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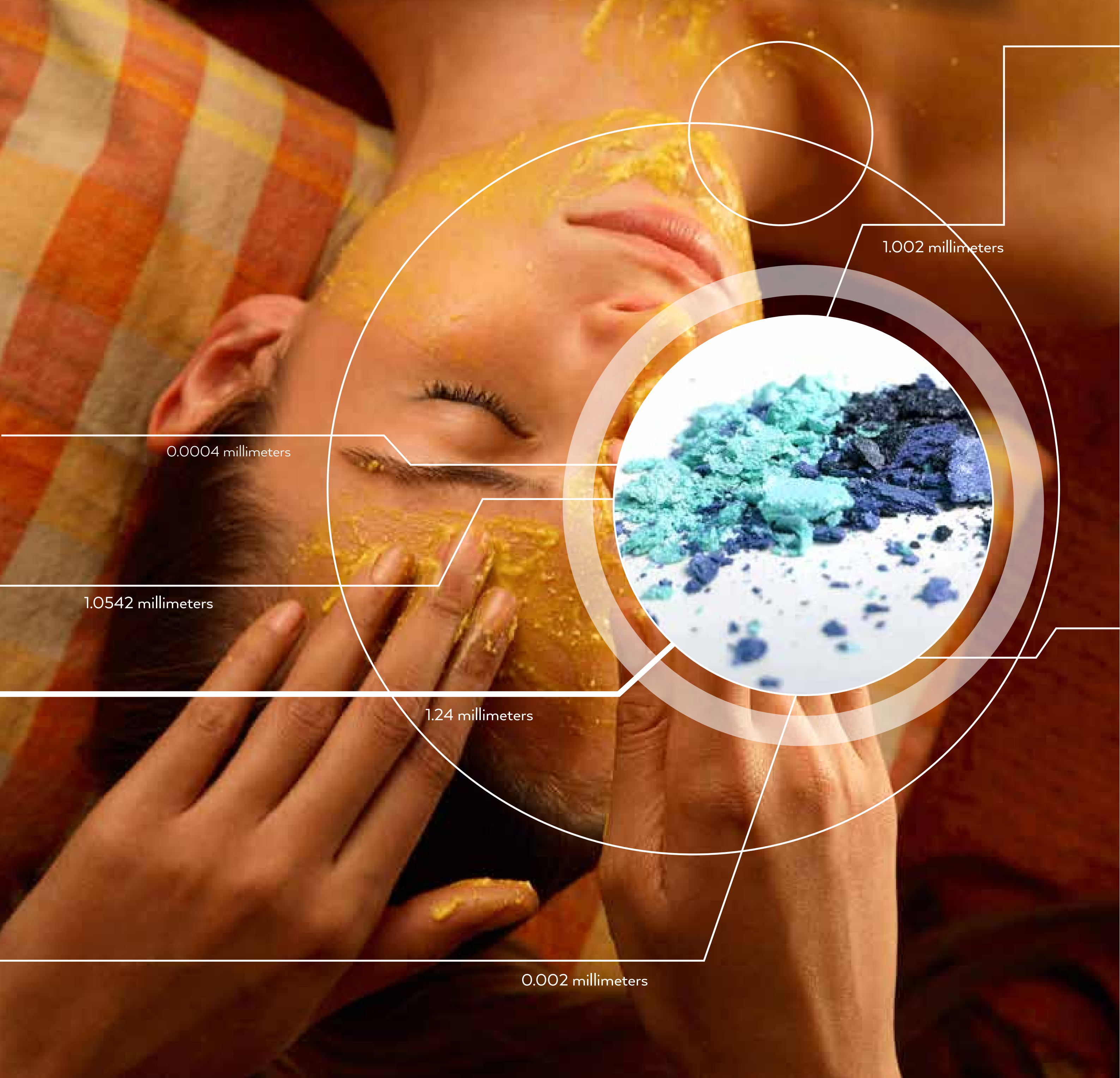
PLASTIC IN COSMETICS

ARE WE **POLLUTING** THE ENVIRONMENT THROUGH OUR PERSONAL CARE?

PLASTIC INGREDIENTS THAT CONTRIBUTE TO MARINE MICROPLASTIC LITTER



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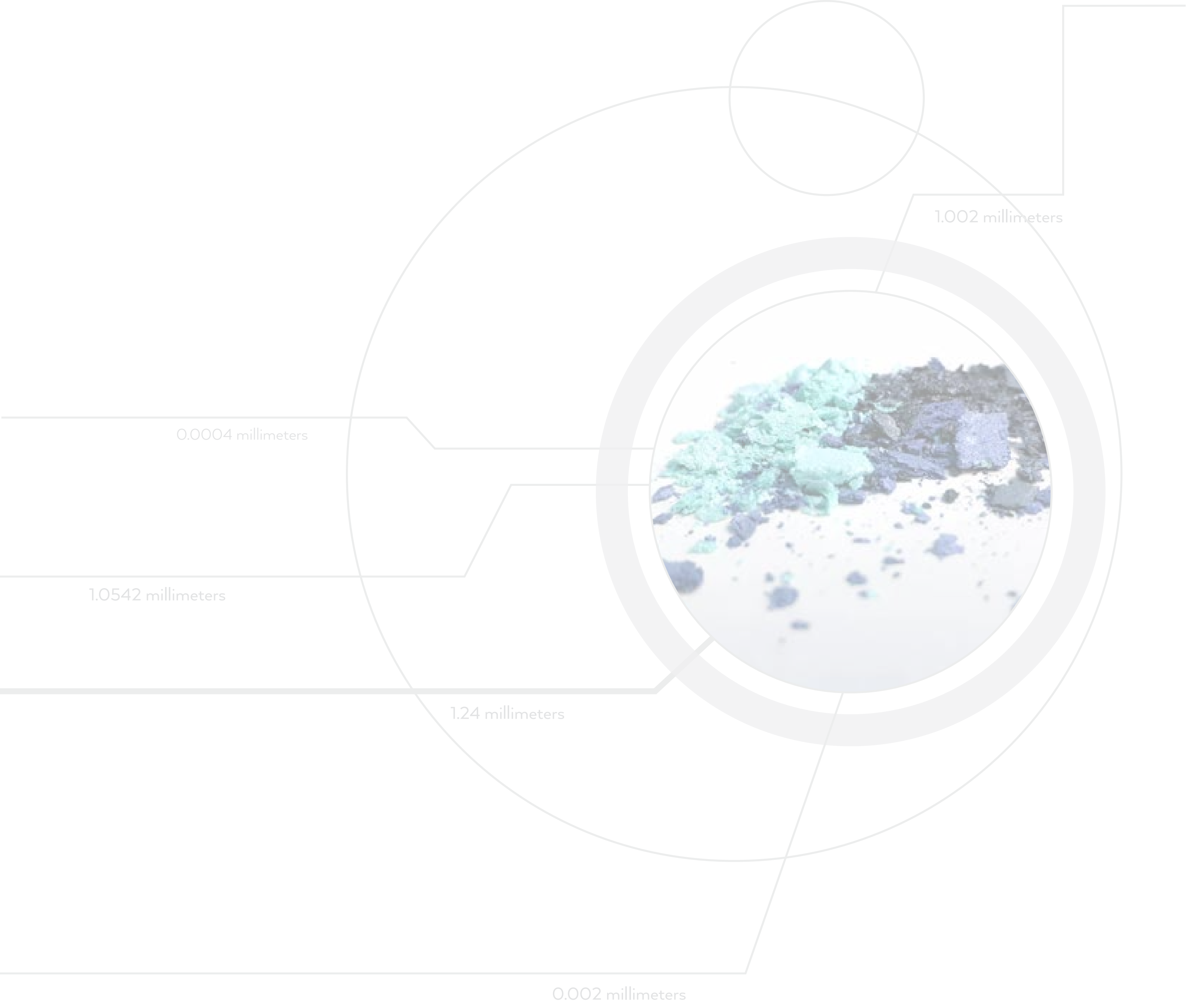

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Contents

Summary

- Spherical or amorphic plastic particulates are used as ingredients in personal care and cosmetic products (PCCPs) for a variety of purposes such as sorbent phase for delivery of active ingredients, film formation, exfoliation, viscosity regulation and many others. 'Microbead' is one of many terms applied to plastic PCCP ingredients; they may also be called microplastics, microspheres, nanospheres, plastic particulates etc.

- A large number of plastic materials are currently being applied in PCCPs. Since their appearance in cosmetics 50 years ago, plastics have become widespread in cosmetic and personal care formulations.

- PCCP microplastics, as discussed here, are synthetic solid materials made from various types of polymers[1] and chemicals (e.g. additives). Water-soluble materials and liquid synthetic polymers fall outside the definition of 'microplastics' applied in the marine litter field because they are not particulates (solids) and therefore fall outside the scope of this paper.

- Most of the plastic ingredients in PCCPs contain nondegradable polymers. These plastics may take hundreds of years to completely degrade via oxidative or photodegradation routes. Replacing plastic ingredients with biodegradable plastics such as Polylactic acid (PLA) is not advisable as PLAs only degrade when subjected to high temperatures in industrial settings.

- Plastic ingredients are applied in a variety of leave-on and rinse-off formulations such as: deodorant, shampoo, conditioner, shower gel, lipstick, hair colouring, shaving cream, sunscreen, insect repellent, anti-wrinkle creams, moisturizers, hair spray, facial masks, baby care products, eye shadow, mascara etc.

- There is more to 'microbeads' than meets the eye – while some are large enough to be easily visible to the naked eye, other microbeads on the market for PCCP formulations are as small as 1 µm. Others are even smaller than that (nano-particulates).

- The size of the particulates applied depends on the function in the cosmetic formulation. Many of the particulates in PCCPs today are between 1 and 50 µm in size.

- Microbeads and other plastic ingredients are present in different products at different percentages, ranging from less than 1% to more than 90% in some cases. For example, a typical exfoliating shower gel can contain roughly as much microplastic in the cosmetic formulation as is used to make the plastic packaging it comes in.

- A total amount of 4360 tonnes of microplastic beads were used in 2012 across all European Union countries plus Norway and Switzerland according to a survey by Cosmetics Europe, focusing on the use of microplastic beads, with polyethylene beads representing 93% of the total amount equaling 4037 tonnes.

- Plastic ingredients in PCCPs that are poured down the drain after use, cannot be collected for recycling (unlike the packaging, which can be recycled). The plastic ingredients do not decompose in wastewater treatment systems, which can be lacking in large parts of the world. The ingredients are emitted via raw sewage, treated effluents or with sewage sludge applied as fertilizer (biosolids) on agricultural land, landfilled or dumped at sea.

1 Polymers are macromolecules made up of repeating subunits of low molecular weight molecules called monomers.

- The global PCCP industry was worth 433 billion USD in 2012 – even if a fraction of those products contain small percentages of plastic ingredients, the total emission from this source is still quite significant.
- Knowledge is emerging about the ubiquitous occurrence of microplastics throughout the world's marine environment and their potential for secondary health impacts via the food chain, including to humans who consume seafood. This coupled with emerging knowledge about the toxic effects such particles have on biological organisms including mammals has led to concern and actions to monitor and reduce microplastics emissions.
- Taking the potential impact of product ingredients on the natural environment into account during the design phase and achieving cleaner production of PCCPs could eliminate microplastic (and also packaging) pollution from PCCPs.
- The power of information to help drive mitigation activities is considerable. Consumers, policymakers, industry and businesses with knowledge provided by scientific communities and propagated by NGOs and other civil society representatives are enabled to make informed choices to protect marine ecosystems and human well-being.
- Further research is needed to better understand the implications of nano- and micro-sized plastics in PCCPs on human and marine ecosystem health, especially through ingestion and chemical transfer through the food chain.
- Given the associated potential risks of microplastics, a precautionary approach is recommended toward microplastic management, with the eventual phase-out and ban in PCCPs. Redesigning products that are more environmentally friendly, less plastic intensive and use safer chemicals can contribute towards reducing potential health threats posed by microplastics in PCCPs.

1

INTRODUCTION

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INTRODUCTION

Litter is found in all the world's seas and the ocean, even in remote areas far from human activities. The continuous growth in the amount of solid waste thrown away and the very slow degradation rate of most litter items result in the accumulation of marine litter at sea, on the sea floor and in coastal areas. Marine litter[2] is a major global environmental problem which the United Nations Environment Programme (UNEP) has been actively addressing through the Global Programme of Action for the Protection of the Marine Environment from Land-based Activities (GPA) as well as the Regional Seas Conventions and Action Plans.

The Global Partnership on Marine Litter (GPML), led by UNEP/GPA, is a voluntary multi-stakeholder partnership launched in 2012, and initially guided by the Honolulu Strategy – a framework for the reduction and management of marine litter. The reduction of waste entering the aquatic environment is a key aspect of the GPML. Plastic materials dominate many marine litter samples, whether the litter is large or microscopic in size. This paper focuses on the emerging issue of **plastic particles in personal care and cosmetic product (PCCP)** formulations as a possible source of micro-sized plastic litter. Known as 'microbeads', when used in PCCPs (see Section 2.2), and by several other terms (microplastics, microspheres etc.), these microplastic ingredients are solid materials that fall under the definition of marine litter when emitted to the marine environment (*UNEP 2005*).

1.1 An emerging global environmental issue

The pollution of the world's oceans with plastic and the international commitments made to take mitigating action to avert further plastic discharges to the ocean has helped focus attention on the various sources and routes through which persistent, potentially harmful plastic materials can be emitted to the marine environment. One notable example of a source is the plastic ingredients in PCCPs (*Zitko and Hanlon 1991; Gregory 1996; Derraik 2002; Thompson et al. 2004; Fendall and Sewall 2009; Arthur, Baker & Bamford, 2009; Leslie, Moester, de Kreuk & Vethaak, 2012; Leslie 2012*). The concern is that plastic ingredients in products that are being used by consumers in households worldwide are contributing to the total abundance of plastic particles smaller than 5 mm – or 'microplastics' as they are called (*UNEP 2011*) – in the ocean today.

Some PCCPs contain as much plastic added as **ingredients** as the plastic they are packaged in. Whereas packaging can potentially be recycled, the plastic ingredients cannot.

Normal use of PCCPs introduces these plastic particles directly into waste water 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 because the materials are too dispersed, the scale is too vast, ecological damage would be caused by the remediation (tiny organisms would likely be removed along with the microplastics), and the costs would be astronomical. Emission prevention is the key mitigation strategy (*STAP 2011*). Replacing plastic ingredients with biodegradable plastics such as PLA is not advisable as PLAs only degrade when subjected to high temperatures in industrial settings. It is impossible to collect plastic ingredients in PCCPs (and any other 'down-the-drain' products) at end-of-life for recycling, which sets them apart from most other plastic materials in the marine litter fraction. In contrast, plastic packaging and other large plastic items have the potential to be collected for recycling or to feed waste-to-energy incinerators.

1.2 Objectives of the paper

What are these plastic materials that are being used as PCCP ingredients and which ingredients are relevant for the marine litter debate? How are they emitted, and what harm can they cause in the environment? This paper addresses these questions, and aims to introduce an emerging marine litter-related issue for further discussion and possible action by stakeholders.

2 The UNEP defines marine litter as *'any persistent, manufactured or processed solid material disposed of or abandoned in the marine and coastal environment'* (UNEP 2005). Another term used for marine litter is marine debris.

2
DEFINING THE PLASTIC INGREDIENTS

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DEFINING THE PLASTIC INGREDIENTS

In order to effectively discuss how to deal with PCCPs as a possible source of microplastics, it is important to define which synthetic polymeric ingredients in PCCPs can be regarded as a 'microplastic', as defined by the international marine litter scientific community (*Thompson et al. 2004; Arthur et al. 2009*).

The plastic PCCPs ingredients of interest to the marine litter debate have the following properties in common with other microplastic litter:

- Synthetic polymers and/or copolymers (plastics)
- Solid phase materials (particulates, not liquids)
- Insoluble in water
- Nondegradable[3]
- Small size (maximum 5 mm, no lower size limit is defined)

When microplastic ingredients of PCCPs are discussed, many people refer to the term 'microbead', but what is a 'microbead'? 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 plastic particulates include: microspheres, nanospheres, microcapsules, nanocapsules, as well as several registered trademark and other product names.

The particulates in PCCPs can also be made of non-plastic materials (such as lipid, cellulose, granulated almond shell) but of interest here are those made of plastic according to the properties list above. The shapes of the plastic particulates that are marketed as 'microbeads' can be spherical but also amorphic (*see cover photo*).

3 Nondegradable refers to the lack of ability of the material to decompose or mineralize at measurable rates. The consequence of being nondegradable is that the material is persistent. No material is expected to last indefinitely.

In short, there are currently many terms for the plastic particulates that are sometimes used as ingredients in PCCPs formulations.

2.1 The plastic materials

Plastics have been applied as ingredients in PCCPs for several decades with early patents dating from the 1960's – today they remain a hotbed of innovation in new PCCPs. Plastic is a term for materials made from certain types of synthetic polymers. Besides polymers, plastics also contain other substances (e.g. additives) which help achieve the desired properties of the material. The plastic materials applied as ingredients in PCCP formulations discussed here include two main categories: thermoplastics e.g. polyethylene, polypropylene, polystyrene, polyamide polytetrafluoroethylene (Teflon), poly (methyl methylacrylate), and thermoset plastics, e.g. polyurethanes and certain polyesters. While many polymers used in PCCPs are water soluble or water-dispersable, some silicone polymers are amorphous solids (without a clearly defined shape or form) and with virtually no water solubility (*Cosmetic Ingredient Review 2011; Perry 2005*).

Ingredients are 'microplastics' when they are:

- solid phase materials
- particulates < 5 mm
- water insoluble
- nondegradable
- made of plastic

All synthetic polymer 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 softer materials. Poly (ethylene) molecules less than about 700 carbons in length are waxy, and alkane chains with less than 20 carbons are liquids or gases. Plastic polymer waxes are smaller macromolecules that result from shorter chain lengths, e.g. polyethylene wax, a popular PCCP gellant available as powder, flakes or granules. Polyethylene waxes are water insoluble, solid materials with melting points well above maximum

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sea temperatures and therefore also fall under the definition of marine microplastic litter. Longer chains produce more rigid materials, e.g. poly (ethylene terephthalate) glitters or styrene/acrylates copolymer colour spheres. Lengthening the chain of ethylene oxide polymers (better known as polyethylene glycols) to 20,000 results in solid materials, e.g. PEG-2M (*Gruber 1999*). Cross-linking tends to decrease water solubility of polymers, e.g. 'water dispersable' polymers known as 'microgels' (*Gruber 1999*).

The plastic PCCP ingredients include homopolymers but also many copolymers (see examples in Table 2.1). Homopolymers are polymer chains of a single monomer[4] type, such as is formed when styrene monomers are polymerized to poly (styrene) (PS). 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*). Poly (ethylene terephthalate) (PET) is increasingly being replaced by copolymers such as poly (butylene terephthalate)/PBT and others (*Cosmetic Ingredient Review 2012*).

Other common solid-phase 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.

According to the definition given at the beginning of this section, for a material to be categorized as belonging to the 'microplastic' fraction of marine litter, it must be a solid phase material. All thermoset plastics are solid phase materials. Thermoplasts are also solid materials but they can be melted into liquids when they are heated to temperatures exceeding their melting point (or glass transition temperature). For the typical examples (Table 2.1), the melting points far exceed the temperatures in the marine environment. It is important to note that 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[5] of materials with the same INCI name may be different.

4 A monomer is a low molecular weight molecule that is the basic unit of polymers.

5 'Phase' refers to whether the material is a solid, liquid, gas, etc.

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).

Table 2.1 Selected examples of solid-phase, water-insoluble plastic ingredients currently applied as particulates in personal care and cosmetics products. Note: some polymers that make up the plastic materials 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.

POLYMER	EXAMPLES OF 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 methylacrylate)	Sorbent for delivery of active ingredients
Poly(pentaerythrityl terephthalate)	Film formation
Poly(propylene terephthalate)	Emulsion stabilizing, 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, sun care)

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.*

Very fine up to large, visible particulates of both thermoplastics and thermoset plastics are found in some personal care and cosmetic product formulations.

2.2 Particle sizes

Plastic particulates applied as PCCP ingredients can be large enough to see with the naked eye (e.g. 50 -1000 µm), while others are fine particulates (low µm-range) or very fine particulates (<2.5 µm). The term 'microbeads' generally refers to solid particles of various shapes, e.g. spherical, amorphic, between 1 and 1000 µm (*see Table 2.2*). '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 very fine 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 - 2 µm) (*Ansaldi 2005; Kvitnitsky et al. 2005*). Micro-sized 'sponge' technology makes use of fine particles between 1 and 50 µm (*Saxena and Nacht 2005*); 'sponge' materials such as cross-linked poly(methyl methylacrylate) (PMMA) are sometimes used because they can sorb active ingredients, especially the more hydrophobic ones (*Lidert 2005*).

PHOTO: © WILLIAM ORLALE

These materials are shaped into particulates between ca. 1 and 200 µm for use as innovative delivery systems for active ingredients (*Saxena and Nacht 2005*). Plastic particles in the size range from 10 to 1000 nm are termed 'nanospheres' and 'nanocapsules' or simply 'polymeric nanoparticles' (PNPs) (*Rao and Geckeler 2011; Hubbs et al. 2011*). Once these small particles enter the environment it is impossible to remove them.

Table 2.2 Microplastic particle sizes

PARTICLE	SIZE RANGE
Microbead	1 – 1000 X 10^{-6} m
Microspheres	1 – 1000 X 10^{-6} m
Microcapsule	1 – 2 X 10^{-6} m
Nanospheres/capsules	10 – 1000 X 10^{-9} m

2.3 Functions in formulations

Plastic ingredients are part of the formulation for a variety of PCCPs such as: toothpaste, shower gel, shampoo, eye shadow, deodorant, blush powders, make-up foundation, skin creams, hairspray, nail polish, liquid makeup, mascara, shaving cream, baby products, facial cleansers, bubble bath, lotions, hair colouring, nail polish 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*).

A total amount of 4360 tonnes of microplastic beads were used in 2012 across all European Union countries plus Norway and Switzerland according to a survey by Cosmetics Europe, focusing on the use of microplastic beads, with polyethylene beads representing 93% of the total amount equaling 4037 tonnes (*Gouin et al. 2015*).

Depending on the polymer type, composition, size and 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),

PHOTO: PIXABAY / CREATIVE COMMONS

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), and 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 scrubbing effect of microbeads.

The functions of plastic ingredients in PCCPs go beyond the well-known 'scrubbing' effect of microbeads.

3
PLASTIC INGREDIENTS AS POTENTIAL ENVIRONMENTAL POLLUTANTS

PHOTO: © SUZANNE LONG/ROBERTHADING LIBRARY

PLASTIC INGREDIENTS AS POTENTIAL ENVIRONMENTAL POLLUTANTS

3.1 Emissions and fate

The main emission route of microplastics is via wastewater, with the ingredients being transferred to surface water directly, in the absence of waste water treatment systems, or via sewage overflows. Treated wastewater effluents are also known to contain plastic particles (*Browne et al. 2011*), including particles similar to those applied in some PCCPs (*Leslie et al. 2012*). Besides effluents, sewage sludge is another important receptacle of microplastics from PCCPs. In some parts of the world sewage sludge containing microplastics is eventually incinerated or emitted to the environment via landfilling or application to agricultural lands as bio-solids (*EPA 2006; Fytili and Zabaniotou 2008*).

Up until the late 1990s, many developed countries 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 can reach the marine environment, travelling freely, as aggregates, floating or in suspension in the water column. There they are mixed with 'secondary' microplastics from crumbling macroplastics, as well as 'primary' microplastics (which are not fragments but were manufactured as particulates such as pellets for industrial feedstock) emitted from other sources.

In the environment plastic particulates are consumed by aquatic organisms. Some of these particulates can potentially enter the food chain (*Wright, Thompson and Galloway, 2013*). Microplastics in the marine environment can travel vast distances floating in seawater, or sediment to the seabed (Fig. 3.1). End-of-life plastic PCCP ingredients are typically

"Estimates of half-lives of microplastics run in the hundreds of years, longer than any **persistent organic pollutant**". PCCP microplastics in wastewater become environmental pollutants.

PHOTO: ©RACE FOR WATER 2015 / PETER CHARAF

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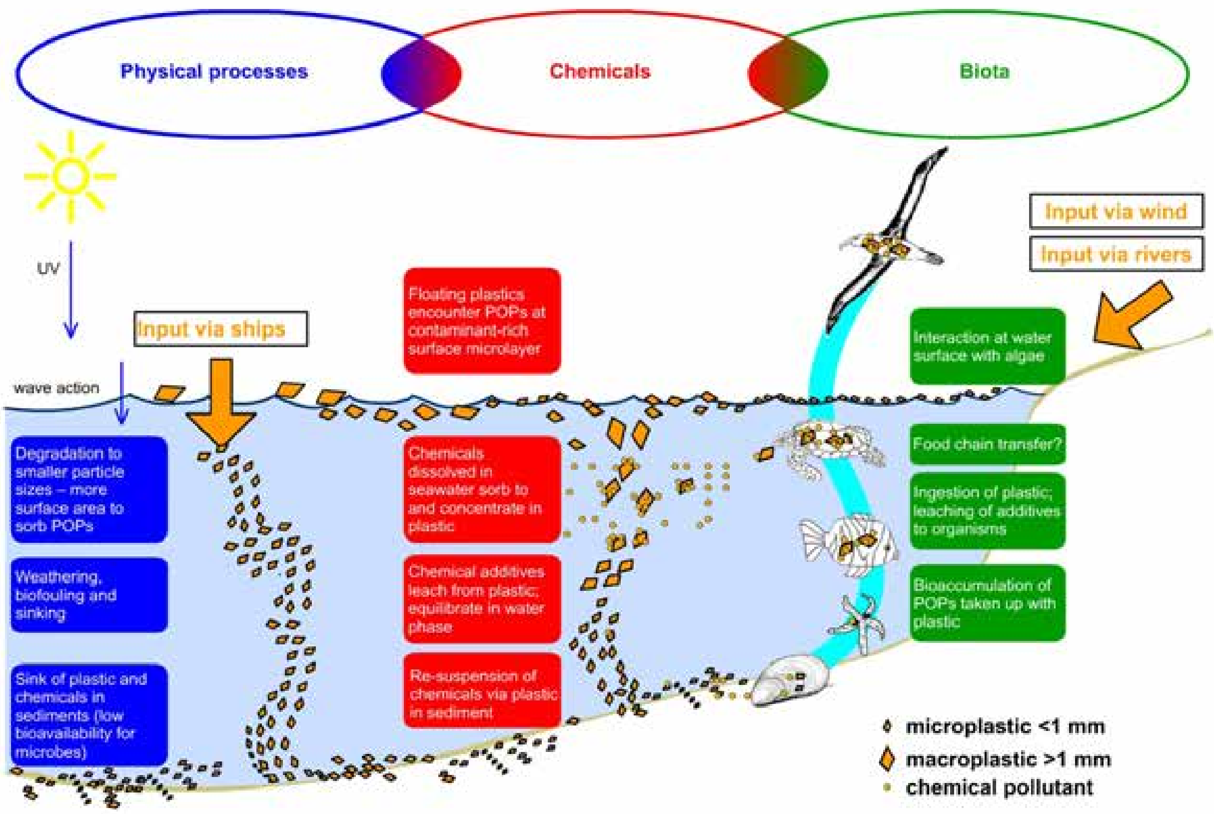
incapable of mineralizing at measurable rates in the environment, either by biodegradation or by photo- or thermal degradation processes; estimates of half-lives run in the hundreds of years (*Andrady 2011; Zeng, Yanful and Bassi, 2005*), longer than any persistent organic pollutant.

1.002 millimeters

Fig. 3.1 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. Plastic PCCP ingredients reach the sea mainly via rivers and directly from ships.



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

0.002 millimeters

3.2 Measured environmental concentrations of microplastics

Plastic powders, microbeads, microspheres, granulates, etc. can contribute to the total microplastic load in the sea, but rarely are these micrometre-sized primary microplastics distinguishable from secondary microplastics when detected in an environmental matrix. The plastics used as ingredients of PCCPs are not unique to these products and they are not distinguishable by shape or other indicators. 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) have been measured in seawater around the globe and have been reported for a growing number of marine sediments (e.g. *Barnes, Galgani, Thompson and Barlaz, 2009; Lavender Law et al. 2010; Browne et al. 2011; Claessens, De Meester, Van Landuyt, De Clerck and Janssen, 2011; Leslie, van Velzen and Vethaak, 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, Dissanayake, Galloway, Lowe and Thompson, 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*), oysters, mussels, common periwinkles and amphipods (*Leslie et al. 2013*) and various species of fish (*Boerger, Lattin, Moore and Moore, 2010; Lusher, McHugh and Thompson, 2013; Foekema et al. 2013*). In summary, the scientific literature is full of reports of microplastics detection in seawater, sediment and biota samples from around the world.

3.3 Environmental risks of microplastics

For a pollutant to pose an environmental risk, there needs to be 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, showing that they can be transported through freshwater and marine ecosystems after being emitted. 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 Poly(methyl methylacrylate) (PMMA) used in hip replacements (*Rudolph, Soyer, Schuller-Petrovic and Kerl, 1999; Requena, Izquierda and Navarro 2001*), and nanotoxicology, to name a few.

The risks of plastic PCCP ingredients are assessed by expert review panels such as the Cosmetics Ingredient Review panel and others. Many of these assessments are voluntary as in large parts of the world most cosmetic and personal care ingredients are unregulated and the onus is on producers to design cosmetic formulations to be safe. The risk assessments tend to 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*)

> Laboratory experiments demonstrated microplastic uptake and adverse effects in marine invertebrates.

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. However, more research remains to be done on characterizing the toxicity of these microplastics to a diversity of biological organisms that potentially come into contact with them 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, etc.) (*Leslie, 2012*).

Nevertheless, several studies of the fate and pathology of ultrafine plastic particles in models using animals, as well as human cells, and human placental perfusion studies (to investigate transfer from mother to foetus) have provided particle transfer and toxicity data, which is useful when assessing the risks posed by microplastics. The emerging field of aquatic nanotoxicological research has many links

Fine plastic particles can pass through **human gastrointestinal tracts** to lymph and circulatory systems, and through human placentas.

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 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 negatively impacting health of bodily cells (*Hopwood et al. 1995; Brown, Wilson, MacNee, Stone and Donaldson, 2001; Kato et al. 2003; Hussain, Jaitley and Florence, 2001; Wick et al. 2010; Berntsen et al. 2010; Fröhlich et al. 2009*).

As for marine species, green algae photosynthesis was observed to be negatively impacted by exposure to nano-sized polystyrene (*Bhattacharya, Lin, Turner and Pu, 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). Von Moos, Burkhardt-Holm and Kohler (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.

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 Polychlorinated biphenyl (PCB) and microplastic exposure (*Besseling, Wegner, Foekema, van den Heuvel-Greve and Koelmans, 2012*). Microplastic particles (ranging from 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*). 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.

3.4 Do PCCP ingredients contribute to the global marine microplastic load?

How much microplastic is there in the sea and how much of the total microplastic load in the sea originates from PCCPs? The answers to these questions are uncertain, in the first place because we do not know the total emissions or load of microplastics in the sea. Most of the sea water surveys focus on microplastics which are greater than 333 µm in size, representing only a fraction of cosmetics formulation plastic particulates and secondary microplastics, i.e. fragments of macroplastic items (*Leslie et al. 2012*). The volumes of plastic ingredients used in PCCP formulations worldwide are not publically available.

An approach to estimating how much microplastic in the sea originates from PCCPs may be to make calculations based on the regional volumes of PCCP applications consumed and a percentage of microplastics contained in them. These data, if available, could be then plugged into a model of emission to aquatic systems, residence time in freshwater systems, and transport to the sea. For instance, consider a European population in which each person uses an average of 2 g of toothpaste a day (*Hall et al. 2007*). If 5% (w/w) of the toothpastes used were a plastic ingredient, then Europeans would be spitting around 74,000 kg of plastic particulates into their sinks on a daily basis. The same calculation could be done for shampoo (average 6 g product used per person per day), and so on, in order to calculate the potential microplastics emissions when microplastics are present in the products.

Despite the collectability and recyclability of macroplastics, a major contributor to the small size fraction of marine litter is currently assumed to be the larger plastic litter pieces in the oceans that break up into microscopic fragments of secondary microplastic, smaller than 5 mm in diameter.

At the same time we are struggling to reduce macroplastic emissions to the sea, we see plastics playing a powerful role in the trend towards novel and innovative cosmetic ingredient delivery systems (*Rosen 2005; Patravale and Mandawgade 2008; Hubbs et al. 2011*). The cosmetic industry is following the lead of the pharmaceutical industry, which is using synthetic polymers including fine and very fine plastic particulates in innovative designs for active ingredient

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In a typical shower gel analyzed in a laboratory for **poly (ethylene) particulates**, there was roughly as much plastic material in the gel by weight as there was in the plastic container it came packaged in

delivery (*Lidert 2005*). As reported by (*Ammala 2013*), there are clear advantages of biodegradable polymers over nonbiodegradable for 'encapsulation' materials in PCCPs, however many applications are still using non(bio) degradable materials such as the plastics under discussion in this paper. Prestigious innovation prizes in the PCCP sector are being awarded to novel plastic ingredient applications, (e.g. PCCP ingredient consisting of a non-biodegradable solid ingredient, polyurethane – a well-known plastic) that are further encouraging their use.

Worth in the order of 433 billion US dollars in 2012, the PCCP market is a global market, growing at 8% per year (*Euromonitor International 2012*). Trends towards increased penetration of new markets include the rural poor of developing countries, e.g. the Global Public-Private Partnership for Hand-washing with Soap in which three leading multinational PCCP producers participate. Whereas the campaign is critical in preventing infectious diseases, it may inadvertently also lead to microplastic pollution, depending on the types of ingredients used. Normal use of PCCPs introduces any plastic ingredients present directly into wastewater streams, as the majority of these products are applied to the skin or hair and then rinsed off with water during bathing and personal care.

If just 5% (w/w) of toothpaste ingredients were plastic ingredients, then Europeans would be spitting ca. **74,000 kg of plastic particulates** into their sinks on a daily basis.

In a typical shower gel analyzed in a laboratory for poly (ethylene) particulates, there was roughly as much plastic material in the gel by weight as there was in the plastic container it came packaged in (*Leslie 2012*). Taking into account the large volumes of wastewater produced globally and the high-volume global consumption of PCCPs, there is potential for significant amounts of plastic ingredients entering (waste) water systems on a planetary scale.

4
POTENTIAL HUMAN HEALTH EFFECTS

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POTENTIAL HUMAN HEALTH EFFECTS

Plastics are synthetic polymers, designed for a multitude of purposes of great benefit to humanity, including health benefits. For example, polymers such as polyethylene are used for targeted drug delivery (*Pillay et al. 2013*). Other polymers are used to provide biodegradable structures for new tissue growth. These same polymers, such as polycaprolactone (PCL), may exhibit completely different properties outside the human body.

4.1 Potential direct health effects

Most published research on PCCPs has focused on non-plastic ingredients and their possible health impacts. The most likely direct route of entry is orally, for nano-particles both in cosmetics and in food, with interest focused on the probability uptake from the gastrointestinal tract (e.g. *Fröhlich and Roblegg 2012, Yada et al. 2014*). In dentistry there has been some suggestion that the presence of residual micro-beads of plastic in crevasses in the gums may lead to periodontal disease but this is not supported by hard evidence. More effort is spent investigating the fate of other constituents such as titanium oxide.

Phthalate esters are added to some types of plastic to improve performance, and they are commonly used in PCCPs. Desirable properties include anti-cracking agents in nail varnish (dibutyl phthalate DBP), skin softeners, colour and fragrance fixers (diethyl phthalate DEP) and anti-foaming agents in aerosols. DEP is the most commonly used in cosmetics. Laboratory tests using animal models have demonstrated an impairment of reproductive function, and some studies have suggested that the ubiquitous presence of phthalates may be causing health impacts in the wider human population. For example, one study suggested that babies whose mothers had recently applied infant care products were more likely to have phthalates in their urine than those whose mothers had not (*Sathyanarayana et al. 2008*). However, phthalates rapidly biodegrade both within the body and the environment, unlike many other contaminants of concern such as PCBs. Regulatory Bodies, such as the Federal Drug Administration (FDA) in the USA, are responsible for ensuring that concentrations of phthalates are within agreed national of international limits (*Hubinger 2010*). The limits themselves are subject to periodic review (e.g. *EC 2007*).

A comprehensive assessment of DBP in nail polish products in the USA, involving independent experts as well as the Centers for Disease Control and Prevention (CDC) and the FDA, concluded that DBP posed little or no risk to humans[6].

4.2 Indirect health effects

Despite a lack of direct evidence, it can be stated with some confidence that the great majority of particles in personal care products will be released to the environment without entering the human body. Current sewage treatment facilities are not designed to remove micro- and nano-sized manufactured particles and these will tend to be released into water bodies.

Once PCCP micro- and nano-plastics enter the marine environment they will join the population of plastic particles from other sources, including particles resulting from fragmentation. Indeed, they will be very difficult to differentiate. One difference is that PCCP plastics will have a limited range of additives compared with some other types of plastic which may contain, for example, flame retardants and UV stabilisers. However, organic contaminants present in seawater, such as PCBs, will be absorbed into the PCCP particles in a similar manner to other plastic particles, according to well-established reaction kinetics.

The routes of possible exposure to humans, as well as other biota, will be similar for all plastic particles of a given size. Particle size is important as it may limit or enhance exposure. For example, nano-sized polystyrene was shown to have one to two orders of magnitude higher affinity for a range of chlorinated biphenyls than micro-sized polyethylene.

6 http://phthalates.americanchemistry.com/Phthalates-Basics/Personal-Care-Products/Diethyl-Phthalate-DEP-in-Cosmetics-Deemed-Safe.html

This is partly due to simple surface area to volume considerations but may also reflect differences in particle properties at these scales (*Velzeboer, Kwadijk and Koelmans, 2014*).

Evidence is emerging both demonstrating the transfer of chemicals originally present in ingested plastic into the tissue of aquatic organisms, and of the presence of particles having a negative physiological consequence (*Rochman 2013*).

Most data come from laboratory experiments using carefully controlled conditions and often with nano-sized particles. Under natural conditions, effects are much more difficult to assess. For example, transfer from plastic is much more difficult to establish due to the ubiquitous presence of contaminants in seawater and natural food stuffs. Some careful studies have been able to demonstrate the transfer of certain flame retardants, presence in relatively high concentrations in

PHOTO: ©JAVIER LARREA/ROBERTHADING LIBRARY some plastic, from ingested plastic into bird and fish tissue. However, whether this is of ecotoxicological significance at the level of individuals or for the population remains unproven. There are no published data indicating transfer of chemicals to humans from ingested plastic, other than trace quantities of phthalates.

Public concern has been expressed about the possible negative impact of microplastics on wildlife and on human health through consumption of seafood, despite the lack of scientific evidence that this is either happening or may be happening. This situation is similar to other perceived hazards which may be difficult for the public to appreciate or visualize, such as radioactive contamination. What matters in terms of direct impact is the level of risk. However, a perceived risk may lead to a change in behavior, such as an unwillingness to consume seafood that may contain microplastics.

Filter-feeding organisms are more likely to ingest micro- and nano-sized plastic particles than many organisms and this is of interest from a human health perspective when such organisms are harvested for human consumption. Laboratory studies have indicated that plastic particles can be taken up by the epithelial cells lining the gut (*Von Moos et al. 2012*) and translocated across the gut wall (*Browne et al. 2008*). A recent study of two bivalve species cultured for human consumption (the blue mussel *Mytilus edulis* and the oyster *Crassostrea gigas*) suggested that European shellfish consumers could ingest 11 000 plastic particles a year per person (*van Cauwenberghe and Janssen 2014*). However, it is not yet possible to quantify the level of risk.

Many studies in the past have focused on single, priority pollutants that can be adsorbed onto the plastic in the marine environment and related ingestion impacts. Emerging science recognizes the importance of viewing microplastics in the marine environment as providing multiple stressors including less food-intake, sorption of chemicals, leaching of chemical additives including phthalates and concern over some polymers present in microplastics that can transfer carcinogenic and estrogenic monomers post-ingestion (*Rochman 2013*).

4.3 Need for further research

Further research is needed to better understand the implications of nano- and micro-sized plastics in PCCPs on human and marine ecosystem health, especially with ingestion and chemical transfer through the food chain. Research opportunities include further examination of the body burden and health impacts resulting from plastic exposure and associated additives in PCCPs such as phthalates to expand upon some of the work which has already been done on this topic (*Koch and Calafat 2009*). Nanoplastics are a growing field of exploration for nanotechnologies, and greater research is suggested for the potential health impacts associated with chemical uptake through nanoplastics.

Where liquid-phase and soluble synthetic polymers have not been traditionally identified as microplastics, whose definition requires a solid-phase and water in-soluble, future research can explore the health impacts associated with these non-solid phase synthetic polymers and related additives. While this paper focuses on plastics in PCCPs, since microplastics in rivers and the marine environment can often be indistinguishable from other primary and secondary microplastics, further exploration into the potential human health impacts associated with other forms of primary and secondary microplastics is recommended.

These may include: (i) microplastic abrasives used in air blasting and other industrial uses, which can become contaminated with heavy metals when used for stripping paint from metallic surfaces and cleaning engine parts; (ii) microplastic fibres arising from synthetic clothing; other sources of secondary nano- and micro-sized plastic particles; and (iii) release of fibers from tires.

Given the associated potential risks of microplastics, a precautionary approach is recommended toward microplastic management, with the eventual phase-out and ban in PCCPs. Redesigning products that are more environmentally friendly, less plastic-intensive and use safer chemicals can contribute towards reducing health threats posed by microplastics in PCCPs.

5
DISCUSSION AND CONCLUSIONS

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DISCUSSION AND CONCLUSIONS

The growing concern about microplastic particles in the marine environment is fueled by emerging knowledge on plastic applications in PCCPs and the micro- and nano-sized plastic particle toxicity to both humans and other life forms. The size range of particulates applied in PCCPs start at polymeric nanoparticles, through the low-micron range to particulates in the visible spectrum up to the millimetre-sized particles that can be found in typical exfoliating shower gels.

Plastic ingredients encompass far more than just the exfoliating plastic beads of scrub shower gels and soaps. The global market is gigantic and growing, and plastic PCCP ingredients typically require high production volumes. The materials are highly persistent, and are not often captured even when wastewater treatment facilities are available. Most of the world does not treat its wastewater or incinerate sewage sludge and most particles will therefore end up in the environment. Considering the ubiquity of plastic ingredients (*Goddard and Gruber 1999*) in the rapidly growing PCCP industry, the potential emissions will remain a concern.

The globally accepted importance of addressing emissions of plastic to the ocean suggests we direct our attention toward cleaner production and including environmental considerations in product design decisions. The potential environmental impact of products is estimated to be largely determined already at the design stage (*German Federal Environment Agency 2000*). Not all PCCPs contain plastics.

Alternatives are available or can be developed to perform the functions that plastics do, enabling the same product qualities that are desirable for companies and consumers alike. The cosmetics and personal care industry has in the past responded to environmental and health concerns associated with its ingredients, evidenced by the fact that many companies have

PHOTO: © COURTESY

Cleaner production in the cosmetics and personal care industry has the potential to completely eliminate a source of environmental pollution by microplastics.

PHOTO: © PIXABAY /CREATIVE COMMONS

moved to eliminate dangerous solvents, volatile organic compounds, heavy metals, and other toxics from their formulations (*Gruber 1999*). Many phase-outs have been implemented despite the absence of regulation of PCCP ingredients in most countries of the world[7]. Therefore it is conceivable that with increasing awareness of producers and consumers, plastic particulate ingredients that can potentially become marine litter may be eliminated from cosmetic and personal care formulations in the future.

The global commitment to reduce emissions of plastic to the ocean at Rio+20, the Honolulu Declaration, and international and national legislation require a dedicated response from all sectors and stakeholders in terms of consumer behavior, waste prevention, cleaner production and the consideration of environmental impacts in product design decisions, including the PCCP industry.

What has been done?

- The "Beat the Microbead" app was launched in 2012, by the North Sea Foundation and the Plastic Soup Foundation – the App allowed Dutch consumers to check whether personal care products contain microbeads by scanning a products barcode. In the summer of 2013, the United Nations Environment Programme and UK based NGO Fauna and Flora International joined the partnership to further develop the App for international audiences. The App, which is available in seven languages. has been very popular, convincing a number of large multinationals such as Unilever, Johnson & Johnson and the Body Shop to announce their intent to stop using microbeads. The App is available at *http://get.beatthemicrobead.org/*
- In the U.S., Illinois became the first state to enact legislation banning the manufacture and sale of products containing microbeads. This two-part ban will enter into effect in 2018 and 2019.
- The Netherlands, Austria, Luxembourg, Belgium and Sweden have issued a joint call to ban the microplastics used in personal care products, saying the measure will protect marine ecosystems – and seafood such as mussels – from contamination. The joint statement[8] that was forwarded to the EU's 28 environment ministers was stating that the elimination of microplastics in products, and in particular, in cosmetics and detergents, "is of utmost priority".

7 In parts of the world where cosmetics ingredients are indeed regulated, such as in the EU with the European Cosmetics Directive, the regulatory assessment criteria are typically limited to human health risks during product use. Environmental impacts or human health effects after product end-of-life are generally not considered.

8 http://register.consilium.europa.eu/doc/srv?l=EN&f=ST%20 16263%202014%20INIT

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ABBREVIATIONS LIST

CDC	Centers for Disease Control and Prevention
DBP	Dibutyl phthalate
DEP	Diethyl phthalate
FDA	Federal Drug Administration
GLOC	Global Conference on Land-Ocean Connections
GPML	Global Partnership on Marine Litter
INCI	International Nomenclature of Cosmetic Ingredients
mm	Millimeter
MW	Molecular weight
NGO	Non-governmental organisation
nm	Nanometer
PCB	Polychlorinated biphenyl
PCCP	Personal care and cosmetic product
PET	Poly(ethylene terephthalate)
PLA	Polylactic acid
PMMA	Poly(methyl methylacrylate)
POP	Persistent organic pollutant
UNEP	United Nations Environment Programme
w/w	Weight per weight
µm	Micrometer

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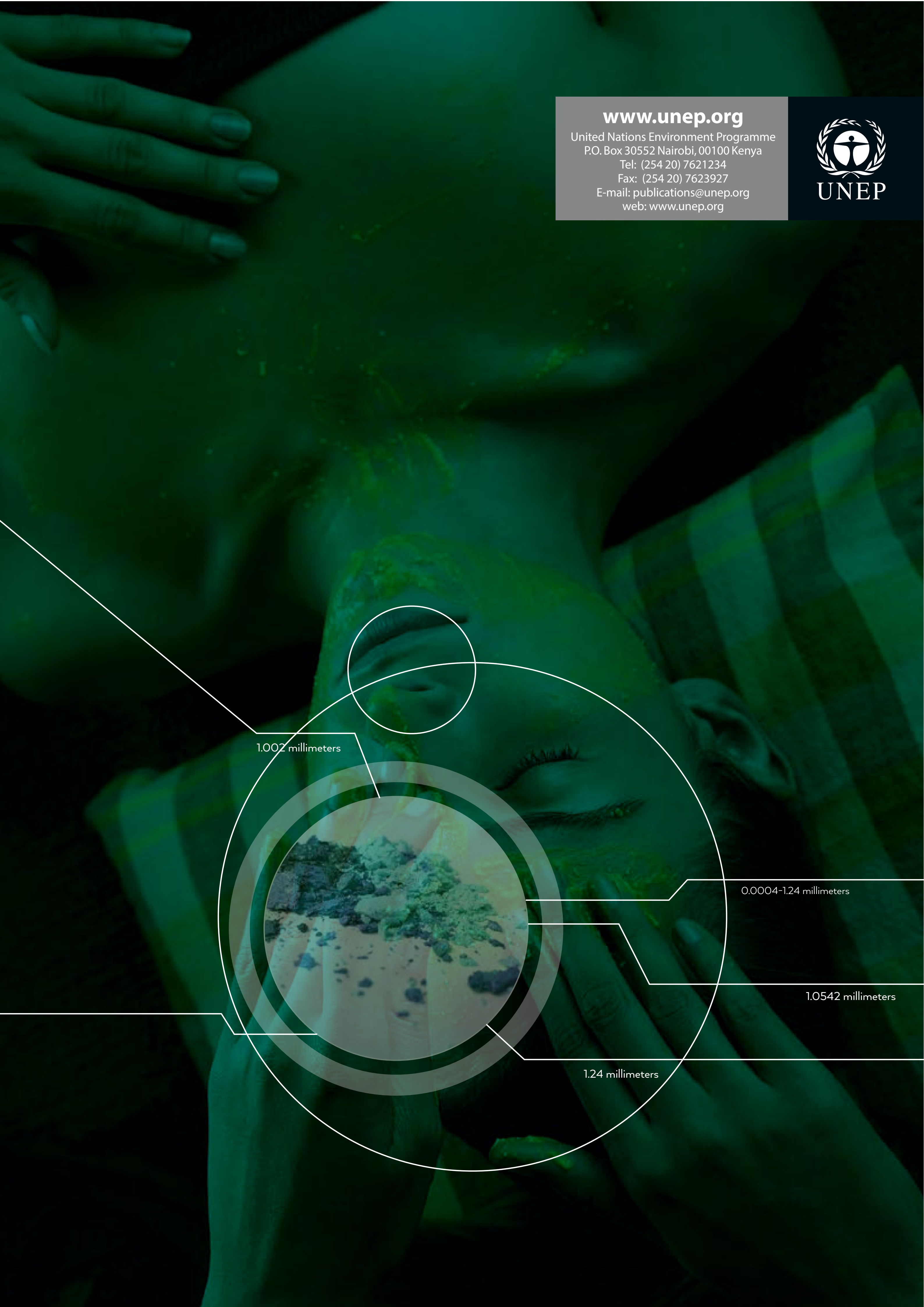

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