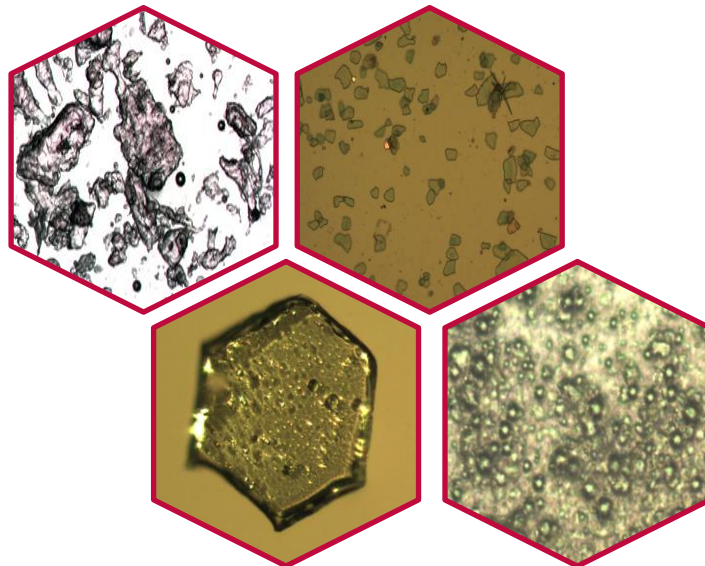


Review of Microplastics in Cosmetics

Scientific background on a potential source of plastic particulate marine litter to support decision-making

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Plastic particulate ingredients detected in cosmetic and personal care products (shower gel, lipstick, bubble bath, toothpaste). Photos © IVM

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Abbreviations

EPA	Environmental Protection Agency of the USA
EU	European Union
FDA	Food and Drug Administration of the USA
INCI	International Nomenclature of Cosmetic Ingredients
IVM	Institute for Environmental Studies, VU University
Ministry I&M	Ministry of Infrastructure and Environment of the Netherlands
mm	Millimeter
MSFD	Marine Strategy Framework Directive of the EU
MW	Molecular weight
NGO	Non-governmental organisation
nm	Nanometer (one billionth of a meter)
PCB	Polychlorinated biphenyl
PCCP	Personal care and cosmetic product
PEG	Polyethylene glycol
PET	Poly(ethylene terephthalate)
PMMA	Poly(methyl methacrylate)
PNP	Polymeric nanoparticles
PS	Polystyrene
POP	Persistent organic pollutant
UN	United Nations
UNEP	United Nations Environment Programme
µm	Micrometer (one millionth of a meter)

Executive Summary

What are microplastics?

‘Microplastics’ are particulates made of plastic material less than 5 mm in size that make up the microsized fraction of plastic marine litter¹. Microplastics represent an emerging topic in pollution research and policy making. Many organisations are currently seeking to identify the various potential sources of microplastic pollution in order to guide decision-making on mitigative actions.

Plastic ingredients are applied in wide variety of cosmetic and personal care products

The cosmetic and personal care industry applies plastic ingredients in a wide variety of products. The plastic materials in question are synthetic, nondegradable², water insoluble, solid materials made up of polymers³ mixed with additives to give the materials the desired properties and functionality. The particles of plastic used in cosmetics are very small (usually not larger than about a millimeter in size, but may be as small as a few tens of nanometers): many are invisible to the naked eye. The particles may be spherically shaped or amorphous. Plastic types applied in cosmetics include thermoplastics⁴ and thermoset plastics⁵ including silicones⁶. Within these plastic categories, many different polymer and copolymer⁷ types are applied in cosmetic formulations. The functions of these materials in the products include film formation, viscosity regulation, skin conditioning, emulsion stabilizing and many others. Plastic ingredients fulfill these functions in a wide range of cosmetic and personal care product types, such as (but not limited to) soap, shampoo, deodorant, toothpaste, wrinkle creams, moisturizers, shaving cream, sunscreen, facial masks, makeup (e.g. lipstick or eye shadow), and children’s bubble bath. It should be noted that many synthetic polymers in cosmetic formulations are not thermoplastics or thermoset plastics and do not fulfill the criteria for microplastic (e.g. polymers that are liquids at normal environmental temperature ranges; water soluble polymeric substances⁸) and that we limit the discussion here to the solid, plastic particles that would be considered to be marine litter if they were to reach the marine environment.

Plastic *en route* from bathroom drain to ocean

Many ‘rinse off’ products that contain plastic obviously end up in household wastewater streams. Research has shown that a portion of the microplastics (from multiple sources) in wastewater streams are retained in sewage sludge and the rest is emitted to surface waters via treated

¹ While 5 mm is the most common upper size limit used among marine litter researchers, some prefer a definition of <1 mm for ‘microplastic’. If one should choose to apply the latter definition, any cosmetic formulation containing particles greater than 1 mm in size would be said to contain ‘meso- or macroplastic’, which are also indicators for marine litter under the Marine Strategy Framework Directive.

² Nondegradable refers here to the property of materials that does not allow them to mineralize under natural conditions over long time periods.

³ A polymer is a large molecule consisting of repeating subunits called monomers.

⁴ The high molecular weight polymers in thermoplastics can be melted at high temperatures and remolded.

⁵ The polymers in thermoset plastics are irreversibly cured upon heating, as covalent chemical bonds are formed.

⁶ Refers to polymerized siloxanes or polysiloxanes, inorganic-organic polymers with a silicon-oxygen backbone.

⁷ A copolymer is a polymer consisting of more than one type of monomer in the polymer chain.

⁸ All plastics consist of polymers but not all polymers are plastic.

wastewater effluents. Some of these effluents are discharged directly to the sea. Other effluents enter river systems, which are known to transport suspended particulate matter (e.g. sediment particles, organic matter, microplastics) with the current to the sea. Depending on hydrodynamics and other factors, a fraction of the particles may also remain in the riverine environment for extended time periods. Microplastics in treated wastewater effluents are thought to originate not only from cosmetic and personal care products, but also many other plastic emissions to wastewater such as fragments of larger macroplastic objects, synthetic polymer textile fibers, and any other ‘down-the-drain’ product containing plastic particulates. It is sometimes possible to determine the polymer type of microplastics detected in effluent samples, but it is not possible to trace the exact origin or source of the particle, since the same polymers are used in very diverse product applications. For example, polyethylene is widely used as an ingredient in cosmetics but it is also the world’s most produced plastic type. Fragments of polyethylene could potentially originate from cosmetics and millions of other types of products.

Environmental fate and toxicity of microplastics from cosmetics

Microplastics in cosmetics long outlast the consumers who use them. Once released into the environment, microplastic materials are expected to persist for centuries before becoming fully decomposed and reentering normal biogeochemical cycles. Due to extremely slow decomposition kinetics of the macromolecules in plastic, it has been postulated that all plastic that has ever been released into the environment still exists today. This means that any adverse effects that microplastics discharged to the environment might have will likely persist for the decades and centuries to come.

It is known from the scientific literature that when exposure to sufficiently high doses occurs, microplastics are able to cause particle toxicity⁹ in diverse biological systems, from primary producers to marine invertebrates to mammalian systems. In the toxicological literature such as the Cosmetic Ingredient Reviews, the effects of plastic ingredients are assessed based on available toxicological data on plastic particulates. Not all studies used have tested plastic particles from the same batches that are applied in the cosmetic products, but if the polymers are the same, the particle toxicity data is considered relevant by these reviewers. The toxicological studies in the literature often examined the fate and effects of plastic particulates in mammalian systems, such as the toxicological implications of fragmentation of plastic implants (replacement hips and heart valves made of plastic). Plastic particle uptake is known to occur in the gastrointestinal tract; typically nano-sized particles (<1 µm) are more mobile than strictly micro-sized particles (1-999 micrometers). Inhalation route studies have linked exposure to fine particulates with allergic reactions, asthma, cancer and heart disease.

In the marine environment, the body of exotoxicological data is growing and indicates that microplastics have a negative impact on the health of marine organisms, affecting the energy availability needed for important life processes. In the blue mussel for example, filtration behavior as well as physiological parameters are affected when certain doses of plastic particles are taken up into tissues. As in mammalian systems, the immunological system can be impacted in the marine animal species studied to date. Continued systematic collection of these types of (eco)toxicological data is warranted given the early warning signals that have been emerging in the literature. The ecotoxicological effect concentrations should be determined and compared to typical exposure scenarios measured in the field to determine the environmental risks. It will take time for the scientific community to build up the body of hazard and environmental exposure data for a full risk assessment of microplastics of the types applied in cosmetics and personal care products formulations.

⁹ Exposure to particles can lead to toxicity (e.g. oxidative stress and inflammatory responses) in various parts of the body that fine particulates can translocate to including bloodstream, brain and other organs.

End-of-pipe vs. cleaner production emission reduction strategy

This report presents information that brings into question the effectiveness of an end-of-pipe¹⁰ type of solution for halting microplastics emissions via wastewater streams. This is because treatment facilities are not designed to retain plastic particulates, and applying further microfiltration is expected to be costly both in terms of energy inputs and financial investments. Furthermore, during high flow periods, wastewater is discharged to the surface water untreated. A significant percentage of households across Europe are not connected to wastewater treatment facilities, meaning microplastics are discharged directly to surface water in many communities. In developing countries over 90% of untreated wastewater is discharged to surface waters, according to UN reports. In Europe, over a third of the total sewage sludge generated is currently being applied agricultural fields as fertilizer ‘biosolids’¹¹; microplastics are expected to enter waterways via runoff from such fields. These facts point to cleaner production¹² as a more universally effective route to achieve a reduction in microplastics emissions from cosmetic and personal care products. The costs and benefits of end-of-pipe options versus cleaner production options should be carefully scrutinized.

The cosmetics industry world-wide is largely a self-regulated industry when it comes to product formulation decisions. For example in the USA, cosmetic product ingredients do not need FDA premarket approval, with the exception of pigments and ingredients deemed to have medicinal activities that could be regarded as drugs (e.g. antibacterial ingredients). The EU Cosmetics Directive is the strictest existing regulation of cosmetic and personal care ingredients in the world, now also addressing nanomaterials as ingredients. The substances in products are regulated in the Cosmetics Directive according to toxicity upon normal use of the product, assuming dermal exposure only. The environmental impact of emissions through normal product use are not compulsory for producers to take into account in the scientific and technical assessments of ingredients, although more and more companies are striving to pay attention to the environmental friendliness of their ingredient lists.

Outlook

The subject of microplastics in cosmetics has drawn world-wide attention due to the increasing concern about marine litter and the microplastic fraction of the litter that is entering food chains and causing adverse effects. These products are one of many sources being scrutinized by the international community of concerned citizens, policy makers, scientists and companies which recognize and take producer responsibility in this area. While packaging and other macro-sized plastic objects have the potential to be recycled, plastic cosmetic ingredients are impossible to recycle because the products are discharged into wastewater at end-of-life. Considering the vast size of the global cosmetic and personal care product market and the vast volumes of wastewater we humans annually produce, if we load these products with plastic particulates, we are creating an automatic source of microplastic environmental pollution. This source, as well as other sources from other sectors are being addressed through various governance activities.

¹⁰ End-of-pipe solutions curb pollution emissions by add-on measures, e.g. filters.

¹¹ Biosolids refers to sewage sludge used as nutrient-rich organic fertilizer for plant crops.

¹² Cleaner production limits pollution at the source through design of cleaner products and/or production methods. Investments in cleaner production lead to innovations that companies benefit from economically, often making it a favourable approach (Frondel et al. 2004).

1 Introduction

Over the past couple of years there has been increased attention to the issue of plastic ingredients in personal care and cosmetic products (abbreviated in this report as PCCPs) as a potential source of plastic pollution in the sea. In the Netherlands it is known as the *plastic soup*, or sometimes the plastic *bouillon*, referring to the tiny plastic particle fraction of the plastic litter¹³. NGOs, PCCP companies, scientists, the EU, UNEP, Rio+20 and many other conferences, policy makers as well as the media¹⁴ have given the subject broad coverage. The EU Marine Strategy Framework Directive (MSFD) specifically refers to microplastics under ‘Descriptor 10’ Marine Litter. Most EU Member States have already submitted initial assessments and are now currently in the planning stages of emission mitigation actions and monitoring programmes¹⁵.

The pollution of the world’s oceans with plastic and the international commitments made to take mitigating action has helped focus attention on the various sources and routes through which nondegradable plastic materials can be emitted to the marine environment. Plastic ingredients in PCCPs have been described as one of the many sources (Zitko and Hanlon 1991; Gregory 1996; Derraik 2002; Thompson et al. 2004; Fendall and Sewall 2009; Arthur et al. 2009; Leslie et al. 2012a,b). The concern is that plastic ingredients in products that are being used by consumers in households worldwide are contributing to the total abundance of particulate plastic litter – or ‘microplastics’ as they are generally called – in the ocean today. Normal use of PCCPs introduces these solid, plastic materials directly into wastewater streams since the products are for the most part washed or rinsed down the drain during or after use. Remediation of widespread microplastic contamination in the marine environment is futile due to the complications of continuously removing diffusely distributed, persistent pollution from vast affected areas; emission prevention is the key mitigation strategy (STAP 2011).

It is impossible to collect plastic ingredients in PCCPs (and any other ‘down-the-drain’ products) at end-of-life for recycling, setting them apart from many other items in the plastic litter fraction of marine litter. Plastic packaging and other large plastic items have the potential to be collected for recycling (or may otherwise be contained in landfills or feed waste-to-energy incinerators).

With the interest in microplastics comes a need to understand what the characteristics of the plastic ingredients in PCCPs are, what their function is, and what the environmental effects are. Environmental effects of microplastics in general are being studied using microplastic particles in the laboratory, such as in the EU FP7 CleanSea Project, a 17-partner consortium led by IVM (www.cleansea-project.eu). On the specific topic of ‘microplastics in PCCPs’ however, there is no overall review in the scientific literature (both gray literature and peer reviewed) and the relevant information that can be found in scientific literature is fragmented and spread over diverse journals that are not accessible to a broad readership.

The discussion of mitigating microplastics in cosmetics requires clear definitions of which materials are under discussion and which are not: stepping away from collective terms like ‘microplastic’ and moving towards unambiguous substance identification of plastic particles that are solid, water insoluble, synthetic, nondegradable, polymeric materials that are important to the marine litter issue and the MSFD. The information should be available and broadly disseminated to all interested stakeholders. A common understanding of the PCCP ingredients in question is critical to finding practical solutions and establishing policies that mitigate microplastic pollution emissions. This review makes clear that microplastics as viewed in the scientific community

¹³ Volkskrant (Dutch daily newspaper) article ‘Plasticbouillon à la Noordzee’, 30.06.2012

¹⁴ For example, <http://edition.cnn.com/2013/01/07/health/microplastics-soap-unilever> and www.theguardian.com/environment/2012/dec/09/microbeads-microplastics-cosmetics-ocean-pollution

¹⁵ Reporting Obligations Database for Deliveries for MSFD reporting on Initial Assessments <http://rod.eionet.europa.eu/obligations/608/deliveries>

refers to a vast amount of different polymer materials. If regulators opt to regulate any of the plastic particles in cosmetics, unambiguous substance identification would be helpful¹⁶ as a first step.

1.1 Scope and objectives

This report aims to bring together existing information in the public domain to give a concise overview of key aspects regarding the microplastics that are applied in PCCPs and what the consequences of this practice are in terms of environmental pollution and marine litter.

Specifically, the following questions were posed by the Ministry I&M:

- i. What is understood to be a microplastic in cosmetics? What types of plastics are used in cosmetic formulations?
- ii. What is the function of microplastic in cosmetics?
- iii. Which plastic ingredients enter wastewater streams and thereby can potentially contribute to the solid-phase microplastic fraction of marine litter? What are the environmental effects of microplastics in cosmetics?

The report begins with defining what is scientifically understood to be ‘microplastics’ in the context of PCCPs and the marine plastic litter research field, which focuses on solid, plastic waste materials (including fine particulates) that are not water soluble (Chapter 2). In the same chapter, a description of the types of plastic ingredients used in such products is given. Chapter 3 presents the variety of functions which plastic fulfils in the product formulations, according to the scientific literature, regulatory bodies and publicly available cosmetic industry producer information. Examples of product types that sometimes contain plastic ingredients are given. The environmental fate and effects of microplastics are discussed in Chapter 4, drawing on data sources which are often not specific for the PCCP sector (with the exception of the effects data in the Cosmetic Ingredient Review publications), but which provide a brief overview of the current state of knowledge on fate and effects of the same plastic materials that could also be applied in PCCPs. What is known about the emission routes of microplastic ingredients in cosmetics is described, the widespread environmental distribution of microplastics in general is highlighted, and existing hazard or toxicity data regarding plastic particulates is briefly reviewed. Concluding remarks in the final chapter summarize and synthesize the findings.

¹⁶ Unambiguous substance identification is generally a first step in the processes under REACH, the regulatory framework for chemicals in Europe. See <http://echa.europa.eu/regulations/reach>

2 Plastic ingredients in personal care and cosmetic products

In this chapter we provide an explanation of the materials that are applied in cosmetic and personal care product formulation that fall under the definition of ‘microplastics’ as understood for the environmental pollutants described under Descriptor 10, marine litter, of the MSFD. The complexity of the polymers¹⁷ currently in use in PCCPs will become apparent.

2.1 Defining microplastics

In order to properly discuss how to deal with the possible source of microplastics in PCCPs, it is important to define which synthetic polymer ingredients in PCCPs can be regarded as a ‘microplastic’ as it is understood by the international marine litter scientific community (Thompson et al. 2004; Arthur et al. 2009 etc.). Plastic particles are described as ‘primary’ when they are originally manufactured as particulates and ‘secondary’ when they are fragments of larger plastic items. Plastic PCCP ingredients fall into the category ‘primary’ since they are manufactured and applied as plastic particulates. The plastic PCCPs ingredients of interest to the marine litter issue have the following properties in common with other microplastic litter:

- Solid phase materials (i.e. solid particulates, not liquids)
- Insoluble in water
- Synthetic
- Nondegradable (e.g. according to standardized tests)
- Made from plastic
- Small size (up to 5 mm, although they can be even smaller than 1 μm , i.e. nano-sized)

It should be noted that many synthetic polymers in cosmetic formulations do not fulfill the criteria for microplastic (e.g. polymers that are liquids at normal environmental temperature ranges; water soluble polymeric substances) and that we limit the discussion here to the solid particles that would be considered to be marine litter if they were to reach the marine environment.

What’s in a name

‘Microplastic’ is a common term in the environmental science literature referring to a wide range of plastic particulate sizes between the low nm range and a particle size of 5 mm. Therefore the term covers particulates in the milli range (1-5 mm), micro range (1-999 μm) and in the nano-range (1-999 nm). Nanotechnology applies engineered particles in the 10-100 nm range. Scientists studying plastic particulates in the environment refer to particles $<1 \mu\text{m}$ as ‘nanoplastics’ because it is in the nanometer range (not necessarily in the engineered nanoparticle range). In the PCCP industry, the word ‘microbead’ refers to solid particulates that are applied to products for a variety of functions. Other general terms for such particulates include: microspheres, nanospheres, microcapsules, nanocapsules, as well as several registered trademark and other product names. The particulates in PCCPs are sometimes made of non-plastic materials (such as lipid, cellulose, granulated almond shell) but of interest here are only those made of synthetic plastic particulates according to the properties list above. The shapes of the plastic particulates that are marketed as ‘microbeads’ can be spherical but also amorphous. In this sense they look similar to many of the microplastics detected in the oceans: beads and irregularly shaped fragments.

¹⁷ A polymer is a large molecule consisting of repeating subunits called monomers.

2.2 Plastic ingredient types in personal care and cosmetic products

Plastic ingredient types

Plastics consist of synthetic polymer materials that are mixed with a variety of chemicals ('additives') in order to achieve an end product plastic material with properties that are appropriate for the function. Plastic materials have been applied as ingredients in PCCPs for several decades with early patents dating from the 1960's¹⁸ – today they remain a focus of innovation in new PCCPs (e.g. Lochhead 2007; Patil and Ferritto 2013; Patil and Sandewicz 2013). The plastic materials applied as ingredients in PCCP formulations discussed here include the two main categories of plastics typically made from petroleum carbon sources: thermoplastics e.g. polyethylene, polypropylene, polystyrene, polytetrafluoroethylene (Teflon), poly(methyl methacrylate), polyamide, and thermoset plastics, e.g. polyester, polyurethanes. These polymers are carbon-based¹⁹ and have i) carbon atom chains, such as polyethylene, polypropylene, polystyrene, ii) hetero-atom chains including both carbon and oxygen, e.g. poly(methyl methacrylate), polyester or poly(ethylene terephthalate), polycarbonate, or iii) heteroatom chains containing carbon and nitrogen, e.g. polyamide, polyurethane. Another type of plastic is the silicones, also relevant to the PCCP plastics discussion. These are polymerized siloxanes or polysiloxanes, inorganic-organic polymers with a silicon-oxygen backbone. Most of these types of polymers are water soluble or water-dispersible, and therefore are not solid materials as described under section 2.1. 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.

Macromolecules in plastic

All plastic materials are made up of mixtures of macromolecules of different chain lengths and thus different molecular weights (MW), known as a 'polydisperse' MW. The MWs of these solid-phase macromolecules are generally large, as shorter chains (as well as branching of the chains) leads to increasingly softer materials. Polyethylene molecules up to about 700 carbons in length are waxy solids, and alkane chains with less than 20 carbons are liquids or gases (Peacock 2000). Polymer waxes are macromolecules that result from shorter chain lengths, e.g. polyethylene wax (Cosmetic Ingredient Review 2007), a popular PCCP gellant available as powder, flakes or granules. Polyethylene waxes that are nondegradable²⁰, water insoluble, solid materials with melting points well above maximum sea temperatures and also fall under the definition of marine microplastic litter. Longer polymer chains produce more rigid materials, e.g. poly(ethylene terephthalate) glitters or styrene/acrylates copolymer²¹ colour spheres. Lengthening the chain of ethylene oxide polymers (better known as polyethylene glycols) to 20,000 results in

¹⁸ For example, US Patent 3196079 filed in 1959 for polyethylene as ingredient in bath powder, baby powder, rouge and various other cosmetic products.

¹⁹ The carbon source for plastic in production today is predominantly fossil-fuel based. Biomass can also be used as a carbon source to make identical plastic products with the same functions, benefits and end-of-life environmental impacts as a plastic product made from fossil-fuel feedstocks. Biomass carbon plastic is referred to 'biobased plastic' or 'bioplastic', although the name does not infer sustainability. Plastic made from biomass carbon is as equally nondegradable as plastic made from petroleum carbon. Europe has a limited capacity to produce ecologically sustainable biomass and meeting future demands for biomass will be challenging (see PBL 2012).

²⁰ The biodegradability of even relatively low molecular weight linear paraffin waxes (n-alkanes) is low and dependent on finding the right species organism under specific conditions (e.g. high temperatures). Chain branching further negatively impacts biodegradability. Most plastic needs to be pretreated in order to prime the material for biodegradation. See Pott et al. 1972, Albertsson et al. 1986 and Bonhomme 2003).

²¹ A copolymer is a polymer consisting of more than one type of monomer in the polymer chain, e.g. styrene and acrylate monomers.

solid materials, e.g. PEG-2M (Gruber 1999). 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).

Identifying solid polymeric materials by way of INCI names

To determine whether a PCCP ingredient is a liquid (not defined as litter) or a solid (potential litter) the International Nomenclature of Cosmetic Ingredients (INCI) name is sometimes insufficient, as the phase of materials with the same INCI name may be different. This is because the phase depends not only on the monomers that make up the polymer or copolymer, but also on properties like chain length, degree of crosslinking and MW. Sometimes the ratio of different monomers in copolymer materials determines the phase, e.g. the random copolymers of ethylene oxide and propylene oxide, INCI name PPG-N-Buteth-M, are water insoluble if they contain <50% ethylene oxide (Gruber 1999). To determine if a polymeric ingredient is a solid particle or not, additional chemical information on top of the INCI name may be required in such cases.

Homopolymers, copolymers and blends

The plastic PCCP ingredients include homopolymers but also many copolymers (see examples Table 2.1). Homopolymers are polymer chains of a single monomer type, such as is formed when ethylene terephthalate monomers are polymerized to poly(ethylene terephthalate) (PET).

Copolymers are made by polymerizing different monomers in the same chain, either in random order, alternating monomers, or as ‘block’ copolymers (i.e. monomers clustered into blocks in the polymer chain of the copolymer molecule). Copolymers are developed to enhance material properties in PCCP applications, such as resistance to degradation (Guerrica-Echevarría and Eguiazábal 2009). The homopolymer PET is increasingly being replaced by copolymers such as poly(butylene terephthalate)/PET and others (Cosmetic Ingredient Review 2012). Other common solid-phase synthetic plastic copolymer blends used in PCCPs are ethylene/propylene styrene copolymers, butylene/ethylene styrene copolymers, acrylates copolymer and many others (Cosmetic Ingredient Review 2002; 2012).

Blends are made by combining different polymer materials after the polymerization process. Copolymer design and blending enables formulators to combine desirable properties from individual (co)polymers in one material, without the expense and effort required for developing an entirely new polymer type. This results in dynamic growth in the number of plastic materials available for application in PCCP formulations (Gruber 1999).

Particulate sizes

Plastic particulates applied as PCCP ingredients can be large enough to see with the naked eye (e.g. 50 -1000 μm), fine particulates (low μm -range) or very fine particulates (<2.5 μm). The term ‘microbead’ generally refers to solid particles of various shapes, e.g. spherical, amorphous, between 1 and 1000 μm . ‘Microspheres’ are of similar particle sizes 1-1000 μm (Lipovetskaya 2010), however microspheres are by definition spherical and often are hollow, enabling them to be loaded with an active ingredient (Lidert 2005). The typical 1-50 μm microspheres on the PCCP market are desired for their ‘ball-bearing’ effect on the formulation, giving products an ‘extra silky texture and good skin adhesion’ (Patravale and Mandawgade 2008). Some plastic particulates marketed as ‘microspheres’ are available, according to commercial websites, as small as 10 nm in diameter.

‘Microsphere’ is also used interchangeably with the term ‘microcapsule’ (1 μm - 1 or 2 mm) (Ansaldi 2005; Kvitnitsky et al. 2005). Micro-sized ‘sponge’ technology makes use of fine particles between 1 and 50 μm that are used as innovative delivery systems for active ingredients

(Saxena and Nacht 2005); ‘sponge’ materials such as cross-linked poly(methyl methacrylate) (PMMA) are sometimes used because they can sorb active ingredients, especially the more hydrophobic ones (Lidert 2005). Polymeric particles in the size range from 10 to 1000 nm are termed ‘nanospheres’ and ‘nanocapsules’ or simply ‘polymeric nanoparticles’ (PNPs) (Guterres et al. 2007; Rao and Geckeler 2011; Hubbs et al. 2011). For cutaneous application cosmetic products, nanoplastics in the 200-300 nm size range may be observed (Guterres et al. 2007).

Table 2.1 Selected examples of plastic ingredients currently applied as particulates in personal care and cosmetics products (PCCPs).

Polymer name	Functions in PCCP 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
Poly(ethylene terephthalate)	Adhesive, film formation, hair fixative; viscosity controlling, aesthetic agent, (e.g. glitters in bubble bath, makeup)
Poly(methyl methacrylate)	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)

Note: some polymers may be available in various forms, as dispersions in solvents, or as partially water soluble polymer forms. International Nomenclature for Cosmetic Ingredient (INCI) names for polymers given. The functions given are examples and not an exhaustive list. Sources: 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.

3 Functions of microplastic in product formulations

Plastic ingredients are part of the formulation for a variety of PCCPs 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. Plastic ingredients are present in different products at different percentages, ranging from a fraction of a percent to more than 90% in some cases (Cosmetics Ingredient Review 2012).

Depending on the polymer type, composition, size, shape, the plastic ingredients have been included in formulations with a vast number of functions including: viscosity regulators, emulsifiers, film formers, opacifying agents, liquid absorbents binders, bulking agents, for an 'optical blurring' effect (e.g. of wrinkles), glitters, skin conditioning, exfoliants, abrasives, oral care such as tooth polishing, gellants in denture adhesives, for controlled time release of various active ingredients, sorptive phase for delivery of fragrances, vitamins, oils, moisturizers, insect repellents, sun filters and a variety of other active ingredients, prolonging shelf life by trapping degradable active ingredients in the porous particle matrix (effectively shielding the active ingredient from bacteria, which are too big to enter particle pores). The functions of these polymers clearly go beyond the well-known, well-publicized scrubbing effect of microbeads.

4 Environmental fate and effects of microplastics

4.1 Plastic ingredients in wastewater streams *en route* to the oceans

Thermoplastics and thermoset plastics in PCCPs are expected to outlast the consumers who use them. Where do PCCP microplastic materials go after they are used by consumers and can they potentially contribute to the marine litter problem?

Emissions via treated and untreated wastewater streams

The ingredients in ‘rinse-off’ PCCPs (but also in some cases ‘leave-on’ PCCPs) are typically discharged to wastewater streams, the main global emission route. From wastewater the ingredients can be transferred to surface water directly in the absence of wastewater treatment systems or via sewage overflows. A significant percentage of households across Europe are not connected to wastewater treatment facilities, meaning microplastics are discharged directly to surface water in many communities. In the Netherlands the percentage of the population connected to urban wastewater treatment facilities is around 99%, in the EU as a whole the average percentage of the population benefiting from wastewater treatment is about 70% (EuroStat 2012). In developing countries, 80-90% of untreated wastewater is discharged to water bodies and coastal zones (UN Water 2008).

Treated wastewater effluents are also known to contain plastic particles, up to 100 particles per liter with current methods (Browne et al. 2011; Leslie et al. 2012b, 2013; Brandsma et al. 2014), including particles made from the same plastic type which are also the same size and shape as particles applied in some PCCPs (see Leslie et al. 2012b). Note that many other fragments, (such as textile fibers and particles near or >5 mm in size) detected in wastewater that are not attributable to PCCP sources. This demonstrates that wastewater treatment plants have not been designed to degrade plastic particulates or retain 100% of plastic particles in the sludge fraction. In turn, the products containing plastic particles were not designed to be degraded in the wastewater treatment plant, in the effluents or in the natural environment.

Emissions via sewage sludge or biosolids

Besides effluents, sewage sludge is another important receptacle of microplastics from PCCPs, with hundreds of particles per kg wet weight typically detected there with current methods (Brandsma et al. 2014). In the Netherlands, most sewage sludge containing microplastics is eventually incinerated, however emission to the environment via landfilling or application to agricultural lands as biosolids is common practice in many developed countries (e.g. EPA 2006; Fytili and Zabaniotou 2008). In Europe, over a third of the total sewage sludge generated is currently being applied on agricultural fields as fertilizer, called ‘biosolids’, around 40% is being landfilled, and 12% is used for forestry, land reclamation, etc. (Fytili and Zabaniotou 2008). Microplastics introduced to the agricultural land from biosolids applications or landfill site applications are expected to enter waterways via runoff²² eventually, since the particles are unlikely to degrade. Up until the late 1990s, many developed countries including the Netherlands dumped sewage sludge at sea, which is still a common practice in other areas of the world (UNEP 2005). Via runoff and emissions to freshwater systems, microplastics from PCCPs and other sources can potentially reach the marine environment, travelling freely, as aggregates, floating or in suspension in the water column, depending on several factors such as size and polymer material type, river water current velocity and many others.

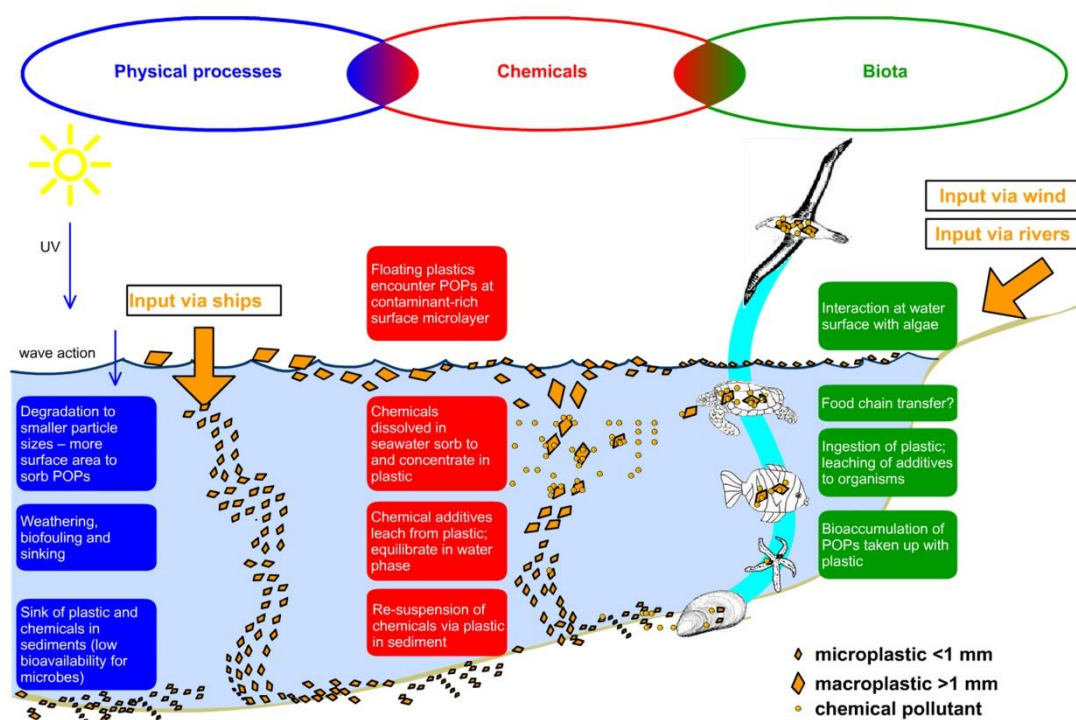
²² Runoff consists of water that is not absorbed by the land but can transport soil (and other) particles, nutrients and contaminants to surface water.

PCCP plastic ingredients are one part of total microplastics fraction

Once emitted, microplastics from PCCPs are mixed with other ‘secondary’ microplastics from crumbling macroplastics, as well as ‘primary’ microplastics (which are not fragments but were manufactured as particulates) emitted from other sources, e.g. transport spill. The vast number of different categories of microplastic sources are hardly distinguishable from one another based on the appearance of microplastics. Polymer type can be determined using Fourier Transform Infra Red (FTIR) or Raman spectroscopy, as long as particles are large enough (on the order of ca. 10 μm thick) and the appropriate reference materials are available. Polymer types give clues as to the chemical identity of the particle, however pinpointing the *source* based on polymer type alone is normally not possible. For example, polyethylene is widely used as an ingredient in cosmetics but it is also the world’s most produced plastic type. Fragments identified as polyethylene could potentially originate from PCCPs and millions of other types of products.

4.2 Environmental consequences of cosmetic microplastics

The body of toxicity data for microplastics such as those applied in PCCPs comes largely from fields outside the cosmetic chemistry field. Such data are also cited in Cosmetic Ingredient Reviews. These data are briefly reviewed here with special attention to some of the plastic types that are applied in PCCPs.



Graphic by H.A. Leslie and P.J. Kerstens

Figure 4.1 Microplastics from sources on land and at sea are emitted to the marine environment where they are distributed among the various environmental compartments such as sea surface layer, water column, sediments and biota. Biota may bioaccumulate hydrophobic chemicals associated with plastic; organisms that can excrete ingested plastic may also undergo plastic-facilitated depuration under certain conditions (see Koelmans et al. 2013). Plastic PCCP ingredients that reach the sea are expected to do so via runoff, fluvial transport (rivers) and directly from ships. Plastic particles may be transported with other particulates through the air.

PCCP microplastics are persistent, transported and potentially enter food chains

Plastic particulates entering the environment for example, via wastewater or biosolids runoff, can potentially be consumed as food by aquatic organisms and enter the food chain (Wright et al. 2013). Another possible uptake routes is via the gills. Microplastics in the marine environment can potentially travel vast distances floating in seawater (Barnes et al. 2009), or sediment to the seabed (Figure 4.1). End-of-life plastic PCCP ingredients are typically incapable of mineralizing at measurable rates in the environment, either by biodegradation or by photo- and or thermal degradation processes; estimates of half-lives run in the hundreds of years (Andrady 2011; Zeng et al. 2005), longer than any persistent organic pollutant.

Environmental distribution

Plastic powders, microbeads, microspheres, granulates, etc. can contribute to the total microplastic load in the sea, but as mentioned above, once in the sea, these micrometre-sized primary microplastics are not readily distinguishable from secondary microplastics in the same environmental matrix. In the environment, PCCP microplastics will behave similarly to microplastics from other sources of similar sizes and polymer types. Other than plastic preproduction pellets and small plastic objects <5 mm that are still recognizable, the exact origins of the plastic particulates in the sea are untraceable. Nevertheless, to assess exposure, concentrations of microplastics (both primary and secondary combined) have been measured in seawater around the globe and have been reported for a growing number of marine sediments (e.g. Barnes et al. 2009; Lavender Law et al. 2010; Browne et al. 2011; Claessens et al. 2011; Leslie et al. 2013). It has been demonstrated in a variety of laboratory experiments that marine invertebrates take up microplastics e.g. lugworms, amphipods and barnacles (Thompson et al. 2004), blue mussels (Browne et al. 2008), sea cucumbers (Graham and Thompson 2009), and others. In field-collected biota, microplastics have also been detected, for example Northern Fulmar seabirds (Van Franeker et al. 2011), Norwegian lobsters (Murray and Cowie 2011) and various species of fish (Boerger et al. 2010; Lusher et al. 2013; Foekema et al. 2013). Plastic particles may adhere to microalgae (causing disturbances at very high concentrations, e.g. Bhattacharya et al. 2010) and very tiny biological organisms can also colonize microplastic particles (Zettler et al. 2013). In summary, the scientific literature is full of reports of microplastics detection in water, sediment and biota samples from around the world. A recent Dutch pilot study reported thousands of microplastics per kg (dry weight) in suspended particulate matter sampled from major rivers in the Netherlands (Brandsma et al. 2014).

Assessment of (eco)toxicological risk

For a pollutant to pose an environmental risk, there needs to be a combination of exposure to the pollutant and the pollutant must be hazardous. In the previous section, current knowledge of plastics particulates in the marine environment was briefly summarized for microplastics as a whole. We saw that organisms are known to take up microplastics when exposed. It is clear that exposure is possible in various habitats, including water bodies receiving wastewater or treated wastewater effluents, in rivers (present in suspended riverine particulates), in seawater and in marine sediments. What is known about the hazards of the plastic particles that are being detected in the environment and in field collected biota?

The knowledge about the (health) hazards of plastic particulates are emerging from a number of fields, including drug delivery, marine ecotoxicology, fragmentation of polymer implants such as PMMA or PE used in hip replacements (Rudolph et al. 1999; Requena et al. 2001; Cooper et al. 1992), heart valve implants (Ghanbari et al. 2009; Claiborne et al. 2013), inhalation toxicology (Carthew et al. 2006), nanotoxicology (Bergin and Witzmann 2013) and oral exposure nanotoxicology (Mahler et al. 2012).

The risks of plastic PCCP ingredients are specifically assessed by expert review panels such as the PCCP-industry funded Cosmetics Ingredient Review (www.cir-safety.org) and others. These assessments take the entire body of toxicological literature into account in principle, such as the studies cited here, as well as data provided by the PCCP industry. These panels focus largely on human health impacts during use of the product, i.e. via dermal uptake. This leads to the situation where an ingredient is considered safe (for dermal application), even if it is implicated in tumour formation when implanted inside the body (e.g. Cosmetic Ingredient Review 2012). Normal use of the product would not lead to internal exposure for the consumer, but should the product enter the environment as a consequence of using the product normally, organisms may encounter this material and become exposed via other uptake routes (such as via gills or ingestion).

Many of these assessments are voluntary as in large parts of the world most cosmetic and personal care ingredients are unregulated. The onus is on producers to design cosmetic formulations to be safe. For example in the USA, cosmetic product ingredients do not need Food and Drug Administration (FDA)²³ premarket approval, with the exception of pigments and ingredients deemed to have medicinal activities that could be regarded as drugs (e.g. antibacterial ingredients). The Cosmetics Ingredient Review in the USA has set up review procedures²⁴ for its independent efforts 'to determine those cosmetic ingredients for which there is a reasonable certainty in the judgment of competent scientists that the ingredient is safe under its conditions of use'. The EU Cosmetics Directive²⁵ is the strictest existing regulation of cosmetic and personal care ingredients in the world, now also addressing nanomaterials as ingredients. The substances in products are regulated in the Cosmetics Directive according to toxicity upon normal use of the product, assuming dermal exposure only. The environmental impact of emissions through normal product use are not taken into account in the scientific and technical assessments of ingredients.

Importance of particle size and shape in particle toxicity

From particle toxicity studies outside the cosmetics formulation area there is evidence of the toxicity of plastic particulates in diverse biological systems, from marine invertebrates to mammals to human tissue systems. Most of the research has focused on particle hazards in mammalian systems (a toxicological model for humans), and scientific research remains to be done on characterizing the toxicity of these plastic particles to the broader diversity of biological organisms that potentially come into contact with plastic particulates originating from PCCPs via various exposure routes. What is known is that particle toxicity is size- and shape-dependent but may also be dependent on the specific chemical make-up of the microplastic particle (polymer, monomer, additives, possible sorbed contaminants) (reviewed in Leslie et al. 2011).

Microplastics and mammalian systems

Several studies of the fate and pathology of ultrafine plastic particles in animal models and human cells, and human placental perfusion studies (to investigate transfer from mother to foetus) have provided particle transfer and some toxicity or hazard data, which is useful at least as a starting point when assessing the risks posed by microplastics. The emerging field of aquatic nanotoxicological research has many links to the study of microplastics toxicity. In mammalian systems, the uptake and toxicity of several types of plastic nanospheres have been studied, indicating that fine particulate plastic may in principle be transported through human gastrointestinal tracts to lymph and circulatory systems, through placentas to unborn foetuses, absorbed in lungs when inhaled, causing a variety of biological responses from the immune system and impacting health of bodily cells (Hopwood et al. 1995; Brown et al. 2001; Kato et al. 2003; Hussain et al. 2001; Wick et al. 2010; Berntsen et al. 2010; Fröhlich et al. 2009). The

²³ <http://www.fda.gov/Cosmetics/default.htm>

²⁴ <http://www.cir-safety.org/sites/default/files/pdf1.pdf>

²⁵ <http://ec.europa.eu/consumers/sectors/cosmetics/documents/directive/>

hazards have to date often been identified for nano-size ranges of plastic particles normally using known particle sizes, and often high doses, with many study designs identifying a hazard and not a dose response relationship that is directly useful for risk assessments. At the same time, we are lacking environmental exposure data in this nano-size range due to limitations in current analytical methods to determine unknown polymeric nanoparticles in environmental matrices.

Microplastics and marine species

As for marine species, green algae photosynthesis was observed to be negatively impacted by exposure to nano-sized polystyrene at very high concentrations (Bhattacharya et al. 2010). In another exposure assay using blue mussels, no toxicity of microplastics was observed, although translocation of microplastics to the haemolymph of the organism was reported (Browne et al. 2008) and microplastics in edible tissues of mussels have been reported at levels of one particle per gram (Van Cauwenberge et al. 2012). Von Moos et al. (2012) demonstrated negative effects of microplastics (1-80 μm) on marine mussels when they measured a variety of physiological endpoints after exposure to microplastic, such as granuloma formation (inflammatory response), decreased lysosome stability and an increase in haemocytes. Data for a dose-response relationship was not presented. In the marine lugworm, negative effects on feeding in the presence of microplastics were also observed, as well as weight loss in exposed animals in a study that also examined combined PCB and microplastic exposure (Besseling et al. 2012). Research on the impact of microplastic exposure on energy budgets of organisms is ongoing within the marine litter research community. Microplastic particles (1.7-30.6 μm) were observed to be taken up by 17 species of marine organisms, and it was shown also to reduce algal feeding by copepods when they were acutely exposed to high concentrations of microplastic (Cole et al. 2013). Lee et al. (2013) reported effect concentrations for exposures of copepods to polystyrene (PS) particles of different sizes. They observed chronic copepod mortality from exposure to 0.05 μm PS at concentrations $>12.5 \mu\text{g/ml}$. Reduced fecundity was reported for exposure to 0.5 and 6 μm PS particles at all concentrations tested. The hazards of microplastic exposure are currently under study in many research initiatives around the world and the body of microplastic toxicity knowledge is growing. A great deal of data would still need to be collected, including dose-response relationships for the various materials and particle sizes and shapes, for the purpose of a future risk assessment of this large class of particulate materials.

5 Concluding remarks

Plastic is one of thousands of types of ingredients that are currently applied in personal care and cosmetic product formulations around the world. A survey of the literature and industry websites shows that a wide variety of plastic types, particle sizes and shapes are used to achieve the functionality and ‘feel’ that cosmetic chemists and marketing directors aim to achieve in the products they sell. An unintended consequence of PCCP plastic ingredients is that they are emitted to the environment via wastewater. In most parts of the world, the emission is directly to surface water, but emission can also proceed via treated effluents, or the application of sewage sludge (biosolids) on agricultural lands, landfills, or sewage sludge dumping at sea. Impossible to collect after product use, the PCCP microplastics become potentially available for accumulation by vast number of species, may be transported long distances, may enter the marine food chain and may ultimately end up on our plates as well. This could eventually have not only a toxicological impact but also economic implications if consumption of certain food types in the future would need to be limited (as we have seen with the persistent organic pollutants in fish from the Baltic Sea for example).

The toxic consequences of microplastic exposure have been studied in discrete research fields and published in a variety of journals covering topics ranging from drug delivery, plastic medical implants, occupational health and epidemiology to marine ecology and ecotoxicology. A growing body of evidence is showing that microplastics (including very fine nano-sized plastic particles) are not harmless but can have clear adverse effects on biological systems representing a wide variety of taxonomic levels when the exposure level exceeds threshold levels. Exactly what those levels are would be useful to further investigate. Linking the types of microplastic exposures in the field to existing and forthcoming laboratory toxicity data would help characterize the risk. A major knowledge gap is how to assess plastic nanoparticle exposure in the field situation. International interest and commitments towards a reduction in marine litter including microplastics in the marine environment (such as the EU’s MSFD legislation) has spurred attention to more research and development in this area, as well as examining potential sources. These PCCPs are one of many sources being scrutinized by the international community of concerned citizens, policy makers, scientists and the companies which choose to recognize and take producer responsibility in this area.

An *end-of-pipe*²⁶ type of solution to halting PCCP microplastics emissions via wastewater streams is challenging because treatment facilities are not designed to fully retain plastic particulates, and applying further nano- or microfiltration is expected to be costly both in terms of energy inputs and financial investments. Furthermore, during high flow periods, wastewater is discharged to the surface water untreated, bringing persistent microplastics into the environment anyway. When looking further at the situation in Europe and beyond, it should be noted that an appreciable percentage of European households and up to 90% of households in the developing world are not connected to wastewater treatment facilities of any kind, meaning PCCP plastic ingredients can be discharged directly to surface water in many communities worldwide. Data regarding exactly how many PCCP products on the market contain microplastics as defined here and where they are sold are not (publically) available.

The UN World Water Development Report (UN 2003) estimated global wastewater production to be 1500 km³ per day. Based on all wastewater observations so far, wastewater throughout the world is expected to contain PCCP plastic ingredients to various degrees. How much of the microplastics problem in the sea would be solved by a cleaner production approach to PCCPs? The contribution of PCCP plastic ingredients to the total load of microplastic from all sources in the oceans has not been measured, but to get an indication of the scale of emissions for toothpaste

²⁶ End-of-pipe solutions curb pollution emissions by add-on measures, e.g. filters.

alone, consider a European population in which each person uses an average of 2 g of toothpaste a day. If a hypothetical 5% (w/w) of the toothpaste they use were to be a plastic ingredient, then Europeans would be spitting around 74,000 kg of plastic particulates into their sinks on a daily basis. The global PCCP industry was worth 433 billion USD in 2012 (Euromonitor International 2012) – even if a fraction of these mountains of products were to contain small percentages of plastic ingredients, the total emission from this source is not likely to be negligible. Using the same database, Gouin et al. (2011) estimated that Americans were emitting 263 tonnes of exfoliating beads on an annual basis. There are also many examples of larger sources – a single accidental spill of plastic preproduction pellets apparently created 150 tonnes of microplastic pollution near Hong Kong in 2012 (six shipping containers were lost in Typhoon Vincente). Sewage sludge is made into biosolids and used as fertilizer making it undesirable to retain highly persistent microplastics in sewage sludge, as they will be emitted to the land and end up in run-off.

Taken together these facts point to *cleaner production*²⁷ as a more universally effective route to achieve a reduction in microplastics emissions from cosmetic and personal care products on the short term. While packaging and other macrosized plastic objects have the potential to be recycled, plastic cosmetic ingredients are impossible to recycle because the product is discharged into wastewater at end-of-life. There is a great potential to reduce environmental impact in the product design phase (German Federal Environment Agency 2000) however that requires the decision-makers in companies to select cleaner production designs. This is in principle possible, and has been done in the past when eliminating dangerous solvents, toxic metals and ozone depleting gasses from PCCP formulations (Gruber 1999). Not all PCCPs contain plastics today - many PCCP formulations currently on the market do *not* contain any toxic, nondegradable plastic materials. Alternatives are available or could be developed to perform the functions that plastics do, enabling the same product qualities that are desirable for companies and consumers alike. The costs and benefits of end-of-pipe options versus cleaner production options should be carefully scrutinized. Future transitioning to plastic-free ingredients would be in line with Europe's ambition to promote a circular economy in which (inter alia) eco-design is applied, the loss of valuable materials is prevented, thereby moving us 'towards zero waste' and reductions in environmental impacts (COM 2014, <http://ec.europa.eu/environment/circular-economy/>).

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²⁷ Cleaner production limits pollution at the source through design of cleaner products and/or production methods. Investments in cleaner production lead to innovations that companies benefit from economically, often making it a favourable approach (Frondel et al. 2004).

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