THE FORGOTTEN SYNTHETIC POLYMERS AND THEIR ENVIRONMENTAL AND HUMAN HEALTH CONCERNS

A perspective-review on water-soluble, liquid, semi-solid & biodegradable polymers and engineered nanoplastics

The scientific community's and the public's attention towards environmental contamination by synthetic polymers has increased substantially over the past decade[1–3]. Great effort has gone into investigating the extent of the pollution through data collection and the development of analytical tools. As a result of these efforts, we now know that plastic pollution stretches from the Himalayas[4] to the deepest ocean trenches[5], to our homes[6]. Most of this research has focussed on water-insoluble polymers, in the macro- and micro-size ranges. Currently, various legislative developments to reduce plastic pollution are underway at the European level. For example, the Single-Use Plastic (SUP) directive aims to reduce the use of certain plastic products[7], and the European Chemicals Agency (ECHA) has proposed a restriction on intentionally added microplastics (>100 nm) in among others agricultural products, detergents and cosmetics[8]. An EU-wide restriction of microplastics is considered justified due to concerns similar to those of persistent, bioaccumulative and toxic (PBT) substances[8]. Some groups of synthetic polymers have however been exempted in this restriction proposal, even though PBT concerns and similar hazards can exist for these polymers too. With this statement we address the most relevant and pressing concerns for these exempted synthetic polymers and stress the need for hazard assessments to identify hazardous polymers within these groups. Whilst knowledge gaps concerning PBT characteristics of synthetic polymers still exist, we argue that the precautionary principle should apply.

Water-soluble, liquid and semi-solid polymers

Water-soluble polymers
Synthetic water-soluble polymers (WSPs) are "substances that dissolve, disperse or swell in water and, thus, modify the physical properties of aqueous systems in the form of gelation, thickening or emulsification/stabilization"[9]. Annual production volumes of major WSPs such as polyethylene glycol (PEG) and polyacrylic acid (PAA) are estimated in the millions of tons range in Europe alone[10]. Many WSP applications in, for example, paints, building materials, agricultural products, personal care products, pharmaceuticals and oil & gas extraction[11] enable direct or indirect discharge into the environment (see references within[10]). This combination results in a high potential for the increasing presence of WSPs in the environment. Scientific studies indicate that concerns regarding persistency and toxicity that exist for insoluble polymers are also true for some WSPs. Some WSPs are very resistant to degradation and therefore persistent in the environment[12–14], which in itself is a cause of concern. With the continuous release of a persistent substance, environmental concentrations will inevitably increase, as will the probability of adverse effects. Once adverse effects are observed, reversing contamination could take centuries, or even longer[3,15]. Moreover, as WSPs are considered desirable for a wide range of applications, for example as soil conditioners or flocculent agents in wastewater treatment (e.g., polyacrylamide), they may act as such flocculants and soil conditioners

in areas they were not intended for. When present in the environment in sufficiently high concentrations, this could potentially lead to long-lasting changes to natural ecological processes[16].

Other WSPs are more prone to degradation and will degrade into various transformation products (e.g., smaller and more mobile polymers, oligomers, monomers and other chemical by-products)[16], which may exhibit persistent and toxic characteristics[17]. Polyacrylamide (PAM) is a prominent example of this as its monomer acrylamide is a known neurotoxin and potential carcinogen[10,11] and has been included in the 'Substances of Very High Concern' candidate list[18]. The bioavailability of WSPs seems to be of less concern as WSPs themselves are often too large to cross biological cell membranes[11,13], however, their transformation products may behave very differently[11].

WSPs can enter drinking and waste-water treatment plants and are often deliberately added to these plants to flocculate colloids and organic matter to a WSP-rich sludge. WSPs that do not end up in the sludge, and potentially in soils later as fertilizers, will then enter drinking water or surface waters. The extent of WSPs and degradation products that are present or accumulating in drinking water, surface water, sediments or soils is not investigated due to lacking analytical tools. Therefore, despite increasing exposure to WSPs, the environmental and health risks resulting from them remains highly uncertain[10,17].

Liquid and semi-solid polymers
Some synthetic polymers are used in a liquid or semi-solid phase when being applied in products. Depending on their molecular structure, these polymers can be water soluble -- for example polyethylene glycol (PEG) -- and readily dispersed in water. They can also form insoluble droplets, such as polydimethylsiloxane (PDMS) oils, also called dimethicone oils[19,20]. The phase of a polymer depends not only on the monomers that make up the (co)polymer but also on properties like chain length, degree of crosslinking and molecular weight. For instance, the longer the chain length of the dimethicone, the larger the viscosity, and the higher the melting point. Therefore, dimethicones can span from liquids to semi-solids to amorphous, rubbery solids. Lastly, the ratio of different monomers in copolymer material can also determine the phase of the polymer[21]. The use of liquid and semi-solid polymers is widespread; every year 23,700 tonnes of soluble, semi-solid and liquid polymers are used in cosmetic products in Germany alone, compared to 922 tonnes of solid synthetic polymers (<5 mm). Many of these soluble, liquid and semi-solid polymers are released into the wastewater[22]. An example of a liquid polymer of concern and commonly used in personal care products is dimethicone. Dimethicone meets the persistent criteria as described in REACH legislation, has been identified as a CMR (carcinogenic/mutagenic/reprotoxic) substance and exhibits endocrine disrupting properties[23]. Moreover, some dimethicones have been identified as a potential risk to the environment[20,23]. Thus, liquid or semi-solid polymers should not be presumed to be benign, as they can be of environmental concern as well.

Engineered nanoplastics

Though some debate exists about the exact cut-off between a microplastic and nanoplastic, nanoplastics are often defined as 1 to < 100 nanometres[8,24]. Plastics of 100 to <1000 nm can be referred to as submicron plastics and plastics > 1000 nm can be referred to as microplastics[24]. Engineered nanoplastics (ENPs) are nanoplastics that are deliberately produced at the nanoscale to allow for specific product characteristics. While not all applications of engineered nanoplastics or plastic nanomaterials will facilitate discharge into the environment (e.g., medical devices), others will. In cosmetic products for example, these engineered nanoplastics can be as small as 10

nanometres and can be directly or indirectly emitted into the environment via wastewater[21]. Secondary nanoplastics which have fragmented from larger plastic objects are another source of environmental pollution[25]. Substantial knowledge gaps about the presence of nanoplastics in the environment, however, still exist, particularly because analytical tools to detect these particles in environmental matrices are still in the development phase[25,26]. Concerns have been expressed by the scientific community with regards to the environmental and human health hazards of nanoplastics[26,27]. These hazards are related to the physical *and* chemical characteristics of nanoplastics and are outlined below.

Hazardous chemicals
Environmental contaminants are known to be absorbed into the surface of plastic (references in[2]). Nanoplastics have a large surface-volume ratio and the resulting high surface area allows for greater chemical reactivity and sorption[26]. Organisms can thus be exposed to hazardous chemicals after the ingestion or inhalation of plastic particles.

Moreover, plastics contain a complex mixture of chemicals including additives (e.g., plasticizers, flame retardants), starting materials and side products from processing. When plastics are ingested or inhaled, these chemicals can migrate from the plastic into the exposed organism. A continuously growing body of scientific evidence has demonstrated that plastic chemicals can result in a wide range of adverse health effects[28,29]. The role of (engineered and secondary) nanoplastics in mediating chemical effects has, however, been insufficiently investigated.

Bioavailability and translocation
Experimental studies with nanoplastics have demonstrated that these particles are taken up via inhalation or ingestion by different organisms, for example rats[30,31] and scallop[32]. Uptake in plants via their roots has been demonstrated for plastic particles in the micrometer, submicron and nanometer-size ranges[33–35]. Animal studies have shown that plastics can pass barriers present in the gut[31], lung[36], placenta[37] and brain[38], a process referred to as 'translocation'. Translocation of nanoplastics and small microplastics (< 10 micrometer) across important human barriers such as the gut[39,40], lung[40], placenta[41] and brain (personal communication R. Westerink) has also been demonstrated in human cell models. It has moreover been shown that smaller particles more easily translocate than larger particles[32,37,42]. Consequently, nanoplastics have a higher potential to penetrate tissues and reach organs compared to larger plastic particles[31,43,44].

Particle toxicity
While a material may be chemically inert, the particle itself may exert toxicity. An example of this is black carbon, where exposure to the particle has been linked to the development of lung diseases including cancer[45]. Particle toxicity has also been demonstrated for small plastic particles and includes immune responses, inflammation, DNA damage (see references in[46,47]) cellular damage[46,48] and behavioural changes[38,49] among others. In humans, exposure to micro- and nanoplastics can induce oxidative stress and an increased vulnerability to develop neuronal disorders[50]. Moreover, interstitial lung disease has been demonstrated for workers processing nylon and other synthetic fibres, indicating a link between plastic fibrous dust inhalation and respiratory problems[51–53]. Lastly, plastic is a constituent of airborne particulate matter[47,54] and air pollution is estimated to cause 4.2 million deaths annually[55]. This, along with understanding of the particle toxicity of plastics has led to the hypothesis that plastic particles may in part be responsible for these deaths[56].

Many features of plastics such as size, shape and chemical make-up ultimately determine the extent of particle toxicity. It has, for example, been suggested that toxicity increases with decreasing particle size[44,57]. However, what features of plastic contribute most to particle toxicity remains to be further investigated.

Biodegradable polymers

A relatively new group of polymers are biodegradable polymers. Biodegradable plastics are designed for conversion into CO_2, methane, biomass and mineral salts by the action of microorganisms (a process called mineralization) under specified conditions. The speed and degree of biodegradation in the environment, however, is largely dependent on the prevailing conditions (e.g., temperature, humidity)[58] and polymer fragments may therefore remain in certain environments over long time scales. Biodegradable plastics can be made from renewable feedstocks (bio-based) or fossil fuels[59], and should not be confused with bio-based plastics or compostable plastics. Bio-based plastics are derived from biological raw materials and can, but do not necessarily have biodegradable properties. Compostable polymers biodegrade according to defined standards yet require very specific conditions present in industrial composting facilities. Though developed as an "environmentally-friendly" alternative to conventional plastics, various questions regarding the persistency and toxicity of biodegradable polymers remain.

Standards for assessing the biodegradability of plastics
Various standardized laboratory tests have been developed to assess the biodegradation of plastics in different environmental compartments, for example in water[60], aquatic sediments[61] or soil[62]. Depending on the plastic application, different tests have been approved by ECHA. For engineered microplastics, a tiered approach is used, distinguishing between a 'screening tier' and a 'higher tier' assessment[8]. The latter is only required when a plastic fails to meet any biodegradability criteria of the screening tier. Biodegradability criteria differ between tests, yet all require partial mineralization within a specified time frame, and most of these tests are conducted at average temperatures of at least 20 °C and in oxygen rich conditions. While these experimental conditions may be useful for determining the maximum degree of biodegradability, they poorly reflect relevant environmental conditions such as colder climates and low oxygen availability. In those conditions, microbial activity may be much lower and hence biodegradation rates are also lower[58,63]. Consequently, these plastics can still be persistent in the environment. In addition, any standardized lab test will always present an oversimplification of real-world conditions and many variables affecting biodegradation rates such as nutrient availability or weather conditions are not included in the approved tests. While it is applaudable that standards are available, the current standards are not rigorous enough and it has been argued that degradation under actual field conditions should be studied[58,64]. By comparing results from field studies to the standardized test results, the standard can be calibrated.

Toxicity of biodegradable polymers
Apart from limited understanding of the real-world biodegradation of these materials, concerns also exist with regard to the presence of hazardous chemicals. Chemicals used in biodegradable plastics can have similar toxicity to conventional plastics[65,66], showing that "bio-based and biodegradable material, despite being marketed as better alternatives, is not necessarily safer than conventional plastics"[66]. Chemicals and micro-sized particles of biodegradable materials have been shown to adversely affect marine[67] and freshwater organisms[68], crop growth[69] and soil quality[70] as well as bacteria[71], among others.

Precautionary principle

With their restriction proposal, ECHA aims to address the environmental and human health risks posed by microplastics[8]. By including only solid, non-biodegradable plastics between 5 mm and 100 nm we argue that ECHA is overlooking other potential sources of plastic pollution. As outlined above, the presence of WSPs in the environment is expected based on their production volumes and high potential for environmental discharge. The understanding of their distribution, concentrations, and impact is unfortunately still highly uncertain, in part due to the lack of suitable analytical methods to investigate them. Moreover, little is known about the transformation products of many WSPs, and their persistency and toxicity. With regards to liquid and semi-solid polymers, the example of demethicone illustrates that these polymers can be of environmental concern as well. Regarding engineered nanoplastics, such as those applied in personal care products, these can cross biological barriers and exert toxic effects. Moreover, little is known about the environmental behaviour (e.g., stability), fate and health risks of the new generation of nanoplastic materials (plastics with nano-scale additives that give the material extra properties), a currently booming industry. ECHA justifies the lower limit of 100 nm by arguing that a lower size limit (of 1 nm) cannot be enforced. However, an interdisciplinary group of scientists recently argued that this claim is invalid and that "intentionally added plastic particles in the nano-range (<100 nm) could be reintroduced into the restriction proposal"[72]. With respect to biodegradable polymers, real-world conditions are poorly reflected in current standardized biodegradation tests and various concerns about their toxicity exist.

While not all synthetic polymers within the discussed groups may be harmful to environmental and human health, too many knowledge gaps currently exist to determine which ones pose a risk, and which ones do not. Future research and hazard assessments will hopefully provide new insights and identify sub-groups requiring strong regulation. Until that time, we call upon policymakers to adopt the precautionary principle for all synthetic polymers and encourage them to consider the hazards these materials pose when developing new regulatory measures. In the case of ECHA's restriction proposal, the exemption of above-mentioned groups is particularly unjustified for applications for which ample non-synthetic polymer alternatives already exist (e.g., personal care products). Additionally, regulations can be introduced that will require more short-term and long-term hazard toxicity testing before authorizing synthetic polymers to be placed on the market at certain volumes. One avenue for this would be to include the registration of polymers under REACH[72,73]. Moreover, we call upon industry to move away from these potentially "regrettable substitutions" that are currently not covered by planned regulations. We furthermore encourage industry to seek materials and substances for which safety has been established. Lastly, we call upon the scientific community to focus their efforts on filling the knowledge gaps presented here.

Signatories

The signatories below have signed in their personal capacity as independent scientific experts and are not representing the views or endorsement by their home institutions.

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This statement continues to be open for signatories. This document will be updated with additional signatures at a later date.

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