

What comes after the sun? – The integration of soil biogeochemical weathering into microplastic experiments

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10 **Abstract.** Recent studies have been engaged in estimating the adverse effects of
microplastic (MP) on soil quality parameters. Mass concentrations of MP as found in highly
contaminated soils were shown to weaken the soil structure, and parts of the edaphon are
adversely affected by mainly the < 100 µm MP fraction. However, the vast majority of these
studies used juvenile particles, which have surface characteristics different from that of
15 environmental MP. Exposed to UV radiation, plastic undergoes photochemical weathering
with embrittlement and the formation of surface charge leading to an alteration of
physiochemical behavior. When plastic particles then enter the soil environment, a second
phase of alteration seems possible. This little explored soil biogeochemical phase includes
biofilm cover, decay with enzymes, as shown in laboratory experiments, biotic and abiotic
20 acids, oxidants as well as further physical fragmentation by bioturbation and feeding of the
soil fauna. This perspective article encourages to consider the weathering history of
microplastic in soil experiments and highlights the need for reproducing the surface
characteristics of soil MP to conduct laboratory experiments with close-to-nature results.

Did we use too young plastic?

25 Since the mass production of plastic articles of daily use started in the early 1950th
(Thompson et al., 2009), a number of processes cause the contamination of ecosystems such
as inland and coastal waters, sediments, the open and deep seas, soils and even the
atmosphere with MP (e.g. Cole et al., 2011; Woodall et al., 2014; Wu et al., 2018; Büks and
Kaupenjohann, 2020; Trainic et al., 2020). The formation of soil MP pools occur through
30 littering and dispersion from landfills, the application of wastewater, contaminated surface
water, sewage sludge, composts, digestates, mulching foils, seed and fertilizer coatings, road
dust as well as atmospheric deposition (Eerkes-Medrano et al., 2015; Huerta Lwanga et al.,
2017a; Weithmann et al., 2018; Corradini et al., 2019; Dierkes et al., 2019; He et al., 2019;
Edo et al., 2020; Huang et al., 2020; Bertling et al., 2021; Katsumi et al., 2021; Szewc et al.,
35 2021).

Today, we are faced to a global contamination of soil ecosystems with MP, that averages
1.7 mg kg⁻¹ dry soil in agricultures (Büks and Kaupenjohann, 2020), exceeds this value by
several orders of magnitude in heavily contaminated soils such as at road sides and industrial
areas (Fuller and Gautam, 2016; Dierkes et al., 2019), and reaches even remote areas
40 (Abbasi et al., 2021). Several laboratory studies showed adverse effects of high MP
concentrations on the soil fauna (Büks et al., 2020a) and soil structure (e.g. de Souza
Machado et al., 2018; de Souza Machado et al., 2019; Liang et al., 2019; Lozano et al., 2021)
and underlined the relevance of especially the small-sized fraction (MP<100 µm) (Büks et al.,
2020b).

45 Although these results are rightly alarming due to the function of soil structure and the
edaphon as soil fertility parameters (Bronick and Lal, 2005; Thiele-Bruhn et al., 2012), their
informative value is limited by the fact, that the vast majority of experiments used juvenile
plastic and a short run time that does not allow for further weathering (e.g. de Souza
Machado et al., 2018; de Souza Machado et al., 2019; Liang et al., 2019; Büks et al., 2020a;
50 Lozano et al., 2021). In the environment, however, the bulk of MP is exposed to weathering
for years including both photooxidative and soil biogeochemical alteration. The surface
characteristics and role of MP within soil ecosystems thereby likely change.

The photooxidative weathering of microplastic

In a microscopic perspective, the surfaces of juvenile plastic items are normally smooth and
55 uniformly structured with nearly no surface charge (e.g. Fotopoulou and Karapanagioti, 2012;
Fotopoulou and Karapanagioti, 2015). When exposed to sunlight, which is mainly the case in
the “use and dispose” phase of the product life cycle, the weathering of plastic is largely
driven by photooxidation or indirect photolysis. This phase of weathering is well researched
and reviewed (e.g. Kokott, 1989; Pickett, 2018). The incoming solar photons need to hit weak
60 bonds within the polymer structure with wavelengths in the UV and blue spectrum to initiate

photooxidative aging (Pickett, 2018). This generates radicals, which cause chain scission and reactions with nearby polymers and O₂, resulting in crosslinks and a wide spectrum of carbonyl groups that increase surface polarity (ter Halle et al., 2017; Dong et al., 2020). From the point of view of the macroscopic observer, the plastic becomes less hydrophobic, stiff and more prone to fragmentation. Further additives such as inks, plasticizers, flame retardants, UV absorbers and HALS (hindered amine light stabilizers) are degraded in parts also by longer wavelengths of the UV-vis spectrum. The underlying reaction rates, except for the initial radical formation, increase with temperature and are also accelerated with advancing depletion of chemical UV protection.

Today, standardized approaches are applied by materials science to test the resilience of plastic items in face of artificial photooxidative stress, but are also newly used in soil science to produce weathered MP (BMBF initiative “Plastik in der Umwelt”, e.g. Büks et al., 2021). These treatments include an imitation of solar radiation by an UV or full-spectrum lamp, controlled temperatures and artificial irrigation with at least one of these factors enhanced compared to natural conditions (Pickett, 2018). Treatments of several weeks cause severe weathering leading to enhanced crystallinity, density and cracked surfaces (Gulmine et al., 2003). Whereas formerly used carbon arc lamps are outdated because they emit a spectrum unlike natural sunlight (Howard and Gilroy, 1969), many industrial weathering protocols advice xenon arc lamps with borosilicate filters, that adjust the emitted spectrum tighter to the natural UV spectrum (DIN EN ISO 4892-2), or fluorescent UV lamps (DIN EN ISO 4892-3). The performance of these approaches is enhanced by use of modern daylight filters, a steady temperature of 38°C, relative air humidity of 25 to 50 % and regular washing of the sample surfaces by artificial rain (Pickett, 2018). Beside the use of UV, the γ -irradiation is reported to imitate the carbonyl stretch in PE samples similar to a long-term UV-B exposition (Johansen et al., 2019). Furthermore, Zhou et al. (2020) could demonstrate that discharged plasma oxidation (DPO) is likewise suitable to increase surface area, crystallinity and carbonyl indices of plastic particles within hours.

Although lamps and other techniques have been shown to properly imitate natural photooxidation, we do not know if these results are comparable with those induced by belowground weathering. However, we can assume, that different surface characteristics arise from these two types of aging.

The soil biogeochemical phase

When plastic is exposed to the dimmed world of soil fauna, microorganisms, roots and frequent leaching, the composition of weathering parameters changes significantly (Table 1). The plastic is now faced to new mechanical stresses such as (bio)turbation, largely moist conditions and exposed to a variety of biogeochemical processes.

One of these factors is the diverse and active soil fauna, that has been shown to ingest, digest and excrete plastic particles (Büks et al., 2020). It is an ensemble of small, mobile bioreactors, that incubate soil particles including MP within a habitat of high microbial diversity – their gastrointestinal tract – and distribute them throughout the soil by excretion. A well known example for this multifaced functionality is the earthworm. Some taxa like woodlice, termites, mealworms and earthworms were additionally found to comminute plastic by gnawing and, hence, actively produce MP (e.g. Lenz et al., 2012; Zhang et al., 2018; Büks et al., 2020a). There are also indications that the mealworm microbiome is able to degrade PE and PS to an eminent degree beyond the proportion of additives, but with yet no information on the underlying reactions (e.g. Brandon et al., 2018).

While moisture evaporates quickly on sun-exposed, heated plastic surfaces and is then not an important factor of weathering (Pickett, 2018), in soils it is the ubiquitous condition for microbial life, extracellular metabolic processes and the release of chemical agents that react with the plastic outside the fauna. The microbial colonization and biofilm formation on surfaces of MP particles has been shown in studies on various aquatic ecosystems (e.g. Zettler et al., 2013; McCormick et al., 2014; Oberbeckmann et al., 2015; Dussud et al., 2018; Jiang et al., 2018). Much scarcer in number, recent studies on soil ecosystems found surfaces of differently originated MPs inhabited by microbial communities, whose composition differs widely from the soil matrix (Chai et al., 2020; Zhang et al., 2019). The altered soil microbial community (Ng et al., 2020; Wang et al., 2020) is thereby not only determined by the physiochemical properties of the surrounding soil, but also by the type of plastic and its additives (Chai et al., 2020; Wiedner and Polifka, 2020; Yan et al., 2020; Yi et al., 2020).

A biofilm, in turn, causes the alteration of the plastic surface. Not only a viscous matrix, that protects bacteria against mechanical stress, predators, desiccation and irradiation, it is also an extracellular reaction space that facilitates the concentration and metabolization of nutrients and the recycling of dead cell material (Flemming and Wingender, 2010). For this purpose, manifold extracellular enzymes are produced by the biofilm community to decompose food sources or modify the biofilm matrix in face of e.g. oxygen or nutrient gradients (Flemming and Wingender, 2010). Among these are esterases, proteases and amidases that target on substrates like polysaccharides, proteins, extracellular DNA, lipids and urea, but also allow cometabolism of artificial polymers such as diverse polyesters, ester-based PU and PET in laboratory experiments (Shimao, 2001; Wei and Zimmermann, 2017; Danso et al., 2019).

Given a poor biodegradability of polymers with C-C backbones and no hydrolysable functional groups such as juvenile PE, PP, PS and PVC, Yoon et al. (2012) showed an unexpected degradation of PE by a bacterial alkan hydroxylase, and, beyond this, Yoshida et al. (2016) found the specific targeting of PET with a bacterial PETase. In contrast, unspecific lignin-degrading enzymes such as laccases, manganese peroxidases, hydroquinone peroxidases and lignin peroxidases produced by actinomycetes, other bacteria as well as fungi, were

shown to depolymerize even plastics such as PE, PS and PA, that were considered recalcitrant (Bhardwaj et al., 2013; Wei and Zimmermann, 2017). Beside the direct proof of enzymatic degradation pathways there are numerous references on the metabolization of (bio-)plastic samples by bacterial and fungal strains (e.g. Bhardwaj et al., 2013; Kale et al., 2015; Raziya-fathima et al., 2016; Roohi et al., 2017). In contrast, for PP and PVC neither degrading enzymes nor observed biodegradation were reported (Danso et al., 2019). In consequence, it is reasonable to assume slow biodegradation of plastic surfaces in soil, even if many studies worked with commercial polymers, that have concealed compositions (Danso et al., 2019), so there is poor insight to what degree the measured mass loss is caused by microbial/enzymatic decomposition of the polymer or additives.

Table 1: Development of surface characteristics during the three phases of aging (juvenile, photooxidative and soil biogeochemical phase). Data of soil biogeochemical weathering are only known from aquatic systems. (?) marks assumptions based on soil biogeochemical processes found in soils. Some references are: ¹Fotopoulou and Karapanagioti (2012), ²Fotopoulou and Karapanagioti (2015), ³ter Halle et al. (2017), ⁴Dong et al. (2020), ⁵Pickett (2018), ⁶Andrady et al. (1993).

characteristic	juvenile phase	photooxidative phase	soil biogeochemical phase
topography	smooth ^{1,2,4}	rough ⁵	rough ^{1,2,4}
surface charge, carbonyl index	no ^{1,2,3,4}	yes ⁶	increasing ^{1,2,3,4,(?)}
crystallinity, crosslinks, chain scissions	low ³	high ⁵	increasing ^{3,4,(?)}
biofilm cover	low	low	growing or mature ^{2,5,(?)}
aging factors	no	UV radiation ⁵ blue/violet spectrum ⁵ frequent leaching ⁵	enzymes ^(?) organic acids ^(?) inorganic acids ^(?) bases ^(?) oxidants ^(?) bioturbation ^(?) feeding by the edaphon ^(?) frequent leaching ^(?) freeze-thaw-cycles ^(?)

Beside the soil biome, soil pH and oxidants are expected to directly influence the belowground alteration of plastic surfaces. While there is – to the best of our knowledge – no systematic examination of the effect of soil born acids, bases or oxidizing agents within natural ranges of concentration and time of exposure, the treatment of plastic fragments with concentrated reagents caused damaging effects from color leaching and expansion to total dissolution (Enders et al., 2017). However, pre- and post-treatment with oxidants such as

155 H₂O₂ are common parts of the extraction of MP from soil samples with density fractionation (Büks and Kaupenjohann, 2020).

160 In winter, when the mechanical treatment through biota is reduced, freeze-thaw-cycles might be an additional factor of fragmentation. Studies on the effect of alternating freezing and thawing on the structure of plastic surfaces are sparse and only focus on composite materials that include non-plastic components (Wang et al., 2007; Adhikary et al., 2009; Zhou et al., 2014). However, water, that has already entered the cracks of weathered plastic with reduced hydrophobicity, most likely contributes to the fragmentation of the brittle material by freezing and expansion leading to an increase of exposed surface.

Pre-weathering in recent and future experiments

165 The alteration of plastic surfaces during weathering shows, that future experiments have to be conducted with pre-weathered instead of juvenile MP. But it is still open, if there is effective soil biogeochemical aging beyond the photooxidative phase and, thus, if a DIN EN ISO 4892-2/3 approach, as applied in recent work, is sufficient to imitate soil weathering conditions. Currently there are no studies that show the development of MP surfaces in soil ecosystems over time, and only a few studies integrated soil biogeochemical factors into pre-weathering of artificial MP (Table 2, Büks et al., 2020a), alas fragmentary, heterogeneous and often directly applied to juvenile plastic:

175 Tsunoda et al. (2010) heated plastic items within a water bath at 90 °C for 3 weeks and abraded the surface prior to feeding experiments with termites. This treatment was aimed to make the surface more accessible for gnawing and might also extract soluble additives from the juvenile plastic. In another experiment, the formation of biofilms on MP surfaces was provoked by four weeks of incubation in seawater to make the material more attractive as a food source for the lugworm *Arenicola marina* (Gebhardt and Forster, 2018), an approach that can be likewise applied with soil solution. With the intention to clean up artificial MP from soluble substances and fine particles, juvenile plastics were also treated with organic solvents (Huerta Lwanga et al., 2016; Huerta Lwanga et al., 2017b; Rodrigues-Seijo et al. 2018; Rodrigues-Seijo et al., 2019; Wang et al., 2019; Yang et al., 2019). If the plastic type is prone to the solvents, the surface is roughened by the dissolution of oligomers and, thus, increased. However, these techniques are not assumed to increase carbonyl groups and surface charge. Thus, they do not change the interaction with the soil matrix and the soil fauna, and were never tested on the similarity with natural weathering.

185 Some authors avoided artificial weathering and instead applied natural aging over shorter periods between two weeks and 12 month, which can be used as a kind of “plastic nursing” (e.g. Martin-Closas et al., 2016; Zhang et al., 2018). This treatment changes the physiochemical characteristics of plastics similar to environmental short-term weathering

190 belowground and is suitable for aging large amounts of plastic, but might be very costly in terms of time when the production of strongly weathered MP is needed.

To better understand the dynamics of surface alteration of soil MP and to identify the important aging factors, long-term weathering experiments will be extremely helpful. These experiments must take into account not only ecosystem parameters (e.g. humidity, edaphon
195 activity and soil organic carbon) and start conditions such as plastic type, particle surface and protection by specific additives. Increasing surface area and charge density over time might also cause a non-linear aging, and biofilm-cover might cloak the real MP surface characteristics, an issue that should be carefully included into the experimental design.

Should there be a significant soil biogeochemical phase, there is a great incentive to design
200 pre-weathering approaches containing full chains of aboveground and in-soil aging factors to create designer-MP for laboratory experiments. The quality of future studies, that work on the effects of soil MP on the edaphon and the direct and indirect influence on soil structure, will profit from those protocols. Their results will help us to better understand and forecast short- and long-term effects of environmental soil MP concentrations, that have been the result of
205 decades of contamination and are still growing.

Table 2: Approaches of surface (pre-)weathering in recent experiments with soil microplastic. The abbreviations used in this table are as follows: UV – ultraviolet, TBBPA – tetrabromobisphenol A, FE – feeding experiment. Polymers: BD – biodegradable plastics, OP – oxodegradable plastics, PA – polyamide, PE – polyethylene, PO – polyolefins, PP – polypropylene, PVC – polyvinyl chloride, TCE – thermoplastic copolyester elastomers. NA denotes that information was not available.

aging factor	applied plastic type	aging time (d)	resulting characteristics	experimental focus	reference
UV radiation (climate chamber)	diverse	variable	photooxidative aging	diverse	DIN EN ISO 4892-2, DIN EN ISO 4892-3
gamma irradiation (⁶⁰ Co source)	PE, PP	NA	photooxidative aging	cation adsorption	Johansen et al. (2019)
discharged plasma oxidation (DPO)	PVC	0.02	photooxidative aging	TBBPA adsorption of and toxicity to algae	Zhou et al. (2020)
water bath (90°C) + abrasion	PO, PA, PE, TCE	21	extraction of additives, increased accessibility for feeding organisms	feeding experiment with termites	Tsunoda et al. (2010)
incubation in seawater	PA, PS	28	surface biofilm formation	FE lugworms	Gebhardt and Forster (2018)
incubation in aquatic systems	PE, PP	19	surface biofilm formation	cation adsorption	Johansen et al. (2019)
methanol treatment	PE, PS	NA	extract soluble additives	FE earthworms	Wang et al. (2019)
ethanol treatment	PE	NA	extract soluble additives	FE earthworms	Rodrigues-Seijo et al. (2018)
pentane + octane treatment	PE	NA	extract soluble additives	FE earthworms	Rodrigues-Seijo et al. (2019)
		NA	extract soluble additives	FE earthworms	Huerta Lwanga et al. (2016)
		NA	extract soluble additives	FE earthworms	Huerta Lwanga et al. (2017b)
plastic nursing (soil)	BD, OD, PE	NA	extract soluble additives	FE earthworms	Yang et al. (2019)
		~150	belowground weathering	mulch foil degradation experiment	Martin-Closas et al. (2016)
		14-365	belowground weathering	feeding experiment with earthworms	Zhang et al. (2018)

Data availability

All of the data are published within this paper and in the supplement.

210 **Author contributions**

FB developed the article concept, collected data and prepared the manuscript. MK supervised the study by participating in structural discussions on the idea and concept of the paper and the final corrections.

215 **Competing interests**

The authors declare that they have no conflict of interest.

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