National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Microplastics in soil systems, from source to path to protection goals

State of knowledge on microplastics in soil

RIVM report 2021-0224

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Synopsis

Microplastics in soil systems, from source to path to protection goals
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Microplastics are small plastic particles less than five millimetres across. More and more microplastics are ending up in the environment. They are everywhere: not just in water and air but also in soil, including in urban areas, on farms and in nature reserves. There are indications that microplastics pose a risk to the health of humans, plants, animals and soil life. An increasing number of publications on microplastics in the soil have featured in the scientific literature recently. And yet there is still a lack of clarity on whether or not there are potential risks.

This is why the National Institute for Public Health and the Environment (RIVM) deems further clarification important. Consequently, it has summarised the available knowledge for policymakers and risk assessors. This information proves to be fragmentary, incomplete and sometimes even contradictory, as a result of which it cannot (or cannot yet) be used to arrive at a reliable risk assessment. For example, there are no measurements available of microplastics in Dutch soils. This is largely down to the fact that no reliable, practical, standard techniques exist to analyse microplastics in the soil. These need to be developed.

More knowledge will be required to reliably gauge the environmental risks presented by microplastics. RIVM recommends better charting of the sources from which microplastics are released and how they are dispersed in the environment. In addition, a greater degree of insight is needed into the rate at which various types of plastic break down as well as the rate at which microplastics break down into harmless compounds. This is necessary for the purposes of gauging exposure. To support efforts to this end, the RIVM has created a 'conceptual model'.

Another recommendation is to simultaneously assess the risks posed to the soil by various types of microplastic. The substance risk assessment frameworks currently in place are unsuitable for this as microplastics occur in various forms and compositions. Finally, a list has been drawn up of what is known about measures geared towards reducing the release of microplastics. Examples include a ban on free plastic bags and deposits on PET bottles.

Keywords: microplastic, biodegradation, ecological risks, human risks, review, conceptual model, mitigation, framework.

Publiekssamenvatting

Microplastics in de bodem: van bron, via pad naar beschermdoelen
Kennisoverzicht van microplastics in de bodem

Microplastics zijn kleine kunststof deeltjes; kleiner dan vijf millimeter. Er komen steeds meer microplastics in het milieu terecht. Ze zitten overal: niet alleen in water en lucht, maar ook in de bodem, en dus ook in de bodem van stad-, landbouw- en natuurgebieden. Er zijn aanwijzingen dat microplastics schadelijk kunnen zijn voor de gezondheid van mensen, planten, dieren en bodemorganismen. De laatste tijd verschijnen er steeds meer publicaties over microplastics in de bodem in de wetenschappelijke literatuur. Toch is het nog onbekend wat de mogelijke risico's zijn.

Het RIVM vindt het belangrijk dat er meer duidelijkheid komt. Het heeft daarom voor beleidsmakers en risicobeoordelaars de beschikbare kennis samengevat. Deze blijkt gefragmenteerd en niet volledig te zijn en bevindingen zijn soms zelfs tegenstrijdig. De informatie is daardoor (nog) niet te gebruiken voor een betrouwbare risicobeoordeling. Er zijn bijvoorbeeld nog geen metingen beschikbaar van microplastics in Nederlandse bodems. Dit komt vooral omdat er geen betrouwbare, praktische (standaard)technieken bestaan om microplastics in de bodem te analyseren. Deze moeten verder ontwikkeld worden.

Meer kennis is nodig om risico's van microplastics voor het milieu betrouwbaar in te schatten. Het RIVM beveelt aan beter in kaart te brengen door welke bronnen microplastics worden uitgestoten en hoe ze zich in het milieu verspreiden. Verder is meer inzicht nodig in de snelheid waarin verschillende soorten plastic gefragmenteerd worden en als microplastic afgebroken worden tot onschadelijke verbindingen. Dat is nodig om de blootstelling te kunnen schatten. Als ondersteuning hiervoor heeft het RIVM een 'conceptueel model' gemaakt.

Een andere aanbeveling is om de risico's van verschillende soorten microplastics voor de bodem tegelijk te beoordelen. De bestaande kaders voor risicobeoordelingen van stoffen zijn er niet voor geschikt, omdat microplastics verschillende vormen en samenstellingen hebben. Tot slot is op een rij gezet wat bekend is over maatregelen om de uitstoot te verminderen. Voorbeelden zijn een verbod op gratis plastic tassen en statiegeld op petflessen.

Kernwoorden: microplastic, biodegradatie, ecologische risico's, humane risico's, review, conceptueel model, mitigatie, raamwerk

Contents

Summary

Microplastics research is mainly focused on freshwater and marine environmental compartments, but soil systems are expected to receive a far larger amount of these particles annually. For this reason, it is crucial to increase our understanding of the sources, the release and emission pathways, and the further fate and transport of microplastics in soil systems (see figure 1). Knowledge on all of these topics is required to assess the potential risks on human and environmental health

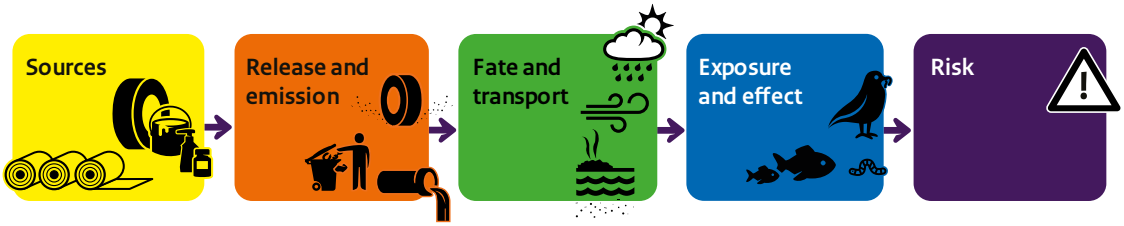

Figure 1 Diagram showing the relevant topics to address in order to assess risk of microplastics and other substances and materials. This study mainly addresses knowledge leading up to assessing exposure, thus not focusing on the available knowledge about effects or risk.

In this report, we summarize the current knowledge of microplastics with respect to sources, release and emission, and fate and transport, with specific attention given to (bio)degradation and potential mitigation options. These aspects are discussed using the main issues highlighted in 45 recent review studies, most published in 2020 and 2021, and the requirements for any future risk assessments of microplastics. Microplastics are defined here as solid polymers with a size smaller than 5 mm.

A major issue hampering current research on microplastics in soil is the large range in the chemical, morphological, physical and biological characteristics of polymers that are considered microplastics, the analytical challenges to separate particles from the complex solid soil matrix, and the detection and quantification of different microplastic types. Nevertheless, several measurement methods have been developed. It is likely that a combination of some contrasting methods would provide the most reliable data. It is essential, for this research field and for the implementation of policies, to work towards a harmonized protocol, describing which (set of) method(s) are best to use for which purpose, taking into account required quality standards.

Once emitted to terrestrial ecosystems, microplastics accumulate in soils and can migrate to marine and fresh waters through runoff, erosion and atmospheric transport, and to the groundwater and deep soil through, for example, leaching, bioturbation and farming activities. In this dynamic environment, microplastics' physical, chemical and biological properties can change through different processes, such as fragmentation, (bio)degradation and other interactions with the soil matrix.
Some quantitative estimates of the release of microplastics to soils are available, but these do not cover all major sources and data is only

available for a few regions. This means an overview of emissions to soil systems in the EU is missing and data for the Netherlands are virtually absent.

Modelling will play an important role in coping with the complexity of microplastics due to the range of MP properties (e.g. polymer type, size, shape) and the limitations of measurement methods. Several modelling approaches are already available for estimating the emission of microplastics to the environment from different sources. Modelling approaches are readily applicable to microplastics, but data on source volumes and release rates from different events (e.g. painting, driving and weathering of plastics) are lacking. After estimating the emission of microplastics to the soil system, exposure to organisms is often assessed using fate and transport modelling approaches. Several models are available that include fate in soil and are at least partially applicable to microplastics. However, there are several key issues that need further attention since most of these models were designed for much smaller sized nanomaterials. Input data such as fragmentation and degradation rates are often also highly uncertain.

The degradation and fragmentation of microplastics is important for estimating the fate of microplastics in soil systems. On the one hand, it means a reduction of microplastics due to mineralization or a complete breakdown to harmless compounds. On the other hand, degradation can occur partially, resulting in the fragmentation of microplastics to smaller particles, e.g. nanoplastics. Fragmentation is also thought to be one of the most important processes resulting in microplastics: formation of secondary microplastics due to fragmentation of meso- and macroplastics. Complete or incomplete degradation depends on three major aspects: 1) the chemical properties (e.g. polymer type and additives) , 2) the physical properties (e.g. shape and crystallinity) of a microplastic and 3) the environmental abiotic (e.g. temperature, light, oxygen) and biotic conditions (e.g. microbial composition). The first steps have been taken to determine the biodegradation rates for different plastics. However, there are still many uncertainties with respect to determining these degradation rates reliably under different environmental conditions.

It is essential to assess the rate and degree to which microplastics degrade in order to adequately model the fate of microplastics in soil systems. There are several existing test guidelines for assessing the (bio)degradability of chemicals, e.g. by ISO or OECD. However, none of these were created with (micro)plastics in mind, which could cause some uncertainty regarding the applicability of the tests and use of the results. Nevertheless, a testing strategy has been proposed by the European Chemicals Agency Risk Assessment Committee (RAC) for the restriction of intentionally added microplastics in order to determine the biodegradability of microplastics. In the restriction, biodegradable microplastics are derogated. The proposed strategy consists of screening level tests and more advanced tests in soil, water and sediment in order to test under environmentally realistic conditions. The restriction has yet to be adopted by the European Commission. Within other regulatory domains, e.g. related to fertilizers, mulch films or composting, standardized test guidelines have also been proposed and/or have been

recently developed. While available data from biodegradability studies often does not fit within fate models, data generated by standardized test methods is not yet available. Consequently, further investment in research into the biodegradation of microplastics is required.

Since the persistence of microplastics and their large-scale emission to soil are evident, and as there are indications of potential microplastic risks, it is clear that action should be taken to mitigate these emissions. Preferably, these actions should aim to reduce emissions based on source-oriented or end-of-pipe solutions. As a last resort, soil remediation type solutions can be applied to reduce microplastic exposure. However, there are many gaps in the state of knowledge and information needed to fully assess the behaviour, fate and effects of microplastics in soil, making it unfeasible to derive risk-based soil quality criteria for microplastics in this study. It is advised that, in parallel to work on mitigation options, our understanding on several broad knowledge gaps should be further developed, such as:

- Data on sources and emissions of microplastics to soil is still scattered, meaning there is a lack of data on microplastic emissions, including fragmentation of macroplastics, to the environment.
- Consistent data on microplastic concentrations and particle abundance in different soil types and land uses are absent and are needed to better understand the current extent of microplastic pollution in Dutch soils.
- Robust, reliable, standardized analytical methods are not yet available. These are needed to gather and compare data on microplastic concentrations in soils, including their characteristics, such as polymer type, size and shape.
- The knowledge about the exposure and potential effects of microplastics, additives and other adsorbed compounds in soil systems is very scattered and largely unknown.
- There is a lack of refined understanding of the link between microplastic properties and soil fate, exposure and uptake in plants and organisms. Insight is missing on the interaction of microplastics with the soil matrix. This should inform safe-by-design approaches.
- The (bio)degradation mechanisms and degradation rates of microplastics are largely unclear, also due to the lack of validated, standardized (bio)degradation test methods. Research conducted to understand biodegradation mechanisms and the development and use of standardized tests should result in improved quantification of degradation rates for fate modelling.
- The long-term implications of microplastic persistence and mobility in soil systems (worldwide spread/detection) are as yet unknown.

In conclusion, there is a need for:

1. Filling all of the above-mentioned knowledge gaps according to smart research approaches, e.g. using read-across and modelling.
2. Quantification of sources and emissions to soil is needed for the prioritization of mitigation options and the assessment of exposure as part of risk assessment.

3. Exploring possibilities for a policy and management framework for (micro)plastics in soil systems (or more broadly) in order to minimize the emission of microplastics where possible and to mitigate ecological and human health risks where needed.

The environmental issue of microplastics in soils is complex and much more research is needed to reduce uncertainties and build up scientific evidence. It is, however, certain that microplastics (and nanoplastics) are present ubiquitously in urban, natural and agricultural soils worldwide and that, due to their persistence, spreading, the continuous emissions, and society's dependency on plastic, microplastics will be present in the environment for a long time. Steps can be taken to mitigate and reduce microplastic emissions into the environment, adapted to the latest knowledge developments, to reduce the growing amount of pollution while improving our overview and understanding of the problem.

1 Introduction

The Dutch Ministry of Infrastructure and Water Management (IenW) wants to develop adequate policies to counteract current and future problems for human health and ecosystems. In line with the European Circular Action Plan and Plastic Strategy, microplastics are one of the prioritized environmental issues that needs to be addressed and goals are being set to minimize and mitigate emissions to the environment.[1] The soil system used as a sink for microplastic accumulation was acknowledged in the recently published EU soil Strategy for 2030.[2]

The accumulation of microplastics in soil and water systems has been considered an environmental concern from different perspectives (Waaijers-van der Loop et al., 2022). For instance, the release of microplastics:

1. can create a hazard for human health and ecosystems in the long term (the risk-based perspective), as is described in many reviews on microplastics; or
2. should be avoided at all reasonable costs because of violating the intrinsic value of our environment (the zero-pollution based perspective); or
3. is considered as material losses in a circular economy.[3]

The optimal mitigation measures and policy actions to deal with microplastic emissions can be different, depending on the perspective. In this report, we focus on microplastics in soil systems and aim to provide an overview of the available knowledge. This knowledge can serve as a basis to establish mitigation options from any perspective.

The state of knowledge will reflect the current understanding of microplastic sources, release and emission pathways, transport, behaviour, and fate in the environment. This structure fits the commonly used source-path-receptor diagrams, which can be used to assess the potential risk of microplastics (Figure 1). Most research so far has been on (micro)plastic litter in the water compartment, ephemerally labelled the 'plastic soup' in oceans (Hammer et al., 2012; Andrady, 2011; McFedries, 2012). Research was subsequently expanded to cover other compartments, such as air, soils, groundwater, surface waters and sediments (viz. Verschoor et al., 2014; Verschoor, 2015; Horton et al., 2017; Xu et al., 2020a; Xu et al., 2020b; Quik et al., 2021). In this report, we look at the state of knowledge about microplastics in soils, since it is still limited.

We will follow the same order of topics as in the source-path-receptor diagrams to structure this review, with the aim of identifying specific information gaps. The focus will be on the source-path, with a brief

[1] EU plastics strategy. ec.europa.eu/environment/strategy/plastics-strategy_en

2 EU Soil strategy for 2030, COM(2021) 699. https://ec.europa.eu/environment/publications/eu-soil-strategy-2030_en

[3] IenW (2018) Kamerbrief, betreft Gezamenlijke aanpak plastic zwerfafval Kenmerk IENW/BSK-2018/232541 Datum 6 november 2018, Den Haag

reflection on receptor (effects). We also use this approach for the construction of a more detailed conceptual model in Chapter 6.

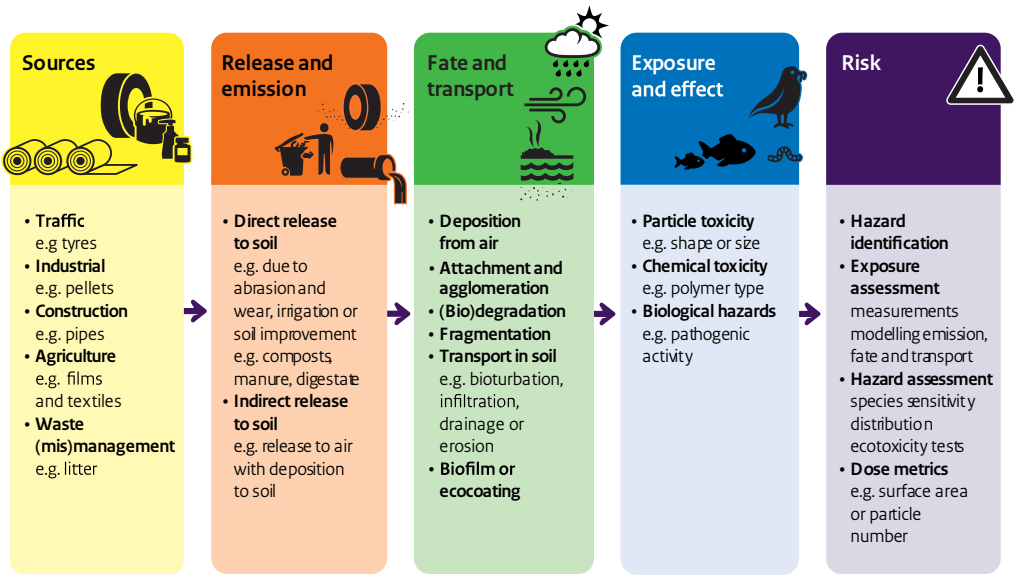

Figure 1 Research topics for microplastics in soil systems classified in a source-path-effect-risk model approach. Explanation to a few terms: ***Dose metrics*** *is the effective description of the dose to which organisms are exposed.* ***Eco coating*** *is the attachment of organic macromolecules and biofilms onto the particle surface.* ***Natural attenuation*** *is a variety of physical, chemical, or biological processes as part of fate and transport processes that reduce the mass, toxicity, mobility, volume, or concentration of contaminants*

1.1 Classification of microplastics

Plastics can be made of different types of polymers (e.g. polypropene, polyethylene, polystyrene, polyvinylchloride) and can be divided based on their size into macro-, meso-, micro-, and nanoplastic. In the last decade, several slightly differing definitions for microplastics have been proposed (Verschoor, 2015). In this report, the recent definition used in the context of the proposed restriction on intentionally added microplastics is considered to be most relevant. The European Chemicals Agency (ECHA) Risk Assessment Committee (RAC), in its opinion on the restriction, defined microplastics as 'particles containing solid polymer to which additives or other substances may have been added, and in which ≥ 1% w/w of particles have (i) all dimensions ≤ 5 mm, or (ii) a length of ≤ 15 mm and a length to diameter ratio of >3 (ECHA, 2020). The definition does not discriminate on the type of polymer used and sets no lower size limit. While the upper size limit for microplastic is widely accepted, the lower size limit is still under debate (Möller et al., 2020). This is also the case for the definition proposed in the restriction; ECHA initially proposed 100 nm as the lower size limit and, while RAC recommended setting no lower size limit in its opinion, the sister committee for Socio-economic Analysis (SEAC) recommended defining a lower limit size of 1 nm, and a temporary limit of 100 nm in order to ensure the enforceability of the restriction (ECHA 2020).

1.2 Objectives and scope

One objective of this report is to identity and summarize the current knowledge on microplastics in the soil system with respect to sources, types, emissions, fate, accumulation, attenuation (including degradation) and potential mitigation options. This information can be used in current and future research to start estimating the potential risks of microplastics in soils. Also, the information and quantitative data can be used in modelling to better understand the (relative) extent of microplastic sources and emission to soils.

Consequently, the objectives and scope for this report are:

1. To summarize sources and emissions of microplastics in the soil system for major microplastic types.
2. To compile the current distribution and dispersal of the microplastic types in soil systems by different mechanisms of fate, such as transport routes and degradation mechanisms.
3. To describe gaps in our knowledge and available information that hamper a reliable analysis and assessment of the risk of microplastics in soil systems.
4. To collect ideas to mitigate the effects of microplastics in soil systems through source or effect-oriented solutions and current criteria for (micro)plastic-contaminated soil.

1.3 Current state of publications

Over the last 20 years, the number of publications on the issue of plastics in the aquatic environment has steadily increased and reached more than 5,000 publications per year in 2020 (data not shown). For soil, the number is considerably smaller (Figure 2). In contrast, the environmental topic ‘microplastic’ (including the hyphenated expression) has received significant interest only in the last 7 years, which demonstrates a rapid exponential increase. He et al. (2020) noticed the same trends. With respect to the environmental compartment of interest, the yearly number of publications focused on water systems outcompeted the yearly number on the soil system (Figure 2). Having said that, in 2021 there was a considerable flow of publications focused on microplastics in soil systems, i.e. more than one per day.

At the moment, nanoplastic research is also rapidly increasing and is obviously linked to the broader environmental issue of ‘nanomaterials’ and ‘nanotechnology’ (data not shown).

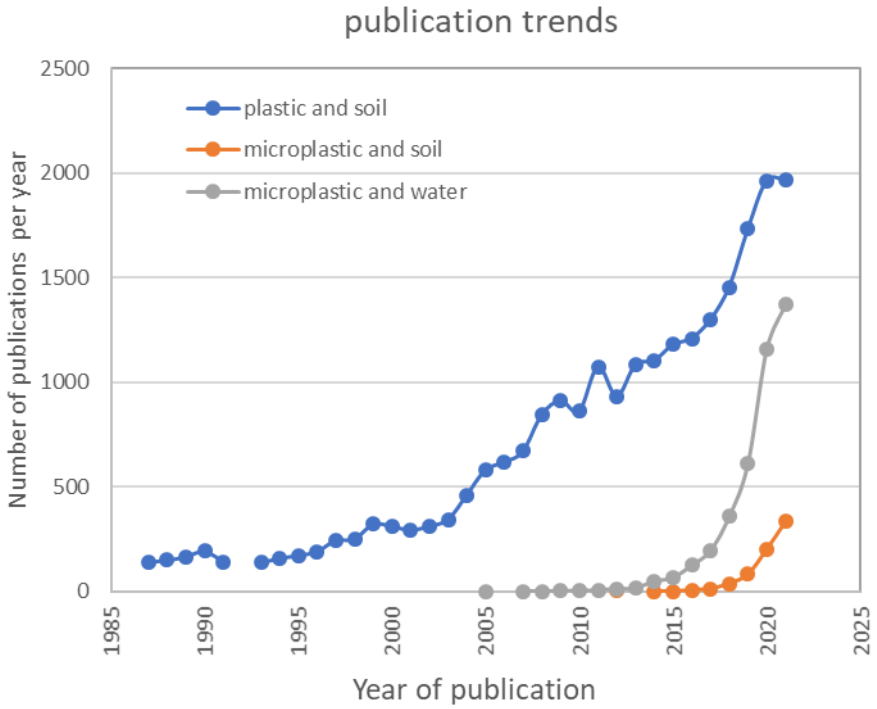

Figure 2 Number of scientific publications per year showing the publication trends (Scopus 25 October 2021): 'Microplastic and soil' (orange), 'microplastic and water' (grey) and 'plastic and soil' (blue) in the title, keywords and abstract

An interesting observation was the large number of literature reviews (45 reviews in our search) that were published in 2020 and 2021, covering the issue of microplastics in the soil system. Of the 45 publications that were found with 'microplastic' and 'soil' in the title and 'review' in the field 'title/keyword/abstract', five reviews have already received more than 50 quotes (Scopus 8 August 2021). Considering that these reviews were produced in parallel, it is inevitable that repeated information on the issue is presented by the different publications. About 85% of all publications on microplastics in soil systems is produced in only two regions: 65% in Asia and 20% in Europe (Xu et al., 2020b, He et al., 2020).

Zhang et al. (2021b) provided an overview of the locations and type of research in which microplastics in soil were studied (Figure 3). The map shows that Asia and Europe are the dominant regions for research of microplastics in soils. Arable systems are represented most (Pie diagram in Figure 3).

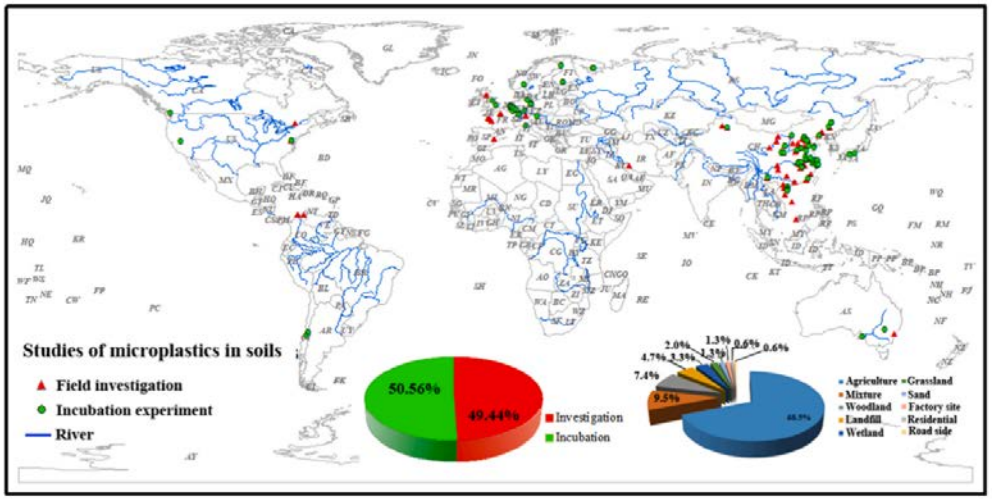

Figure 3 Studies on microplastics in the soil under controlled conditions in laboratories (green part of the pie) and from field samplings (red part of the pie). The majority of the publications dealt with arable soils (blue part of the pie). Figure reproduced from Zhang et al. (2021b).

Recently, 45 reviews dealing with microplastics in soil were produced. It was considered not effective to comprehensively review the available literature again in order to collect the current state-of-the-art. Instead, we provide a general overview of the state of knowledge, based primarily on the published reviews and focused on the interpretation of the results and some subsequent steps needed for risk assessment. When needed, the original publications were consulted. This was further supplemented with information from interviews of experts working on microplastics research.

In this report, we have viewed the current knowledge from the perspective of how it can be used to improve our understanding of microplastics and their behaviour in the environment. We collected generalized information on emissions, transport, and the behaviour of microplastics in soil. The focus of the latter was specifically on natural attenuation, and specifically the (bio)degradation of microplastics in soil (Figure 1), as these processes are considered critical to conducting a meaningful fate analysis of microplastic accumulation in soil. We reflect on the state of this knowledge and on what this may mean for further research. Finally, mitigation options aimed at emission reduction and pathways to risk or quality-based criteria for soils are discussed.

1.4 Readers guide and highlights

Several of the published reviews provided key messages on the topic of microplastics in soil systems (viz: Li et al., 2020a; Kumar et al., 2021; Horton et al., 2017; Dioses-Salinas et al., 2020; Hurley and Nizzetto, 2018; Sun et al., 2022; Wang et al., 2019; Wang et al., 2020; Wang et al., 2021a; Wang et al., 2021b; Xu et al., 2020b; Yang et al., 2021; Zhang et al., 2020; Zhang et al., 2021a; Zhang et al., 2021b; Zhou et al., 2020; Zhu et al., 2019; Rillig and Lehmann 2020). To assist in reading this report, we used already published key messages and supplemented them with the key findings from our research to yield ten

highlights. These highlights provide a reading guide, as they present a synopsis of each new topic discussed in Chapters 2 to 8. Each new topic starts with a highlight (in bold font), followed by a detailed analysis of the specific topic, including the knowledge gaps. After discussing the results in the first chapters, we reflect on these findings and give an outlook and perspectives for action in Chapters 6 and 7. The complete set of highlights is given below:

1. The past decades, microplastics in soil systems have been largely overlooked. Nowadays, microplastics are detected in all environmental compartments worldwide: in air, marine waters, fresh waters, sediments and terrestrial systems. Considering release and migration to agricultural, urban and industrial soils and the analytical data, the yearly global release of microplastics to soils is likely much higher than that to rivers and oceans.
2. Microplastics are a very diverse group of synthetic particles, consisting of different polymer types and additives, and having different sizes, densities, shapes and colours. Microplastics can also acquire biological characteristics, caused by the attachment of microorganisms, including pathogens, on the (weathered) particles.
3. The absence of a suitable method for analysis of the type and quantity of microplastics in soil is a big hindrance to research. Although a number of analytical methods have been developed, an efficient, robust, fast and low-cost analytical method for soils is still not available, especially for the isolation of the polymers and particles from the complex, organic matter containing a solid soil matrix (sample clean-up). Data on the abundance and distribution of microplastics in soils are still limited, and results obtained from studies differ significantly.
4. In a variety of studies, microplastic emission sources are identified, such as tyres, agricultural plastic films, textile, seed coatings, personal care products, cosmetics, paints (e.g. on roads and buildings), artificial turfs, landfill, litter, packaging and construction materials. Both primary and secondary microplastics are released to the environment, but secondary microplastics are the dominant source.
5. Once emitted to terrestrial ecosystems, microplastics can migrate to marine and fresh waters through runoff, with erosion and by atmospheric transport, and to the groundwater and deep soil through leaching, bioturbation and farming activities, to name a few. After microplastics enter the soil system, microplastics' physical, chemical and biological properties change through fragmentation and (bio)degradation.
6. Polymer type, shape, and particle-size-dependent modelling of microplastics' fate and behaviour is important for coping with the complexity of microplastics in the environment. Studies for the modelling of microplastics' emissions, transport and fate in water are available, however there are not yet adequate for the investigation of the soil system.
7. Published biodegradation data for (micro)plastics differ greatly between studies. The (bio)degradation potential of microplastic particles depends on three major aspects, namely 1) their chemical composition, 2) their physical properties and 3) the environmental conditions to which they are exposed to. The

limited data on (bio)degradation and monitoring show that microplastics can accumulate in the environment, including soil systems. However, the actual accumulation rates are uncertain, not only due to the scarcity of data in combination with a large variation between measured (bio)degradation rates, but also simply due to the lack of standardized methods suitable to assess the fragmentation and degradation of microplastic particles under environmentally relevant conditions, as well as the vast range of polymer types and identities.

8. High amounts of microplastics in soils can alter the fundamental physicochemical and biological properties of the soil system. Soil health and function may be impaired and trophic transfer in food chains may also occur. This may lead to adverse effects on ecosystems. To address the potential ecological impact on soil systems, (further) research is needed on the (direct and indirect) effects of microplastics on the viability, development, fertility and mortality of soil organisms. In addition to such effect studies, improved insights into the exposure, bioavailability and uptake of microplastics by organisms and humans are necessary. The current information on the impact of microplastics on the functioning of soil organisms and the ecosystem is incomplete and is sometimes conflicting.
9. Overall, research on the emissions to and fate of microplastics in soil systems, and their effects on terrestrial ecosystems, is still limited but is increasing rapidly, demonstrated by increasing publication rates. This motivates an active monitoring of the progress of the knowledge required for risk assessment.
10. Some studies provided specific recommendations for policy development and land quality management. The need to act on the prevention and reduction of microplastic accumulation is motivated based on the persistence, the potential adverse effects and the widespread detection of microplastics. Preventing and reducing the impact of microplastic soil contamination requires a large set of measures for a large variety of stakeholders. In several countries, measures are being implemented to reduce the release (of mostly primary microplastics) at the source (source control) and to reduce emissions at end-of-pipe (emission control), both of which results in the reduction of emissions to terrestrial ecosystems.

2 Origin and types of microplastics in soil

In past decades, microplastics in soils have been largely overlooked (highlight 1). Nowadays, microplastics are found in all environmental compartments worldwide: in air, marine waters, fresh waters, sediments and terrestrial systems. Considering the release and migration to agricultural, urban and industrial soils and the analytical data, the yearly global release of microplastics to soils is likely much higher than the release to oceans and rivers.

2.1 Definitions and types

Microplastics are a very diverse group of synthetic particles, consisting of different polymer types and additives, and having different sizes, densities, shapes and colours (highlight 2). Microplastics can also acquire biological characteristics, caused by the attachment of microorganisms, including pathogens, on the (weathered) particles.

'Plastic' is defined as being synthesized from solid materials that can be moulded and that contain a considerable number of carbon-based polymers. Reversible moulding refers to rubber as a specific plastic material. One of the more recent definitions of 'microplastic' is: 'particles containing solid polymer, to which additives or other substances may have been added, and in which ≥ 1% w/w of particles have (i) all dimensions 0.1µm ≤ x ≤ 5 mm, or (ii) a length of 0.3µm ≤ x ≤ 15mm and a length to diameter ratio of >3 (ECHA, 2020).[4]

Referring to this definition, microplastics are:

- synthetic materials containing polymers with carbon in their structure;
- solid at ambient temperature;
- smaller than 5 mm;
- insoluble in water; and
- relatively inert, persistent, poorly or non(bio)degradable (this varies among the types of the polymers).

Microplastics are universally classified as primary and secondary substances (e.g. Fu and Wang 2019):

- Primary microplastics are particles that are synthesized intentionally at defined small sizes for commercial applications. Microbeads from cosmetics and personal care products, seed coatings, and pre-production pellets used as intermediates in plastic production are all significant sources for primary microplastics worldwide.
- Secondary microplastics are fragmentation products of larger plastic debris, which are broken down by UV-radiation and mechanical abrasion (Wagner et al., 2014). The sources of secondary microplastics are composed of fishing nets, films,

[4] Note that the definition is proposed by RAC (Risk Assessment Committees) with regard to the restriction on intentionally used microplastics. SEAC (Committee for Socio-Economic Analysis) has proposed a different definition (no lower limit size for the particles). The final definition has not been chosen yet by the European Commission.

household items, tyres, textile, paints, littering, industrial raw materials and other discarded plastic debris (e.g. Eerkes-Medrano et al., 2015; Verschoor and de Valk, 2018).

The most widely used plastics according to Shah et al. (2008) consist of one or more of the following polymers: polyethylene (PE: LDPE, MDPE, HDPE and LLDPE), polypropylene (PP), polystyrene (PS: PS and EPS), polyvinyl chloride (PVC), polyurethane (PUR), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), and poly amide/nylons (PA). Examples of biodegradable plastics are: polylactide (PLA), poly-3-hydroxybutyrate (PHB), polypropiolactone (PPL), poly-ε-caprolactone (PCL), polyethylene succinate (PES), polybutylene succinate(PBS), poly-3-hydroxybutyrate-co-3-hydroxyvalerate (PHBV) and polyester carbonate (PEC).

Zhang et al. (2021b) reviewed 170 publications on microplastics and soils extracted from the Web of Science covering the period 1950–2020. They found that these studies were mainly carried out in Asia (60%) and Europe (23%), and most focused on arable soils (69%) (Figure 3). The microplastics most frequently found in soils were: PE (79% of the studies), PP (79%), and PS (46%), with particle sizes ranging between 20 – 5000 µm. Of the soil samples taken, 64% contained microplastics: 1,000 – 4,000 particles per kg soil. This does not imply that only these polymer types where present, as there may be bias or a limitation in sampling, extraction and/or detection methods (see Annex 3).

2.2 Polymers identified in soils

The widespread use of plastics in all possible applications has resulted in a wide dispersion of polymers in ecosystems around the globe, especially in inhabited areas. Since these polymers have different sources, identities and behaviour, the composition of the accumulated material varies strongly. For instance, agricultural use of land occupies the largest share of managed soil systems worldwide. Consequently, microplastics associated with agriculture management also demonstrate the highest appearance level in managed soil systems worldwide, i.e. the classic plastics such as PA, PP, PE, PS, PET and PVC. It should be noted that some other plastics did not appear very often, such as fluoropolymers and natural or synthetic rubbers. Obviously, rubber from abrasion of tyres is found or estimated to locally have high concentrations along roads (Baensch-Baltruschat et al., 2021; Sieber et al., 2020).

2.2.1 Chemical identity

Figure 4 illustrates estimates of relative abundance of polymer types for a microplastics mass flow modelling study in Switzerland for seven common polymer types (Kawecki and Nowack, 2019). Soil microplastic measurements indicate globally that PA (Polyamide nylon), PP, PE and PS accounted for most polymers found while PVC and PET were found to a lesser extent (Xu et al., 2020b). For example, PA and PP were two major polymer types in soils from the farmland of Wuhan, with percentages of 33% and 29% (from Chen et al., 2020). PP, polycarbonate and acrylonitrile butadiene styrene were confirmed as major microplastics in soils from Guiyu, an E-waste dismantling zone in China (Chai et al., 2020). PP and PE represented 51% and 43% in shallow and deep soils in

the farmland of Shanghai, respectively (Liu et al., 2018). Likewise, PE covered 63% of microplastic polymers in agricultural farmland in southeast Germany, while PP and PS accounted for 25% and 13%, respectively (Piehl et al., 2018). This was consistent with soils from rice-fish coculture stations in Shanghai. PE was predominantly present (61%) followed by PP (36%) and PVC (3.5%) (Lv et al., 2019).

In addition to field observations, a modelling approach can be taken to identify the dominant polymer types expected in soils. Such modelling is based on Material Flow Analysis, which follows material flows through the technosphere by quantifying the material flow from the application of plastics, processes generating microplastics (e.g. sawing) up to the eventual releases of microplastics to the environment (Kawecki and Nowack, 2019).

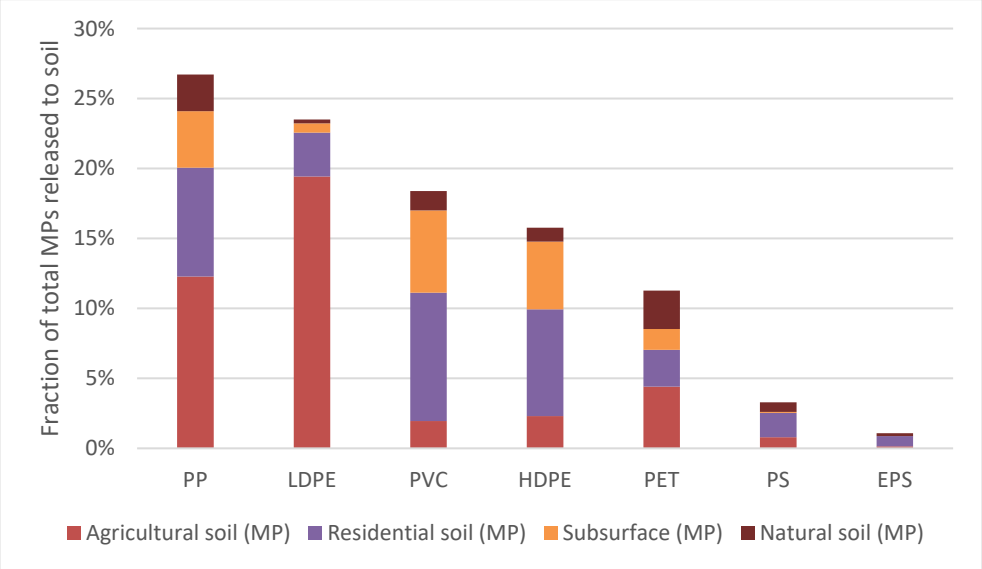

Figure 4 Relative mass flow of microplastics estimated to be released to three soil types and the soil subsurface in Switzerland, for seven common polymer types (data from Kawecki and Nowack, 2019).

In one of the first studies that models the release of microplastics to the environment, Kawecki and Nowack (2019) followed the application of seven common polymer types. This resulted in estimates of microplastic releases divided over four different soil compartments: agricultural soil, residential soil, natural soil and the soil subsurface[5] based on the (micro) plastic application (Figure 4). The data show trends for MP release per polymer type to different soil types for Switzerland. Although this cannot literally be translated to other locations, it is likely that the largest sources and waste management processes are similar to other locations, such as those in the Netherlands, resulting in a similar division of polymer types between soil types.

It is not strange that PP, LDPE, HDPE and PVC are polymer types estimated to be most abundantly present in the environment because

[5] For instance the soil in which construction pipes are laid under a road.

these polymers are estimated to have the highest production volumes (Geyer et al., 2017).[6]

2.2.2 *Morphological characteristics – physical identity*

The physical identity is characterized by the dimensions, density, shape and colour of the microplastic particles. Overall, fibres were the dominant shape of microplastics in the majority of the soils (Xu et al., 2020b). Of the four dominant shapes in the arable soils of Shanghai, the average percentage of fibres was highest (53%), followed by fragments (38%), film (6.7%) and pellets (2.1%) in shallow soil. In deep soil, the average percentage of fibres was still highest (38%), but close to films and fragments (34% and 28% respectively), while pellets were present to a small extent (0.32%) (from Liu et al., 2018). Fibres made up 92% of soil samples from Yunnan China and fragment and film only accounted for 4.0% and 3.0%, respectively (Zhang and Liu, 2018; Lv et al., 2019).

The accumulation of microfibres may result from the application of sewage sludge and waste water from household washing processes as fertilizer. Synthetic fibres have been proposed as an indicator of sludge application (Zubris and Richards, 2005). The abundance of fibres in sewage sludge could reach 38,080 particles/kg dry weight and waste water containing clothing fibres may be discharged directly for irrigation (Zhang and Liu, 2018). 1,000–4,000 particles of microplastics per kg of dry mass was found in arable sites in European countries (Zubris and Richards, 2005). In other studies, microbeads and fibres accounted for 48% and 37% of the accumulation in the arable land of Wuhan, China (Chen et al., 2020). To a large extent, microplastics <1 mm are present: for instance sizes between 0.03−1 mm occupied 49% and 60% of shallow and deep soils in arable land of China (Liu et al., 2018). Microplastics with sizes of less than 0.2 mm represented over 70% found in soil samples from Wuhan, China, followed by 0.5−1 mm (13%), 0.2−0.5 mm (9%) and 1.0–3.0 mm (7%) (Chen et al., 2020). Ninety-five per cent of sampled plastic particles from planted soils in Yunnan were in the size of 0.05−1 mm, only 5% were larger than 1 mm (Zhang and Liu, 2018)

Most microplastic particles were reported to have different colours. Transparent and black particles were dominant in both rice-fish co-culture soils and farmland soils in Shanghai, China (Liu et al., 2018; Lv et al., 2019). Red, black, green, blue, brown and transparent particles were observed in the suburbs of Wuhan (Chen et al., 2020). White, transparent, and blue were dominant colours for the particles found in arable land from southeast Germany (Piehl et al., 2018). Besides other characteristics such as shape and dimensions, information on the colour of particles can be useful for things such as determining the source or whether ingestion by organisms as a result of active foraging is induced by certain colours.

2.2.3 *Biological characteristics*

In the case of microplastic in soil, it might be useful to define the biological characteristics as well. Microplastics consist of particles of

[6] https://ourworldindata.org/grapher/plastic-production-polymer

different sizes and shapes to which organisms, such as bacteria, and, as in the case of larger particles, (multicellular) organisms like nematodes and algae can adhere (Wang et al., 2021a). Also, microplastic particles can adhere to the roots of plants and may affect the uptake of water and nutrients (Zhang et al., 2021b). Microplastics can also be taken up by plants and accumulate in plant tissue (Li et al., 2020b; Zhang et al., 2021b). However, not much is known about this and methods to determine this characteristic are not broadly available.

2.3 Methods for the extraction, separation, identification and quantification of microplastic in soil

The absence of a suitable method for the analysis of the type and quantity of microplastics in soil is a big hindrance for research (highlight 3). Although a number of analytical methods have been developed, an efficient, robust, fast and low-cost analytical method for soils is still not available, especially for the isolation of the polymers and particles from the complex, organic-matter-containing solid soil matrix (sample clean-up). Data on the abundance and distribution of microplastics in soils are still limited and results obtained from studies differ significantly.

There is currently no standardized method for separating microplastics from a soil matrix, nor a standardized analysis and identification protocol. In Annex 3, we provide a brief overview of the methods developed and used in scientific literature for the different steps of measuring microplastics in complex environmental samples with a special focus on their applicability for the analysis of soil samples. These steps comprise sampling, extraction, separation, purification, identification and quantification (detection). Some steps have several possible methods to use. Each method has its benefits, its specificity for certain polymers, differential applicability for solid matrices and some uncertainty attached.

The biggest hurdle is the extraction and clean-up method. This is a challenging step because of the presence of various, abundant and complex organic substances in the soil organic matter fraction (Bläsing and Amelung, 2018; Li et al., 2020a; Ruggero et al., 2020; Möller et al., 2020). Often applied techniques are based on the extraction of microplastics with a salt solution through density differences (Table 1) between the microplastics, the other soil constituents and most of the organic matter. Different salt solutions can be used for different polymers. With saturated NaCl, the light polymers can be extracted, while with NaI or NaBr, for example, the whole range can be extracted. It is, however, difficult to distinguish organic compounds from microplastics by conventional salt solution density separation, irrespective of the soil type and structure (Li et al., 2020a). For samples with organic matter, such as soil and sediment samples, a digestion step is often required (Ruggero et al., 2020).

The second hurdle is to find a suitable analytical instrument for different types or purposes (reviews e.g., by Ruggero et al., 2020 and Möller et al., 2020). Depending on the scope of a study, one method may be more suitable than another. For monitoring purposes, determining mass quantities of polymers or volumes may be the best way to measure

concentrations in the field. Whereas a toxicity laboratory test may require another analytical methodology to determine elasticity and aspect-ratio (both related to potential uptake). Currently, few methods are available to provide sufficient insight into the morphology of microplastics. No one method fits all purposes.

Combinations of methods have also been tested (see Annex 3) and seem to be the way forward. However, most studies are still limited to one analytical methodology. To be able to combine and compare test results, there is a need for a harmonized or even unified protocol describing which (set of) method(s) can best be used for which purpose, with the needed quality standards.

Table 1 Densities of widely used plastics (data from Ruggero et al., 2020).

Plastic class	**Acronym**	**Density**
		(g/cm3)
Expanded Polystyrene	EPS	0.03–0.15
Acrylonitrile butadiene styrene	ABS	0.90–1.53
Polypropylene	PP	0.91–0.95
Low density polyethylene	LDPE	0.92
High density polyethylene	HDPE	0.96
Mater-Bi	MB	1.02
Nylon (Polyamide)	(Nylon) PA	1.02–1.06
Polystyrene	PS	1.05
Polymethyl methacrylate	PMMA	1.09–1.20
Polyurethane	PU	1.2
Polycarbonate	PC	1.20–1.22
Polyhydroxy butyrate	PHB	1.25
Polylactic acid	PLA	1.25
Polyvinyl chloride	PVC	1.35–1.39
Polyethylene terephthalate	PET	1.38–1.41
Polyester	PES	Unknown

3 Source, emission, behaviour and fate of microplastics in soil

3.1 Source and emission

In a variety of studies, microplastic emission sources are identified, such as tyres, agricultural plastic films, textile, seed coatings, personal care products, cosmetics, paints (e.g. on roads and buildings), artificial turf, landfill, litter, packaging and construction materials. Both primary and secondary microplastics are released to the environment, but the secondary microplastics are the dominant source (highlight 4).

3.1.1 Introduction

The distribution of microplastics in soil systems is widespread, which has drawn the attention of many scientists (e.g., Rillig and Bonkowski, 2018, reviewed by Sun et al., 2022 and Xu et al., 2020b). Microplastics can enter terrestrial systems in various ways, such as sewage sludge[7], plastic mulch from agriculture, tyre abrasion, littering, illegal waste dumping and fertilizers (Horton et al., 2017; Bläsing and Amelung, 2018; Li et al., 2019; Weithmann et al., 2018). Based on measurements of microplastic abundance in soils (see Table 2), plastic mulch and compost are considered the most significant sources for microplastic accumulation in arable soil (Xu et al., 2020a). Another, often reported source is sewage sludge (Corradini et al., 2019; Wang et al., 2019). In the Netherlands, the application of municipal sewage sludge on agriculture soil is currently restricted, but with the introduction of a new environmental act in 2022 this ban will be partly withdrawn.[8] This could lead to a future increase of emissions of microplastics to arable soil. In recent decades, the increased use of single-use grocery bags and plastic mulch has led to a predominant contamination of arable soil with polyethylene (Ramos et al., 2015). However, recent restrictions on the single use plastics has proven effective, e.g. in the EU[9].

Overall, the amount of microplastics discharged to soil is estimated to be much higher than that discharged to oceans (Horton et al., 2017; Nizzetto et al., 2016; Hurley and Nizzetto, 2018), but a consistent comparison is lacking. It was estimated that roughly 107,000–730,000 tons of microplastic entered arable lands annually in North America and Europe.

[7] In the Netherlands, WWTP sludge is mostly burned as the sludge does not meet strict quality standards.

[8] Aanvullingsregeling Bodem OMGEVINGSREGELING consolidated and updated 08-09-2021 afdeling 9.6 specifieke voorschriften omgevingsvergunning milieubelastende activiteit – het op of in de bodem brengen van zuiveringsslib. Https://iplo.nl/regelgeving/stelsel-omgevingswet/omgevingswet-in-ontwikkeling/hoofdlijnen-omgevingsregeling/

[9] Directive (EU) 2019/904 of the European Parliament and of the Council of 5 June 2019 on the reduction of the impact of certain plastic products on the environment. https://eur-lex.europa.eu/eli/dir/2019/904/oj

Table 2 Microplastics present in different soils from around the world as reported by Xu et al. 2020b.

Location	**Abundance (particles/kg)**	**Polymer identification**	**Shape**	**Size (mm)**	**Colour**	**Reference**
Farmland soil in Shanghai, China	Shallow soil: 78.0±12.9	PP (50.5%), PE (43.4%), PES (6.1%)	Shallow soil: fibres (53.3%), fragments (37.6%), films (6.7%), pellets (2.1%)	Shallow soil: 0.03-1: 48.8%	Shallow soil: black (39.4%)	(Liu et al., 2018)
	Deep soil: 62.5±12.9		Deep soil: fibres (37.6%), films (33.8%), fragments (28.3%), pellets (0.32%)	Deep soil: 0.03-1: 59.8%	Deep soil: transparent (46.3%)	
Rice-fish co-culture ecosystems, Shanghai, China	10.3±2.2	PE (61.4%), PP (35.5%), PVC (3.5%)	Fibres and microbeads predominant	0.02-5	Transparent and black dominant	(Lv et al., 2019)
Farmland around suburbs of Wuhan, China	320-12560	PA (32.5%), PP (28.8%), PS (16.9%), PE (4.2%), PVC (1.9%)	Microbeads (48%), fibres (37%), fragments (15%), foams (1%)	<0.2 (70%), 0.5-1 (13%), 0.2-0.5 (9%), 1.0-3.0 (7%), 3-5 (<1%)	red, black, green, blue, brown and transparent	(Chen et al., 2020)
Tree-planted soils in Yunnan, China	7100-42960	Not mentioned	Fibres (92.0%), fragment (4.0%), film (3.7%)	0.05-1 (95%), > 1 (5%)	Not mentioned	(Zhang and Liu, 2018)
Agricultural soil from Chilean central valley	1210±250	Not mentioned	Fibres (97.0±3.0%)	Not mentioned	Not mentioned	(Corradini et al., 2019)
Municipal solid from waste facility in Sydney, Australia	300-67500	PE, PS, PVC	Not mentioned	Not mentioned	Not mentioned	(Fuller and Gautam, 2016)
Floodplains in Swiss nature reserves	~593	PE (88%), PS, SBR, PVC	Not mentioned	0.125-0.5	Not mentioned	(Scheurer and Bigalke, 2018)
Agricultural farmland in southeast Germany	0.34±0.36	PE (62.5%), PP (25.0%), PS (12.5%)	Films (43.8%), fragments (43.8%), fibres (12.5%)	1-5	White (62.5%), transparent (18.75%), blue (12.5%), green (6.25%)	(Piehl et al., 2018)

± reflects the standard deviation

3.1.2 *Soil specific estimates*

Only recently has knowledge about the sources of microplastics in soils grown to such an extent that quantitative estimates of release related to specific sources can be made using modelling approaches (SAPEA, 2018). Two such studies using state-of-the-art Material Flow Analysis approaches are briefly described here, one focusing on seven common polymers (Kawecki and Nowack, 2019) and one on rubber from tyres (Sieber et al., 2020) for a Swiss scenario. From these studies, the main inputs seem to come from microplastics that originate from secondary sources (see Table 3), e.g. from abrasion wear of tyres, construction pipes, agricultural films and agrotextiles. But also from the sawing of construction pipes, resulting in sawdust. Furthermore, manufacturing (excluding textiles) releases plastic pellets, flakes and dust to residential soils. Microplastic present in compost is expected to be a main source for its presence in agricultural soils and gardens, but data for the Netherlands do not exist yet (see Table 3 for data from Switzerland,).

Table 3 Distribution of microplastics sources of seven commodity polymers in Switzerland (Kawecki and Nowack 2019).

Source	**Fraction**
Construction pipes	22.9%
Agricultural films	18.9%
Outdoor air	15.6%
Agrotextiles	10.7%
Non-text. manufacturing	6.6%
Compost	5.4%
Other 19 categories	19.9%

From all these sources, it is estimated that rubber released from tyres makes up more than 10 times the microplastic release estimated for the seven commodity plastics (PET, PP, LDPE, HDPE, PVC, PS and EPS) in Switzerland (Sieber et al., 2020). This figure might in reality be lower, as the fragmentation of macroplastics into microplastics is not accounted for in the study conducted by Kawecki and Nowack (2019), as well as some other sources which might still need to be quantified, such as paint. Additionally, these figures do not account for spatial distribution of these sources. For instance, tyre wear particles are thought to end up mainly in roadside soils, whereas the abrasion of agricultural films would mainly end up in arable land.

Verschoor et al. (2016) estimated microplastic emissions from abrasive cleaning agents, paints and tyre wear to different environmental compartments in the Netherlands. The authors note that the Dutch Pollutant Release and Transfer Register (Dutch: Emissieregistratie), which registers and calculates regionalized emissions of 375 pollutants in the Netherlands, allocate 40% and 90% of tyre abrasion emission to soil in urban areas and non-urban areas, respectively. For paints no distribution was provided.

3.1.3 *Overall environmental estimates*

Within the REACH restriction on intentionally added microplastics, estimates have been made regarding the release of (primary)

microplastics to the environment (Table 4). Although the releases are not specified for each receiving environmental compartment, for certain sources it is to be expected, based on literature, that some of the emitted microplastics will end up in soils. This will primarily be the case for emissions from products considered under the categories 'agriculture and horticulture', 'paints and coatings' and 'construction products'. With respect to agriculture and horticulture, based on the applications noted, it is expected that most microplastics will end up in arable soils. Several publications showed that microplastics can subsequently end up in crops, as plants are able to take up microplastics via the roots (Zhang et al. 2021b; Li et al., 2020b). For paints, RIVM estimated an average fraction of 57% of microplastics end up in soils as a result of maintenance and wear of paints in rural and urban areas (Verschoor et al., 2016). Although the estimate is made for secondary microplastics, primary microplastics (beads) incorporated in the matrix of paints will also likely end up in soils via this route, although amounts are expected to be low compared with the first estimate.

Estimated emissions of primary microplastics to soils are, however, expected to be small compared with the secondary microplastic emissions. The combined release of primary microplastics from agriculture, horticulture and paints and coatings in the whole EU/EEA (~12.700 tonnes/year – ECHA, 2020)) is only slightly higher than expected secondary microplastic emissions to water from only land-based litter fragmentation in the Netherlands alone (~10,000 tons/year – Verschoor and De Valk, 2018). It is unclear what amounts end up in soils, but the method used by Verschoor and de Valk (2018) is derived from OSPAR, which estimates 15-40% fraction of litter reaching the sea in its catchment area. This suggests that a substantial amount of plastic litter ends up in soils. For tyre wear particles, it was estimated that 74% is released to road side soils and 4% to other soils (Sieber et al., 2020).

Table 4 Use and releases of intentionally added microplastics in the EU/EEA. Derived from data from ECHA (2020).

Sector/ Product Group	**Use (tonnes/year)**	**Eventual release to the environment (tonnes/year)**
Cosmetic products	8,700 (4,100-13,100)	3,800 (1,800-5,900)
Detergents and maintenance	17,000 (11,100-23,000)	8,500 (5,600-11,600)
Agriculture and horticulture	10,000 (3,500-18,000)	10,000 (3,500-18,000)
Oil and gas	1,200 (300-2,000)	270 (~0-550)
Paints and coatings	5,300 (10,200)	2,700 (5,200)
Construction products	Unknown	Unknown
In vitro diagnostic devices	50 (0.5-100)	0.27 (0.25-0.29)
Medical devices	Unknown	Unknown
Medicinal products	2,300 (800-3,700)	1,100 (400-1,800)
Food additives	Unknown	Unknown
Infill material for synthetic pitches	100,000 (15,400-184,800)	16,000 (2,000-52,000)

3.1.4 *Data gaps*

The main sources of microplastics emitted to soils seem to have been identified, but quantitative data is not complete. Based on data for other environmental compartments (Verschoor and de Valk, 2018; ECHA 2020), it seems that an adequate dataset of emission rates of microplastics to soils is still missing. The soil-specific data seems to lack important information, such as paints as part of construction products and microplastic release due to the abrasion of macroplastics (litter). Manure and compost might be another source of microplastics in soils based on measurements taken in China (e.g., Yang et al. 2021). Furthermore, the data is estimated for different locations, some sources quantified for Switzerland, the EU and the OSPAR region. So a full overview of emissions to soil for the EU or the Netherlands is still missing.

3.2 Observations in soil – behaviour and fate

Once emitted to terrestrial ecosystems, microplastics accumulate in soils and can migrate to marine and fresh waters through runoff, with erosion, and by atmospheric transport, and can migrate to the groundwater and deep soil through, for example, leaching, bioturbation and farming activities (highlight 5). After microplastics enter the soil system, the microplastics' physical, chemical and biological properties change through fragmentation and (bio)degradation.

Although information on sources can be improved, it is clear that the microplastics are found almost ubiquitously. In a survey of Swiss floodplains, 90% of soil samples contained microplastics (Scheurer and Bigalke, 2018) and polyethylene particles accounted for 88% of all microplastics found. In addition to MPs being directly emitted to soils, it is thought that indirect sources also play a role. Several studies explained the detection of MPs in natural areas as likely coming from flooding and air deposition (Scheurer and Bigalke, 2018; Chen et al., 2020; Liu et al., 2018; Lv et al., 2019). Although there is some data available on microplastics in soils, the data remains limited at the moment and data on microplastics in soil specific to China is relatively abundant compared with measurements taken in other locations (Xu et al., 2020b; Table 2). There is currently no data found in scientific literature for soils in the Netherlands. As reported in the overview by Xu et al. (2020b) (Table 2), the abundance of microplastics in 20 arable fields in Shanghai were 78.0±12.9 and 62.5±12.9 particles/kg (sizes of 0.02 − 5 mm) in shallow and deep soils, respectively (Liu et al., 2018). Sewage sludge is used as fertilizer on arable fields, which is considered a primary driver of soil microplastic pollution. Ng et al. (2018) reported a mass of microplastic derived from sewage sludge application of up to 2.8×10^3 – 1.9×10^4 tons annually in Australia. Microplastic counts found in 31 agricultural fields from the Chilean central valley (600 – 10,400 particles/kg) increases upon successive sludge applications (Corradini et al., 2019). Another study from Wuhan, China revealed 320–12,560 particles/kg in horticulture (Chen et al., 2020). By now, the highest abundance of microplastics in agricultural soils was observed in soils in Yunnan (7,100-42,960 particles/kg) (sizes of 0.02−10 mm) (Zhang and Liu, 2018). It is currently unknown whether these amounts are high or not and what the related potential risks are.

As an example of the fate and behaviour of microplastics in arable soil, we have selected the investigations of Yu et al. (2021), who studied microplastic distribution in a greenhouse vegetable production base in Northern China (Shouguang City, Shandong Province). Samples of greenhouse agricultural soil, open-field agricultural soil, and agricultural plastic mulch film were collected to investigate the distribution characteristics, influencing factors, and discharging sources of microplastics.

The abundance of microplastics at all sampling sites ranged from 310 to 5,698 items/kg, with an average value of 1,444 ± 986 items/kg. The main size category of microplastics was less than 0.5 mm and the contribution of microplastic particles with sizes <0.5 mm in the 10–25 cm layer of greenhouse agricultural soils was significantly higher ($p < 0.05$) than that found in the 0–5 cm soil layer, which indicated that small microplastic particles tended to migrate to deeper soil layers. The prevailing shapes of microplastics were fragments, while polypropylene, ethylene-propylene copolymer, and polyethylene dominated among chemical compositions with a total of 85%. This correlates to the widespread use of these plastics in agriculture in China (Zhang et al., 2019) and similar results have been found in other studies (Liu et al., 2018; Ding et al., 2020). Other microplastics measured were: polystyrene (PS), polyester fibres (PES), cellophane, polyurethane (PU), rayon, acrylonitrile-butadiene-styrene copolymer (ABS), and polymethyl methacrylate (PMMA).

The fractions of silty and sandy particles in soils were correlated with the abundance of MPs in those soils. The microplastic abundance in sandy loam was significantly higher ($p < 0.05$) than that found in silty loam or loam. Therefore, the soil texture may affect the distribution of microplastic particles in local agricultural soils. In addition, the age of the greenhouse was positively correlated with microplastic-particle abundance in the soil, while there was no significant difference in the abundance of microplastics in greenhouse agricultural soils under different irrigation regimes.

3.3 Modelling microplastic transport and fate in soil

Polymer type, shape and particle-size-dependent modelling of microplastics' fate and behaviour is important for coping with the complexity of microplastics in the environment (highlight 6). Studies for the modelling of microplastic emissions, transport and fate in water are available, but these are not yet sufficient for an investigation of the soil system.

Modelling can play an important role in coping with the complexity of microplastics due to the range of MP properties (e.g. polymer type, size, shape), affecting their fate in soils and the environment. However, studies modelling the fate of microplastics in soil are not yet sufficient. For instance, recent research provides modelling approaches to deal with the various sizes, shapes and densities of microplastics (Kooi and Koelmans, 2019). There are various types of models, each with particular benefits and shortcomings, but each of them are optimized to typically answer one question. Here we focus specifically on those

models aimed at estimating microplastic release, transport and fate in the soil system. Typical models available today are able to estimate:

- The emission to the environmental compartments (air, water, soil) of substances and materials, for instance Material Flow Analysis (MFA) models (Kawecki and Nowack, 2019; Sieber et al., 2020).
- Background concentrations of substances (Hollander et al., 2016) and materials (Meesters et al., 2014) at a screening level.
- Spatially and time resolved concentrations of substances and materials (Besseling et al., 2017).

Some of these models have already been applied and adapted for use with microplastics, others still require some work to account for the specific needs that come with modelling microplastics in the environment. Here we will briefly discuss some of the modelling tools already available and the remaining knowledge gaps.

3.3.1 Applicability of existing models

Existing models fit for organic compounds or metals need to be adapted in order to be fit for use with microplastics. That changes are required is partly based on existing knowledge about the behaviour of particles (nanoparticles or colloids) compared with dissolved compounds in the environment (Meesters et al., 2013). And on existing knowledge about the behaviour of microplastics in the environment (Koutnik et al., 2021). For this reason, the first modelling studies conducted on microplastics (Besseling et al., 2017) were based on models developed for nanomaterials (Quik et al., 2015). However, none of the modelling studies on microplastics included the fate in soil and they focused largely on the aquatic environment. At most, the release to soil is quantified (Kawecki and Nowack, 2020; Sieber et al., 2020), but not the fate and behaviour in order to estimate exposure levels in soil. This is a significant knowledge gap, because it has been estimated that the emission of microplastics to soil is likely to be almost tenfold the emission to rivers and oceans (Kawecki and Nowack, 2019; Sieber et al., 202; Zubris and Richards, 2005).

Models aimed at describing nanomaterial fate can, for the most part, already be applied to microplastics and the following models include fate in the soil compartment:

- SimpleBox4nano (Meesters et al., 2014);
- MendNano (Liu and Cohen, 2014);
- NanoFATE (Garner et al., 2017);
- NanoFASE WSO (Lofts et al., 2019).

These models are developed for materials at the nanoscale (1-100 nm) with an applicability domain of 1 nm to 10 µm sized particles. This falls within the stokes regime of particle movement in another medium (e.g. sedimentation/deposition). For larger microplastics (>10 µm), it might be necessary to test some of the underlying theories, such as the application of stokes law for describing the movement of particles in another medium. However, this particular issue is probably not as relevant for particle movement in soil compared with air and water. In soil, larger particles will get trapped more easily in soil pores, thereby greatly reducing their mobility. Another uncertainty is the parametrization of

microplastic degradation because many nanomaterial models were initially developed with metal particles in mind. As such, dissolution to an alternate form is sometimes included, but in practice degradation and, in particular, fragmentation is not included in these models.

Some studies on microplastics have already started to indicate that the fragmentation and degradation of microplastics is an essential process for modelling microplastics in the environment (Koelmans et al., 2017; Koutnik et al., 2021; Kaandorp et al., 2021; Chamas et al., 2020). Soil, together with water and sediment, is considered a sink for microplastics, and therefore it is a relevant process which may largely define the accumulation level of microplastics in soil. As an example of how the full degradation can be described for modelling, degradation rate constants from Chamas et al. (2020) are further examined in Annex 2.

3.3.2 Next steps and knowledge gaps

In order to estimate microplastic exposure concentrations in soil using modelling tools, the basic steps are first to estimate the emission to soil, and then to estimate the microplastic concentration in soil. Microplastic-relevant models that estimate the emission to soil, as well as to air and water, are already available based on material flow analysis (Kawecki et al., 2019; Sieber et al., 2020). The currently missing link is an extrapolation of the modelling study from Switzerland to other regions of interest, such as the Netherlands and the entire EU. In addition, further effort might be needed to estimate emission in a spatially resolved manner in order to estimate local emissions. This means the major shortcoming is not the lack of models or tools, but rather data to include all important sources and material flows. Gathering these data takes a lot of effort, but the inclusion of uncertainty by applying a probabilistic approach can be very useful.

Following a sufficient estimation of the emission rates (tonnage per year), the further fate in the soil compartment can be modelled. To do this, existing models fit for nanomaterials can be applied first. These models can then be optimized based on validation and calibration with microplastic measurements/observations to include potentially relevant microplastic specific fate processes and physico-chemical descriptors, such as approaches to cope with the large variety in shapes, sizes and polymer types.

These modelling efforts will result in essential exposure data for soil-dwelling organisms, plants, but also for further studies on human exposure through the environment.

Overall, the main knowledge gaps for modelling the fate of microplastics in soil are related to:

- Estimating emissions to soil at the EU level by applying the existing framework applied in Switzerland and including some other major sources, such as emissions from macroplastic litter and paint, in addition to already included sources (see Section 3.1.2).
- Quantifying the fragmentation and degradation process (see Chapter 4 and Annex 2).

- Validating and improving existing fate modelling tools for use with microplastics.

Some of these issues are likely to be tackled in some of the recently started EU research projects, such as LEON-T, Papillions and MINAGRIS. (see Section 6.4)

4 (Bio)degradation of (micro)plastics

Published biodegradation data for (micro)plastics differ greatly between studies (highlight 7). The (bio)degradation potential of microplastic particles depends on three major aspects, namely 1) their chemical composition, 2) their physical properties and 3) the environmental conditions to which they are exposed. The limited data available on (bio)degradation and monitoring data show that microplastics can accumulate in the environment, including soil systems. However, the actual accumulation rates are uncertain, not only due to the scarcity of data in combination with a large variation between measured (bio)degradation rates, but simply also due to the lack of standardized methods suitable to assess the fragmentation and degradation of microplastic particles under environmentally relevant conditions, and the vast range of polymer types and identities.

4.1 Introduction

One of the advantages of using plastics is their resistance to environmental influences. Nevertheless, the degradation of macro- and microplastics is shown to occur over time depending on the intrinsic properties of the (micro)plastic (e.g. size, type of polymer and additives) and the extrinsic properties of the environmental and biological conditions (e.g. microbiome, acidity, temperature and UV radiation) (EC, 2020; Maier, 2018; Tokiwa et al., 2009). Processes that induce degradation can be physical (e.g. UV-light), chemical (e.g. hydrolysis) or biological (e.g. microorganisms, such as bacteria and fungi) in nature (Klein et al., 2018).

(Bio)degradation is a process that causes the breakup of the polymer structure and any of the additives present in the microplastic particle through biotic and abiotic processes. This can yield smaller sized (micro)plastic particles whose polymer structure is still largely intact, a process termed fragmentation. It is this process of the fragmentation of macroplastics which is found to be the largest contribution to microplastic emissions to the environment (Verschoor and de Valk, 2018) (see Chapter 3.1 on sources and emissions). Fragmentation can be the first step in a (bio)degradation cascade, as fragmented particles have been shown to be more susceptible to further (bio)degradation (Mohanan et al., 2020). However, there have been types of plastics designed that only fragment to a certain degree and that have been banned for that reason (e.g. oxo-degradable plastics – conventional plastics containing additives which promote the oxidation of the material).[10]

(Bio)degradation can also lead to a more complete disintegration of the polymer structure, yielding oligomers, monomers and other degradation products. While the microplastic particles are no longer present, other chemicals become available. The process whereby the polymer and additives are fully metabolized, yielding carbon dioxide, is termed

[10] DIRECTIVE (EU) 2019/904 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 5 June 2019 on the reduction of the impact of certain plastic products on the environment. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019L0904&from=EN

mineralization. Mineralization is also known as complete or ultimate degradation.

There is an increasing interest in (bio)degradable (micro)plastics as a solution for certain uses of (micro)plastics (e.g. single use products, seed coatings). As such, the development of biodegradable plastics is considered one of the technological innovations which could reduce the potential effects of future emissions of microplastics (EC, 2020; Maier, 2018). However, caution is needed, as accumulation in the environment can still occur if the (bio)degradation does not occur under environmentally relevant conditions and within reasonable timeframes. Different terms, such as partially or completely biodegradable, degradable and compostable, are used simultaneously to describe the (bio)degradability, i.e. the ability of a material to breakdown, of materials and products. In practice, these claims can mean various (bio)degradation events occurring under different biotic and abiotic conditions. In that respect, it is important to understand what the drivers of (bio)degradation are and to have suitable testing methods and standards to characterize the biodegradability of microplastics.

In the remainder of this chapter, we discuss in more detail the main determinants for (bio)degradation (4.2), the issues associated with allowing (bio)degradable microplastics for certain uses (4.3) and finally the implications for estimating microplastic fate in the environment (4.4).

4.2 Determinants for (bio)degradation

The (bio)degradation rates of plastics depend on three major aspects: 1) the chemical properties of a material, 2) the physical properties of the material and 3) the environmental abiotic and biotic conditions to which the material is exposed. Each aspect includes multiple variables, the most important of which – and thus not the extensive list of all parameters – are discussed below.

4.2.1 Chemical properties

Polymers

Polymers contained in microplastics often have long backbones with many carbon-carbon bonds, but can also contain other atoms such as oxygen, hydrogen, fluoride and silicon. Carbon-carbon bonds are difficult to break, as the dissociation energy required for it is high, with increasing energy needed for increasing bond order (Blanksby and Ellison, 2003). The addition of fluoride creates even more stable structures (Reddy, 2015). (Bio)degradation by microorganisms or environmental processes will therefore be slow. Some polymers are (undesirably) prone to deteriorate due to abiotic processes (e.g. the colouring of polyurethane of mattresses from white to yellow over time through photo-oxidation) which initially leads to superficial breakdown effects. Plastics can also be designed to deteriorate under certain conditions, but care should be taken that the plastics really (bio)degrade and not only fragment as is the case with oxo-degradable plastics (Abdelmoez et al., 2021). While some structures, e.g. esters, are expected to be easily broken down (Iwata, 2015), their (bio)degradation potential can vary greatly under environmentally relevant conditions, with PET being hardly degraded (Hiraga et al., 2019), PLA being compostable through hydrolysis that occurs under the

prevailing high temperatures (Gorassi and Pantani, 2017), and the microbial polyester PHA being enzymatically degradable by bacteria and fungi under ambient conditions (Ong et al., 2017). This illustrates that theoretical considerations alone are not sufficient to underpin the degradation potential of substances. It also suggests that naturally occurring polyesters might be more prone to (bio)degradation than synthetic polyesters. Moore and Saunders (1998) further note that the addition of hydrophilic groups foster biodegradation. These functional groups promote the attachment of bacterial cells and, simultaneously, the accessibility of secreted enzymes to the surface of polymers (Wilkes and Aristilde, 2017).

Additives and contaminants

Plastics often contain additives to optimize the material properties and to stabilize the polymers. These additives are blended in the polymer during production processes. Commonly used additives include stabilizers, UV-blockers, fillers, plasticizers, colouring agents and flame retardants (ECHA, 2021; Hahladakis et al., 2018). During the lifetime of a plastic material, other contaminants can also adhere. For example, during usage, as well as afterwards when it has become waste and/or ends up in the environment. The presence of additives and contaminants generally hampers breakdown processes, but additives could be added that accelerate the breakdown (e.g., for PLA; Gorassi and Pantani 2017).

4.2.2 Physical properties

Shape

Primary microplastics are produced in a manner that enables them to serve their purpose for the intended production processes. They can be produced in different sizes and shapes, and will have similar physical properties in each batch. Secondary microplastics are formed due to weathering and the fragmentation of larger plastics. These plastics will be heterogeneous and can have all kinds of shapes and sizes. This directly affects the potential to degrade, as roughness of the surface area and size of the surface area are positively related to (bio)degradation (Moore and Saunders, 1998). If it is assumed that microplastics are impenetrable for organisms, biodegradation can only occur on the surface of a particle. The ratio between surface area (SA) and volume (V) will therefore determine how fast a particle can biodegrade. A high ratio will lead to a large surface area available for biodegradation and a relatively fast degradation rate, compared with a low ratio for which the reverse applies. This also applies for abiotic degradation.

Molecular weight

The molecular weight of a polymer depends on the degree of polymerization (the length) and the weight of the repeating unit (Balani et al., 2015). In general, the molecular weight is inversely related to the degradability. During (bio)degradation, polymers are converted into simple, low-weight molecules, such as monomers, CO_2 and H_2O. High-weight polymers contain greater mass and have a higher degree of entanglement with other polymers, making them more rigid but also (chemically) more resistant, and as such more effort is needed to degrade these polymers down to these basic molecules.

Crystallinity
In crystalline polymers, the polymer chains are regularly aligned, in contrast with amorphous polymers, in which the chains are unstructured. The crystallinity of a polymer influences the density and hardness of a material. Aligned polymers are dense and rigid (e.g. HDPE), while semi-crystalline polymers contain both amorphous and aligned polymers and are less dense and more flexible (e.g. LDPE). Crystallinity lowers the rate of microbial degradation, as it limits the movability of polymer chains and as such the accessibility of polymer chains to microbes (Mohanan et al., 2020; Moore and Saunders, 1998; Tokiwa et al., 2009).

Density
The density of plastics varies between around 0.8 to 2.3 kg/dm^3 (Omnexus, 2021). As the density of water is around 1 kg/dm^3, plastics < 1 kg/dm^3 will primarily float, while plastics > 1 kg/dm^3 will sink. Based on these differences, plastics end up in different parts of the environment and are exposed to different influences. Sinking plastics, such as PET, PVC and PLA, will likely end up directly in sediment after entering water systems. These plastics will be affected by the abiotic characteristics and the microbiota present in these sediments. Floating plastics, such as HDPE, LDPE and PP, may be transported longer distances and are vulnerable to weathering processes such as photo-oxidation, storms and the turbulence caused by waves and tides. The fate of microplastics in soils is less apparent based on its density.

In soils, the fate of microplastics is less determined by the density of the material, while agglomeration/aggregation plays an important role, as it affects the surface area and bioavailability.

4.2.3 Environmental conditions

Temperature
Biological activity is positively related to temperature, as temperature induces microbial growth. Each microbe has a range of temperatures in which it is active and, within this range, an optimal temperature to thrive in. Pischedda et al. (2019) showed that, for a commercial plastic (containing biodegradable polyester (65%), starch (28%) and a natural plasticizer (~6%)) in soil, the mineralization rate increased with temperature (28°C>20°C>15°C). Around 80% of mineralization was achieved after 400 days at 28°C, while around 40% and 30% was achieved at 20°C and 15°C, respectively. Also, for films made from the polymer PLA, it was observed that increasing temperature from 25°C to 37°C and 50°C significantly enhanced the degradation rates in soil and compost, on top of that abiotic chemical hydrolysis that occurs at these temperatures (Karamanlioglu, 2013). Increasing temperature may also limit biodegradation in soil, as was observed by Nishide et al. (1999); the bioplastic PHB/PV underwent a slower degradation at 52°C compared with 30°C under aerobic conditions.

pH and soil organic matter
Although no studies could be found that have investigated the effect of pH on biodegradation rates, the pH of soils will most likely impact these rates. For example, the pH affects the availability of nutrients, toxins and metals and will have an influence on the composition of micro-organisms

present and subsequently the biodegradation rates. A study of Zhao et al. (2021) demonstrated that the presence of microplastics can alter the soil pH as well. The results indicate that the pH of soils containing different types and shapes of microplastics (0.4% w/w) in most cases increases over time. The rise in pH is explained as a potential increase in porosity and soil aeration after the addition of the microplastics to the soil and due to an alteration of the biota after the leaching of chemical compounds from the microplastics (Zhao et al., 2021).

Light

Light and other types of electromagnetic radiation can induce degradation (Yousif and Haddad, 2013), as it is a source of energy. When polymers absorb photons (small light particles), photochemical reactions can occur, leading to breakdown. This type of degradation is also known as photodegradation. UV-blockers are frequently added to plastics to prevent photodegradation. In the environment, photodegradation will occur mainly due to exposure to UV radiation. The process is primarily explained by two mechanisms, i.e. photoionization and chain scissions (Niaounakis, 2015). These mechanisms lead to a shortening of polymer chain sizes and the formation of oxidized groups, which facilitate biodegradation (Ghatge et al., 2020). Lee et al. (1991), for example, showed that UV-treated polyethylene films were biodegraded (by different bacteria) to a larger extent than heat-treated films and control films. Taghavi et al. (2021) demonstrated increased biodegradation of PS and PE after UV pretreatment. Note that in soils there is limited exposure of microplastics to UV light and photodegradation will therefore be a minor factor.

Microbial composition

There is great variation between microorganism community compositions in environmental compartments (marine, freshwater, sediment, soil), but also within an environmental compartment. The microbial composition depends on the characteristics of the environment, with important soil parameters being the oxygen level, the carbon content, the presence of (trace) metals, the pH, the moisture level, the C:N:P ratio, salt concentration and the temperature range. Besides, environmental constraints and the large variation in microorganisms can at least partially explain why degradation rates can differ so much between environments. It should also be noted that microorganisms compete with each other, but also with other organisms (e.g., plants), for the available nutrients. Microorganisms being able to use (micro)plastics as carbon and an energy source can give a competitive advantage to other organisms. Under rate-limiting conditions, adaptations could occur, but this is rather speculative and has so far not been shown to occur for conventional plastics (PE, PET, PS) under environmentally relevant conditions, while during the last 50 years increasing levels of plastics can be found in the environment. More information on the ability of microorganisms to degrade microplastics can be found in Annex 2.

4.3 Biodegradable plastics

4.3.1 Biodegradation standards

Whether or not a material is considered biodegradable, differs between legal frameworks. In most frameworks, internationally standardized ISO

and OECD test guidelines are laid down to assess and justify the biodegradability of materials and products. For plastics as a diverse group of materials, it is still difficult to assess biodegradability. Most frameworks have not (yet) established (definitive) criteria to earmark plastics as biodegradable. Nevertheless, there is broad consensus on the overall definition of biodegradable plastics, that being plastics that are either mineralized to CO_2 and converted to biomass, or are degraded to small molecules that do not exhibit hazardous properties.

Reach restriction
The ECHA proposed restriction on intentionally added microplastics limits the use of primary microplastics in products in the EU/EEA (ECHA, 2020). It is expected that the restriction will reduce primary microplastic emissions significantly. The restriction does not restrict all microplastic uses and derogates biodegradable polymers from the restriction, as they do not meet the concerns raised on persistency. Within the restriction, a testing strategy has been proposed to assess the biodegradability of microplastics as put on the market. The tests are grouped in five categories, with the most stringent test methods in the higher groups. In Annex 2, an overview of the groups and test guidelines is available.

The lower groups (1-3) consist of screening level tests (ready, enhanced ready and inherent biodegradability tests according to OECD test guidelines). These tests are considered conservative and if full mineralization occurs within the timeframe of a test, it can be assumed that the microplastics will also degrade under environmentally relevant conditions within a reasonable time frame (thus not meeting the (very) persistent criteria as laid down in REACH). The drawback of these tests is that they were not designed for particles and, as such, might be less suitable for assessing the biodegradability of microplastics. For PHBV, two screening level biodegradation tests according to OECD TG 301B and 301F have been reported, in which PHBV was found to be readily biodegradable (reported in ECHA, 2020). These tests support the usability of the OECD screening level test for assessing the biodegradability of microplastics.

Group 4 consists of ISO test methods designed for testing the biodegradability of plastics in water, soil and sediments. In these tests, biodegradation is assessed by comparison with a reference item. Within the proposed restriction, these tests are similarly considered screening level tests, meaning that passing the criteria of any of the tests rules out the persistency of a (micro)plastic. It is, however, unclear how the outcomes of these studies can be compared to the persistence criteria since, for example, the duration of the tests is longer (up to 24 months in soil) and because degradation is assessed relative to a reference material.

Group 5 consists of OECD simulation degradation tests that are used to assess the degradability of substances under environmentally realistic conditions. These test guidelines allow the derivation of degradation rates in surface waters, sediment water systems and soils. However, as with the OECD screening level tests, there is some uncertainty regarding the testing of particles and how this affects the comparison between compartments. These uncertainties have been noted in the RAC opinion on the proposed restriction.

The RAC opinion (ECHA, 2020) examined how the above tests can be best used to determine if microplastics are biodegradable. Out of the eight presented scenarios, the following was considered to be the most suitable. When the criteria of the screening tests (groups 1-3) are not met, tests in multiple environmental compartments from either group 4 or group 5 should be performed (and passed) to justify a derogation. Considering the uncertainties, both the dossier submitter and the RAC proposed a review of the conditions of the derogation be conducted five years after entry of the restriction. It is expected that once the restriction is adopted and tests are conducted, more knowledge will be gained on the usability and comparability of the different tests. Whether substantial biodegradation data in soils will be generated has yet to be seen. The proposed restriction foresees that screening level data (group 1-3) will be sufficient to justify a derogation. Only if these OECD screening level tests are not passed, ISO tests (group 4) and OECD simulation tests from group 5 can be performed, whereby all relevant environmental compartments have to be tested, including soil (EN ISO 17556:2019 (Group 4) and OECD TG 307:2002 (Group 5).

Fertilizer product regulation

Biodegradable polymers are also exempted in fertilizer products by the upcoming regulation (EC) No 2019/1009.[11] This regulation lays down rules to make fertilizers available on the European market and will come into force in July 2022. Each fertilizer has to contain one or multiple materials that belong to a component material group (CMC).[12] One component material group is on polymers, i.e. CMC 9: Polymers other than nutrient polymers. It allows the addition of biodegradable polymers from this group to a fertilizer, however these polymers have to fulfil a certain purpose, e.g. increasing the water retention capability of a fertilizer.[13,14] A recent amendment has further allowed the presence of biodegradable polymers in CMC 1 (Virgin material substances and mixtures), CMC 8 (Nutrient polymers) and CMC 11 (By-products within the meaning of Directive 2008/98/EC) (EC, 2021).

Criteria and test methods used to assess biodegradability have not yet been established in the regulation. These will be assessed by 16 July 2024 and, where appropriate, laid down in criteria. It is stated in the regulation that future criteria must ensure that 90% of the organic carbon has to be converted to carbon dioxide within 48 months after the claimed functionality, as compared with an appropriate standard. This matches the criteria of the proposed ISO test for soils in group 4 of the restriction on intentionally added microplastics. In addition, the

[11] Regulation (EU) No 2019/1009, 2019. REGULATION (EU) 2019/1009 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 5 June 2019 laying down rules on the making available on the market of EU fertilising products and amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009 and repealing Regulation (EC) No 2003/2003, Official Journal of the European Union p. 114.

[12] In total 11 CMC are laid down in Regulation (EC) No 2019/1009. An initiative has been set out to extend the list with two additional CMC. https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12163-Fertilising-products-precipitated-phosphate-salts-and-derivates_en

[13] An EU fertilizing product may contain polymers other than nutrient polymers only in cases where the purpose of the polymer is (Regulation (EU) No 2019/1009, 2019).
(a) to control the water penetration into nutrient particles and thus the release of nutrients (in which case the polymer is commonly referred to as a 'coating agent'),
(b) to increase the water retention capacity or wettability of the EU fertilizing product, or
(c) to bind material in an EU fertilizing product belonging to PFC 4 (Growing medium).

[14] By 16 July 2024, the Commission shall carry out a review in order to assess the possibility of determining biodegradability criteria for mulch films, and the possibility of incorporating them into component material category 9 in Part II of Annex I. (Regulation (EU) No 2019/1009, 2019).

regulation specifies that the polymer ultimately needs to decompose and that it does not accumulate in the environment.[15]

Polymers from CMC 9 (except for those that serve as binder) have to comply with the biodegradability criteria by 16 July 2026 and onwards. If no criteria are established by then, fertilizers are not allowed to contain these polymers when placed on the market.

Mulch films
Plastic mulch films are used on agricultural lands to 'modify soil temperature, limit weed growth, prevent the loss of moisture, improve crop yield and precocity' (BASF, 2021). In 2018, the European standard EN17033:2018[16] for biodegradable mulch films for use in agriculture and horticulture was introduced. The European standard replaced national standards, although these were only available for France (NFU 52-001:2005) and Italy (UNI 11495:2013). At that moment, no ISO standard to determine the biodegradability of mulch films was available yet. In 2021, an ISO standard was published as well (ISO 23517:2021).

In order for mulch films to comply with these standards, 90% of the organic carbon in the material has to be converted to CO_2 (absolute or relative to a positive control) within two 2 years under ambient soil conditions (Hayes and Flury, 2018). The time frame is similar to the ISO test in soil, proposed in the restriction on intentionally added microplastics.

In the fertilizer product regulation (Regulation (EU) No 2019/1009), it is stated that by 16 July 2024 a review shall be carried out to 'assess the possibility of determining biodegradability criteria for mulch films, and the possibility of incorporating them into component material category 9 in Part II of Annex II'. Currently, the methods to assess biodegradability have not been decided on.

Other test standards for plastics in soils
Beside the aforementioned standards, two more standards were found that describe how to determine the biodegradability of plastics in soils. The first, ASTM D 5988-12, was developed by ASTM International to determine the aerobic biodegradation of plastic materials in soils. The methodology is considered to be equivalent to the ISO 17556 test (ASTM, 2018). The second is UNI 11462:2012, published by the Ente Nazionale Italiano di Unificazione (UNI). This document specifies test methods to assess the biodegradability of plastics mostly used in agriculture (UNI, 2012).

Composting standards
The terms biodegradable and compostable are often used interchangeably when describing the breakdown potential of a product or material, although these terms describe the same process under different environmental conditions. Biodegradable indicates the capability of a

[15] 1) The polymer is capable of undergoing physical and biological decomposition in natural soil conditions and aquatic environments across the Union, so that it ultimately decomposes only into carbon dioxide, biomass and water. 2) the use of polymers does not lead to accumulation of plastics in the environment.
[16] 17033:2018 – Plastics – Biodegradable mulch films for use in agriculture and horticulture – requirements and test methods

material or product to be decomposed by microorganisms under environmentally relevant conditions in the environment (e.g., surface water, sediment and soil), compostability implies that this process occurs under specific conditions that are discussed below (EEA, 2020; European Bioplastics, 2016). Standards are available for composting processed at two typical locations and corresponding environmental conditions: 1) industrial and 2) home. These are also discussed below.

Industrial composting
ISO 16929:2021 describes a test method used to determine the aerobic compostability of plastic materials on a pilot scale (ISO, 2021). This test is part of an ISO evaluation scheme (ISO 17088:2021) used to assess the industrial compostability of plastics. The latter includes four aspects that are assessed in combination.

Standard EN 13432:200017 has been established to assess the compostability or anaerobic treatability of packaging materials. For plastic materials, the standard EN 14995:200718 has been established. Both standards have been harmonized in Europe and materials that fulfil the requirements of these standards are considered processable in industrial composting facilities.

The capability of the plastics to biodegrade is measured relative to the biodegradation potential of cellulose. At least 90% of the organic carbon has to be converted to CO_2 or 90% biodegradation compared with cellulose has to be achieved within six months after start of the test. In addition, 70% of the cellulose has to be converted to CO_2 within six months in order for the test to be valid (Degli-Innocenti, 2002). It is also required that the material disintegrates by 12 weeks after starting the test and more than 90% of the material should be smaller than 2 mm (BPF, 2021). Environmental conditions such as humidity, temperature (58 ± 2° C) and oxygen level are kept constant during the test period in order to resemble industrial composting conditions (BioBasedEconomy, 2021; Degli-Innocenti, 2002). While tests are performed under these standardized conditions, composting processes in industrial composting facilities can occur under divergent environmental conditions. In the Netherlands, the composting process takes place during 2-3 weeks, which is less than 1/6th of the duration of the test protocol (Vrins and Costenoble, 2019). This implies that, although materials can be certified as compostable, materials can be composted to a limited degree in practice. An example is compostable cutlery, which is discarded with the residual waste in the Netherlands as these are unwanted in the composting process.[19]

Home composting
There are different certification schemes used to assess whether plastics are compostable at home at a national and continental level. Most of the standards have pass criteria similar to the industrial composting standards. Environmental conditions deviate from industrial composting standards; test temperatures are lower and test durations are shorter in

[17] EN 13432:2000 – Packaging – Requirements for packaging recoverable through composting and biodegradation – Test scheme and evaluation criteria for the final acceptance of packaging
[18] EN 14995:2007 – Plastics – Evaluation of compostability – Test schema and specification
[19] https://www.delta.tudelft.nl/article/what-happens-your-coffee-cup

order to resemble the composting conditions at home. An overview of a number of certification schemes can be found in Annex 2.

4.3.2 Uncertainties associated with biodegradation testing

As discussed above, the biodegradability of (micro)plastics can be assessed differently between frameworks. Regardless of the framework, standardized test guidelines are needed to obtain comparable and regulatory relevant results. These tests should enable a differentiation between fragmentation into smaller sized plastic particles and degradation into (hazardless) substances and complete mineralization. Exempting biodegradable plastics which do not (bio)degrade in the environment must be avoided. A number of uncertainties regarding the aforementioned frameworks and biodegradation test methods are therefore summarized below:

1. The restriction on intentionally added microplastics still has to be adopted. In the final RAC and SEAC proposal, a passed OECD screening level test suffices for a derogation based on biodegradability. These tests might not be particularly suitable for particles and might be too stringent (as can also be the case for the OECD simulation degradation tests). For the ISO test methods, it still has to be decided whether these are considered screening tests and how their outcome can be used to determine the biodegradability of (micro)plastics. For example, the ISO soil test (EN ISO 17556:2019) foresees a testing duration of 24 months, which greatly exceeds the persistence and very persistence criteria used under REACH.
2. As field conditions are inherently variable, test results will likely not represent all the different field conditions that can occur. This is inherent to all environmental fate and toxicity testing, as testing is performed under controlled conditions (needed for standardization and comparison) rather than realistic environmental conditions.
3. Some standards allow tests to be conducted with the plastic base material instead of a product or material containing additives (e.g. as in the case of standard EN17033:2018 for plastic mulch films). Tests with the base material will likely result in different biodegradability rates due to differences in physical and chemical composition.
4. In case of industrial composting, and potentially home composting, standards do not always characterize the processes that take place at composting sites. As described earlier, the duration time prescribed by the industrial composting standard is significantly longer than the actual full-scale industrial composting time in the Netherlands, resulting in materials that are labelled ‘compostable’ while actual degradation may be limited.
5. For some tests, only partial biodegradation is required for plastics to be regarded as biodegradable. The ISO tests proposed in the restriction on intentionally added microplastics note as pass criteria that 90% biodegradation is achieved in comparison with the reference material. This implies that when a minimal required biodegradation of 60% for the reference material is achieved during the test, only 54% of the test material has to be mineralized during the test.

4.3.3 *Which plastics are biodegradable according to literature?*

A categorization often used for plastics, as presented in Iwata (2015), is that plastics are either biodegradable or non-biodegradable and either bio-based or mineral-based. While mineral- and bio-based plastics can be distinguished based on the source material, no information is given by Iwata (2015) on the criteria to assign a plastic as biodegradable. The presence of an ester group is often considered as an inherent property making polymers biodegradable. While this appears to be the case for certain polymers, e.g., PLA and PHA, this is certainly not always the case, for example, for PET.

PLA

Kjeldsen et al. (2018) report a broad overview of biodegradation data for PLA, showing that results vary greatly in soil as well when composted. The compostability of PLA ranged from 100% biodegradation in 28 days to only 13% after 60 days. In both cases, temperature was kept constant at 58°C and it is unclear what underlies the large difference. Parameters such as microbiological composition, pH, humidity, the availability of nutrients and energy sources or other factors could have all influenced the outcomes (see Chapter 4.2 for the determinants for (bio)degradation). In addition, most tests have not been performed in accordance with standardized test protocols, which means that results could have also been influenced by the applied methods and measurement techniques. In this study, these differences are not further explored, but it demonstrates that interpreting and comparing study results should be done with caution.

Several biodegradation tests were performed with PLA in soils (Kjeldsen et al., 2018); however, most test materials contained an additional substrate/material such as fertilizer, sisal fibres and/or starch. Additional substrates can hamper or enhance the degradation rate of polymers as these substrates may affect the microbiological composition. Test conditions have to be standardized or at least similar in order to compare degradation rates, even though other studies may still be valuable and maybe even more representative when testing actual environmental conditions. One test performed with solely PLA resulted in 10% degradation after 98 days in a soil with a moisture content of 30%, while information on temperature was not given (Wu, 2012). Tests with powdered PLA in soils resulted in 14% biodegradation in 28 days at 25°C and 60% humidity (Adhikari et al., 2016). While PLA is regularly referred to as a biodegradable plastic, the available biodegradation data does not support this.

PHA

Most tests with PHA-based plastics are performed with PHB (polyhydroxy butyrate). Composting of PHB resulted in around 80% biodegradation in 28 days at 55°C and humidity levels of 70% (Tabasi and Ajji, 2015). Comparable results were achieved in another study, however, at 58°C in 110 days and 50% humidity (Weng et al., 2011).

Under lab-scale conditions, more than 64% degradation was recorded after 6 months in soil (Jain and Tiwari, 2015). In tropical soils under real environmental conditions, up to 98% biodegradation was achieved for PHB films, though over a longer time span (300 days) (Boyandin et al.,

2013). But for pressed PHB pellets, degradation in tropical soils was limited to a maximum of 55%, demonstrating the importance of the physical properties for the biodegradation rate.

Mc Donough et al. (2017) reported 66% of mineralization after 28 days for PHBV microparticles. The result for the copolymer of PHB was achieved in a screening test in the laboratory. For the polymer PHB/PHBV, 79.2% biodegradation in 28 days was reported in the microplastics restriction (ECHA, 2020).

For PHA, lower biodegradation results were recorded than were for PHB, with 35% biodegradation in 60 days in soil at a temperature of 35°C and 35% moisture content and 48.5% biodegradation in 280 days at 20°C and a moisture content of 60% (Gómez and Michel Jr, 2013; Wu, 2014). Similar to PLA, for PHA-based plastics the degradation under environmentally relevant conditions is rather slow and PHA should not be regarded as biodegradable (compared with the P and vP criteria as laid down under REACH).

4.4 Implications for estimating microplastics fate

As noted in Section 4.2, there are many determinants which influence (bio)degradation. To model the fate of microplastics in soils, these determinants should be incorporated in relevant modelling tools. While the importance of fragmentation and degradation for modelling has been emphasized by different researchers (Kaandorp et al., 2021; Koelmans et al., 2017), implementation of the underlying processes is as important for the model to represent real-world conditions.

Initial steps have been taken to determine the biodegradation rates for different plastics (See Annex 2 for elaboration). However, there are still many uncertainties concerning a reliable determination of these degradation rates under different environmental conditions. Based on the information gathered on biodegradation, there are some issues that have implications for estimating microplastics' fate under relevant environmental conditions and these issues need to be addressed for modelling:

- The number of biodegradation tests performed with microplastics is limited. Furthermore, only a very small fraction of these tests were performed under environmentally relevant conditions (simulation tests) and even fewer in soils.
- Tests performed at a screening level will not allow the derivation of degradation rates, but the outcome (ready/not ready/inherently biodegradable) can be used as input for modelling tools. The usability of ISO tests remains questionable at this stage.
- There is an urgent need for the standardization of (bio)degradation tests that are suitable to assess the degradability of microplastic particles. Options have to be scrutinized and existing tests have to be adapted or benchmarked to be comparable to available persistence criteria (under REACH, but also other frameworks).
- A great number of variables have been determined which influence the biodegradation rates of microplastic, including their

chemical and physical properties, as well as the abiotic and biotic conditions to which the particles are exposed. As for (initial) modelling, it will be impossible to take into account all of these factors. The focus should be on the variables which impact biodegradation rates the most. Note that these variables could also indirectly enhance biodegradation, for example, by the fragmentation of (micro)plastics.

- The size and shape of microplastics are an significant aspect influencing the biodegradation rate. Information on size distributions and the shapes of microplastics in the environment, as well as degradation rates for a heterogenous set of particles, should be determined to assess biodegradation in modelling tools. For the time being, one could also use worst-case assumptions for surface area (linked to size and shape) as a first tier, and refine them when necessary based on particle size.

Note that list above is not exhaustive, but are the first steps to implement and improve biodegradation in microplastics fate modelling.

5 Impacts and risks of microplastics in soil

High amounts of microplastics in soils can alter the fundamental physicochemical and biological properties of the soil system (highlight 8). Soil health and function may be impaired and trophic transfer in food chains may also occur. This may lead to adverse effects on ecosystems. In order to address the potential ecological impact on soil systems, (further) research is needed on the (direct and indirect) effects of microplastics on the viability, development, fertility and mortality of soil organisms. Next to such effect studies, improved insights are necessary on the exposure, bioavailability and uptake of microplastics by organisms and humans. The current information on the impact of microplastics on soil organisms and ecosystem functioning is incomplete and sometimes conflicting.

5.1 Introduction

There are a lot of publications focused on studies of microplastics in soil, but their usefulness for risk assessment is not yet clear. Initial results published show that microplastic particles can be taken up into crops from the soil. It follows that the ingestion of soil particles and the consumption of primarily vegetables could form a potential health risk due to the presence of microplastics in the soil. The seriousness of the health risk due to the exposure to microplastics through soil is not yet clear.

In studies focused on ecosystem effects, unrealistically high concentrations of microplastics and short incubation times are often used. It is expected that these ecosystem impact estimates will soon start to become more reliable and reflect real field conditions, in view of the high rate of studies being published and the recent or imminent start of new projects on effects of microplastics on the ecosystem. Here we present some scattered observations on this issue made by the authors of several recently published literature review studies, illustrating the mosaic contribution of different studies.

5.2 Ecosystem effects of microplastics

5.2.1 Microplastic particles

It was reported that microplastics may adversely affect soil properties, protist communities[20], plants and even agroecosystems (Li et al., 2020b). Several studies found that bulk density, water holding capacity, soil fertility and the stable water aggregates of soils could be affected by microplastic contamination (de Souza Machado et al., 2018; Ma et al., 2018), and polyester fibres exhibited the most apparent effect. A case focused on soil protists (Rillig and Bonkowski, 2018) indicated that protists were likely to ingest microplastic particles smaller than 1 µm. This means microplastics could be transferred higher up the food chain of soil organisms, e.g. by earthworms feeding on soil protists (Rillig et al., 2017). Another study reported that microplastics could induce damage to earthworms' tissues and immune systems (Rodriguez-Seijo et al., 2017). Recently, long-term adverse effects on earthworms of

[20] Protists are single-celled organisms of the kingdom Protista such as protozoan or algae.

polystyrene microplastics were reported using environmentally relevant concentrations (Sobhani et al., 2022).

5.2.2 *Uptake by plants*

Micro- and nanoplastics are of particular concern due to their ability to be taken up by plants (Li et al., 2020b). Recent studies have observed the adverse effects of fragments of (biodegradable) mulches on plant growth and rhizosphere bacterial communities (Qi et al., 2020; Serrano-Ruiz et al., 2021). In a toxic effect investigation on wheat, low-density polyethylene and starch-based biodegradable plastic mulch film were selected as microplastic residues (Qi et al., 2018). The result showed that the growth of wheat was more affected by plastic mulch film than it was by its polymer, polyethylene. Specifically, both the vegetative and reproductive growth of wheat were inhibited. Plant height, the number of fruiting bodies and the root biomass of wheat were also significantly reduced. Microplastics also showed a high adsorption capacity for heavy metals and antibiotics (Li et al., 2018a; Li et al., 2020b) in soil environments. Polyamide particles could even serve as a carrier for antibiotics. By absorbing heavy metals or antibiotics, microplastics affect microbial communities directly.

5.2.3 *Leaching of chemicals from microplastics*

Most plastics found in soil also contain a variety of additives that may result in enhanced ecological toxicity (Xu et al., 2020b). Plasticizers such as phthalic acid ester (PAE) have been widely used in plastic mulch films, which have contaminated Chinese agricultural soil (Niu et al., 2014). Since it has been proved that they have negative effects on soil enzyme activity and microbial diversity and are often regarded as endocrine disrupting chemicals (Ye et al., 2010), there is a need to study the fate of PAE in microplastics in residue arable film and the risks of exposure through the food chain (Qi et al., 2020).

5.2.4 *Interactions of microorganisms with microplastics*

In the review of Xu et al. (2020b), significant differences in bacterial communities from different soils related to microplastics were reported. Consequently, microplastics can provide new ecological niches in soils supporting microplastic-associated bacteria with surface-related life strategies. Besides having major ecological implications, this life strategy can theoretically support pathogenic bacteria as a disease vector that favours infection (Chai et al., 2020).

5.2.5 *A Trojan Horse*

Finally, a 'Trojan Horse' effect is described in the literature, which means that microplastics can act as carriers and accumulators for other substances associated with plastics, such as organic substances and metals. This results in increased exposure to other pollutants in comparison with systems with a low abundance of microplastic particles (Hildebrandt et al., 2021). However, a recent review of current work has not found much evidence to substantiate the claim that this Trojan Horse effect actually brings about chemical risks under present natural conditions (Koelmans et al., 2021). Furthermore, cascade and food chain effects can potentially play a role, but are largely unknown (Abdolahpur Monikh et al., 2021; Li et al., 2020b).

5.3 Conclusion on ecosystem effects

There is evidence that microplastics can affect ecological endpoints, directly or indirectly, although there is uncertainty in relation to the actual risks present in current natural conditions. In general, the quantitative relationship, e.g. in a dose-effect relationship, between microplastic pollution in soil and subsequent adverse effects on community and other ecosystem properties is unclear (Abdolahpur Monikh et al., 2021; Li et al., 2020b). Furthermore, systematic knowledge about microplastic properties – such as polymer type, degradation or transformation state, size and shape – affecting soil organisms is largely missing as many of the studies lack a proper physicochemical characterization of the tested microplastics, especially those with outdoor experiments under field conditions. This implies there are severe knowledge gaps in the final steps of performing a risk assessment based on the source-path-receptor framework. Many publications demonstrate potential effects, but a quantitative underpinning under realistic exposure conditions that capture both direct and indirect effects remain uncertain, partly unknown and therefore should receive more attention.

6 State of knowledge and perspectives for action

Overall, research on microplastic emissions to and the fate of microplastic in soil systems, and its effects on terrestrial ecosystems is still limited, but is increasing rapidly, demonstrated by the increasing publication rates (highlight 9). This is motivating an active monitoring of the progress of the knowledge required for risk assessment.

6.1 Introduction

Microplastics in the soil environment exhibit a wicked problem for society worldwide, including the Netherlands. Large volumes of plastics are produced that are an integral part of our daily life: they are managed by a very diverse group of stakeholders, and they represent versatile and diverse applications and emissions. Microplastics accumulate everywhere in the environment because of slow or even the absence of degradation. Understanding the degradation process in terms of full mineralization compared with partial degradation or fragmentation is important, but it is rather complex due to test guidelines that have not been designed with microplastic particles in mind. Microplastics contain a lot of different substances for which the specific fate, behaviour and toxicity is largely unknown. Microplastics are also very difficult to quantify, especially in soil matrices. Currently, research on microplastics in the soil system has reached a publication rate of one scientific article per day and is still increasing. Consequently, more data will become available over time, although it is expected that interpretation in a risk assessment context will remain a challenge.

To address the complexity of and streamline research on microplastics in soil systems, we introduce a conceptual model (6.2) based on the source, to path, to receptor approach for the assessment of health and ecological risk. In addition, we have identified data and knowledge gaps (6.3). This should hopefully guide recently started research projects on these topics (6.4) in order to provide knowledge for policy and management options to mitigate microplastic pollution. We summarize some options for mitigation (6.5) and discuss current criteria for plastic in soil in the Netherlands and Belgium (6.6).

6.2 Conceptual model of microplastics in the soil system

To responsibly manage microplastics, we need knowledge of the links between sources, the environmental exposure and the (potential) adverse effects on receptors in order to mitigate potential risks to acceptable levels. A conceptual model showing the procession of source through pathway to receptor (see Figure 1 in Chapter 1 for a simple diagram) can help to provide a structure to interpret available data and information and to structure knowledge development. It can also help to uncover information gaps. In Figure 5, in addition to Figure 1, we have illustrated the microplastic mass flow between microplastic sources, release and emission to soil, microplastic fate in soil processes and the transport process in and around soil. This is a so-called conceptual model that promotes discussion and facilitates the recognition of weaknesses, gaps and research needs. We used the findings of the

review in previous chapters to draw the model from a recognition of the most important microplastic sources, release and emission routes and fate and transport processes in relation to soil.

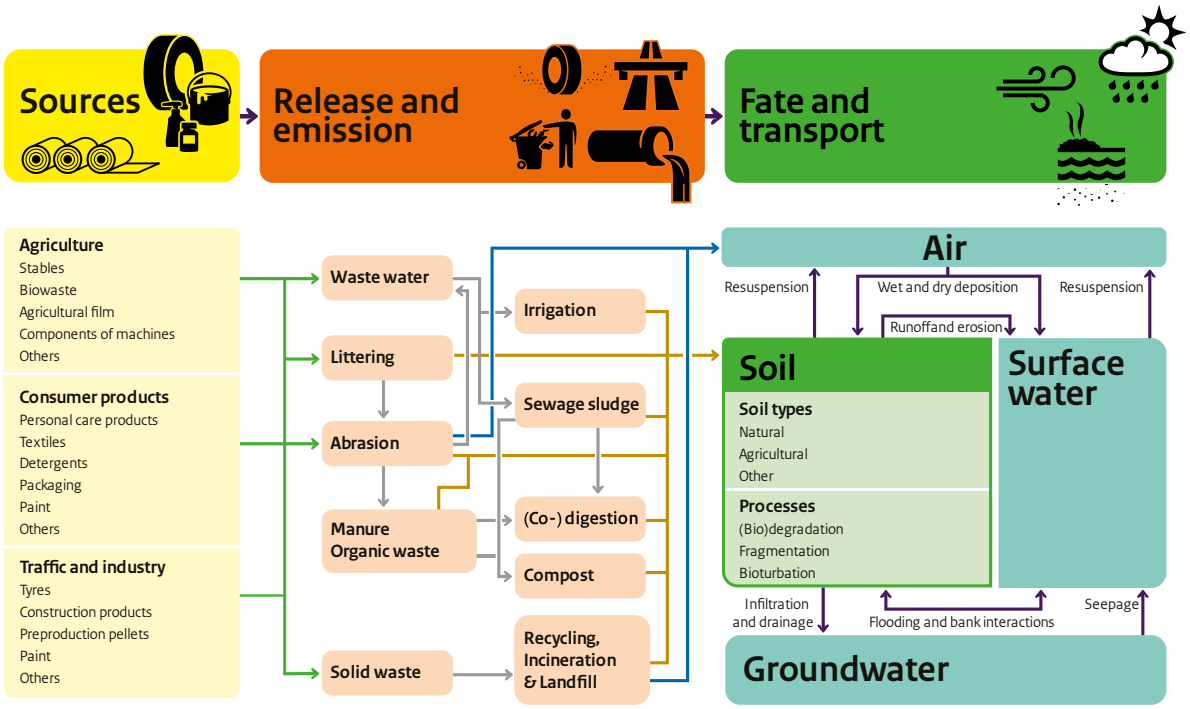

Figure 5 Conceptual model of microplastic sources, release and emission to soil, and further MP fate in soil (processes: (Bio)degradation, Fragmentation, Bioturbation) and transport to and from soil. The main mass flow routes from sources to Soil are indicated, but not prioritized. The green arrows indicate sources linked to waste processing or litter/abrasion that ultimately lead to emission to soil (brown arrows), and (blue arrows) air and water (not indicated here). The purple arrows indicate the main transport processes between soil, groundwater, surface water and air.

Each of the boxes in Figure 5 should in theory be quantified in terms of the mass or abundance of microplastic particles (in a source or in a technical or environmental compartment). The arrows represent the fluxes (mass or other unit per unit time) between the sources and each compartment. The flux of microplastics released from a source (green arrows) to another (specific) temporary or technical compartment (e.g. waste water or compost) via different processes (grey arrows) eventually end up (at least partially) in environmental compartments, soil (brown arrows), Air (blue arrows) and water (not indicated). Within and between these environmental compartments, fate and transport processes (purple arrows) determine the environmental exposure. The accumulation of microplastics in soil depends on the rate of emission and transport to soil and the rate of transport from soil and (bio)degradation. Fate processes such as fragmentation and bioturbation mostly affect the form and mixing of microplastics within the soil compartment.

It is possible to measure mass or abundance in some of the compartments without direct knowledge of the fluxes. This gives an idea of the status of microplastic pollution, but to assess the effectiveness of

specific mitigation options it is beneficial to quantify each flux in order to link it to the rate of microplastic accumulation in soil or other environmental compartments. This provides an understanding of the dominant processes. For instance, sludge from waste water treatment plants is known to contain microplastics as well as composts and manure (e.g. Li et al., 2018b). Furthermore, the fragmentation of films and fabrics and the application of seed coatings containing microplastics is thought to contribute to the emission of microplastics to agricultural soils as well. However, for each country and each region, the relative contribution of different potential sources will be different. Nevertheless, information on all significant fluxes, abundancies and mass of microplastics, specific for the relevant polymer types, is needed for sufficient exposure modelling and to provide valuable perspectives for mitigation and action.

6.3 Knowledge and information gaps

Although it is clearly a cause for concern that microplastics end up in soil and can induce adverse effects, there are many gaps in the current state of knowledge and the information needed to fully appreciate the behaviour, fate and effects of microplastics in soil systems. Some of the reviews stressed the need for action, because of the recalcitrance and persistence of microplastic particles, the current levels and trends of microplastic emission to soil, and the indications of adverse effects on the soil system (e.g., Ng et al., 2018). In the restriction on intentionally added microplastics, proper data was missing to perform of a quantitative risk assessment. But based on observations of the effects on microbiota and their persistency in the environment, sufficient information was available for RAC to consider the microplastics as non-threshold substances, meaning that all releases should be minimized (ECHA, 2020). At the same time, knowledge gaps can be filled in line with the source through pathway to receptor diagram in order to increase insights into and prepare for potential future human and environmental health risks.

The following gaps and visions were recognized in the reviews and previous chapters – overlapping partly with the highlights – as significant hurdles to better understanding the risks of microplastics in soil (e.g., Qi et al., 2020; Horton et al., 2017):

1. **Types and environmental concentrations**: Current knowledge about the extent of microplastic pollution in terrestrial environments is insufficient, although research is progressing. There are few regional and international studies focused on which polymers are most abundant and general data on how this varies across habitats, land uses and regions is lacking. Relevant data on microplastic concentrations in soils, as well as on agricultural soil management for the Netherlands specifically, is needed to better understand the current extent of microplastic pollution in Dutch soils.
2. **Auxiliary substances:** Auxiliary substances such as plasticizers (e.g. phthalic acid esters, PAE) and solvents are introduced into our soils together with microplastic particles. The potential effects of microplastic additives for soil ecosystems are unknown. Microplastics can also act as carriers for other pollutants, the so-

called 'Trojan Horse' effect. However, the meaning of this for risk assessment is still unknown.

3. **Analytical methodology**: There is a lack of robust, reliable, practical and standardized methods for sampling, separating, purifying and quantifying microplastic particles and polymers in solid matrices. These need to be developed to measure microplastics in different types of soils and to compare, combine and repeat scientific findings from different studies.
4. **Properties and behaviour**: There is currently a limited understanding of *how* environmental conditions and microplastic properties (chemical, physical and biological) affect microplastic behaviour and bioavailability for terrestrial environments. If the dominant or deterministic properties and conditions that cause the observed effects are understood, this can be used for emission, effect or exposure prevention and safe-by-design approaches.
5. **Release and emission**: Data on sources and emission is still patchy, meaning there is a lack of understanding of the overall relative and absolute contributions of microplastic emissions to the environment. Measures to mitigate microplastic emissions will be more effective if both relative and absolute fluxes (emissions) are better understood.
6. **Soil-microplastic interaction (fate)**: Insight is missing into the contribution of specific processes that are relevant for the fate of microplastics in soil. Studies on linkage, attachment and other interactions of soil aggregates with microplastic particles are needed to better estimate fate and exposure. Lessons can be learned from nanoparticles.
7. **(Bio)degradation (fate)**: A quantitative understanding in natural attenuation and degradation are pivotal for estimating the fate of microplastics in the soil environment under different scenarios. This understanding is currently insufficient and contradictory. The available methods need to be scrutinized for their suitability in determining the fate of microplastics in soil systems, explicitly making the difference between degradation and fragmentation. With persistent substances, (bio)degradation is negligible (nearly zero) or slow (some degradation observed) but the difference may be crucial for reliable modelling of microplastic fate and exposure.
8. **Effects and impacts**: To understand the impact and potential risks of the presence of microplastics in terrestrial ecosystems, data are needed on the exposure, bioavailability, uptake and dose-effect relationships (source-path-receptor). Data do exist and adverse effects have been identified, but the data are also scattered, limited and/or contradictory. Testing of microplastics that occur in the environment (weathered instead of virgin) is needed. Improved designs of studies and more reliable and specific data can, in turn, provide a better understanding of the (in)direct effects on public health, ecosystem resilience and ecosystem services.
9. **Long-term:** The environmental issue of microplastics is complex and entails several uncertainties, particularly for soil. It may take a lot of research to sufficiently underpin and build up scientific evidence for the adverse effects of different microplastics under

different conditions. Currently, however, it is certain that micro (and nano) plastics are present in soils and agricultural land worldwide and they will, if business continues as usual, be continuously and increasingly emitted. Due to these emissions, spreading and persistence, this is a persistent global contaminant whose long-term effects cannot be fully understood at this stage.

6.4 National and international projects

Several (inter)national programmes are addressing data and knowledge gaps. Microplastics in soil systems, often in agricultural land, are the focus of recent national and international (EU) projects with contributions from research at national institutes and universities. Presented below is a shortlist of current projects with representatives in the Netherlands. This list will not cover all scientific work that is currently being done. These projects are expected to address some of the knowledge gaps elucidated. The projects are:

- SoilVer platform: a soil and land research funding platform for Europe, previously known as SNOWMAN.
- MISSOURI (MIcroplasticS in Soil and grOUndwateR: sources, transfer, metrology and Impacts): a one year EU project with a focus on an interlaboratory survey of microplastics in soil and groundwater.
- SOPLAS Macro and Microplastic in Agricultural Soil Systems: EU Innovative Training Network, Marie Skłodowska-Curie Actions.
- LeonT (Low particle emissions and low noise Tires): a three year project focused on increasing knowledge (and the measurement) of particle and noise emissions from tyres—and their associated effects on public health and well-being—in order to propose effective and efficient mitigating measures through regulation, labelling and tyre design.
- PAPILLONS (Plastic in Agricultural Production: Impacts, Lifecycles and LONg-term Sustainability): an EU Horizons project to elucidate the ecological and socioeconomic sustainability of agricultural plastics (APs) in relation to the releases and impact of micro- and nanoplastics (MNPs) in European soils.
- MINAGRIS (MIcro an Nano plastics in AGRIcultural Soils): an EU Horizons project to assess the impact of plastic debris in agricultural soils on biodiversity, plant productivity and ecosystem services and their transport and degradation in the environment. It aims to provide tools and recommendations for the sustainable use of plastic in agriculture at the levels of farm and field for ensuring safe and economically viable food systems in Europe.

6.5 Looking for potential mitigation options

Some studies provided specific recommendations for policy development and land quality management. The need to act upon the prevention and reduction of microplastic accumulation is motivated by the persistence, the potential adverse effects and the widespread detection of microplastics (highlight 10). Preventing and reducing of the impact of microplastic soil contamination requires a large set of measures for a large variety of stakeholders. In several countries, measures are being implemented to reduce the release of (mostly primary) microplastics at

the source (source control) and to reduce emissions at end-of-pipe (emission control), both of which result in the reduction of emissions to terrestrial ecosystems.

The management of microplastics encompasses the definition of appropriate policy goals: avoid unacceptable risks to human health and ecosystems. These broad protection goals can be further supported by the ambition to move towards a situation with zero-pollution and avoid losses in a circular economy (highlighted in Chapter 1).

In order to advise on policies based on the risk paradigm, a human or environmental risk assessment needs to be conducted. Such a risk assessment can be conducted within the different regulatory frameworks already available for pollutants, but requires important adaptations in order to apply current methods to microplastics as a specific group of substances. The reason for this is the complex effects of microplastics, different from chemical substances, in that they have both a chemical and a physical identity (and potentially biological properties), as well as the difficulty in assessing the fragmentation and degradation of the microplastic particles. Risk assessment approaches for particles need to take their physicochemical characteristics and related effects into account (cf. nanoparticles).

Another approach is to limit the emission of microplastics at the source without considering the actual or potential ecological risk posed to the environment for specific types or groups of microplastics, thus minimizing and preventing further microplastic environmental pollution, and contributing to preventing losses in a circular economy.

Currently, the adverse effects of microplastics on the environment are not yet understood (Figure 1). Particles can, however, be prioritized based on other known criteria, such as their mobility in combination with their persistency, which are characteristics of fate and transport (Figure 5). Within REACH, the release of substances classified as PBT (persistent, bioaccumulative, toxic) and/or vPvB (very persistent, very bioaccumulative) needs to be minimized, with the goal of eventually finding substitutes for these substances. It is expected that substances may also be classified as PMT (persistent, mobile, toxic) and vPvM (very persistent, very mobile) within REACH in the future, as these traits are also considered undesirable. Similar minimalization efforts are likely. As microplastics have similar traits (being persistent as well as mobile in the environment), limiting the emissions could be justified without understanding the exact ecotoxicological effects and performing the conventional threshold-based risk assessment based on a case-by-case assessment. Microplastics are considered non-threshold substances in the restriction on intentionally added microplastics. Similar to PBT/vPvB substances under REACH, this means that all releases are assumed to result in a risk (ECHA, 2020).

Regardless of the exact policy goals, regulatory action will benefit from looking at the whole source through pathway to receptor route (Figure 1 and Figure 5), which allows for more refined mitigation options compared with prioritizing mitigation based only on information on the microplastic emission sources (Mitrano and Wohlleben, 2020).

Overall mitigation to reduce environmental contamination with microplastics can span a large set of measures, since plastics are applied in and originate from a great variety of production processes, applications and uses. Below we reflect on 'source-oriented solutions' to control microplastic release, and on 'end-of-pipe and remediation solutions' to manage microplastic emission to and fate and exposure in soils. Note that the options and measures described are not exhaustive and should be regarded as a starting point for further work on mitigation options when research gaps are filled.

6.5.1 Source-oriented solutions

There is a great variety in the sources of microplastics that end up in soils (Figure 5). To effectively address microplastic emissions, it is important to quantify sources of microplastics to determine the main contributors of the polymers in the environment. Measures to address these sources will likely have most effect. However, it also depends on the emission routes. A source may contribute heavily to the presence of microplastics in the aquatic environment, but to a limited extent in soils, e.g. when emissions enter the aquatic compartment directly. Because microplastics are found in all environments and microplastics can migrate to other compartments and are therefore often cross-border, an integral approach should be pursued to tackle microplastic emissions.

As mentioned before, microplastics found in soils can be divided between primary and secondary microplastics, the latter represents the largest share (see Chapter 3). Currently, knowledge about the magnitude of emissions is mainly derived from estimates and models, while data to support these numbers is very limited. Without reliable numbers, the effects of source control cannot be evaluated. Source control for both types are discussed below.

Primary microplastics

In the past, substantial steps have been taken to address the problem of primary microplastic emissions. One of the first initiatives was the 'Beat the Microbead' campaign, which stimulates producers of cosmetics to voluntarily remove microplastics from personal care[21] products. It also informs consumers about products that are microplastic-free. However, this will mainly impact the emissions to aquatic compartments as the sewage system is one of the main emission routes. More noteworthy is the upcoming ECHA restriction on intentionally added microplastics. The restriction, which according to the planned timetable should be adopted in 2022[22], restricts the placing of intentional (primary) microplastics in the EU/EEA on the market and affects the use of these microplastics in important sources of microplastic emissions to soils, such as products used in agriculture, horticulture and construction. A large decline in primary microplastic emissions is expected, but it should be noted that a number of derogations and the final definition of 'microplastic' may still result in the weakening of measures. Currently, it is unclear whether microplastics will have a lower limit for size; when a lower size limit is attributed, small particles may still be used in products. Another uncertainty is addressed in Chapter 4; biodegradable polymers are

[21] https://www.beatthemicrobead.org/
[22] https://echa.europa.eu/nl/hot-topics/microplastics

exempted from the restriction, but the tests to assess biodegradability were either not designed for microplastic particles or, if designed for plastics, their outcome cannot be compared to the existing persistence criteria. This rather complicates the biodegradability assessment.

Uses at industrial sites, in medicinal products for human or veterinary use, and substances or mixtures containing food additives are examples of derogations. For derogated uses, a reporting requirement will be effective, not of the amounts used (what was initially proposed), but rather the quantities released to the environment. (Downstream) users, excluding consumers and professional users, are responsible for delivering information on the use and emissions of microplastics to ECHA. In addition, suppliers should provide instructions on use and disposal, and the effectiveness of this information should be monitored. All information is used to assess the risks arising from microplastic uses in the future. From a regulatory perspective, action is already being taken to reduce primary microplastic emissions, but the restriction is not yet definitive and could be modified by the European Commission in the adaption process. When data becomes available, the effectiveness of the restriction can be assessed.

Secondary microplastics
Verschoor and de Valk (2018) estimated that microplastic emissions in catchments areas in the Netherlands mainly derive from litter fragmentation, tyre abrasion, and paint abrasion; three sources of secondary microplastics. In earlier research, distribution to different environmental compartments was estimated for emissions from paints (rural and urban areas) and tyres (Verschoor et al., 2016). Based on these estimations, a large proportion of the emitted microplastics end up in soils. For this compartment, it is therefore important to reduce microplastic emissions from these sources. Relevant measures known are discussed very briefly below. In addition, measures to reduce secondary microplastic emissions from sludge and compost, as well as from agricultural films and textiles, are discussed because these are widely recognized as sources in the literature.

Litter fragmentation
In recent years, regulatory measures have been taken against littering (of plastics). Examples of initiatives are the EU ban on single-use plastics (Directive (EU) 2019/904), the EU Plastic Bags Directive (Directive (EU) 2015/720) to reduce the use of lightweight plastic carrier bags, and the introduction of a deposit on small plastic bottles in the Netherlands.[23] All these initiatives were implemented during recent years. The direct results of most measures therefore will only become clear in the years to come. Implementation of the EU Plastics Bags Directive led to a reduction of 70% in issued plastic bags, yet 600 million plastic carrier bags were still issued in 2018.[24] It can be expected that these measures will not completely diminish intentional and unintentional littering, and it is questionable whether this can be achieved. Most of the additional solutions will therefore exist in end-of-

[23] Kamerbrief besluitvorming statiegeld flesjes en uitvoering moties blikjes. Kenmerk IENW/BSK-2020/64322.
[24] Rapport 'Evaluatie verbod op gratis plastic draagtassen'. https://www.rijksoverheid.nl/documenten/rapporten/2020/03/02/bijlage-1-evaluatie-verbod-op-gratis-plastic-draagtassen

pipe solutions (Chapter 6.5.2). In the design phase, there are some options to reduce the effects of microplastic emissions, e.g. the use of biodegradable polymers or the use of alternative materials that do not contain plastics. Another approach is to promote behavioural change by providing information and education. Behavioural interventions, education and raising awareness within the European Marine Strategy Framework Directive, as well as several campaigns[25] have already proved to be effective in reducing litter(ing).[26]

Paints

Recently, RIVM asked stakeholders in the paint sector which measures have been and could be taken to avoid microplastic emissions (Faber et al., 2021). The paint sector believes that polymers are needed in paints to sustain the beneficial features of current products. Relatively few few paint products are available without polymers and these are not (yet) suitable for all paint applications. Indirect measures, such as optimizing durability, have been realized by the paint producers, but the effects of these type of measures cannot be quantified.

The REACH restriction will have limited effects on the use of primary microplastics in paints. Film-forming, which occurs after paints are applied, is derogated as the physical properties are permanently modified and the microplastics are permanently incorporated in a solid matrix. In addition, the restriction does not address secondary microplastics, which is the main source of microplastics from paints. No information on types and amounts used will become available through the ECHA restriction, only the emission of primary microplastics. However, the European Strategy for Plastics in a Circular Economy does specify that policy options for reducing the unintentional release of microplastics from paint (and tyres and textiles) are to be developed.[27]

Tyres

Tyres are made of synthetic rubber. Both rubber and plastics are polymers and abrasion results in the formation of microplastics. Verschoor and de Valk (2018) have investigated the effects of measures to reduce emissions of microplastics. Although the focus was on emissions to water, the study recognizes that most emissions will end up in soils. A number of measures (e.g. legal threshold for tyre abrasion and tyre label with abrasion indicator) for source control were identified and reduction potentials were estimated. In 2020, the EU Regulation on the labelling of tyres was adopted (Regulation (EU) No 2020/740).[28] The need for measures to reduce microplastics, addressed by the European Commission[29], is mentioned in the Regulation. The regulation specifies

[25] e.g. Nederlandschoon (https://www.nederlandschoon.nl/zwerfafvalaanpakken/aanpakken/campagnes/) and Rijkswaterstaat (https://zwerfafval.rijkswaterstaat.nl/actueel/nieuwsbrief/2021/gratis-beschikbaar-campagne-plek-schoon/)

[26] Brief regering – Voortgang beleidsprogramma microplastics d.d. 25th of June, 2021. Tweede Kamer, vergaderjaar 2020–2021, 30 872, nr. 261.

[27] ANNEXES to the Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. A European Strategy for Plastics in a Circular Economy. https://ec.europa.eu/environment/pdf/circular-economy/plastics-strategy-annex.pdf

[28] Regulation (EU) No 2020/740, 2020. REGULATION (EU) 2020/740 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 25 May 2020 on the labelling of tyres with respect to fuel efficiency and other parameters, amending Regulation (EU) 2017/1369 and repealing Regulation (EC) No 1222/2009, Official Journal of the European Union p. 31.

[29] A European Strategy for Plastics in a Circular Economy.

that 'once reliable, accurate and reproducible methods to test and measure tyre abrasion and mileage are available, the Commission should assess the feasibility of adding information on tyre abrasion and mileage to the tyre label'. The additional information regarding tyre abrasion and mileage to end-users may reduce microplastic releases when durability becomes an important criterion for selecting tyres.

Agricultural films and textiles
Agricultural films and agricultural textiles have different functions in agriculture. Depending on the function and the environment degradation (or fragmentation), rates differ. In general, agricultural textiles have a longer lifespan than films. One of options to reduce the impact of these materials is to apply biodegradable films. However, until now, biodegradable films do not meet the criteria for mechanical strength. For a few years now, a biodegradability standard for mulch films has been available (see Section 4.3.1).

Infill of synthetic materials in soccer sport fields
Artificial turf sports fields are increasingly enriched with synthetic infill materials – often granulated rubber from tyres – in order to enhance performance and endurance. There is concern about the health and ecosystem risks from substances leaching from the particles. [30] Microplastic emissions are also an issue for these fields. Potentially, the use of these infill materials will also be banned by the upcoming restriction on intentionally added microplastics.

6.5.2 *End-of-pipe and remediation solutions*

End-of-pipe mitigation options can, in addition to source control, reduce emissions of microplastics to soil. Microplastics can be removed biologically, physically and/or chemically. As an ultimate mitigation option, soil remediation might be able to lower microplastic abundance, however this often comes with negative trade-offs, such as high-costs, high energy usage and (temporal) associated damage to the ecosystem. The best known methods to remove microplastics from a matrix are related to the aquatic environment. Examples are waste water treatment plants (WWTPs), the bubble barrier[31] and the shoreliner.[32] As examples of End-of-pipe measures two tyre wear related options and sludge and compost treatment are briefly discussed below. Although the authors currently do not know many pragmatic methods to remove microplastics from soils, some techniques for dealing with contaminated soil may also lead to a reduction in microplastics in soil, both for contaminated sites and for the reuse of (excavated) topsoil, briefly discussed below.

Tyre wear
Verschoor and de Valk (2018) have proposed two measures that may reduce emissions to the environment: 1) Replacement of top layer of asphalt of motorways by another type which retains more wear particles

[30] ANNEX XV REPORT: AN EVALUATION OF THE POSSIBLE HEALTH RISKS OF RECYCLED RUBBER GRANULES USED AS INFILL IN SYNTHETIC TURF SPORTS FIELDS Substance Names: Substances in recycled rubber granules used as infill material in synthetic turf. Submitter: ECHA. Version number: 1.01 Date: 28 February 2017. https://echa.europa.eu/documents/10162/13563/annex-xv_report_rubber_granules_en.pdf/dbcb4ee6-1c65-af35-7a18-f6ac1ac29fe4
[31] https://thegreatbubblebarrier.com
[32] https://www.tauw.nl/op-welk-gebied/circulaire-economie/shoreliner-plastic-soup-oplossing.html

and 2) street cleaning campaigns in urban areas. According to the calculations, both measures will have effect on a limited fraction (<25%) of the microplastic emissions to soil.

Sludge and compost
In the literature, sludge is also mentioned as an important source of microplastics in agricultural soils. In many countries, sludge from waste water treatment plants (WWTP) is applied as fertilizer. A large proportion of microplastics is removed in the WWTP and end up in this sludge. In the Netherlands, WWTP sludge is mostly burned as the sludge does not meet regulatory quality standards. In some cases, 'clean' sludge from WWTP can be used as fertilizer (e.g. some WWTP of food manufacturers). As the processed materials are food-grade, it is expected that these sludges will not contain (substantial amounts of) microplastics. However, no standards are available for microplastics in food while recent research has demonstrated that plants are able to take up microplastics (Li et al., 2020b) and food products can contain microplastics (Rainieri and Barranco, 2019). In contrast to WWTP sludge, compost is applied by farmers and consumers and can contain microplastics. Although (macro)plastics are removed as far as possible, in the Netherlands, microplastics can still be found in compost (van Schothorst et al., 2021). The sources of microplastics in compost are most likely to be plastic materials that are discarded by households. In the past, some issues were identified with compostable plastic materials; these materials were labelled as compostable while the Dutch compost installations are not able to degrade these materials. Without prior removal, these plastic materials can fragment and in time result in the formation of microplastics.

Soil remediation of a microplastic-contaminated site
Since 1987, soil protection, soil quality management and remediation have been integrated into Dutch legislation. Many soil remediation techniques and soil management schemes have been developed to reduce the risks of contaminated sites for human and environmental health (Pijls and Marnette, 2020). Some of these will be applicable in the effort to reduce microplastics in soils as well. For instance, excavated soil can be treated with heat, which may result in oxidation of microplastics (Brand et al., 2021). However, soil remediation often comes with negative trade-offs, such as high-costs, high energy usage and (temporal) associated damage to the ecosystem. Trade-offs should be weighed against each other to determine whether remediation is preferred. Contaminated soils can be transferred to a waste disposal site, can be applied under controlled conditions or can be treated, for instance, with heat or incineration in order to end up as thermally cleaned soil. Additionally, soils can be treated on the spot, although available remediation techniques might not be feasible to remove microplastics. One remediation technique that does not remove microplastics, but may lower their impact, is covering contaminated soil with a layer of clean topsoil, a so-called living layer. Such a layer limits unwanted exposure of the ecosystem to microplastics. Concerning microplastics, no standards have been included in Dutch soil regulations. Microplastics are not included in the list of substances for soil management and investigations. This means that microplastic

contaminated soil can only be remediated on the basis of the 'due diligence' (in Dutch: 'zorgplicht') article in the Soil Quality Decree.

Restrictions on the reuse of microplastic-contaminated soil
Parallel to the development of remediation techniques, restrictions on the reuse of excavated soil were developed. Slightly polluted soil can only be reused when the level of the contamination and the sensitivity of the land use do not lead to an unacceptable risk for human health and the ecosystem. This can be made instrumental through so-called soil quality criteria for the reuse of soil containing microplastics. This is described in the next section.

6.6 Pathway to quality criteria for microplastic in soils

A soil quality criterion (e.g. standard) is a policy instrument that contributes to a sustainable management of soil systems. With soil quality standards, the protection of ecosystems and human health as the two overarching endpoints can be put in place. However, the current state of the art in microplastic research is insufficient for reliable criteria setting. There is an urgent need for reliable and practical analytical methods for microplastic determination in soils, and for data on the impact of microplastics on different ecosystem parameters.

Today there is a growing awareness about the harmfulness of microplastics for human health and ecosystems. Societal concerns are about the potential effect that these large-scale, accumulating substances can have on ecosystem integrity and human health. Consequently, there is a plea for action in reducing the emission of plastics and particularly microplastic to vulnerable ecosystems, including the soil. We summarize current policies to arrive at (criteria for) acceptable levels of plastics in soil in the Netherlands and Flanders.

6.6.1 Criteria for microplastic in soil and sediments

6.6.1.1 Netherlands

An excavated top layer of soils and sediments generally contains materials such as timber, rubble and manmade fragments, and other evidence of human activities. In many cases, these materials, although not entirely from a natural origin, do not pose problems for ecosystem quality or human risks. Nevertheless, it is necessary to limit these materials to a certain level. In the Soil Protection Act of the Netherlands, this is incorporated in Article 34 of the Soil Quality Decree[33] and the second part of Article 1.1. in the Directions for Soil Quality.[34] All substances that are intentionally added to the soil or sediment are excluded from the Soil Quality Decree.
When applying soil or sediment, a maximum of 20% (w/w) of non-natural material may be present: i.e. wood and stony fragments. These fragments must already be contained in the soil or sediment before excavating the soil or collecting the sediment, and it must not be possible to remove these materials. Under the act, local authorities can apply stricter maximum percentages in local legislation.
Substances other than wood or stony fragments, such as plastic and expanded polystyrene, may only be present when it is "sporadically"

[33] Besluit bodemkwaliteit: https://wetten.overheid.nl/BWBR0022929/2021-01-01
[34] Regeling bodemkwaliteit: https://wetten.overheid.nl/BWBR0023085/2021-01-21

present in the soil or sediment before excavating and harvesting, and only when it is not reasonable to demand clean-up of the soil or sediment before application. The term 'sporadically' is not stringently defined. The term is explained in the overview of changes to the Soil Quality Regulation.[35] In that context, the term 'sporadically' indicates that small amounts of other foreign material may be present in the excavated soil or dredging sludge to be used, because this cannot always be prevented. The term sporadically was chosen as there are no precise criteria, such as a certain percentage by weight, to determine the maximum amount of other foreign material that may be present in excavated soil or dredging sludge. (Expanded) polystyrene, for example, is a light material for which the percentage by weight is not a suitable criterion. In addition, the content of this term is partly determined by what can reasonably be removed during careful excavation or before application. In practice, however, it is not necessary to define boundary limits for foreign materials; what is important is that effective action can be taken in cases where it is evident that more than sporadic foreign matter occurs in soil or dredging sludge.

In the policy letters of the Ministry of the Environment, VROM and IenW[36], it is stated: *"… this means that non-natural substances or materials other than woody and stony fragments in the soil are not allowed, such as plastic and expanded polystyrene"* and in the Soil Quality Decree[34] and the Guidance on soil quality[35]: *"The amount of plastic should be restricted to a minimum. This is accomplished through a visual estimation."* Note that this visual examination is not meant to accurately assess microplastic content, but is used as a rough inspection method.
When the criteria cannot be met, application of the excavated soil and sediment is restricted by the Soil Quality Decree. As a solution to overcome the restriction, soil sieving is proposed to reduce the number of artificial substances. This solution may help for the larger wood and stony fragments but will not be practically applicable to remove the majority of microplastic in contaminated soils or sediments.

The second policy letter (IenW 2018 (37)) was added by the government because of multiple applications of contaminated soil in deep lakes with large amounts of plastics. This is in violation of Article 7 (due diligence or "Zorgplicht" article) in the Soil Quality Decree.

6.6.1.2 Flanders

In Flanders the policies regarding excavated, microplastic-contaminated soils are similar to those in the Netherlands (OVAM/VLAREBO)[37]. There are no practical criteria for microplastic-contaminated soil. However, there is a quantitative criterion for non-natural materials other than stones or stony fragments. These substances include plastic and microplastic: the maximum acceptable level in soil and sediment is 1% (w/w) and 1% (v/v) in mass percentage and volume percentage, respectively.

[35] https://zoek.officielebekendmakingen.nl/stcrt-2018-68042.html.
[36] VROM (2010) Kamerbrief 30 015, Voortgang bodemsanering Nr. 39, brief van de Minister van Volkshuisvesting, Ruimtelijke Ordening en Milieubeheer, Den Haag, en IenW (2018) Kamerbrief, betreft Gezamenlijke aanpak plastic zwerfafval. Kenmerk IENW/BSK-2018/232541 Datum 6 november 2018, Den Haag.
[37] VLAREBO (2008) Bodemsanering. Besluit van de Vlaamse Regering houdende vaststelling van het Vlaams reglement betreffende de bodemsanering en de bodembescherming, afd. III Voorwaarden voor het gebruik van bodemmaterialen. Gebruik van bodemmaterialen als bodem. art 161. https://ovam.be/bodemdecreet-en-vlarebo

6.6.2 *Towards soil quality criteria for microplastics*

At the moment, it seems difficult to establish soil quality criteria for microplastics which are feasible in practice and which are perceived to have only limited risks for humans and ecosystems. There are various issues which need improvement before deriving these standards:

- Improved analytical instruments, (i) for separating polymers from the soil matrix and (ii) for robust and practical quantitative analyses of all types of polymers and (iii) that can practically and reliably determine the shape and dimension of aggregates (see Annex 3 for more information).
- Data that can support the quantification of the fate of microplastics in the environment – polymer and aggregate dependent – such as natural attenuation and transport processes.
- Data on the effects of polymers on ecosystem parameters beyond the standard tests for substances and that are meaningful for ecological endpoints (species, processes and integrity) under realistic field conditions and ready for a risk assessment.
- Data on the impact of microplastics on human health.

Considering the current interest of the scientific community in microplastic research, as seen from the exponential increase of publications on the subject in the last five years, it is expected that some gaps can be partly covered with new innovations in the near future.

7 Conclusion and Recommendations

7.1 Conclusions

Microplastic accumulation in soil systems is a complex phenomenon, due to the heterogeneity in composition and dynamics of both soils and microplastics, and the large amounts emitted, combined with slow or absent degradation. In contrast to air and water, the soil system generally reacts slowly to environmental pressures and recovery is even slower. Combined with obstacles from inadequate analytical techniques, it is no surprise that there is a substantial gap in our knowledge. We are not yet able to perform a proper risk assessment of microplastics in soil for human health or ecosystems.

A particular gap in our knowledge is related to the fate and behaviour characteristics of microplastics in the soil environment, i.e. natural attenuation, especially the processes of fragmentation and (bio)degradation. The pragmatic definition – that microplastics encompass (mainly) non-biodegradable polymers – can be understood from a precautionary point of view, but it is not useful for constructing a risk-based perspective on what the accumulation of polymers means. Secondary microplastics often originate from the fragmentation of meso- and macro-plastic particles. In dedicated experiments, many polymers have been demonstrated to be (bio)degradable, albeit often at slow rates. Still, microplastic particles containing additives and having specific sizes and shapes might display different (bio)degradation potential in the environment. Understanding degradation is important for understanding microplastics in soils. A further use of biodegradable plastics can be part of the solution to reduce the accumulation of microplastics in soils. However, if plastics and polymers are not degraded completely or fast enough under environmentally relevant conditions, microplastics will continue to accumulate.

A suite of degradation tests is available. Some tests have been designed specifically for (micro)plastics, but their outcome is difficult to compare with the available persistence criteria, while tests designed for substances need to be scrutinized for their usability to assess microplastic particles. Furthermore, screening level tests determine (bio)degradation by measuring carbon dioxide formed during the complete break-down (taking into account that a part is converted into biomass) under standardized conditions. In a simulation test, primary degradation can be monitored, but it remains complicated to distinguish between the intact polymer, shorter pieces of polymer, oligomer and monomers. The biotic and abiotic degradation processes resulting in partial degradation need to be explicitly quantified, as this can result in a major source of microplastics. In addition, these processes may foster biodegradation. To what extent these processes and different environmental conditions contribute to degradation, has yet to be investigated.

For modelling of the fate and behaviour of microplastics in soils and cross-cutting environmental boundaries, tools are available, but reliable data to run the models are missing. For instance, models for describing

general particle behaviour are fit for use, although some adaptation to microplastics might be needed, e.g. the inclusion of the fragmentation process.

The conceptual model produced in this report can help organize the information needed to mitigate potential risks to acceptable levels. Firstly, fluxes of microplastics and polymers should be quantified for different sources, (e.g. by measuring release and modelling emissions), and for different soil types and locations (e.g. based on land use and scale: site, a region or continent). This will also improve our knowledge about potential successful mitigation options. At the moment, the 'source control' mitigation options are expected to be more effective than 'end of pipe solutions'. Nevertheless, in a few restricted serious cases, remediation of a site followed by a soil treatment can be opted for.

Secondly, sufficient exposure and toxicity data for human health and ecosystems are absent and therefore it is not possible to perform a risk assessment and to arrive at reliable risk-based soil quality criteria. We need research that pragmatically solves the problem of the complexity of microplastics' fate, behaviour, exposure and toxicity for different soil systems and organisms. We need to work towards a generic approach whereby we study which biological, physical and chemical properties, under which environmental conditions, are responsible for any observed effects. This is only possible if reliable analytical methods are developed to quantify microplastics in soil. This should pave the way for deriving soil quality criteria for (micro-)plastics.

As can be concluded from the trends in the literature, research on microplastics in soil is increasing rapidly and many national and international projects have recently started or will be started soon; many of them with a focus on agricultural soils or on large emissions such as from abrasion of tyres. Therefore, the harmonization of methodologies and data reporting, as well as a common language and overview, is of utmost importance to ensure that new results can be validated and made useful.

7.2 Recommendations and reflections

The following recommendations and reflections were articulated:

Prioritization and filling of knowledge gaps
There is a lack of data and knowledge of almost every aspect on the issue of microplastics in the soil system. Although some initial data have become available, the way to interpret the data for application in risk assessment, or for effective emission reduction, is still a big challenge. It is important to deal effectively with the complexity of microplastics in soil systems, for instance with over-arching approaches, and to set priorities in the details because time and resources are limited.
For more accurate modelling, there is a need for practical, robust, affordable and sensitive analytical methods that take polymer type, form and size into account. For a better understanding of the sources, emissions, fate and behaviour, and for fuelling the risk assessment schemes, interpreted data on microplastics are needed with respect to source, emission, transport rate, polymer type, size and shape, natural

attenuation and (bio)degradation rate in order to establish realistic exposure scenarios for relevant end-points. For the latter specifically, tests have to be scrutinized and optimized to assess the biodegradation of microplastics.

For the hazard estimates, we need data on direct and indirect (toxic, food chain and other) effects of microplastics on ecosystem elements, such as species, processes, resilience and resistance phenomena. Data are especially lacking on the toxicity of 'environmental 'microplastics which have been exposed to physical and chemical stressors, rather than the fabricated particles which are generally tested.

Quantifying sources
Quantitative information on emissions from specific sources is relevant for the prioritization of policy management. Emission modelling by linking human activities (e.g. sawing or driving) to the application of plastics (pipes or tyres) is already applied to estimate secondary microplastic release (Sieber et al. 2020; Kawecki and Nowack, 2019). To obtain a more complete overview for the EU, there is a need to (i) expand these modelling approaches to the whole EU (i.e. studies are for Switzerland only), (ii) extend the modelling with other important release routes, such as 'natural' incomplete fragmentation of plastic products discarded in the environment or applied in the environment for long periods (e.g. building materials) and (iii) include other (potentially) relevant sources coupled with activities that induce wear (e.g. paints and sanding).

Policy and management framework
We have seen during this study that, although data can be scattered and a better overview is needed, results show that microplastics are widespread in soil systems. While improving our knowledge and understanding, steps can be taken now to minimize and mitigate emissions throughout the environment. We thus recommend exploring the possibilities for a policy and management framework for (micro)plastics in soil systems (or broader) in order to minimize microplastic emission where possible and to mitigate ecological and human health risks where needed. To this end, collaboration and knowledge exchange with a wide range of stakeholders is required. Our conceptual model in Figure 5 (see Section 6.2) can help to start discussions, obtain an overview and prioritize microplastic sources and routes that can be addressed. The framework should be sufficiently versatile to accommodate all the different aspects of microplastics in the soil system, including their behaviour as particles, source control for zero emission, source control for avoiding emergent risks, or the mitigation of ecosystem and human health effects. tTo establish such a framework with all stakeholders, requires a multi-actor, multi-effort type of collaborative work.

A long-lasting environmental issue
The environmental issue of microplastics in soils is complex and has many uncertainties, in particular for soil. It may take muchresearch to sufficiently underpin and build up scientific evidence. It is, however, certain that micro- (and nano)plastics are present everywhere in urban, natural and agricultural soils worldwide and that, due to their persistence, spreading, emission rates, and society's dependence on

plastic, microplastics will be a contaminant in our environment for a long time. Steps to mitigate and reduce microplastic emissions to the environment can be taken to prevent pollution, while improving our overview and understanding.

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Annex 1 Abbreviations of polymer types

EPA	expanded polystyrene
HDPE	high-density polyethylene
LDPE	low-density polyethylene
LLDPE	linear low-density polyethylene
MDPE	medium density polyethylene
PA	poly amide/nylons
PAE	phthalic acid esters
PBS	polybutylene succinate
PBT	polybutylene terephthalate
PCL	poly-ε-caprolactone
PE	polyethylene
PEC	polyester carbonate
PES	polyethylene succinate
PET	polyethylene terephthalate
PHA	polyhydroxyalkanoate
PHB	poly-3-hydroxybutyrate
PHBV	poly-3-hydroxybutyrate-co-3-hydroxyvalerate
PLA	polylactide
PP	polypropylene
PPL	polypropiolactone
PS	polystyrene
PUR	polyurethane
PVC	polyvinyl chloride
SBR	styrene butadiene rubber

Annex 2 Biodegradation of microplastics

A2.1 Test methods to assess biodegradation within the proposed restriction on intentionally added microplastics

Table 4 Overview of the test methods proposed by the ECHA restriction focused on intentionally added microplastics to determine biodegradability. Note that not all information is given regarding the test methods. For detailed information, see Table 22 of the Background Document to the Opinion on the Annex XV report that proposes restrictions on intentionally added microplastics, which is the latest document published regarding the restriction (ECHA, 2020)

Group	Determines	Pass criteria	Permitted test methods
1	Ready bio-degradation	60% mineralization measures as evolved CO_2 or consumed O_2 in 28 days	1. Ready biodegradability (OECD TG 301 B,C,D,F) 2. Ready biodegradability – CO_2 in sealed vessels (OECD TG 310)
2	Enhanced / modified ready biodegradation	60% mineralization measured as evolved CO_2 or consumed O_2 in 60 days	1. Ready biodegradability (OECD TG 301 B,C,D,F) 2. Ready biodegradability – CO_2 in sealed vessels (OECD TG 310) 3. Modified biodegradability in Seawater (OECD TG 306)
3	Inherent bio-degradation	≥ 70% mineralization measured as O_2 within 14 days	1. Inherent biodegradability: Modified MITI Test (II) (OECD 302C)
4	(Bio)degradation relative to a reference material	Ultimate degradation of ≥ 90% relative to the degradation of the referen-ce material (60%) within: 1. 6 months in aquatic tests (O_2 consumed) or 2. 24 months in soil (CO_2 evolved or O_2 consumed) or water/sediment tests (CO_2 evolved)	1. Ultimate aerobic biodegradability in an aqueous medium (EN ISO 14852:208 or EN ISO 14851:2004) 2. Aerobic biodegradation in sweater/sediment interface (EN ISO 19679:2016 or EN ISO 18830:2016) 3. Ultimate aerobic biodegradability in soil (EN ISO 17556:2019) 4. Aerobic biodegradation when exposed to marine sediment (ISO 22404:2019
5	Half-life in the environment (under relevant environmental conditions)	1. The degradation half-life in soil is less than 180 days or 2. The degradation half-life in marine, fresh or estuarine sediment is less than 180 days or 3. The degradation half-life in marine, fresh or estuarine water is less than 60 days	1. Aerobic and anaerobic transformation in soil (OECD TG 307:2002) 2. Aerobic and anaerobic transformation in aquatic sediment systems (OECD TG 308: 2002) 3. Aerobic mineralization in surface water – simulation biodegradation test (OECD TG 309: 2004)

Composting of plastics

Table 5 Overview of continental and national certification schemes for plastics compostable at home. Sources: Bastioli (2020); European Bioplastics (2016); Standards Australia (2010); TUV Austria (2019)

Standards	**Country/ continent**	**Target/ product**	**Test criteria**	**Pass criteria**
OK compost HOME	Europe[a]	Plastics	T: 20-30°C Duration: 12 months	Absolute or relative 90% biodegradation (similar to EN 13432, see above)
NF T 51-800:2015	France	Plastics	Very similar to OK compost home	
prEN 17427 (2020)	Europe	Carrier bags	Unknown[b]	Unknown[b]
AS 5810:2010	Australia	Plastics	T: 25±5°C Duration: 12 months	90% biodegradation absolute or relative to suitable reference substance
UNI 11355:2010 and UNI 11183:2006	Italy	Plastics	Very similar to OK compost Home and AS 5810	

a: The label originated in Belgium at the testing institute Vinçotte.
b: The information could not be retrieved from open sources.

A2.2 Microbes and biodegradation

Strains able to degrade plastics have been reported for various environmental compartments, including soil. For example, a polyethene (PE) deteriorating *Aspergillus terreus* strain was isolated from a mangrove tree's (*Avicennia marina*) rhizosphere (Sangale et al., 2019). This strain was reported to be a highly efficient PE deteriorating fungus, reducing the amount of PE microplastics present in the test system by more than 50% at pH 9.5. Other reports of PE degrading microorganisms include isolates from waste coal, a forest and a crater in Poland (Nowak et al., 2011). The dominant microorganisms in waste coal and crater soil were bacteria (*Bacillus mycoides*, *Acinetobacter baumannii*a and *Pseudomonas fluorescens*), while in forest soils filamentous fungi (*Staphylococcus cohnii*) were detected. Experiments with these microorganisms resulted in a weight loss of 0.13% to 17% of the modified PE film within 225 days. Hiraga et al. (2019) isolated microbes from landfills that apparently can synthesize PET under laboratory conditions. Auta et al. (2018) reported that a *Bacillus* and a *Rhodococcus* strain were isolated from mangrove sediments that could degrade PP. The structure and morphology of PP was altered following exposure to the bacteria, as was shown with FTIR and SEM analyses. After 40 days of incubation, weight loss of 4.0 and 6.4% was observed for the bacillus and *Rhodococcus* strains,

respectively. It should be noted that the studies discussed above are just a few examples of microorganisms that have been isolated from soils and that have the potential to degrade different types of plastics, e.g. PE, PET and PP. Many more examples can be found in the literature. It remains unclear though whether these strains also effectively degrade (micro)plastics in the environment, where suboptimal conditions occur and where more readily available carbon sources might be present.

A2.3 Determination of degradation rates

Degradation rates are generally expressed in terms of unit loss per unit time. Theoretically, the rate can be described as the loss of weight, size, polymer structures or other parameters, while it could also be described as a gain in breakdown products, for example, monomers or CO_2.

It is likely that the degradation of microplastics is largely dependent on the available surface area (Chamas et al., 2020). Therefore when it is assumed that degradation only occurs at the surface of a polymer, degradation can be expressed as a specific surface degradation speed ($k_{a.deg}$) in m/s, such that m is the depth of the microplastic (perpendicular to the surface) which is degraded after a certain period of time (s). Note that the (surface) degradation speed can also be expressed as a rate constant (e.g. s^{-1}), which is discussed later on. The $k_{a.deg}$ will differ between polymer types, based on the susceptibility of the material to degradation, but also between similar polymer types due, for example, to additives and environmental conditions that hamper or accelerate degradation. The amount of mass lost over time depends on the density of the polymer (kg/m^3) and the surface area (m^2). Multiplying these parameters with $k_{a.deg}$ will result in the mass-based degradation rate (kg/s). This is the basic approach that can be taken when assuming a constant surface area, for instance for a thin flat surface. However, the surface area of most shapes of plastics will change over time and therefore degradation of most plastics cannot be described by a constant mass-based degradation rate in time.

The recent study conducted by Chamas et al. (2020) elaborated on the surface-based degradation of microplastics and described the degradation rate and time to complete degradation for MPs with a constant surface area (flat, thin plastic), spherical (microbead) and cylindrical (fibre) shapes. By including a few assumptions, the authors derived the following equations to determine the degradation time:

$$t_d\, constant\ surface\ area = \frac{V_0}{k_{a.deg} SA}$$

$$t_d\, spherical = \frac{3}{6^{2/3} k_{a.deg}} \sqrt[3]{\frac{V_0}{\pi}}$$

$$t_d\, cylindrical = \frac{1}{k_{a.deg}} \left(\frac{m}{\pi \rho h}\right)^{1/2}$$

With:
t_d = complete degradation time (s)
V_0 = volume start (m^3)
$k_{a.deg}$ = surface degradation speed (m/s)
SA = surface area (m^2)
m = mass (kg)
ρ = density (kg/m^3)
h = height (m)
* For t_d cylindrical it is assumed that SA = 2 πrh instead of SA = 2 πr (h+r).
** Note that the $k_{a.deg}$ can vary over time, which is not included in the model equations.

Because a mixture of microplastics is present in the environment, modelling degradation will be complex. The above formulas can however be used to estimate the degradation times of individual plastics. To do so, physical properties are necessary, but also the $k_{a.deg}$. Chamas et al. (2020) has published an overview of $k_{a.deg}$ values found in the literature for different plastics in different environmental conditions. Based on the available data, $K_{a.deg}$ values can vary up to a factor of 1,000 for a type of plastic in an environmental compartment (e.g. soil or water). It is expected that the variation in $k_{a.deg}$ may largely be allocated to the different test conditions, especially as the results for landfill, compost and soils are presented jointly. Under these environmental conditions, the range of specific degradation rates seems mostly to be large for LDPE and other plastics, although for these plastics also the most data points were available. As the results are presented jointly for plastics containing fillers and/or tested under accelerating conditions and for those not, it is unclear what the individual effects of these are. On average, the $k_{a.deg}$ for plastics in landfill/compost/soils seems to be around 5-10 µm/year. In Table 6, $k_{a.deg}$ values are given for each plastic type (Chamas et al., 2020). For many plastics no data was retrieved, while for some types of plastics no degradation was observed. It is argued that the lack of degradation is likely a result of the testing conditions. For plastics, generally long periods of time are needed to degrade, which corresponds to low $k_{a.deg}$ values.

Table 6 Estimated specific surface degradation rates for different plastics (min-max, in µm/year) in landfill/compost/soil under normal buried conditions or under influence of UV/heat. 0 = no degradation was observed. – = no published data was found. From Chamas et al. (2020)

	Estimated specific surface degradation rates (µm/year)	
Polymer type	*Buried*	*Accelerated by UV/heat*
PET	0	–
HDPE	1.0 (0.91-1.11)	1.3 (0.55-2.6)
PVC	0	–
LDPE	11	22 (1.6-83)
PP	–	0.51
PS	0	–
Others	270 (20-1400)	320

Several of the currently available fate models (Besseling et al., 2017; Meesters et al., 2014) that are fit for use with microplastics are based

on first-order rate constants for describing the different environmental fate processes, such as degradation. First-order degradation rate constants (s^{-1}) for each type of polymer can be estimated by using the specific surface degradation rates (zero-order degradation rates) to calculate the degradation of a microplastic over time. As an example, a microbead particle is used. The equation to calculate the complete degradation time of a microbead (t_d spherical) takes into account that the particle has completely shrunk over time. Chamas et al. (2020) also formulated the equation to determine the mass of a particle as a function of time (see below).

$$m_t = \left[(m_0)^{1/3} - \frac{6^{2/3}}{3} k_d (\pi\rho)^{1/3} t\right]^3$$

With:
m_t = mass at time = t (kg)
m_0 = mass at time = 0 (kg)
ka.deg = specific surface degradation speed (m/s)
ρ = density (kg/m^3)
t = time (s)

This equation assumes that degradation of the particle increases over time. By modelling m_t for an initial period over time (e.g. until m_t = 0.5*m_0), the slope of the decrease in mass can be used to determine the degradation rate constant.

In the case of a HDPE microbead of 8mm, the following input parameters can be used:
Size: 8 mm
Volume: 2.9 cm^3
ρ: 0.950 kg/m^3
m_0: 0.00275 kg
$k_{a.deg}$: 1.0 µm/year (average $k_{a.deg}$ for HDPE in soil/landfill/compost, from Table 6).

As a result, a slope, and with that a first-order degradation rate constant, of 1E-11 s^{-1} can be estimated for the first part of the degradation in time.

Although such a degradation rate constant provides insights into the degradation rate of an individual particle in a very generic manner, a number of aspects have to be taken into account when applying such a constant in modelling:

- Rate constants are polymer-type-specific and dependent on $k_{a.deg}$. (Estimations for) $k_{a.deg}$ are currently not yet available for all type of polymers in soils, while for some polymers limited information is available.
- Calculated degradation rate constants depend on the different initial particle characteristic which used as input, such as size, density and mass (the latter of which is related to both aforementioned characteristics).
- Besides the physical particle characteristics used as input, other physical/chemical characteristics (e.g. presence of additives,

smoothness of the surface) can also influence degradation and should be considered.
- The calculations do not take into account changes over time. The specific surface degradation rate can vary over time (e.g. as a result of changing environmental conditions or changing surface area), which is not captured.
- The specific surface degradation of each polymer depends on the environmental conditions. As seen in Table 6, $k_{a.deg}$ can vary to a great extent between studies. Note that in addition to environmental factors, particle characteristics could also have influenced the results.

Annex 3 Methods for extraction, separation, identification and quantification of microplastic in soil

A3.1 Introduction

There is no standardized method for separating microplastics from a soil matrix, nor a standardized analysis and identification protocol. In this Annex we provide a brief overview of the methods that have been used in research for different steps of measuring microplastics in complex environmental samples, with a special focus on their applicability for the analysis of soil samples. These methods are described in more detail in other reviews, amongst others by Bläsing and Amelung, 2018; Zhang et al., 2018; Li et al., 2020b; Ruggero et al., 2020; Möller et al., 2020.

Some steps have several possible methods to use. Each method has its benefits, its specificity for certain polymers, differential applicability in solid matrices and uncertainty.

The biggest hurdle to face is the extraction and clean-up method. This is a challenging step because of the presence of various, abundant and complex organic substances in the soil organic matter fraction.

Commonly used techniques are based on the extraction of microplastics with a salt solution based on density differences between the microplastics, the other soil constituents and most of the organic matter. Different salt solutions can be used for different polymers. With saturated NaCl, the light polymers can be extracted, while with NaI or NaBr, for example, the whole range can be extracted. It is, however, difficult to distinguish organic compounds from microplastics by conventional salt solution density separation, irrespective of the soil type and structure (Li et al., 2020a). For samples with organic matter such as soil and sediment samples, a digestion step is often required (Ruggero et al., 2020).

The second hurdle to overcome is to find a suitable analytical instrument for different types or purposes (reviews e.g., by Ruggero et al., 2020 and Möller et al., 2020). Depending on the scope of a study, one method may be more suitable than another. For monitoring purposes, determining the mass quantities of polymers or volumes may be the best way to measure concentrations in the field. Whereas a toxicity laboratory test may require another analytical methodology to determine elasticity and aspect-ratio (both related to potential uptake). Currently, fewer methods are available to sufficiently provide insight into the morphology of microplastics. There is no one method that fits all purposes.

A3.2 Sampling

Soil sampling is the first step in the extraction of microplastic from the environment. Proper soil sampling is very important to obtain representative information, but this is unfortunately sometimes overlooked. Different strategies exist to obtain reliable information from a qualified sampling approach. Möller et al. (2020) summarized the main characteristics of different approaches:

- Judgmental sampling. This is based on pre-existing information, very effective but for that reason subjective.
- Simple random sampling. Effective and flexible with rather homogeneous sites. However, microplastics are often not homogeneously distributed, neither are the covariables in the soil system.
- Systematic grid sampling. Simple to implement, but sometimes too inflexible.
- Unaligned grid sampling. Like grid sampling, but with more flexibility to select sampling locations within a grid cell.
- Transect sampling. This method allows a systematic identification of the distribution of microplastic pollution in straight lines, i.e. according to the distance to a source, parallel to roads and streams, and other gradients in the field.
- Stratified sampling. Based on a division of the sampling area into smaller areas with homogeneous features. This is a recommended approach with significant different strata, where microplastics are relatively homogenously distributed compared to the distribution in the area as a whole.

A dedicated sampling approach is required to obtain a representative set of samples for each type and size of the landscape and for each type of microplastic with respect to polymer composition and sizes of the particles. Very often, constraints in budgets hamper the collection of a robust set of samples with sufficient statistical power to detect the impact of pollution gradients. Sometimes a combination of samples in a composite sample may be a solution in order to reduce the number of samples, without losing the representative nature of the information obtained. However, there is a danger here that small differences between samples disappear and some pollution hot spots may be missed as a result.

More details on sampling and the preparation of samples can be found in the literature (viz. Möller et al., 2020).

A3.3 Extraction

A3.3.1 Introduction

For an analysis of microplastics in complex environments (Möller et al., (2020), it is recommended that the bulk of the solid matrix be removed, preferably isolating the microplastic particles from the matrix and removing adhering substances. For inhomogeneous solid samples such as soils, microplastic extraction is challenging, especially with decreasing grain size of the soil texture (clay) and microplastic particle sizes (Crawford and Quinn, 2017). Soil particles can form relatively stable aggregates, which may enclose microplastic particles and obscure them from analysis (Zhang and Liu, 2018; Blӓsing and Amelung, 2018). For this reason, developing a method for soil aggregate dispersion without risking the destruction or artificial fragmentation of microplastic particles is an important first step for the microplastic analysis in soil.

A3.3.2 Manual extraction

The crudest method for microplastic extraction is sieving and hand sorting, using a stereo microscope to exclude obvious mineral or biogenic matter, such as particles with visible cell structures. This

method can be combined with the 'hot needle test' to further confine the number of putative microplastic particles. However, manual sorting and visual identification is extremely time and labour intensive, restricted to sizes >500 µm and is highly prone to misidentification and bias, rendering a subsequent reliable polymer identification indispensable.

A3.3.3 Electrostatic separation

A rather novel methodology used is the electrostatic separation of microplastic particles from solid matter. While experiments conducted by Hidalgo-Ruz et al. (2012) in this direction failed, Felsing et al. (2018) managed to modify a smaller but fair demonstration device of an electrostatic separation unit commonly used in the recycling industry to isolate microplastics from sand and sediment samples. The method allows a relatively high sample throughput with a mass reduction of up to 99% and recovery rates of 90−100% for pristine microplastic particles ranging from 63 µm to 5 mm. However, to achieve these high recovery rates, the samples underwent the procedure three times, with a temporal effort of 3 to 4 h per 150 g sample. Furthermore, the authors claim that the method is independent of organic matter content, particle density, shape, age, or biofouling. However, this method is not suitable for moist samples (Felsing et al., 2018) and its suitability for cohesive soil samples is questionable, due to the unavoidable formation of aggregates. Furthermore, the applicability for very small particles must still be verified, as the force of adhesion to the metal drum and scraper may be higher than the gravity-force, possibly leading to significant losses of the small microplastic fraction in the final sample.

A3.3.4 Removal of the mineral fraction

Oil extraction

Crichton et al. (2017) developed an oil extraction protocol, a simple approach for the extraction of microplastic from solid samples, taking advantage of the lipophilic surface properties of most plastics.

Dried sediment samples are mixed with water and canola oil, then agitated thoroughly and left to settle until the oil, water and mineral fractions are completely separated. Any microplastic particles coming into contact with the oil should thus be conveyed to the oil layer, which can easily be transferred onto a filter. According to Crichton et al. (2017), recovery rates range from 90 to 100% for all seven tested pristine polymer types compared using density separation methods with NaI and $CaCl_2$. The procedure is simple, safe, cheap and time efficient, but may require an additional step to remove organic substances from a sample (Crighton et al., 2017). A recent study conducted by Mani et al. (2019) tested the oil extraction method using castor oil to separate microplastics (0.3−1 mm) from four complex environmental matrices: fluvial and marine suspended surface solids, marine beach sediments and agricultural soil. The method showed high recovery rates for the four pristine polymers: polypropylene, polystyrene, poly(methyl methacrylate, and glycol modified polyethylene terephthalate The average recovery rates were 99% ± 4% (mean ± SD) and a mean matrix reduction of 95% ± 4% (dry weight). For validation, non-spiked Rhine river water suspended solids underwent the castor oil extraction protocol. Of the 978 microplastic particles found in the five examined samples, 773 microplastic particles were recovered in the upper oil phase, whereas 205

microplastic particles were recovered from the lower aqueous and solid phase, resulting in a recovery rate of 74% ± 13%. For organic-rich sample matrices, an additional H_2O_2 digestion step was required to achieve an adequate sample purification (Mani et al., 2019).

Density separation

The most commonly applied method for the separation of microplastics from solid matrices is density separation. These methods use high-density salt solutions as extraction media (Hanvey et al., 2017).
In an early study conducted by Thompson et al. (2004), a method was devised using a saturated NaCl solution to separate microplastic from sand. After stirring and sedimentation, the plastic-containing supernatant is sucked into an extra flask from where it is transferred onto a filtering system (Crawford and Quinn 2017; Thompson et al., 2004). Losses may occur in the tubing or while decanting, which is why more sophisticated devices have been built since. Furthermore, saturated NaCl solution can only achieve a maximum density of 1.2 g ml^{-1}, and several synthetic polymers, such as polyethylene terephthalate (PET) and polyvinylchloride (PVC), have higher densities and are not extracted by saturated NaCl solutions. Thus, alternative salt solutions such as NaI (ρ=1.8 g·ml-1 ml^{-1}), Na6[H2W12O6] (ρ=1.4 g·cm^{-3}), Zn2Cl (ρ=1.6−1.7 g·cm^{-3}) and NaBr (ρ=1.55 g·cm^{-3}) were recommended, although the costs and hazardousness of these solutions may impose a need for alternatives.

Imhof et al. (2012) developed the Munich Plastic Sediment Separator (MPSS) to separate a diverse range of microplastics from a maximum of 6 L of sediments using a $ZnCl_2$-solution. A removable sample chamber equipped with a filter holder enables a direct transfer of the microplastic particles onto a filter, separating the sample from the density solution. As no decantation or repetitive extraction steps are required, losses and contamination can be avoided. According to Imhof et al. (2012), recovery rates of 95−100% can be achieved. However, Zobkov and Esiukova (2017) evaluated the MPSS and found the recovery rates of pristine plastics to be similarly high, as stated by Imhof et al., but the recovery of aged plastics was significantly lower at only 13−39%. Moreover, $ZnCl_2$ is hazardous and corrosive, has a low pH value and may react with the natural components of sediment, especially carbonates, resulting in bubbling and a foam that significantly hampers the procedure (Zobkov and Esiukova, 2017).

It is also questionable whether soil aggregates will be adequately dispersed in the MPSS, or whether a prior dispersion is required, especially for cohesive soils. Furthermore, the large surface area which may corrode due to the nature of the $ZnCl_2$ may be problematic if small particles get caught on the rough surface and cannot rise upward, possibly causing significant losses. Coppock et al. (2017) saw the need for a cheaper and portable microplastic-sediment separator and devised the Sediment-Microplastic-Isolation unit consisting of PVC tubes, a PVC ball valve and zinc chloride agitated by a magnetic stir bar. The principle mirrors that of the MPSS and shows similarly high recovery rates of 92−98%.
However, PVC must be excluded from the analysis as abrasion from the PVC tubing may contaminate the samples. This may compromise the value of the analytic results, as PVC is a relevant contaminant in

environmental samples, such as sediments and possibly soils, and excluding it from the analysis poses a significant loss of information.

Möller et al. (2020) considered a NaBr solution (1.55 g cm−3) as the most suitable separation medium, due to its relatively high density and low viscosity. Recovery rates of over 90% were reported for 10 types of microplastics (PA, PC, PP, ABS, PE, PS, PMMA, POM, PET and PVC). PE particles of different size ranges (100−500 µm, 500−1000 µm, 100−3000 µm) and shapes (particle, fibre, film) were tested using the device. The recovery rates differed only marginally for the different sizes, whereas the shape of 'particles', that is fragments, showed the highest recovery rate (98.3%), as opposed to films, with the lowest recovery rate (85%). This method appears promising for soil sample analysis for microplastic particles >100 µm and, although particles down to 32 µm were found in soil samples, the recovery rate (i.e. reliability) of the device for smaller particles (e.g. 1−100 µm) must first be systematically determined. One severe drawback of the described system is that the separation cylinder is made of Plexiglas (i.e. PMMA). Abrasion caused by the stirring of coarse soils may lead to an overestimation of the PMMA contamination in the samples. Therefore a non-plastic material should be used or PMMA excluded from the analysis.

In reviewing the above-named publications, no upper or lower size limit for the extraction of microplastic particles using density separation has been established (Möller et al., 2020). In general, the recovery rates of the particles were established for size ranges lying between 40 µm 55 and 5 mm (Liu et al., 2019; Imhof et al., 2012). The applicability for smaller microplastics and nanoplastics have not yet been analysed. However, according to the Stokes equation, small particles have very low settling velocities that are further reduced by high drag coefficients, which are dependent on the particle shape. So an in-depth empirical study of the minimum size of various microplastic particles in common density separation fluids is called for.

Froth flotation

Another method, which is not only dependent on the density of the polymer, but also on the hydrophobicity of its surface, is froth flotation, commonly used in the recycling industry (Alter, 2005; Fraunholcz, 2004; Marques and Tenório, 2000). Air bubbles will selectively attach to the more hydrophobic particles and carry them upward, thus separating them from the less hydrophobic matrix. According to Imhof et al. (2012), however, the mean efficiency of the froth flotation in separating microplastic from sediment is quite low (55 ± 28%), with high differences between polymer types.

Magnetic extraction

Another method that relies on the hydrophobic nature of microplastic surfaces is the method introduced by Grbic et al. (2019). By functionalizing iron nanoparticles with hydrophobic hydrocarbon tails (using hexadecyltrimethoxysilane (HDTMS)), the iron nanoparticles will bind to the microplastic surfaces and allow extraction with a magnet. In addition to water samples, the method was tested for sieved (mesh size 45 µm) benthic sediment spiked with microplastics (200 µm to 1 mm; PP, PVC, PU, PS, and PE). The recovery rates ranged from 49% (PP) to

90% (PE), albeit with high standard deviations within trials. This method is a proof of concept that still has a few disadvantages: for instance, lipophilic substances that are present in a soil's organic matter may result in nonspecific binding, reducing the effectiveness of the method. The authors suggest using the method for sediment samples after density separation or digestion, after which a further extraction method would no longer be necessary. Furthermore, brittle microplastic particles were shown to fragment during the procedure. In environmental samples, where aged microplastics may be more brittle than pristine particles, this could distort the results significantly. Grbic et al. (2019) propose that limiting microplastic contact with the magnet may reduce fragmentation. However, the question of the iron interfering with possible subsequent analytical characterization methods has not been addressed and should be further examined. While the authors describe how sonicating the magnet in a surfactant or acid solution can remove the iron nanoparticles from the microplastic surfaces, this method may lead to further fragmentation of microplastic particles (Löder et al., 2015) and is therefore undesirable.

Vertical density gradient separation
A new methodology for density separation developed for the recycling industry uses a liquid containing colloidal ferromagnetic particles. A magnetic field around the liquid creates a vertical density gradient, with the highest density being at the bottom and the lowest density being at the top of the liquid container, thus a separation of a sample into different strata of various densities can be achieved (Hu, 2014). This method may have potential concerning the isolation of microplastics from environmental samples; however, no research in this field has been conducted yet and the costs may be unjustifiably high.
In general, density separation may be useful to remove the mineral fraction of soil samples, but methods developed for aquatic sediments must be adjusted to the new matrix 'terrestrial soil', which can be extremely heterogeneous. Special care must be taken here that no microplastic losses occur due to enclosure in soil aggregates. Also, a soil's organic matter has a similar density (ρ=1−1.4 g·cm^{-3}) to most plastics and needs to be removed separately, as organic compounds can impede microplastic analysis.

A3.3.5 Removal of the organic fraction

Acidic and alkaline digestion
The treatment of organic matter in soil becomes the first key to detecting microplastics in soil. H_2O_2 and Fenton reagent are still common reagents for the digestion of organic compounds. H2O2 at 70°C improves the digestion of organic compounds (Li et al., 2019). However, studies have found that H_2O_2 will reduce the extraction efficiency of microplastics (Wang et al., 2018). NaOH and KOH are also used in the digestion of organic matter in soil. Although they effectively remove organic matter from soil, these two reagents can cause the discoloration and degradation of microplastics (Maes et al., 2017; Ruggero et al., 2020). A recent study used 98% H_2SO_4 to separate microplastics from farmland soil in order to obtain pure polyethylene (Li et al., 2020b). This method can be widely used in agriculture, although the use of large quantities of acid solution is not preferred. Standard instruments used to detect microplastics in ocean environments can be used in soil instead.

Acidic and alkaline digestion methods are frequently reported in the literature to remove the organic fraction of the sample matrix. Due to the fact that soils contain organic matter as well, a digestive step is imperative for an undisturbed analysis. Claessens et al. (2013) determined that the most efficient chemical digestion of mussel tissue was obtained by treating the sample with 22.5 M HNO_3. Also, the ICES (International Council for the Exploration of the Sea) recommends an acid blend of HNO_3:$HClO_4$ (4:1) to digest marine animal tissue for microplastic analysis. However, several studies state that certain strong acids may destroy specific polymers such as polystyrene and polyamide. Alkaline digestion is similarly discussed by many authors, especially for the digestion of animal tissue. However, treatment with NaOH was found to destroy polyamide and polyethylene (PE) fibres and led to the melding or discoloration of other polymers. Foekema et al. (2013) digested fish intestines with 10% KOH solution at room temperature for 2−3 weeks. While it is apparently successful and non-destructive to synthetic polymers, the procedure is time-consuming and may not be applicable to plant material or stabilized organic matter in soil. Generally, the purification of samples with strong acidic or alkaline solutions will lead to uncontrolled bias in the resulting microplastic composition of the sample (Löder et al., 2017). As stated earlier, the use of large quantities of strong acid or alkaline solutions is producing a lot of harmful chemical waste.

Oxidation with hydrogen peroxide

Oxidation treatment with boiling hydrogen peroxide (30% H_2O_2) is commonly used in soil analysis to remove a soil's organic matter (Pansu and Gautheyrou, 2006). It has also been used at lower temperatures to digest organic matter in the context of microplastic isolation from organic rich sediment matrices: Nuelle et al. (2014) allowed samples to remain in 30% H_2O_2 for 7 days. Around 50% of the biogenic matter was dissolved completely, but polyamide (PA), polycarbonate (PC) and polypropylene (PP) pellets showed visible signs of degradation such as discoloration and size reduction. Liebezeit and Dubaish (2012) claim that a 30% H_2O_2 treatment overnight does not affect plastic particles; however, it is not stated whether pristine or weathered plastic polymers were used, which can affect the results. In a study of microplastics in marine snow, Zhao et al. (2017) observed that 30% hydrogen peroxide treatment of organic-rich samples often results in the formation of a dense foam, which may suspend a significant portion of the microplastics sample, remaining separated and not mixed with the reagent. Many more observations on pristine and weathered microplastics were reviewed by Möller et al. (2020), indicating that care must be taken since the degradation of polymers can happen by long or hot exposure to high concentrations of hydrogen peroxide and Fentons reagent. Nevertheless, according to Möller et al. (2020) some promising results were obtained with Fenton's reagent in removing organic matter from the samples (e.g. Hurley et al., 2018). As this method is relatively cost and time-efficient, it has potential to become an important step in the microplastic analysis of soil samples.

Enzymatic digestion
Organic matter within soils is one of the most difficult fractions to remove without destroying the microplastic particles (e.g. Möller et al., 2020). An adapted enzymatic digestion may therefore be beneficial. However, most reports on enzymatic digestion procedures were developed for aquatic samples, removing soft animal tissue, and exclusively use proteolytic enzymes that will not be able to remove the stabilized plant organic matter contained in soils. For this reason, a more promising approach was derived by Löder et al. (2017). A sequential enzymatic digestion coupled with short-term reactions with H_2O_2. The enzymes used target specific organic compounds and using different enzymes may result in better digestion efficiencies than does, for example, a single oxidation step using H_2O_2. The presented protocol is sufficient for complex aquatic samples, but the removal of terrestrial plant matter and stabilized organic matter in soil will probably require a different set of enzymes; so an adaptation of this protocol may be necessary for terrestrial samples. As proposed by Löder et al. (2017) and Möller et al. (2020), a sequential combination of Fenton's reagent, SDS and specific enzymes can enhance the purification of organic matter in soil compared with the use of Fenton's reagent alone.

Combined methodologies
A few series of research studies depict the advantage of combined methodologies to ensure extraction of microplastics from the bulk sample (Ruggero et al., 2020). In particular, in most of them digestion of the organic residues follows a previous treatment for microplastic separation. This application was done for sediment samples after flotation with NaCl (Nuelle et al., 2014) and after elutriation in water (Quinn et al., 2017). These matrices, in fact, allow a proper separation from plastics due to the high difference in density, but can require a further digestion to remove any organic residues which float due to their lower density. In sludges, matrix digestion is combined with a prior sieving treatment (fractions from 60 µm to 2 mm) which ensures the removal of most of the sludges but leaves bigger size organic residues (Lares and Ncibi, 2018). Finally, the treatment which couples elutriation to sieving has a high efficiency in samples of sandy soils (Kedzierski et al., 2016).

A3.4 Identification and quantification (detection)

This chapter will focus on the identification and quantitative analysis of polymers in the soil sample containing microplastics.

A3.4.1 Visual identification and hot needle test

The earliest determination of microplastics was by means of visual identification under a light microscope, which is rather prone to bias and large error rates. To reduce the shortcomings of visual identification, some publications recommend the 'hot needle test', which makes use of the thermoplastic properties of many synthetic polymers. Zhang et al. (2018) further developed this principle for a simple identification of low-density polymers in soils: After a density separation with water, the residue in the supernatant is analysed by comparing microscope-images taken prior and after heating the sample at 130 °C for 3−5 s. Melted particles are then identified as thermoplastic polymers. While simple and feasible, this form of identification neglects high density and

thermosetting plastics and lacks the possibility of identifying the exact type of polymer. Similarly, the use of polarized light microscopy under which certain synthetic particles appear intensely illuminated, though indiscriminate, is non-destructive. For more details and further reading, the reader may take information from the reviews of Möller et al. (2020) and Ruggero et al. (2020).

A3.4.2 Chromatography

Common and more sophisticated methods, which allow the qualitative and quantitative identification of individual polymer types, include various extraction methods coupled to a chromatographic unit. Recommended methods in the literature include high temperature gel-permeation chromatography (HT-GPC) for the identification of polyolefins in cosmetics, liquid extraction with size-exclusion chromatography (SEC) for the identification and quantification of polystyrene (PS) and polyethylene terephthalate (PET) in soil samples (Elert et al., 2017), and pyrolysis followed by gas chromatography mass spectrometry (Pyr GC-MS). Pyr GC-MS is a sensitive and well-established method for the characterization and quantification of many polymer types and their organic additives.

Pyr GC-MS has been shown to be very suitable for the detection of microplastics in environmental samples. However, it also has several drawbacks. The size of the pyrolysis capsule and, accordingly, the sample amount per run is exceedingly small: 1.5 mm (Fries et al., 2013) and 0.5 mg (Dümichen et al., 2017) respectively, which requires an extensive sample clean-up for matrix-rich samples, making it rather unsuitable for bulk analysis. Additionally, it is prone to contamination or even blockage. To overcome these shortcomings, Dümichen et al. (2017) developed the thermal extraction-desorption gas chromatography mass spectrometry (TED GC-MS) for microplastic detection. Each run can accommodate up to 100 mg of sample, which requires no pretreatment other than grinding and mixing in an attempt to homogenize the samples. The processing time requires 2−3 h, which is less than most of the current spectroscopic methods available. This new analytical method seems suitable for the fast analysis of many different environmental sample types, including soils. Eisentraut et al. (2018) confirmed the possibility of identifying tyre wear in environmental samples using TED GC-MS, an achievement that is very important to monitor the massive introduction of microplastic particles into the environment by traffic. However, if sample grinding is necessary, quality controls should take place, ensuring that the risk of microplastic losses and contamination remain at a minimum. One significant drawback of TED GC-MS, as with all extraction-chromatography methods, that it is destructive. Hence, information on the number, size and morphology of the plastic particles cannot be obtained, although this information may be crucial in the context of assessing the effects of microplastics on organisms and eco-systems: size and form may affect exposure and bioavailability. In addition, the morphology of the microplastics may also be important in the context of their influence on the soil structure and function.

A3.4.3 Thermogravimetric analysis

A different thermal analysis method is the thermogravimetric analysis (TGA). Majewsky et al. (2016) coupled TGA with differential scanning calorimetry (DSC) for the microplastic analysis of waste water. However,

in that study, only polyethylene (PE) and polypropylene (PP) could clearly be identified. David et al. (2018) attempted to couple TGA with mass spectrometry (MS) in order to quantitatively analyse polyethylene terephthalate (PET) in soil samples without sample pretreatment. While successful, the method still requires further development and, to the best of the author's knowledge, is still restricted to the analysis of PET only. Thermogravimetric analysis and chromatography bear several promising approaches for fast mass-quantitative identification of microplastics in soils and other complex matrices (Huppertsberg and Knepper, 2018). However, they are limited insofar as subsequent analyses are impossible due to the destructive nature of the methods and the fact that the number, size and form of the particles remain unknown, resulting in the drawbacks mentioned above in the chromatography section.

A3.4.4 Vibrational spectroscopy

Vibrational spectroscopy, such as Raman or Fourier transform infrared (FTIR) spectroscopy, are the most commonly used state-of-the-art analytical methods in microplastic research, because they enable the precise identification of polymer types, their abundance, shape and size (Möller et al., 2020). Raman microspectroscopy enables the chemical imaging of samples down to a pixel resolution of 500 nm, while focal plane array (FPA) based micro-FTIR spectroscopy enables the identification of particles in a size range from 10 to 500 µm. Larger particles (>500 µm) can be analysed by attenuated total reflectance (ATR)-FTIR spectroscopy (Renner et al., 2017). For general information, the reader is referred to the reviews of Ruggero et al. (2020) and Möller et al. (2020), and for the application modes of FTIR and Raman spectroscopy, to the publications of Renner et al. (2017) and Kappler et al. (2016).

Raman and FTIR-based chemical imaging complement each other and should be chosen in accordance with the specific research questions: the Raman imaging run time is significantly higher than FTIR imaging (Kappler et al., 2016), but is independent of the shape, size or thickness of the measured particles, which can influence the results in micro-FTIR imaging. Black particles often result in unidentifiable FTIR-spectra which is particularly problematic with soil samples rich in organic matter. Consequently, a thorough sample purification is needed prior to placing the concentration on the filter surface on which the analysis takes place (Löder et al., 2017). For solid samples, this fact significantly reduces the amount of sample that can realistically be processed. Therefore, the need for an easy high-throughput technique for a representative amount of solid samples, such as soils, has been recognized and solutions have been proposed (reviewed by Möller et al. (2020) and Ruggero et al. (2020)). In all cases and similar to the chromatographic methods, these techniques are destructive and only allow mass-quantitative analysis, and do not provide information on number, size and shape of the polymer particles. Additionally, multiple polymer types in one sample produce complex absorption spectra that may hamper identification. To address these issues, several trials have been undertaken.

A similar, novel method was introduced by Schmidt et al.116 who used short-wave infrared (SWIR) imaging spectroscopy to analyse surface

water samples taken from the Teltow Canal in Berlin (Germany). The purified (using H_2O_2) samples were filtered onto several glass fibre filters each (diameter 47 mm) and scanned with a SWIR imaging spectrometer – measurement speed: 52 048 mm^2 per hour – resulting in the measurement of 10 filters within 20 min. The lower detection limit is for particles of size 560 µm by 280 µm (2 pixels). The evaluation of the spectroscopic images was done automatically by the "PlaMAPP" algorithm, yielding a 75% true detection efficiency. To achieve more reliable results, additional manual checking was required (Schmidt et al., 2018). While the SWIR imaging spectroscopy can process many sample filters in comparatively little time, its drawbacks are the rather large lower size limit and the need for improvement for correct particle detection in the automated analysis algorithm

A3.4.5 Proton nuclear magnetic resonance spectroscopy (1H NMR)

A completely new approach for size-independent microplastic analysis was recently described by Peez et al. (2019). Using quantitative 1H NMR spectroscopy, model samples of polyethylene (PE) particles, polystyrene (PS) beads, and polyethylene terephthalate (PET) fibres could successfully be qualitatively and quantitatively analysed using a calibration curve method. This method has the severe drawback of having to completely remove organic matter from the sample to avoid signal overlays (Peez et al., 2019). As a 100% removal of organic matter from soil samples is not possible, the 1H NMR method is unsuitable for microplastic analysis in soil samples.

A3.4.6 In-situ identification

Several attempts were undertaken to develop in-situ identification and quantification methods. One such approach was developed by Paul et al. (2019), who attempted to combine near-infrared (NIR) spectroscopy (opposed to the mid-infrared range used in conventional FTIR spectroscopy) with chemometrics to identify microplastics in soils. Near-infrared radiation can penetrate deeper than mid-infrared radiation, enabling analysis of particles even when coated with a thick biofilm, and the method is generally not sensitive to water. Paul et al. (2019) were not able to achieve a sensitive quantification of the plastic content in soil samples. However, Corradini et al. (2019) developed a method to 'rapidly assess microplastic concentrations in soils without extraction' using a portable visible NIR spectroscope. Here the microplastic concentration is to be estimated directly in the field, without time- and labour-consuming extraction and detection procedures. The current low accuracy and high detection levels are hampering the limit (which may be much higher than realistic concentrations in natural soils), as well as the need for a training set to accurately predict the polymer concentrations, which has to be adapted to different soil types. Hyperspectral imaging or visible NIR spectroscopy for microplastic analysis in environmental samples are methods under development and have the potential for becoming rapid automated identification methods in soil samples in the future (Corradini et al., 2019; Karlsson et al., 2016; Serranti et al., 2018).

A3.5 Conclusion

Despite considerable efforts made and although many methods have shown their applicability, the search for reliable, practical, robust and

affordable methods for measuring microplastics in complex solid matrices such as soils continues. This is not unexpected since microplastics comprise a broad group of different synthetic polymers, with different shapes and dimensions, which are difficult to separate from the soil matrix. It should therefore be acknowledged that there will be no single method that fulfils all the requirements for the quantification of microplastics in the soil system.

Instead, one recommendation might be to combine sets of contrasting methods, which individually do not demonstrate a fully reliable performance, but in combination with other methods can produce a weight of evidence for quantification.

Combinations involving sets of separation, recovery and identification methods have also been tested and seem to be the way forward (Ruggero et al., 2020). However, most studies are still limited to one analytical methodology for each step. To be able to combine and compare test results, there is a need for a harmonized protocol describing which (set of) method(s) is/are best to use for which purpose, with the necessary quality standards.

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