Dear Referees.

Thank you very much for your mindful reviews and your very helpful comments. It has helped us to see some points which still need clarification. In the following, we want to explain how we propose to adjust our article based on the comments and also explain, why in some cases we do not agree with the proposed changes.

First – and most important – we deeply agree with the referees, that the manuscript contains different topics, that are

- already known (introduction, photochemical weathering of plastic)
- already reviewed (influence of MP on the soil fauna)
- or not widely researched (MP and soil structure, biogeochemical weathering)

and that the focus of this work should be on the latter, but by use of a shorter, more perspective-based manuscript type.

Our response is split into two parts: This first part contains a restructured manuscript in the format of a forum article with focus on soil biochemical weathering of microplastic surfaces and the possible need for soil-like pre-weathering of experimental microplastic. The second part (influence of microplastic on soil structure) will be re-submitted separately and is not part of this answer.

In the following you can find a list of all your points addressed (with numbers representing the order within the old document, excluded numbers no longer are part of the revised manuscript):

[5] Line 20-23: chemical reactions and physical processes are not clearly delineated; also, how do soil enzymes «weather» conventional plastics? The latter typically are chemically highly inert and it's not clear which enzymes can act on these materials
→ We tried to clarify this within the abstract and discuss it later within the main text.

[7] Line 37: maybe fragmentation is the better term than comminution?
→ done across the manuscript

[9] Line 80: Why were studies on biodegradable plastics excluded? Why were papers on polymer photooxidation excluded (by requiring that the term "soil" was included in the search).

→ We focused on the named non-biodegradable polymers that represent …% of the plastic produced since the 1950s and their still growing legacy in soils. On the other hand, biodegradables are relatively new and only a small part of the plastic introduced into soils.

[14] Line 140: The wording makes it sound as if the plastic is either "juvenile" or aged. However, the juvenile plastic will age when in soils. Also, it seems that the terms polymer and plastic are not clearly defined and used. They are not the same.

→ That's correct. When juvenil plastic is added to the experimental soil ist starts aging. However, in short-term experiments it unlikely that even with the initial formation of biofilm cover there is an extended aging of surface characterists. To focus in the process, we replaced "aged" by "aging" in some cases. After shortening the text, "polymers" and "plastic, however, are now used in the correct way without any text modifications.

[24] Line 253: smoothness must depend on how the microplastic is formed/generated. Are the authors therefore sure that the microplastic is always "smooth"?

→ We never had contrary impressions from REM/light microsopic images of juvenile commercial plastic items.

[25] Line 257: Chromophores are no "flaws". Also, there is indirect photolysis in which the polymers must not directly absorb light. Finally, most plastics that have exposure to sunlight are photostabilized. Photostabilizers slow down these reactions. This is not mentioned here.

→ Replaced by "weak bonds" and "indirect photolysis" added.

[26] Line 254: which of the conventional polymers contains NH groups?

→ deleted.

[27] Line 260: Carbonyls are uncharged

→ For soil environments, that's incorrect. Depending on the environmental pH, carbonyl groups (e.g. -COOH) are subject to (de-)protonation, which leads to variable charges. This is a process well known for soil organic matter and the soil mineral matrix strongly controlling adsorption of molecules and interaction with other particles in soil.

[29] Line 270: what are "biogeochemical attacks"? And moisture is also present during the use period of the plastic.

→ Replaced: "The plastic is now faced to new mechanical stresses such as (bio)turbation, largely moist conditions and exposed to a variety of biogeochemical processes."

[35] Line 339: Terms "decay" and "degradation" remain poorly defined. Aren't they describing the same overall loss of plastic integrity (either in terms of physical or chemical changes) / Line 384: the term "decomposition" is not defined. This is a general problem as the authors do not clearly define any of the terms. It seems that "weathering", decay, degradation, decomposition are all used interchangeably. Also, the term "biodegradation" is not defined

→ Both, decay and degradation describe the breakdown of organic matter by the soil (micro-)biome, while in other parts of the text the word was replaced by "aging", "depletion" or otherwise clearyfied.

[32] Line 340: "weight loss" is misleading. Because there is also Mw (molecular weight). The reviewer assumes the authors refer to mass loss?

→ Yes, thank you very much.

[37] Line 345: These polymers certainly decay. The authors mean that they don't biodegrade?

→ We instead used the term "biodegradation".

[38] Line 395: Why would one expect similar reactions? Photochemical reactions often trigger radical chemistry and needs light absorption and electron promotion to occur. This is not the case for subsurface reactions. So it seems very unlikely that the very different reactions result in the same products (unless, of course, the chemistry is looked at in a blunt manner, eg: increase in "oxygen" content)

→ That’s exactly our point: Pre-weathering of plastic for laboratory experiments is mainly conducted by use of climate chambers with photooxidation, but do not the outcome of aging in soil (that we do not know). We now have emphasized that point.

[39] Line 396: The reviewer cannot understand why “photochemistry” is separated from “geochemical” reactions. Aren’t photochemical reactions also “geochemical”? For instance, according to Wikipedia (quick check, and not a scientific source, but most likely accurate here):·Photogeochemistry is the study of light-induced chemical reactions that occur or may occur among natural components of the Earth's surface.

→ For clarification we added “soil (bio)geochemical” throughout the document.

[40] Line 402: “early material science”. What is meant by “early”? Photochemical aging of plastics is extremely well studied but does not seem to be “the early days” of material sciences

→ Deleted.

[41] Line 423: “dimmed world”? Why dimmed? Is this not “dark”?

→ Not necessarily. There can be faint light within the upper centimeters. And you can dim something until its dark.

[42] Line 473: Is it reasonable to develop “THE” standard aging method for plastics in soils? See previous point

→ A standard approach, that includes influcenes by plastic type and additives but also so respective soil environment (e.g. arid/humid, active soil fauna, Corg).

What comes after the sun? – The integration of [39]soil biogeochemical weathering into microplastic experiments

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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 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 [5]seams possible. This little explored [39]soil biogeochemical phase includes biofilm cover, [5]decay with enzymes, as shown in laboratory experiments, biotic and abiotic acids, oxidants as well as [5]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?

Since the mass production of plastic articles of daily use started in the early [9]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 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., 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 (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).

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; Lozano et al., 2021). In the environment, however, the bulk of MP is exposed to weathering for years including both photooxidative and [39]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 [24]normally smooth and 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 [25]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 [25]weak bonds within the polymer structure with wavelengths in the UV and blue spectrum to

initiate photooxidative [35]aging (Pickett, 2018). This generates radicals [26], which cause chain scission and reactions with nearby polymers and O_2, resulting in crosslinks and a wide spectrum of [27]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 [7]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 [35]depletion of chemical UV protection.

Today, standardized approaches are applied by [40]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 photoxidation, 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.[38]

The [39]soil biogeochemical phase

When plastic is exposed to the [41]dimmed world of soil fauna, microorganisms, roots and frequent leaching, the composition of weathering parameters changes significantly (Table 1). [29]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; Raziyafathima et al., 2016; Roohi et al., 2017). In contrast, for PP and PVC neither degrading enzymes nor observed [37]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 [36]mass loss is caused by [35]microbial/enzymatic decomposition of the polymer or additives.

Table 1: Development of surface characteristics during the three phases of aging (juvenile, photooxidative and [39]soil biogeochemical phase). Data of [39]soil biogeochemical weathering are only known from aquatic systems. (?) marks assumptions based on [39]soil biogeochemical processes found in soils. Some references are: [1]Fotopoulou and Karapanagioti (2012), [2]Fotopoulou and Karapanagioti (2015), [3]ter Halle et al. (2017), [4]Dong et al. (2020), [5]Pickett (2018), [6]Andrady et al. (1993).

characteristic	juvenile phase	photooxidative phase	[39]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

H_2O_2 are common parts of the extraction of MP from soil samples with density fractionation (Büks and Kaupenjohann, 2020).

In winter, when the mechanical treatment through biota is reduced, freeze-thaw-cycles might be an additional factor of [7]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 [7]fragmentation of the brittle material by freezing and expansion leading to an increase of exposed surface.

Pre-weathering in recent and future experiments

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 [39]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 [39]soil biogeochemical factors into pre-weathering of artifical MP (Table 2, Büks et al., 2020a), alas fragmentary, heterogeneous and often directly applied to juvenile plastic:

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.

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

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 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 carefuly included into the experimental design.

Should there be a significant soil biogeochemical phase, there is a great incentive to design 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 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 – tetrabromibisphenoal A, FE – feeding experiment. Polymers: BD – biodegradable plastics, OP – oxodegradable plastics, PA – polyamide, PE – polyethylene, PO – polyolefins, PP – polypropylene, PVC – polyvenyl 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 adsoprtion	Johansen et al. (2019)
discharged plasma oxidation (DPO)	PVC	0.02	photooxidative aging	TBBPA adsorption of and toxicity to algae	Zhou et al. (2020)
wather 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 adsoprtion	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)
	PE	NA	extract soluble additives	FE earthworms	Rodrigues-Seijo et al. (2019)
pentane + octane treatment	PE	NA	extract soluble additives	FE earthworms	Huerta Lwanga et al. (2016)
		NA	extract soluble additives	FE earthworms	Huerta Lwanga et al. (2017b)
		NA	extract soluble additives	FE earthworms	Yang et al. (2019)
plastic nursing (soil)	BD, OD, PE	~150	belowground weathering	mulch foil degradation experiment	Martin-Closas et al. (2016)
plastic nursing (soil, compost)	BD, PE	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.

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.

Competing interests

The authors declare that they have no conflict of interest.

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