions in solvents, or as partially water soluble polymer forms. The functions given are examples and not an exhaustive list. Sources (as cited by Leslie 2014): EU Cosmetic Ingredient 'CosIng' Database (http://ec.europa.eu/consumers/cosmetics/cosing); Goddard and Gruber 1999; Cosmetic Ingredient Reviews, the Cosmetics & Toiletries Bench Reference (https://dir.cosmeticsandtoiletries.com) and various manufacturer websites.

### Example of distribution by product group

In recent years, microbeads in personal care and cosmetic products have gained attention from many NGOs and public authorities and NGOs have put an increasing pressure on manufacturers to phase out microplastics. The "Beat the Microbead" campaign, led by the Dutch NGO Plastic Soup Foundation, is supported by 62 NGOs from 31 countries (April 2015) and by UNEP. As part of the campaign, an app for mobile phones was developed where, by scanning the barcode information, the presence of plastics microbeads in the product is indicated, and it is indicated whether the manufacturer has provided data for most EU Member States. The app also contains lists of tested products. The results for the Netherlands, which is among the most comprehensive country data set, are shown in the table below. About 40% of all scrub products contained microbeads and for half of these, the manufacturers had reported that they were in the process of phasing out the plastic microbeads. The data indicate that a higher percentage of the cleansing and scrub products than assumed in the Cosmetics Europe survey may contain microbeads. The percentage for "douche" products is lower but this product group probably covers a wider range of products than the "shower gel" included in the Cosmetics Europe survey.

 $\frac{\text{TABLE 35}}{\text{BEAT THE MICROBEAD APP RESULTS FOR THE NETHERLANDS FEBRUARY 2015 (ROUNDED FIGURES)}}$ 

Product group	Total number of products	Without micro- plastics, per- centage	With microplastics, in the process of being phased out according to manu- facturer	With microplas- tics, other
Cleansing (facial)	69	67%	25%	9%
Douche	495	97%	2%	0.2%
Scrub	272	60%	19%	21%
Toothpaste	124	90%	9%	2%
Total	960	84%	10%	7%

### Appendix 2 OSPAR and HELCOM action plans for marine litter

TABLE 36
PLANNED IMPLEMENTATION FOR COMMON ACTIONS OSPAR CONTRACTING PARTIES WILL TAKE TO COMBAT MARINE LITTER IN THE NORTH EAST ATLANTIC ACCORDING TO THE OSPAR MARINE LITTER REGIONAL ACTION PLAN (OSPAR 2014)

Regional action plan (RAP) § no.	Action	Lead Party / Parties	Implemen- tation year
29	Develop and agree regionally coordinated SMART reduction/operational targets linked to relevant actions as contained in this implementation plan, starting from 2015, including those linked to sources.	OSPAR Litter Expert Group (ICG-ML)	To be decided at ICG-ML
	Theme A: Actions to combat sea-based source		
30	Ensure regional coordination on the implementation of EU Directive 2000/59/EC in relation to MARPOL Annex V ship generated waste. Such coordination could: a) deliver a cost recovery system, ensuring the maximum amount of MARPOL Annex V ship generated waste is delivered to port reception facilities; b) not solely focus on reception facilities, but also other relevant differences; c) analyse the implementation of compulsory discharge of waste in each port for all ships leaving the OSPAR maritime area for non-EU ports, in line with EU Directive 2000/59/EC.	Belgium, Germany, Netherlands, With assis- tance from Seas at Risk	2017
31	OSPAR will assist the European Commission in the ongoing revision of EU Directive 2000/59/EC.	Sweden and Germany, with assistance from Seas at Risk and the Secretariat	2014
	Enforcement of international legislation/regulation regarding all sectors		
32	Identify best practice in relation to inspections for MARPOL Annex V ship generated waste, including better management of reporting data, taking into consideration the Paris MOU <sup>43</sup> on port state control.	Tbc ICG-ML	
33	Seek dialogue with the Paris MOU to take the risk of illegal waste discharges into consideration for the prioritisation of port state control inspections.  Incentives for responsible behaviour/disincentives for littering		2016
34	Improve implementation of the ISO standard 201070:2013 in relation to port reception facilities.	Belgium and Netherlands	2016
35	Identify the options to address key waste items from the fishing industry and aquaculture, which could contribute to marine litter, including deposit schemes, voluntary agreements and extended producer responsibility.	France, Belgium and EU, with participation from Portugal	2015
	Develop best practice in relation to fishing industry	_	
36	Through a multinational project, together with the fishing industry and competent authorities develop and promote best	Sweden and the United	2016

<sup>43</sup> Paris Memoradnum of Understanding on Port State Control

Regional	Action	Lead Party /	Implemen-
action		Parties	tation year
plan			
(RAP)			
§ no.	practice in relation to marine litter. All relevant aspects (includ-	Kingdom, with	
	ing e.g. dolly rope44, waste management on board, waste man-	participation	
	agement at harbours and operational losses/net cuttings)	of Germany,	
	should be included.	the Nether-	
		lands and	
0=	Investigate the prevalence and impact of dolly rope (synthetic	Norway Netherlands	2016
37	fibre). Engage with competent authorities (such as National	Netherlands	2010
	Authorities, EU, North East Atlantic Fisheries Commission,		
	etc.) and the fishing industry in order to work together to re-		
	duce the waste generated by dolly rope on a (sub) regional		
	basis.		
38	Fines for Littering at Sea  Analyse penalties and fines issued by Contracting Parties for	Germany	2015
30	waste disposal offences at sea to highlight the differences,	Germany	2015
	trends, problem areas and issues to relevant organisations,		
	such as the North Sea Network of Investigators and Prosecu-		
	tors.		
	Theme B: Actions to combat land-based sources		
39	Improved waste prevention and management Highlight those waste prevention and management practices	Germany, with	2016
39	that impact significantly on marine litter. Engage with the	participation	2010
	industry and other authorities, at the appropriate level, in order	of the Nether-	
	for them to be able to develop best environmental practice,	lands	
	including identification of circumstances where litter "escapes"		
	into the marine environment. Encourage the recyclability of plastic products (e.g. through reduction of additives).		
40	Share best practice on waste management, e.g. on landfill bans	Germany, with	2016
	of high caloric wastes (especially for plastics).	participation	
		of the Nether-	
41	Exchange experience on best practice to prevent litter entering	lands Netherlands	2015
41	into water systems and highlight these to River or River Basin	with the assis-	2015
	Commissions.	tance of Ger-	
		many and	
		Belgium	
40	Reduction of sewage and storm water related waste Investigate and promote with appropriate industries the use of	Ireland, Nor-	2017
42	Best Available Techniques (BAT) and Best Environmental	way and Swe-	2017
	Practice (BEP) to develop sustainable and cost-effective solu-	den	
	tions to reduce and prevent sewage and storm water related		
	waste entering the marine environment, including micro parti-		
	cles.  Incentives for responsible behaviour/ Disincentives		
	for littering		
43	Assess relevant instruments and incentives to reduce the use of	Germany,	2016
	single-use and other items, which impact the marine environ-	Ireland and	
	ment, including the illustration of the associated costs and	Portugal	
4.4	environmental impacts.	Intercoggican	2015
44	Reduce the consumption of single use plastic bags and their presence in the marine environment, supported by the devel-	Intersessional Correspond-	2015
	opment of quantifiable (sub) regional targets, where appropri-	ence Group on	
	ate, and assist in the development of relevant EU initiatives.	Marine Litter	
45	Encourage international environmental certification schemes	Netherlands	2016

 $<sup>^{\</sup>rm 44}$  Bunches of polyethylene threads used to protect the codend of demersal trawlnet from abrasions

Regional	Action	Lead Party /	Implemen-
action plan (RAP)		Parties	tation year
§ no.	to include the management and appropriate of manine littonia		
	to include the management and prevention of marine litter in their lists of criteria.		
	Elimination, change or adaptation of the products for environmental benefits		
46	Evaluate all products and processes that include primary micro plastics and act, if appropriate, to reduce their impact on the marine environment.	Belgium, Germany and Netherlands	2015
47	Engage with all appropriate sectors (manufacturing, retail etc.) to explore the possibility of a voluntary agreement to phase out the use of micro plastics as a component in personal care and cosmetic products. Should a voluntary agreement prove not to be sufficient, prepare a proposal for OSPAR to call on the EU to introduce appropriate measures to achieve a 100% phasing out of micro plastics in personal care and cosmetic products.	Germany and Netherlands with the par- ticipation of Belgium, UK and SAR	Ongoing
48	Evaluate the potential harm caused to the marine environment by items such as cigarette filters/butts, balloons, shotgun wads, cotton buds and bio-film support media used in sewage plants. Based on this evaluation, proposals can be made on the elimination, change or adaptation requirements for these other potentially problematic items.	Germany	2015
49	Investigate the prevalence and impact of expanded polystyrene (EPS) in the marine environment, and engage with industry to make proposals for alternative materials and/or how to reduce its impacts.	Portugal with support from IE	
	Development of sustainable packaging		2015
50	Engage in a dialogue with industry aimed at highlighting the top marine litter problem items based on OSPAR beach monitoring surveys and/or other evidence on impacts.	Germany	
51	Explore with industry the development of design improvements to assist in the reduction of negative impacts of products entering the marine environment in order to better inform industry on alternative solutions		
	Zero pellet loss		2015
52	Promote initiatives and exchange of best practice aiming at zero pellet loss along the whole plastics manufacturing chain from production to transport.	France, with participation from Germany, Netherlands and Seas at Risk.	
	Theme C: Removal Actions		_
52	Application of Fishing for Litter activities  Strengthen the existing OSPAR Recommendation 2010/19 on		2016
53	the reduction of marine litter through implementation of fishing for litter initiatives, including by reviewing the option that any vessel involved in the scheme can land non-operational waste at participating harbours in OSPAR countries.  Cleaning environmental compartments and keeping		
	them clean		
54	Establish an exchange plaVorm on experiences on good cleaning practices in beaches, riverbanks, pelagic and surface sea areas, ports and inland waterways. Develop best practice on environmental friendly technologies and methods for cleaning.	ICG-ML with assistance from Germany and Seas at Risk	2016
55	Develop sub regional or regional maps of hotspots of floating litter, based on mapping of circulation of floating masses of marine litter, and identification of hotspots of accumulation on coastal areas and the role of prevailing currents and winds.	Portugal	2018

Regional action plan (RAP) § no.	Action	Lead Party / Parties	Implemen- tation year
	Reduction of abandoned, lost and otherwise discarded fishing gear (ALDFG)		
56	Identify hot spot areas through mapping of snagging sites or historic dumping grounds working with other initiatives, research programmes and with fishing organisations.	Norway	
57	Develop a risk assessment for identifying where accumulations of ghost nets pose a threat to the environment and should be removed.	Germany	Ongoing
	Theme D: Education and outreach		
	Education		
58	Develop marine litter assessment sheets to assist Contracting Parties in developing material for education programmes, including those for professional seafarers and fishermen.	ICG-ML	2016
	Outreach		
59	Establish a database on good practice examples of marine litter measures and initiatives and share this database with other Regional Seas Conventions in order to make action more visible to the public.	Germany, ICG-ML and Secretari- at	2016
60	Develop a communication strategy on the Regional Action Plan (RAP) linked in a coherent way with national initiatives/measures. This will include linking the OSPAR website to relevant projects and initiatives.	Secretariat	2015

TABLE 37
CONTRACTING PARTY ACTIONS FROM THE OSPAR MARINE LITTER REGIONAL ACTION PLAN 2015. THE TABLE DETAILS ACTIONS WHICH MAY BETAKEN AT THE NATIONAL LEVEL BY CONTRACTING PARTIES TO IMPLEMENT OSPAR'S REGIONAL ACTION PLAN FOR PREVENTION AND MANAGEMENT OF MARINE LITTER IN THE NORTH-EAST ATLANTIC. CONTRACTING PARTIES WILL PERORT ON THESE ACTIONS EVERY SECOND YEAR FROM 2016. (OSPAR 2014)

RAP	Action Summary	
§ no.	Theme A: Actions to combat sea-based sources of marine litter	
62	Ensuring effective implementation and enforcement of MARPOL Annex V in relation to both fishing and shipping waste.	
63	Investigating markets for plastic waste from the fishing and shipping industry	
	Theme B: Measures to combat land-based sources	
64	Ensuring considerations related to marine litter and actions in this plan are integrated, as appropriate, into the implementation and any future revision of relevant EU Directives.	
65	Seeking cooperation in the river and river basin authorities in order to include impacts of litter on the marine environment in river and river basin management plans.	
66	Promoting and supporting, where appropriate, the inclusion of measures aimed at the prevention and reduction of marine litter in the 2014 revision of the EU's waste legislation.	
67	Including a reference to marine litter, where applicable, in National Waste Prevention Plans and Waste Management Plans.	
68	Entering into dialogue with the waste industry, working towards highlighting waste management practices that impact on the marine environment.	
69	Identifying illegal and historic coastal landfill or dumpsites, including where these might be at risk from coastal erosion, and take action if appropriate.	
70	Promoting Extended Producer Responsibility Strategies requiring producers, manufacturers, brand owners and first importers to be responsible for the entire life-cycle of the product with a focus on items frequently found in the marine environment.	
71	Encouraging the development and implementation of Sustainable Procurement Policies that contribute to the promotion of recycled products	
72	Promoting and enhancing national stakeholder alliances focusing on marine litter.	
	Theme C: Removal measures	
73	Removing barriers to the processing or adequate disposal of marine litter collected in Fishing for	

	Litter initiatives, including landfilling if relevant and in line with waste legislation
74	Encouraging all fishing vessels to be involved in Fishing for Litter schemes, where they are available.
<i>7</i> 5	Ensuring tha any vessel involved in the scheme can land non-operational waste collected at sea at any participating harbour.
76	Undertaking an awareness-raising campaign to make fishermen aware of their obligations under EU Control Regulation (1224/2009) with regard to reporting, marking and retrieval of lost nets
	Theme D: Education and outreach
77	Promoting education activities in synergy with existing initiatives in the field of sustainable development and in partnership with civil society.
78	Promoting curricula for marine-related education, including the recreational sector.
79	Promoting or adopting environmental awareness courses for fishermen and the fishery sector.
80	Encourage participation in International, EU and National Litter Cleanup Campaigns.
81	Promoting the "Adopt a beach" system.
82	Raising public awareness of the occurrence, impact and prevention of marine litter, including micro plastics.
83	Supporting/initiating community/business-based producer responsibility schemes or deposit systems, for example on recycling fishing nets.
84	Developing collective agreements between Contracting Parties, NGOs and industry to tackle particular problems of marine litter.

 $\begin{array}{l} \textbf{TABLE 38} \\ \textbf{REGIONAL ACTIONS} - \textbf{HELCOM COLLECTIVE ACTIONS (HELCOM 2015)} \end{array}$ 

CODE	REGIONAL ACTION	FURTHER SPECIFICATION		
REGION	REGIONAL ACTIONS ADDRESSING LAND-BASED SOURCES OF MARINE LITTER			
General i	improved waste prevention and management			
RL1	Prepare and agree on HELCOM guidelines on marine litter references to be included in national and local waste prevention and waste management plans, i.a. an element highlighting the impacts of marine litter.	Guidelines by 2017		
RL2	Provide HELCOM guidelines on best practice routines with regard to cleaning and collection systems to prevent litter from land entering the aquatic environment.	Guidelines by 2017		
RL3	Share best practice on waste management in order to identify and address loopholes that makes waste turn into marine litter, including the issue of landfills, regulations and enforcement:	Overview report on good waste management and loopholes, taking into consideration similar action within OSPAR by 2016.		
RL4	Improvement of stormwater management in order to prevent litter, including microlitter, to enter the marine environment from heavy weather events.	By 2018 at the latest HELCOM has compiled information to give guidance on improvements of stormwater management on a local level to prevent and reduce stormwater related waste (including micro litter) entering the marine environment, taking into consideration similar action within OSPAR. If appropriate according to findings of the activity and other relevant information, amend HELCOM Recommendation 28E/5 on municipal waste water treatment.		
RL5	Establish a dialogue and negotiate on solutions with business and industry to (i) develop design improvements that reduce the negative impacts of products entering the marine environment, and (ii) reduce over-packaging and promote wise packaging	Initiatives taken by the private sector.		
Measure	Measures to tackle top items			
	Micro particles			

CODE	REGIONAL ACTION	FURTHER SPECIFICATION
RL6	Establish an overview of the importance of the different sources of primary and secondary microplastics. Evaluate products and processes that include both primary and secondary micro plastics, such as fibres from clothing, assess if they are covered or not by legislation, and act, if appropriate, to influence the legal framework, or identify other necessary measures.	By 2017, an overview on what products and processes contribute to the input of micro plastics to the Baltic Sea, taking into account similar action within OSPAR. By 2018, existing legislation is assessed and necessary measures identified together with relevant stakeholders.
RL7	Investigate and promote best available techniques as well as research and develop additional techniques in sewage treatment plants to prevent micro particles entering the marine environment.	By 2018 HELCOM has compiled information, and prepared a report on micro particles removal in sewage treatment plants taking into account similar action within OSPAR. If appropriate according to findings of the search and other relevant information, amend HELCOM Recommendation 28E/5 on municipal waste water treatment.
	Sewage related litter including sanitary waste	
RL8	Assess the importance of the contribution of upstream waste flows to the marine environment and, if needed, identify suitable actions.	By 2017 an assessment of the importance of sewage related waste coming from the upstream waste flow is produced. By 2018 share assessment with River and River Basin Commissions and identify measures including the implementation of related regulations; missing elements are identified and guidelines for improvement are presented.
	Expended Polystyrene (Polystyrene Foam)	
RL9	Compile information on the prevalence and sources of expanded poly- styrene (EPS) in the marine environment, and engage with industry to make proposals for alternative solutions (e.g. use of other materials, establishment of deposits, return and restoration systems, overpackag- ing reduction).	By 2017, an overview of the most significant sources of EPS ending up in the marine environment is produced, in cooperation with OSPAR.  Make recommendations to the Contracting Parties on voluntary agreements with the industry on changes in product design and applying best practices when handling EPS by 2019.
	Plastic bags	
RL10	Define and implement appropriate instruments and incentives to reduce the use of plastic bags, including the illustration of the associated costs and environmental impacts (e.g. establishment of levies, deposit fees, taxes or bans on plastic bags). Support regional coordination in the Baltic Sea of the implementation of the future revised Directive 94/62/EC on packaging and packaging waste to reduce the consumption of lightweight plastic carrier bags, for HELCOM Contracting Parties being EU members.	By 2018 HELCOM Contracting Parties start to coordinate and inform each other about consumption of plastic bags on an annual basis. By 2019 establish a reduction target of plastic bags, taking into account the measures which are implemented nationally.
	Bottles and containers	
RL11	Cooperate on the establishment and/or further development of deposit refund systems for bottles, containers and cans (e.g. glass, plastics and aluminum) in the HELCOM Contracting Parties in accordance with national law as appropriate. Investigate and strive for bilateral and multilateral solutions between the countries for establishment of such systems in relation to passenger ships.	CPs informing in 2017 on the status/plans regarding the deposit refund systems, including on possible solutions regarding passenger ships.
Actions	addressing third parties	
RL12	Encourage, based on existing labels such as the EU Ecolabel and the Nordic Ecolabel, exchange with international environmental certification schemes for information and inclusion of the management and prevention of marine litter in their lists of criteria.	By 2016 initiate an activity on what certification schemes could be addressed, which existing criteria could be promoted for potential inclusion in international certification systems together with ways and means how to help approving these

CODE	REGIONAL ACTION	FURTHER SPECIFICATION
RL13	HELCOM Contracting Parties to seek cooperation with the River and River Basin Commissions, as appropriate, in order to include impacts of litter on the marine environment from riverine inputs, taking into account activities in the context of the implementation of the Water Framework Directive (WFD) and the Bathing Water Directive, and beyond, when applicable. This cooperation should include the exchange of experience on best practice to prevent litter entering into water systems, in line with action RL8.  Remediation and removal actions	HELCOM Contracting Parties will continue cooperation with River and River basin Commissions, as appropriate, in order to integrate measures addressing the reduction of littering in river basins followed up by appropriate information exchange on the implementation of measures.
RL14	Address landfills or dumpsites including historic ones which may eventually pose a risk to the marine environment due to factors such as coastal erosion and vicinity to rivers.	By 2020 a regional-wide map on landfills and dumpsites including historic ones which may eventually pose a risk to the marine environment is produced.
RL15	Establish an exchange platform for spreading experiences on good cleaning practices in beaches, including cleaning beaches actions by local communities, riverbanks, pelagic and surface sea areas, ports, marinas and inland waterways, in cooperation with relevant fora. Develop best practice on environmental friendly technologies and methods for cleaning.	Coordinate with other RSCs in order to set up an exchange platform for spreading experiences on good cleaning practices in the different marine compartments and rivers.
REGION	AL ACTIONS ADDRESSING SEA-BASED SOURCES OF MARINE	LITTER
Actions	addressing shipping related waste	
RS1	Development of best practice on the disposal of old pleasure boats (i.e. intentional disposal of the boats at the ending of their lifetime in the sea and on shore).	Best practice developed by 2018
RS2	Develop best practice in relation to inspections for MARPOL Annex V, including harmonized management of data. Support regional coordination of IMO regulations in accordance with EU requirements for those HELCOM countries which are EU members.	Best practice developed in cooperation with Paris MoU by 2017
RS3	Further work on implementation and harmonization of the no-special-fee system in ports of the Baltic Sea countries, addressing: gaps in existing regulations, enforcement and practices concerning shipping, port reception facilities auditing to assess adequacy of garbage collection, fair waste burden sharing between ports.	Evaluate the implementation of HELCOM Recommendation (28E-10), starting 2016
	Actions addressing waste delivery in ports/marinas	
RS4	Implementation of the ISO standard (ISO 201070:2013) in relation to port reception facilities. Differentiate according to the size of the port. Promote the development of regional statistics on waste collected in ports based on existing information as far as possible.	Assess how many ports are operating according to ISO standards and to propose action as appropriate by 2017.
	Actions addressing waste related to fishing and aquaculture	
RS5	Promote and disseminate best practice in relation to all relevant aspects of waste management within the fishing sector (including e.g. waste management on board, waste management at harbors and operational losses/net cuttings).	By 2018, based on the OSPAR outcome, select best practices to be disseminated in the Baltic Sea.
RS6	Through a multinational project, such as the MARELITT Baltic project, together with the fishing industry and other stakeholders, develop and promote best practice in relation to ALDFG and derelict fishing gear and their removal.	Best Practice developed by 2017, the issues is promoted within HELCOM-EUSBSR cooperation
RS7	Compile information and elaborate guidelines on best practices to reduce the input of ALDFG from commercial and recreational fishing to the Baltic Sea taking into account geographical particularities; utilize UNEP RSC report and FAO on ALDFG as a starting point and focus on regional specifics	Guidelines developed by 2017 taking into account geographical particularities.

CODE	REGIONAL ACTION	FURTHER SPECIFICATION
RS8	Identify the options to address key waste items from the fishing and aquaculture industry, which could contribute to marine litter, including deposit schemes and extended producer responsibility.	Late 2016 assess the use of OSPAR document and in consultation with the Baltic Sea Advisory Council consider and agree on the way forward to address key waste items from the fishing and aquaculture industries.
RS9	Investigate the use and prevalence of dolly ropes (bunches of polyethylene threads used to protect the cod end of demersal trawl nets from abrasions; synthetic fibre) in the areas of the Baltic Sea where they are used and consider the need to act.	Consider the outcome of the study on the impact of dolly ropes currently under development by the Netherlands. Baltic Sea Advisory Council is to be invited to be involved in this activity.
	Remediation and removal measures	
RS10	Mapping of snagging sites or historic dumping grounds and a risk assessment for identifying where accumulation of ghost nets pose a threat to the environment and should be removed.	As part of the assessment to be developed by HELCOM SUBMERGED by 2016. Mapping by 2017. Risk assessment by 2018.
RS11	Based on the risk assessment conducted in RS10 and identification of accumulation areas, initiate removal of ghost nets and their safe management on land.	The aim is to increase the removal and disposal of the nets, and that statistics are available to confirm the increasing trend.
RS12	Enter into the partnership with international and regional organizations (e.g. KIMO, NABU, OSPAR Commission) as well as port authorities, to encourage implementation of passive Fishing for Litter schemes, to collect litter caught in fishing nets during normal fishing activities.	Increasing trends in the number of vessels from the fishing sector involved in the schemes.
REGION	AL ACTIONS ADDRESSING EDUCATION AND OUTREACH ON I	MARINE LITTER
	General improved waste prevention and management	
RE1	To prepare information sheets to assist Contracting Parties in developing material for education programs, especially for professional seafarers including fishermen, highlighting the marine litter problem and including codes of practice in cooperation with relevant organisations including IMO.	Information sheets to be prepared by 2016
RE2	HELCOM website to be updated periodically based on the input from Contracting Parties on marine litter management activities.	2015 initial information uploaded (simplified BSAP follow up system)
RE3	Develop a communication strategy for this Regional Action Plan linked in a coherent way with national initiatives/actions. This will include linking the HELCOM website to relevant projects and initiatives.	2016

TABLE 39 VOLUNTARY NATIONAL ACTIONS (HELCOM 2015)

VOLU	VOLUNTARY NATIONAL ACTIONS ADDRESSING LAND-BASED SOURCES OF MARINE LITTER		
	General improved waste prevention and management		
NL1	National and local waste prevention and waste management plans:		
	* to include a reference to marine litter		
	* to include an element highlighting the impacts of marine litter		
	* to consider the cleaning and cleansing provision/infrastructure in municipalities by the coast or		
	rivers and to make the necessary improvements to prevent sources and pathways of litter from land		
	entering the aquatic environment.		
NL2	Promote Extended Producer Responsibility Strategies requiring producers, manufacturers, brand		
	owners and first importers to be responsible for the entire life-cycle of the product with measures		
	prioritizing the hierarchy of waste management in order to encourage companies to design products		
	with long durability for reuse, recycling and materials reduction in weight and toxicity. Focus to be		
	made on items frequently found in the marine environment.		
NL3	Improvement of stormwater management in order to prevent litter, including microlitter, from heavy		
	weather events and to enter the marine environment.		

Encourage voluntary reporting of companies on their products formulas (i.e. that they do not contain micro particles) towards HELCOM Contracting Parties. Bring in certification schemes, such as Blue Angel, EU Ecolabel, Nordic Ecolabel, etc. Promote a no-littering policy in national parks and protected areas, i.e. visitors should carry out everything they carry in.  Establish an overview of the importance of the different sources of primary and secondary microplastics. Evaluate products and processes that include both primary and secondary micro plastics, assess if they are covered or not by legislation, and act, if appropriate, to reduce the potential impact on the marine environment and to influence the legal framework. This must include the engagement with all appropriate sectors such as manufacturers and retailers.  With regard to the use of primary microplastics in personal care products formulations the possible impact on the marine environment should be reduced by applying substitutes. For other areas of applications appropriate solutions need to be defined.  Sewage related litter including sanitary waste  Clarify and, if needed, carry out research on the importance of sewage related waste in the upstream waste flows (i.e. sewage treatments applied, efficiency of the treatments, existence of untreated sewage, storm water influence, psychology behind people's behavior related to flushing the toilet, identification of missing elements).  Plastic bags  Support local pilot projects phasing out, replacing, and reducing single-use plastic bags. Strive for voluntary agreements with retailers and supermarkets to set an objective of reduction of plastic bags consumption.  Bottles and containers  Establish deposit refund systems for bottles, containers and cans (glass, plastics and aluminum), including the establishment of such systems on passenger ships and related harbors. Encourage refill systems and recycling, e.g. bulk and refill/reusable container for dry food and cleaning products, when applicable.  Cigarett
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Ensure the full implementation of TELECOM Convention Article 8 (Annex 17), especially Regulation 6,
in line with related international agreements such as MARPOL V and related EU legislation
(59/2000/EG) with regard to discharge of wastes to port reception facilities, and Article 9 on adequate
reception facilities for pleasure crafts.
Actions addressing shipping related waste including waste delivery in ports/marinas
Improve and follow-up enforcement of MARPOL Annex V.
Ensure and gather information on the implementation in ports of HELCOM Recommendation
28E/10: Application of the no-special-fee system to ship-generated wastes and marine litter caught in
fishing nets in the Baltic Sea area.
Promotion of garbage collection for pleasure crafts by marinas (i.a. Blue Flag Marinas requirements
related to the availability of pump-out stations and sustainable waste management).
Actions addressing waste related to fishing and aquaculture
Improve enforcement of EU Regulation 404/2011 on gear marking.
Improve enforcement of EU Regulation 1224/2009 on reporting lost gear.
1
Enhance resource efficiency by facilitating markets and applications for plastic waste from the fishing,
aquaculture and shipping industry (e.g. by bringing together producers of waste and recycling compa-
nies) by looking at specific items and differences in materials, including giving value to waste streams
by financial incentives.
Remediation and removal actions  Based on the risk assessment and identification of accumulation areas initiate removal of ghost nets
and their safe management on land.  Promote removal of lost fishing gear
Encourage fishing vessels to be involved in passive Fishing for Litter schemes, where they are availa-
ble.

NE1	Promote and undertake education activities on marine litter in synergy with existing initiatives in the field of sustainable development and in partnership with civil society (including activities related to prevention and promotion of sustainable consumption and production).	
NE2	Identify and promote curricula for marine related education, including both professional seafarer the recreational sector (e.g. diving and sailing schools), which develop awareness, understanding	
	respect for the marine environment and secure commitment to responsible behavior at personal, local, national and global level.	
NE3	Encourage participation in International, EU and National Marine Litter Cleanup Campaigns.	
NE4	Promote the "Adopt a beach" system.	
NE5	Raising public awareness, including for children and youths and consumer campaigns, on the occurrence, and prevention of marine litter (e.g. to use ashtrays in public areas inland and along the coast), including micro particles, taking into account existing materials (e.g. Marlisco Project) and accompanied by image campaigns addressing threats/impact to marine life from various harmful litter items, such as cigarette filters.	
NE6	Enhance cooperation and coordination with global marine initiatives such as:  - The UNEP's Global Programme of Action for the Protection of the Marine Environment from Landbased Activities (GPA-Marine);  - Regional Seas Action Plans;  - The Global Partnership on Waste Management (GPWM); and The Honolulu Commitment and the Honolulu Strategy on marine debris.	

### **Appendix 3** Recommendations from GESAMP for further research

GESAMP (2015) provides the following recommendations for further research:

#### Effects of microplastics on marine biota:

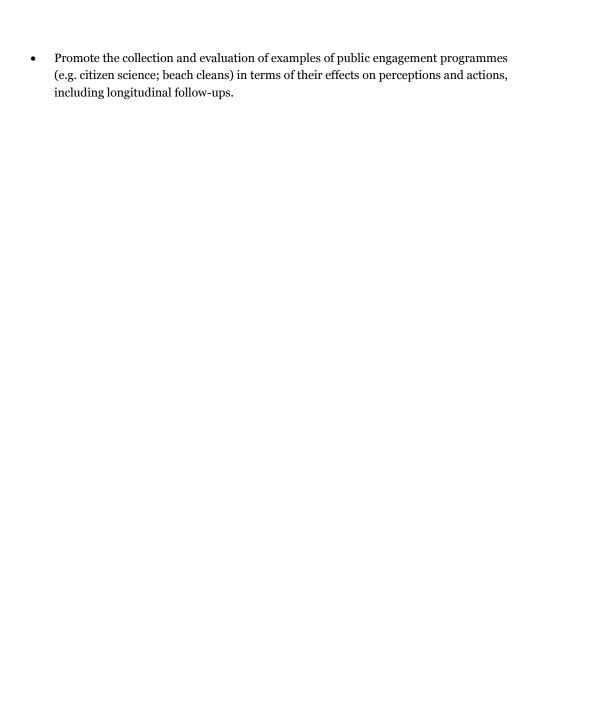
- Examine the extent to which nano-sized plastic particles may cross cell membranes and cause cell damage, under natural conditions, including knowledge and expertise from the medical and pharmaceutical industry (drug delivery).
- Examine the extent to which additive chemicals may cross the gut wall and assess the risk of harm at an individual and population level.
- Examine the extent to which adsorbed organic contaminants may cross the gut wall and assess the risk of harm at an individual and population level.
- Assess the potential health risk of microplastics for humans, including dietary exposure from a range of foods across the total diet in order to assess the contributing risk of contaminated marine food items.
- Examine the potential of microplastics to translocate non-indigenous species, including pathogenic organisms, relative to other transport vectors.
- Examine the potential for accumulations of plastics and microplastics to form additional floating ecosystems.
- Examine species-specific gut conditions that may influence chemical availability and transfer.
- Consider using stable or radioactive labelled compounds (polymers, additive chemicals and absorbed contaminants) to establish the degree of transfer under different conditions.

#### Sources and fate of microplastics in the marine environment:

- Generate data on weathering-induced fragmentation of at least the PE, PP and EPS
  plastics in the marine environment.
- Examine the influence of weathering on particle sorption characteristics.
- Establish improved and validated methods for sampling at the sea surface in sediments and in biota.
- Organize inter-calibration exercises and harmonize reporting units to make future data comparable around the world.
- Design sampling strategies to establish time trends and spatial trends in selected marine areas
- Conduct additional sampling of sub-tidal and in particular deep sea sediment.
- Investigate nano-sized plastic particles in marine organisms as a critical input for future risk assessments.
- Develop more realistic transport models, to incorporate variable particle properties, 3D circulation and

#### Social aspects of microplastics in the marine environment:

- To conduct empirical social research on microplastics to address: a) individuals'
  knowledge and understanding; b) perceived risks; and, c) the associated consequences
  on humans. Social perceptions are linked to behaviour and support of measures addressing the issue.
- To improve the geographical representativeness of this work outside North and South America and Europe – to identify needs and tailor information to account for social, economic and other cultural differences, and promote effective mitigation strategies.
- To analyse the economic impacts of microplastics, in terms of cost-benefit to forecast future effects in response to any changes in microplastic use/input.



## Appendix 4 Further initiatives suggested by the advisory groups and by microplastics seminar, August 2014

#### [In Danish]

Følgende kommentarer og konkrete forslag til nye tiltag fremkom på det afholdte seminar d. 18. august 2015 eller er efterfølgende fremkommet fra følgegruppen.

#### Plastic Change (uddrag af præsentation på seminaret):

#### Videns tiltag:

- Adfærdsanalyser.
- Mere viden om bildæk og tekstiler.
- Fjordmodel, hvor man undersøger betydningen af mikroplast i de forskellige compartments, og derved få information om stofstrømme og hvad der sker i miljøet.
- Vesterhavet; potentiel indsamling af marint affald ved hårdtvejrshændelser.
- Renseanlæg; undersøgelse af polymersammensætning og skæbne

#### Politiske tiltage:

- Pluk frugterne og forbyd mikroplast som er bevidst tilsat produkter
- Forbud/afgifter på de mest oplagte forureningsemner
- National handlingsplan om plastforurening: oplysning, kampagner, forskning og opgraderet EU indsats
- DK bør arbejde for at polymerer omfattes af REACH og anden EU regulering.

**Plastindustrien** pointerede, at det er væsentligt at skelne mellem primær og sekundær mikroplast, da det er to forskellige fronter man kan/skal sætte ind på. Makroplast er f.eks. et affaldsproblem, hvor man kan tage fat i et på EU niveau og f.eks. være med i tiltagene fra OSPAR og HELCOM. Plastindustrien spurgte desuden, hvorvidt det ville være relevant at kigge på spildevand fra hospitaler og professionelle vaskerier, hvor man bruger store mængder af syntetiske materialer til rengøring m.v. og spørger om de ikke har egne rensningsanlæg mv? Ifølge kommentar fra BIOFOS ryger spildevand fra hospitaler og renserier generelt direkte i det kommunale renseanlæg.

**BIOFOS** spurgte hvorfor der i rapporten foreslås test på ferskvandsorganismer, og nævner, at man ikke på nuværende tidspunkt ved meget om effekter og hvorfor man antager at organismer behandler mikroplast anderledes end andre stoffer/materialer, men som man som regel ikke antager er farlige (træ, sand mv.). IVL svarer at det bl.a. skyldes den langsomme nedbrydning af plast, samt det faktum at der kommer mere og mere plast i havene. DTU nævner at fokus er på ferskvandsorganismer, da disse er standard test organismer bl.a. i OECD test regi, og at der mangler standardiserede test af mikroplast til at kunne sige noget om hvor farligt det er.

**DANVA** nævnte, at organisationen har fået udarbejdet en rapport der omhandler forekomst, skæbne og effekt af mikroplast i renseanlæg<sup>45</sup>. Rapportens anbefaling for yderligere tiltag var et måleprogram, i samme tråd som det der er lavet i Sverige. Her skal man se på hvad er det for noget mikroplast, der ledes ind og ud af renseanlæg. Rapporten anbefaler derudover, at man udarbejder nogle standardiserede monitoreringsmetoder der gør det muligt at sammenligne tal fra de forskelli-

http://www.danva.dk/Admin/Public/DWSDownload.aspx?File=%2fFiles%2fFiler%2fUdgivelser%2f2o15%2fMikroplast renseanl%c3%a6g 2015 rapport DHI DANVA.pdf

<sup>45</sup> 

ge studier. **Teknologisk Institut** pointerede i den forbindelse, at det er vigtigt at man ser på, hvordan man tager prøverne i renseanlæggene og efterlyser bedre analysemetoder.

**Krüger A/S** foreslog, at man laver en anbefaling af, hvor man skal sætte ind i forhold til de største kilder.

**Miljøstyrelsen** efterlyste en prioritering af forslagene til yderligere initiativer og undersøgelser, der er udarbejdet i rapporten, således at man kigger på, hvilke forslag der er de vigtigste i relation til hurtigst at komme fremad med handlingerne.

Efter seminaret har **Dansk Erhverv** foreslået, at der udarbejdes en national handlingsplan med udpegede relevante interessenter – industri, organisationer, institutioner, NGO, myndigheder. Udarbejdelsen af handlingsplanen skal være en platform for diskussioner og igangsættelse af frivillige initiativer - noget der kan skabe mere opmærksomhed omkring problemet. En DK platform, men på sin vis også en plastform for handlinger på Nordisk plan og via Nordisk Ministerråd. DK platform kunne være noget som igangsættes allerede i år og som ministeren så med danske erfaringer kan folde ud på nordisk plan i regi af Nordisk Ministerråd.

**Plastindustrien** har efter seminaret udtrykt at organisationen hellere end opstilling af standardvilkår, tager dialogen med MST/kommunerne om at være ekstra opmærksom på at se efter kilder til spild og at virksomhederne bakker det op med Operation Clean Sweep.

# Microplastics - Occurrence, effects and sources of releases to the environment in Denmark

This report contains a review of existing knowledge on issues related to contamination by micro-plastic with a focus on the use and release of micro-plastics in Denmark and the occurrence of micro-plastics in the surrounding waters.

Micro-plastic defined as small plastic pieces of 1µm to 5 mm.

Micro Plastics are detected in organisms at all levels of the marine food chain as well as in water and sediment.

There are potentially three types of adverse effects of the micro-plastic: (1) physical effects related to the intake, (2) toxic response by the release of hazardous substances in the plastic and, (3) toxic reaction to the pollutants which are adsorbed to micro plastic. These effects are shown in laboratory experiments but not proven to occur in the environment.

Although the majority of the micro plastics in the waste water end up in the sewage sludge, wastewater treatment plants are important potential sources of emission of the micro-plastics in the ocean.

The most important sources of release of micro-plastics to the environment are tires, paints, road markings, textiles, etc. The report estimates that only minor significance to micro plastic used directly in certain products (for example, in cosmetics or for use in blowing agents).

