

Deposition and in-situ translocation of microplastics in floodplain soils

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Key Points:

- Microplastics have accumulated frequently in the upper floodplain soils deposited by flood dynamics starting in the 1960s.
- Microplastics can be relocated within floodplain soils through natural processes and reach depths up to 2 meters.
- Plastics have become part of earths geological cycle and could therefore be used as an Anthropocene marker for soils and sediments.

Abstract

Microplastic (MP) contamination of freshwaters and soils has become one of the major challenges within the Anthropocene. MP is transported in large quantities through river systems from land to sea. However, the question is whether there is transport only or also deposition within the system? Floodplains and their soils as part of the river system are known for their sink function for sediments, nutrients, and pollutants. The present case study analyzes the spatial distribution of large (L-MP, 2,000–1,000 μm) and medium (M-MP, 1,000–500 μm) MP particles in floodplain soils of the Lahn River (Germany). Based on a geospatial sampling concept, the MP contents in floodplain soils are investigated down to a depth of 2 meters through a holistic method approach. The analysis of the plastic particles is carried out by density separation, visual fluorescence identification, and additional ATR-FTIR analysis. In addition, grain size analyses and ^{210}Pb and ^{137}Cs dating was performed to reconstruct the MP deposition conditions in floodplains. The results prove a spatial frequent accumulation of MP in upper floodplain soils (0–50 cm) deposited by flood dynamics since the 1960s. MP detection over the entire soil column to a depth of 2 meters and below recent (>1960) sediment accumulation indicates MP relocation and in-situ vertical transfer of mobile MP particles through natural processes (e.g., preferential flow, bioturbation). Furthermore, the role of MP as a potential marker of the Anthropocene is assessed based on the findings. This study advances our understanding of the deposition and relocation of MP at the aquatic-terrestrial interface.

Plain Language Summary

The occurrence of plastic as a man-made material in the environment is widely known for different natural areas (e.g., oceans, rivers, soils). Nevertheless, the question arises with respect to the routes through which plastic is transported across the environment, especially between land and sea. Therefore, the study focuses on the content of microplastics (i.e., plastic particles smaller than 5 mm or, in this case, between 2 and 0.5 mm) in floodplain soils accompanying rivers as main global transport routes of the plastic. Visual analysis of plastic particles (microscopic) and soil parameters revealed that microplastic particles are found very widely in floodplain soils. They occur at all depths between soil surface and down to 2 meters, with the highest contents in the upper 50 centimeters. These upper soil sections correspond to very young soil layers in deposited sediments of the river during floods since the 1960s. However, because plastic particles also occur below this level and plastic has only been increasingly released into the environment since the 1950s and 60s, there must be natural processes that lead to a vertical displacement of the particles, which are thus to be regarded as movable in soils.

1 Introduction

Global plastic pollution of marine, freshwater, and terrestrial ecosystems is one of the major challenges in the Anthropocene (De-la-Torre *et al.*, 2021). Rising scientific efforts have resulted in the fact that nowadays plastics and microplastics (MP) can be detected in all ecosystems worldwide (Cole *et al.*, 2011; Karbalaei *et al.*, 2018; Zhang *et al.*, 2020). Those purely anthropogenic contaminants, defined as particles with shape and surface and classified about their size, are an emerging threat to global ecosystems (Machado, Anderson A. de Souza *et al.*, 2018). Conventionally, MP particles are plastic particles formed by polymerization (polymers) that have a size between 1 and 5000 μm . In addition, larger particles are called *mesoplastics* (MEP, $> 5000 \mu\text{m}$) and smaller particles *nanoplastics* (NP, $> 1 \mu\text{m}$) (Andrady, 2017). The exponential growth of global plastic production since the 1950s (1950: 1.7 Mt) resulted in an annual production of about 368 Mtonne in 2019 and so for more than 70 years have entailed huge potential for plastics entering the environment (PlasticsEurope, 2018, 2020).

Plastics entering the environment, regardless of its size, can be broken down further and further over time by physical and chemical processes (Napper & Thompson, 2019; Chamas *et al.*, 2020). A property of polymers is that they decompose slowly and do not dissolve. For this reason, modeled half-life times for buried plastic particles can exceed 2,500 years, showing a long residence time in the environment (Chamas *et al.*, 2020). If, despite insufficient knowledge, we consider a global "plastic cycle" as comparable to a geological cycle, sooner or later all plastic residues end up in the oceans (Zalasiewicz *et al.*, 2016). Plastic pollution in the marine environment has been the starting point of today's research on MP since the 1970s and has also been one of the main focuses of the scientific community to date (Carpenter & Smith, 1972; Hidalgo-Ruz *et al.*, 2012; Wright *et al.*, 2013). Nevertheless, the question arises regarding how plastic residues reach the oceans. Because global plastic flows, rivers, and freshwater systems play an important role regarding the land-to-sea transport (Siegfried *et al.*, 2017; Alimi *et al.*, 2018; Lechthaler *et al.*, 2020), it is estimated that up to 91% of the global plastic waste entering the environment could be transported by rivers (Lechthaler *et al.*, 2020). Although these findings are models, the quantification of plastic loads in small to large rivers illustrates that most plastics can be found there. This assumption is supported by the fact that a large proportion of global plastic is produced and consumed further away from oceans (Barnes *et al.*, 2009; Ellen MacArthur Foundation, 2017). However, freshwater systems cannot be regarded as transport routes only. Going back to the idea of a global plastic cycle as pendant to geological cycles, temporary deposition of e.g., sediments in river systems is known. This concerns especially floodplains as the surrounding areas of rivers in their morphological transfer zone and part of the river system. Build up from fluvial deposits, which are also known exactly to be temporary sinks for sediments (Bridge, 2003; Brierley & Fryirs, 2007; Fryirs & Brierley, 2013).

Floodplains are important ecosystems and natural habitats. They have important functions for the water balance (flood retention, groundwater recharge), and they are the connecting space between the river and its catchment. They are also the drainage system of landscapes (Bridge, 2003). They join and follow the river through various landscapes, like geological bedrocks, and are known as important deposition and accumulation sites for fluvial sediments, nutrients, and pollutants (e.g., heavy metals) (Opp *et al.*, 1993; Kalias *et al.*, 2003; Houben, 2012; Fryirs & Brierley, 2013; Martin, 2019). Floodplains are a transition zone between fluvial and terrestrial environmental systems, resulting in the formation of semiterrestrial floodplain soils. In addition, they have an important function for humans in several parts of the world in that they have suitable conditions for agricultural use and are often subject to intensive cultivation (Blettler *et al.*, 2017; Scheurer & Bigalke, 2018). The worldwide floodplain area consists only of 0.61% of the total terrestrial continental area, which accounts for approximately 806,525 km^2 in Europe (Nardi *et al.*, 2019). In a national comparison, the floodplain areas of rivers with a catchment area $> 1,000 \text{ km}^2$, accounting for around 4.4% of the national area in Germany (Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit, 2009).

In contrast to the scope of research in the field of freshwater systems, the number of studies on microplastics in floodplains and floodplain soils is still very limited, with only three studies focusing on the quantification of MP concentrations within floodplain soils. In principle, Scheurer and Bigalke (2018) were able to show, based on the example of Swiss floodplain soils, that MPs are found in 90% of the investigated soils and that the amount of MPs found (0 to 55.5 mg kg^{-1}) is clearly related to the population density of the respective river catchment. Another study from 2020 was able to show, using the example of the floodplains of the River Inde (North Rhine, Westphalia), that microplastic contents increase with increasing flow length, particularly with respect to sliding slopes of meandering bends that act as microplastic hotspots (Lechthaler *et al.*, 2021). Furthermore, a first reconstruction of the floodplain chronology based on MP was achieved within this study. However, both studies focus on riparian soil areas and do

not consider any or restricted depths under 5 cm (Scheurer & Bigalke, 2018) or 110 cm at riparian sites and slopes (Lechthaler *et al.*, 2021). Considering floodplain soils as a potential temporary sink for microplastics, this explorative view is insufficient but understandable because of the early stage of the research field. For a spatial quantification and a deeper understanding of the deposition processes of MPs within floodplain systems and soils, more systematic studies, in contrast to the established “explorative studies” in the field of soil science, are required (Weber *et al.*, 2020). The third study, which is a preliminary study to the one presented here, showed that MEP and coarse microplastics (CMP, > 2,000 μm) with average loads of 2.06 p kg^{-1} ($\pm 1.55 \text{ p kg}^{-1}$) and 1.88 p kg^{-1} ($\pm 1.49 \text{ p kg}^{-1}$) are widespread, but are spatial heterogenous distributed in floodplain soils, down to depths between 75 and 100 cm (Weber & Opp, 2020). MEP and CMP particles reaching those depths within floodplain soils indicate that not only depositional processes but also in-situ vertical transfer takes place within these soils (Weber & Opp, 2020).

The current state of research proves that plastics and MPs are present in floodplain soils and that these soils can be considered a potential temporal sink for MP. However, input processes like the deposition by flood dynamics or other sources, as well as the mobility of introduced and deposited MP within floodplain soils, still remains unclear. Both questions are of particular relevance, however, given that plastics in soils are an increasing threat to terrestrial ecosystems. Previous studies suggest a wide range of impacts of MP on soil properties (physical and chemical), soil organisms, and plant growth (Huerta Lwanga *et al.*, 2017; Rillig *et al.*, 2017; Hüffer *et al.*, 2019; Zhang *et al.*, 2019). Moreover, 95% of global food production is obtained directly or indirectly from soils (Food and Agriculture Organization of the United Nations [FAO], 2015). If soils are vulnerable to plastic, and if initial evidence shows that MP can also enter the food chain and the human body, the consequences for global food security and the ecological state of soils will be unacceptable for the society in the Anthropocene (Lahive *et al.*, 2019; Rillig *et al.*, 2019; Selonen *et al.*, 2020; Ragusa *et al.*, 2021).

This paper aims to improve the understanding of the deposition and potential in-situ transport of MP in floodplain soils on the basis of a geospatial research approach with respect to landscape characteristics and a holistic method approach. The present study focuses on the quantification and spatial distribution of large MP (L-MP, 2,000–1,000 μm) and medium MP (M-MP, 1,000–500 μm) because particle size is the decisive factor in determining deposition properties and potential mobility within floodplain soils (Kooi & Koelmans, 2019; Waldschläger & Schüttrumpf, 2019a).

Based on a holistic research approach containing preceding sampling site selection and systematic soilscape survey, analyses of MP, physical soil properties, and recent sediment dating, the following key issues should be addressed: (1) Which concentrations of L-MP and M-MP occur in floodplain soils? (2) Is it possible to trace the lateral and vertical spatial distributions of MP back to a specific environmental driver (e.g., land-use or flood dynamics)? (3) Can the conditions and processes of MP deposition in floodplain systems be reconstructed with the use of MP as a stratigraphic marker? This is intended to achieve the goal of an improved understanding of the spatial dynamics of MP within the three-dimensional system of floodplain soils and to enable targeted research on responsible processes for MP distribution in floodplain systems. In addition, the role of MP in the sedimentary budget and the geological cycle of the Anthropocene will be assessed on the basis of the questions presented.

2 Materials and Methods

2.1 Study Area

The implementation of a geospatial sampling approach took place within the floodplain area of the Lahn River (Hesse, Germany). The Lahn River, with a total length of 245.6 km and a catchment area of 5,924 km², is located within the central German low mountain range (Figure S1) (Regional Council Giessen, 2015; Meschede & Warr, 2019). Based on catchment geology, resulting landscape properties, and hydrology, the Lahn valley consists of different zones: (A) upper course (Rhenish Slate Mountains, smaller floodplain), (B) middle course (a sequence with wide, basin-like and narrow valleys with changing wide and narrow floodplains), (C) narrow valley (almost without distinctive floodplain), and (D) lower course with individual valley widenings and floodplains (Weber & Opp, 2020). The upper floodplain sediments and soils within the Lahn River floodplain are formed by the deposition of organic-rich silt and loams during the late Holocene and latest Pleistocene on older Pleistocene gravel and sand deposits (Rittweger, 2000; Bos & Urz, 2003). The floodplain soils within the direct surrounding area of the Lahn River comprise a total area of 88.9 km² within the federal state of Hesse. Major soil types are Fluvisols (70.7 km²),

Gleyic Fluvisols (6.2 km²), Fluvic Gleysols (2.7 km²), and Stagnic Fluvisols (6.4 km²) (Hessian Agency for Nature Conservation, Environment and Geology, 2020).

The Lahn River valley is subject to frequent flood events, despite of the anthropogenic changes like partwise canalization or river conversions in the case of flood management measures (Gleim & Opp, 2004). Even if very strong floods, such as the widespread medieval flood events (e.g., 1255, 1552) have become rarer, the last century's high-flood event occurred only 37 (1984) years ago (Gleim & Opp, 2004). Therefore, a steady record of sediment deposition and local bank erosions through floods can also be observed in the present (Martin, 2015, 2019). The investigated floodplain soils are partwise under agricultural usage (crop- or grassland), except for riparian strips and settlements. In general, the Lahn River catchment could be named called rural, with only 8.4% of strong anthropogenic land use (urban, traffic, industry) and a population density of 266 inhabitants per km² (Regional Council Giessen, 2015). Urban parts are restricted to four medium-sized cities along the river course (Martin, 2012).

2.2 Geospatial Sampling Approach

For a deeper understanding of the spatial dynamics of microplastics in floodplain soils, systematic studies with a focus on spatial representativeness in contrast to established explorative studies are required (Weber *et al.*, 2020). A first introduction of the approach presented here was made by Weber and Opp (2020), following the suggestions of Weber *et al.* (2020) and Weihrauch (2019). The common approach in environmental science studies soils and microplastics within soils at individual sites (e.g., independent profiles, topsoil samples) without a wider spatial context as a result of logistical limitations. However, many key processes for soil formation act not only locally but in the context of a wider landscape (Weihrauch, 2019; Weber *et al.*, 2020). The soils in floodplain areas are especially the result of processes and, despite anthropogenic influence, are subject to processes that not only are local but also affect the entire floodplain and are significantly determined by river systems, catchment sections, or the entire catchment. To record and study the environmental processes and drivers that are responsible for the distribution and spread of microplastic in floodplain soils, it is necessary to consider a larger part of the landscape. Floodplain soils have to be understood as part of a landscape in which environmental processes take place and thus as part of a "soilscape" (Willgoose, 2018). In the case of the floodplain soils studied here, and with regard to microplastic pollution, various processes such as flooding, groundwater recharge, or land use are important because they could have an influence on the deposition, accumulation, and mobility of plastic particles in the soils itself. To understand these processes, not only on a soil profile itself but also in the spatial context of the floodplain landscape and its soilscape, a geospatial approach was applied to identify suitable and representative sampling sites. Each sampling site must therefore be representative for a larger part of the river area (Lahn River zones, Chapter 2.1) and its floodplain in the case of soil formation, morphology, flood dynamics, and land use.

To identify suitable sampling sites within in the floodplain landscape of the Lahn River, the following criteria were set: (1) location within a natural flood retention area with potential for frequent flood events (recent), (2) Extended floodplain area with a sequence of different morphological units and soil differentiation, (3) land use differences in floodplain cross-section and undisturbed floodplain cross-section (e.g., no railway dams), and (4) no direct potential anthropogenic MP source like highways or industrial sites (Weber & Opp, 2020).

After identifying suitable sampling sites and an initial soil mapping to obtain an overview on soilscape properties, four floodplain cross-section transects were selected that are representative of a larger floodplain area with comparable soil properties (same locations as Weber & Opp, 2020). The transect sites corresponded to the valley sections A: site ELM; B: site ROT and STD; and D: site LIM (Chapter 2.1). At each transect, two sampling plots (ELM, ROT) or four sampling plots (STD, LIM) were established between the river bank and the floodplain edge (Figure S2). Two soil profiles with a distance of 5 m to each other were drilled to a depth of 2 m via pile core driving (100 mm and 80 mm core diameter) and sampled according the following depth sections: 10 cm sections (for 0–0.5 m), 25 cm sections (for 0.5–1.5 m), and 50 cm sections (for 1.5–2.0 m). Sample material with an average sample mass of 1150 g (in total 120 samples, 111 for MP analyses) (Figure S3) was transported in cornstarch bioplastic bags (Mater-Bi bags, Bio Futura B.V., Rotterdam, Netherlands). The comparatively large amount of sample material was taken because previous studies have shown very heterogeneous occurrences of microplastics in soils (Liu *et al.*, 2018; Scheurer & Bigalke, 2018; Corradini *et al.*, 2019). Soil samples for dating were taken at the transect sites ELM (upper course, section A, core ELM-D) and LIM (lower course, section D, core LIM-D) at the beginning of the plain floodplain (behind the riverbank, not under landuse). For this purpose, a drill core (80 mm, 1 m depth) was drilled and then excavated to obtain a drill core free of disturbances as far as possible. The core was subsequently divided into 2 cm sections, which were collected in the field and transported in PE-bags. In this way,

42 samples from core ELM-D (0–84 cm soil depth) and 45 samples from core LIM-D (0–90 cm soil depth) could be obtained. Stratigraphy and pedogenesis of each soil profile were documented according the German soil classification (Ad-hoc AG Boden, 2005), the FAO Guidelines for soil description (FAO, 2006), and the WRB 2015 (IUSS Working Group, 2015). Soil properties (horizon sequences) and soil type according to WRB are documented in Table T1 (Supplementary). Contamination prevention during sampling process was performed by avoiding plastic tools and using stainless-steel spatula, steel drill equipment, and bioplastic bags.

2.3 Sample Preparation, Soil Analyses, and Dating

Sample preparation followed the sample preprocessing process introduced by Weber and Opp (2020) for the analysis of MEP and CMP. Basically, the sample preprocessing was carried out without using plastic tools or materials (including cotton lab coats) and by reducing the exposition time for each sample as much as possible to avoid air contaminations. Soil samples for microplastic analyses were transported in cornstarch bags and dried at 45°C in a drying chamber for a maximum of 4 days. Subsequently, the samples were carefully mortared to solve soil macroaggregates because microplastics can be attached within those aggregates (Zhang & Liu, 2018). This process was carried out manually to ensure a careful dissolution of the macroaggregates with minimum impact on plastic particles. Afterwards, the sample material was dry sieved according to the size classes of MEP (> 5,000 µm), CMP (> 2,000 µm), and MP (< 2,000 µm) through stainless-steel sieves (Retsch, Haan, Germany). During the sieving process, sieves were covered with stainless-steel plates. To homogenize the sample and simultaneously obtain a representative subsample (approx. 12.5 % of total sample volume) for standard soil analyses, the sample was divided by means of a rotary sampler (Retsch, Haan, Germany). Each size fraction and subsample was afterwards weighed and stored in fresh cornstarch bags.

Soil samples for dating purposes were processed in a comparable manner: The samples were transported in PE-bags, weighed wet, and then dried at 50°C in a drying chamber for 4 days. Afterwards, they were ground and sieved to 2 mm through stainless-steel sieves. The coarse soil fraction (> 2 mm) and the fine soil fraction (< 2 mm) were dry weighed. A subsample (approx. 27 g of fine soil material each) was placed in 50 mL PE-containers and sent to the laboratory for further analyses. Soil moisture, coarse soil fraction, and soil density was calculated.

Standard soil analyses included the determination of organic matter (OM) and texture analyses. The content of OM was measured by loss of ignition (DIN ISO 19684-3:2000-08). Soil texture was determined for each sample with the Integral Suspension Pressure Method (Durner *et al.*, 2017) after samples had been prepared according to DIN ISO 11277:2002–08.

Samples for dating purpose were analyzed at the Gamma Dating Center, Department of Geosciences and Natural Resource Management, University of Copenhagen for measurements of the activity of ^{210}Pb and ^{137}Cs via gamma spectrometry. The measurements were carried out on a Canberra ultralow-background Ge-detector. ^{210}Pb was measured via its gamma-peak at 46.5 keV, ^{226}Ra via the granddaughter ^{214}Pb (peaks at 295 and 352 keV), and ^{137}Cs via its peak at 661 keV. The chronologies were calculated using a constant rate of supply (CRS) model in which the activity in the lower portion of the cores was calculated on the basis of a regression of the activity of unsupported ^{210}Pb versus cumulated mass depth (Appleby, 2001; Andersen, 2017).

2.4 Microplastic Analyses

Presented analyses of microplastic particles within floodplain soils consisted of a three-step procedure: (1) separation, (2) staining, and (3) identification. Because soil is an environmental medium containing different materials or substances, the major components, namely (a) mineral component and (b) organic component, must be separated from the searched component, (c) microplastics (Hurley *et al.*, 2018; Möller *et al.*, 2020; Ruggero *et al.*, 2020; Thomas *et al.*, 2020).

Separation of mineral components was performed within the “MicroPlastic Sediment Separator” (MPSS) (Hydro-Bios Apparatebau GmbH, Kiel-Altenholz, Germany) as a separation unit and the only commercially available. The advantage of handling a large amount of sample against the background of a heterogeneous distribution and unknown concentrations of plastic particles in the environment must be highlighted at this point, as should the disadvantage of a long separation time due to the size of the device (Standpipe). Based on the principle of density

separation, organic sample components and the searched plastic can rise through the comparable material density within the unit, while mineral components remain at the bottom (Imhof *et al.*, 2012). Recovery rates of the MPSS are estimated with 100% for CMP and large MP (> 1 mm) and 95% for smaller MP (1,000–40 μm) (Imhof *et al.*, 2012). Potential plastic contaminants (e.g., room air) were controlled by blank samples (Stock *et al.*, 2019). In a total of five blanks (during 41 MPSS runs with a maximum of three instruments), 7 potential plastic particles (fragments and filaments) with a mean size of 341.28 μm were found. Because of their size, visual identification of the particles was not possible through ATR-FTIR.

The MPSS unit was filled with a sodium chloride (NaCl) solution, density adjusted to approximately 1.2 g/ml and sieved with a 300 μm stainless-steel sieve. Solution density was controlled before and after separation process via pipetting and weighing, along with the help of an aerometer (Figure S3). With revolving rotor, sample material was added, and the MPSS unit was closed (dividing chamber). The rotor was left running for 60 minutes, and separation process left for another 14 hours (Imhof *et al.*, 2012). After separation time, the integrated ball valve was closed and the dividing chamber was removed to allow the separated material to be rinsed into glass beakers using filtered (< 300 μm) NaCl solution. After the separation process, the remaining sample material was separated into the following size classes using stainless-steel sieves (\varnothing 75 mm, Atechnik, Leinburg, Germany) and deionized water: $> 1,000$ μm (L-MP), > 500 μm (M-MP), and 500 to 50 μm (S-MP). After sieving, the sieve residues were filtered on pleated cellulose filters (LLG-Labware, Meckenheim, Germany) and for S-MP via vacuum filtration on round cellulose filters (LLG-Labware, Meckenheim, Germany). The S-MP fraction was saved for later analyses, currently in progress.

To separate the remaining sample material of organic material and potential plastic particles in L-MP and M-MP fraction, a Nile Red staining procedure and visual fluorescence identification was applied (Maes *et al.*, 2017; Konde *et al.*, 2020). Following the suggestions of Konde *et al.* (2020) a Nile Red solution with a concentration of 20 $\mu\text{g/mL}$ Nile Red (Sigma-Aldrich, Taufkirchen, Germany) solved in an ethanol-acetone (1:1) mixture was dropped on each filter, using a pipette and subsequently sprayed on (using a spray bottle). The initial dropping prevents the loss of particles due to the subsequent spraying with uniform coverage of all particles on the filter. Filters were stained for 10 minutes at 50°C within a drying chamber before visual analyses (Konde *et al.*, 2020). Stained filters were afterwards visual detected under a stereomicroscope (SMZ 161 TL, Motic, Hong Kong) with fluorescence setup (Excitation: 465 nm LED; Emissions 530 nm color long pass filter: Thorlabs, Bergkirchen, Germany) (Konde *et al.*, 2020). Filters were detected systematically to observe the entire filter surface under fluorescent and white light (Figure S4 and S5). Each fluorescent particle and other potential plastic particles (matching the criteria according to Norén, 2007) were collected and stored in microplates (Brand, Wertheim, Germany). Potential plastic particles were classified according to surface characteristics, which were photographed (Moticam 2, Motic, Hong Kong) and measured (Motic Images Plus 3.0, Motic, Hong Kong).

For final identification and to avoid overestimations, each potential plastic particle was analyzed with a Tensor 37 FTIR spectrometer (Bruker Optics, Ettlingen, Germany) combined with a Platinum-ATR-unit (Bruker Optics, Ettlingen, Germany). In some cases, adherent soil or organic material was removed with stainless-steel tweezers. The Platinum-ATR-unit was cleaned with 2-propanol ($\text{CH}_3\text{CHOHCH}_3$) between each measurement. Measurement was performed with 20 background scans followed by 20 sample scans for each sample (Jung *et al.*, 2018). Spectral resolution was set to 4 cm^{-1} in a wavenumber range from 4,000 cm^{-1} to 400 cm^{-1} (Primpke *et al.*, 2017; Primpke *et al.*, 2018).

2.4.1 Limitations and insecurities

Despite the continuous development of analytical methods for microplastics in soils, no standardized method has been established to date (Bläsing & Amelung, 2018; Möller *et al.*, 2020). The present work focuses on the combination and adaptation of methodological approaches already presented. First, in the case of density separation within the MPSS unit, the recovery rates were reported to be $> 95\%$ for particles down to 40 μm in size by the developers (Imhof *et al.*, 2012). However, the construction and size of the device involves considerable time and expense, which is only profitable when larger sample volumes are used (Coppock *et al.*, 2017). In contrast to other concentrated salt solutions (e.g., ZnCl_2), the flotation medium that was used (NaCl) is cost effective and environmentally friendly (Coppock *et al.*, 2017). A limitation arises from the fact that only plastic particles with a density of < 1.2 g/ml can be separated. Common polymer types such as Polyethylene terephthalate (PET) or Polyvinyl chloride (PVC) may not be separated. Second, concern regarding the Nile Red staining procedure and

recovery rates up to 96.6% from spiked marine sediments in combination with density separation are documented (Maes *et al.*, 2017). In contrast to other methods such as the oxidation of organic matter (e.g., by acid digestion), Nile Red staining offers a completely particle-preserving approach. Depending on the surface of organic material, but especially on calcium-containing shells (e.g., isolated freshwater mussels in alluvial sediments), Nile Red can also bind these materials and thus hinder the visual distinction between plastic and non-plastic (Konde *et al.*, 2020). From 225 selected particles during the Nile Red staining procedure, 46 particles were too small for ATR-FTIR analysis ($> 300 \mu\text{m}$, adherent to organic components); and of 179 analyzed samples, 21 particles (11.73 %) were classified as natural (non-plastic) material. This limitation can and must be lifted by (a) systematic examination of the sample under fluorescent and white light, (b) collection of all potential particles, and (c) a subsequent identification of the particles using analytical methods such as ATR-FTIR to prevent overestimation. Because the application of an FTIR with ATR unit (like Tensor 37, Bruker Optics, Ettlingen, Germany) is carried out by hand only, particles smaller than $500 \mu\text{m}$ or $300 \mu\text{m}$ should not be investigated because they are difficult to handle and as the sample and because ATR crystal often have insufficient contact area. Furthermore, in the case of heavily degraded and weathered particles, there may be no match with different spectral databases because the quality of the spectra is insufficient (Primpke *et al.*, 2018).

2.5 Data and Statistical Analyses

Data processing of FTIR spectra was performed in OPUS 7.0 (Bruker Optics, Ettlingen, Germany) and Spectragryph (Version 1.2.14; Menges, 2020; Oberstdorf, Germany). Spectra identification and polymer type assignment was carried out according to a previously defined scheme (Figure S6). Because the available OPUS database (OPUS 7.0 internal database) contains only insufficient entries of plastic products (only industrial polymers) and no entries of environmental materials, the following procedure was performed with each spectrum: Particles with a identification hit quality higher than 700 within OPUS 7.0 was counted as identified polymer type or group. In the case of particles that show an identification hit quality between 700 and 300, the absorption bands were manually checked for polymer identification according to the criteria of Jung *et al.* (2018). If a sufficient identification of absorption bands were possible, then the particles were also counted as identified polymer type or group (Jung *et al.*, 2018). Spectra of particles with a hit quality less than 300, as a limit for satisfactory identification (Primpke *et al.*, 2017; Lorenzo-Navarro *et al.*, 2018; Primpke *et al.*, 2018), were automatically compared with spectra databases for natural materials provided by Spectragryph (Kimmel_Center: Collection of 363 FTIR absorbance of natural and biogenic material of archeological interest. Provided by S. Weiner from Kimmel Center for Archaeological Science, Weizmann Institute of Science, Israel). In the case of no match in both cases, the spectra were finally matched with the database of “Open Specy” (Cowger *et al.*, 2020). In this case, a satisfactory match was always achieved. In case of a match with natural or biogenic material, potential plastic particles were counted as natural material and excluded from further analyses.

Basic statistical operations were performed in Microsoft Excel 2013 (Microsoft; Redmond, WA, USA), in R (R Core Team, 2020), and RStudio (Version 3.4.1; RStudio Inc.; Boston, MA, USA). Data visualization, tests for normal distribution (Shapiro-Wilk), linear regression analyses, Pearson or Spearman correlation analyses, and variance analyses (ANOVA) were conducted with the standard R-packages and “graphics,” “stats,” “vioplot,” “ggplot2,” “ggirides,” and “scatterplot3d.” We interpreted statistical analysis results as significant with a p-value < 0.05 .

3 Results and Discussion

3.1 Microplastics and their characteristics in floodplain soils

In total, 149 particles from 111 soil samples could be successfully and clearly identified as plastic particles. There is a positive rate (plastic containing samples) of 71.2% within the samples from topsoils down to 2 m deep soil layers. Nile Red staining shows a false positive rate of approximate 12.0 %. However, even if only the definitely identified particles are used for further data evaluation, it should be clear that in all cases the lower range of the actual plastic pollution is concerned. The identified particles result in average plastic concentrations of 2.14 mp/kg^{-1} (number of particles in particle size class per soil dry weight), composed from $2.13 \text{ L-mp/kg}^{-1}$ and $1.91 \text{ M-mp/kg}^{-1}$ (Figure 1a). With the exception of the plastic-free samples (28.8 %), the values vary between 0.37 mp/kg^{-1} up to 13.54 mp/kg^{-1} for the entire MP amount, with absolute maximum value of $16.93 \text{ M-mp/kg}^{-1}$ including MEP and CMP amount from the preliminary study (Weber & Opp, 2020). In comparison to other studies that reported the MP concentrations for

soils by particle/per kg, the concentrations found are clearly below those in agricultural topsoils (Liu *et al.*, 2018; Zhang & Liu, 2018; Corradini *et al.*, 2019). A sufficient comparison with the first study on microplastics in floodplain topsoils, reported for Swiss nature reserves by Scheurer and Bigalke (2018), is not feasible because of the different used units (mg/kg). In the case of the investigation of bank profiles and topsoils of the Inde River (North Rhine-Westphalia) by Lechthaler *et al.* (2021), the corrected average concentrations with 47.9 mp/kg⁻¹ (depth profiles) and 25.4 mp/kg⁻¹ (surface samples) also exceed those available here.

Comparable to the results for larger plastic particles (MEP, CMP) (Weber & Opp, 2020) and to the work of Lechthaler *et al.* (2021), the contents in topsoil (0–30 cm) are clearly above those in subsoil (30–200 cm) (Figure 1b). Average values of 3.33 mp/kg⁻¹ in topsoils and 1.34 mp/kg⁻¹ in subsoils are significantly different (p-value = 0.0002).

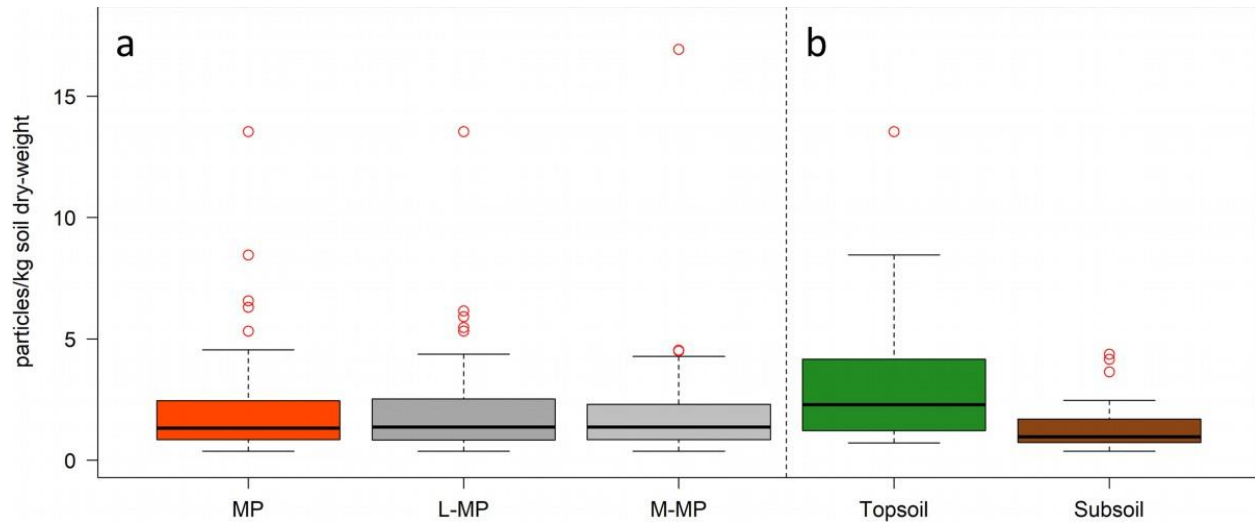


Figure 1. Overview of microplastic concentrations in floodplain soils. (a) Concentrations auf total microplastics (2,000–500 μm , MP), large microplastics (2,000–1,000 μm , L-MP), and medium microplastics (1,000–500 μm , M-MP) given in particles (p) per kg soil dry-weight. (b) Concentrations of total microplastics (2,000–500 μm , MP) within topsoil (related soil A-horizon, depth: 5 to max. 30 cm) and subsoils given in particles (MP) per kg soil dry weight.

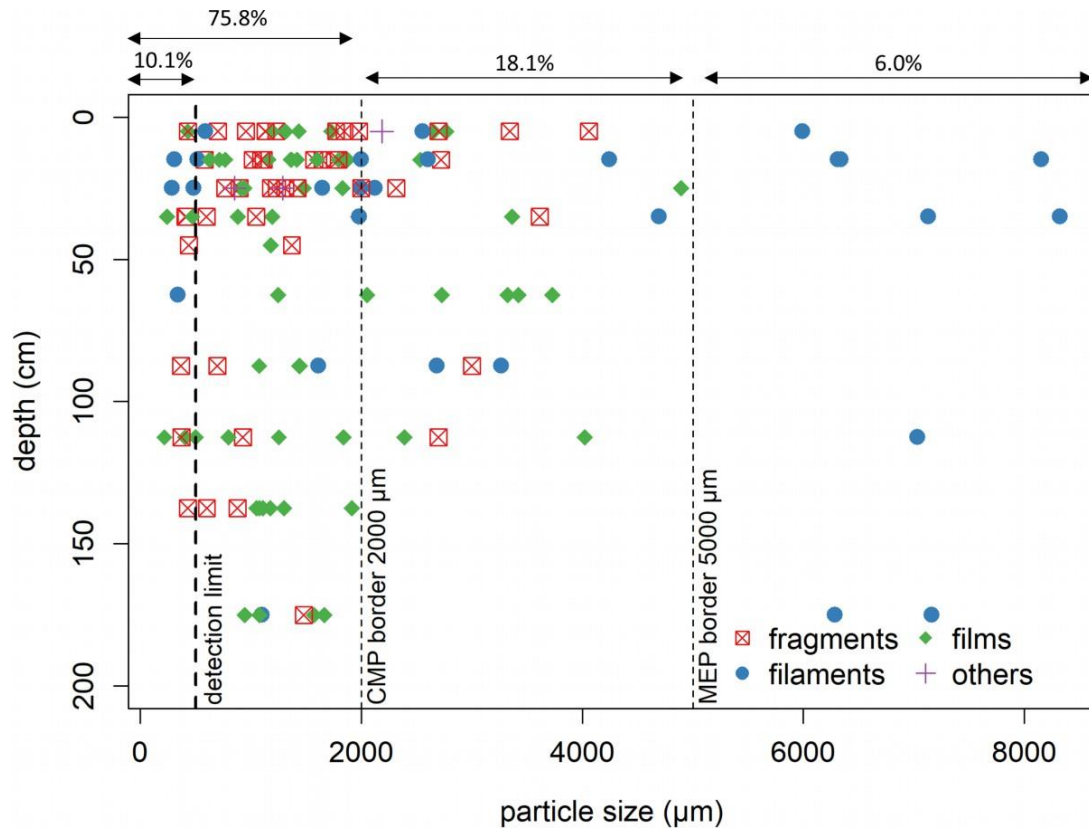


Figure 2. Depth distribution of plastic particles and size classified by plastic shape, including size borders of the ATR-FTIR detection limit (500 μm), CMP (2,000 μm) border, and MEP (5,000 μm) border.

Particle size ranges between 219 μm to 8,321 μm , with an average of 1,171 μm ($n = 149$) for identified particles. Therefore, 10.1% of all ATR-FTIR-analyzed particles were smaller than the fixed detection limit of 500 μm (Figure 2). Major share of particles size lies between the minimum value and 2,000 μm (MP to CMP border) within the upper 50 cm of soil column, including fragments, films, and some filaments. Against expectations, 24.1% of identified particles show a size larger than the CMP- and MEP-size border, despite the previous sieving procedure. Because the longest diagonal of the particles was consistently measured, the width of these particles can be less than 2,000 μm . This is also clear from the fact that only filaments (measurement of filament length) occur in the size range above 5,000 μm .

The characteristics of detected plastic particles also correspond to the findings of other studies. Major particle types are films (42.3%), fragments (31.5%), and filaments (24.2%) with usually irregular or degraded shapes, except in the case of filaments, which show regular surfaces (Figure 3, S7). This result clearly corresponds to the findings within the Inde River catchment (Lechthaler *et al.*, 2021). More than half of all particles detected show a weathered (59.7%) or incipient alteration (11.4%) surface according to visual criteria, whereas fresh surfaces occur for 20.1% of all particles.

Typical polymer types like low-density polyethylene (LDPE) or high-density polyethylene (HDPE), polystyrene (PS), and polyamides (PA, including Nylon-6) could be identified by ATR-FTIR analysis. The results correspond to the most frequently produced and used plastics in Europe (PlasticsEurope, 2018). One surprising finding was the high number (20.1%) of resins (synthetic or polymer resins, grouped as resins) in contrast to other current studies on microplastic in soils (Liu *et al.*, 2018; Piehl *et al.*, 2018; Scheurer & Bigalke, 2018; Zhang & Liu, 2018; Corradini *et al.*, 2019; Lechthaler *et al.*, 2021). The particles classified as “resins” could be significantly identified only by the OpenSpecy database. The resin entries contained in the database are from Primpke *et al.* (2018) and include epoxides, polyurethane acrylic, phenoxy, and polyamide resins, which explains the frequent assignment in OPUS 7.0 to PA (with low hit quality). In principle, however, having resins as the largest group of plastic types is plausible

because these are used in countless objects, and nonfiber plastics contain 97% polymer resins in addition to other additives (7%) (Zink *et al.*, 2018).

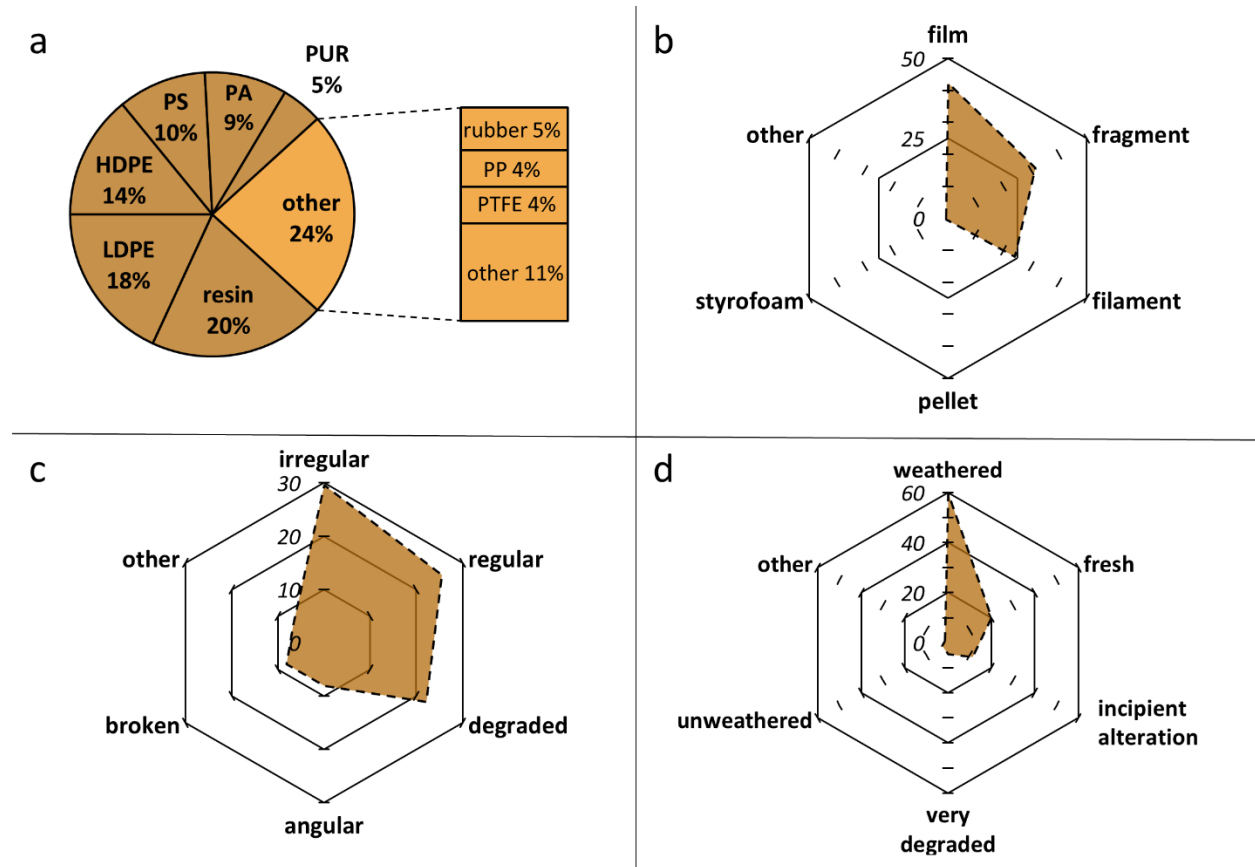


Figure 3: Microplastic particle characteristics. (a) Identified polymer types through ATR-FTIR analyses (class other includes polymers such as PET, CSM, ABS). (b) Percentage share of particle types. (c) Percentage share of surface forms. (d) Percentage share of surface conditions.

3.2 Lateral microplastic distribution

The evaluation of the lateral distribution of microplastics in floodplain soils can be conducted at two spatial scales: Level one follows the catchment scale and the course of the river (metric measure: river-km), and level two follows the floodplain cross-sections (transect sites) with increasing distance to the watercourse (metric measure: distance in m). Figure 4a shows the range of total MP concentrations at each transect site from upstream (ELM, river km: 202) to the downstream site (LIM, river km 57). Average values are significantly different between site ELM and ROT, as well as ROT and STD. Median values including negative samples (zero-values) are 0.75 mp/kg^{-1} (ELM), 0.20 mp/kg^{-1} (ROT), 0.76 mp/kg^{-1} (STD), and 0.40 mp/kg^{-1} (LIM). An increase along the course of the river cannot be concluded on this basis. However, except for the upper reaches (ELM), maximum MP concentrations increase continuously from STD (upper middle reaches) to LIM (lower reaches). The simple assumption of an increasing accumulation of microplastics in floodplains with increasing flow length of the river, caused by the also increasing number of potential plastic sources and available water quantity and sediments, cannot be upheld unequivocally (Scheurer & Bigalke, 2018; Xiong *et al.*, 2018; Liu *et al.*, 2019). Moreover, local phenomena, instead of a superordinate accumulation, ensure a heterogeneous distribution. Nevertheless, the significant increase in maximum

concentrations seems to indicate that, with increasing river length and thus reduced flow velocity and higher frequency of floods, more microplastic particles can be deposited (Bridge, 2003; Brierly and Fryirs, 2007). Regarding the lateral level two, maximum concentrations are clearly reached next to the river bank and within a short distance from the river (Figure 4b). Independent of this, a widespread distribution of plastic contents around or above the mean value of 2.14 mp/kg^{-1} can be determined for the entire study in the floodplain cross-sections. According to the general knowledge that lateral sediment deposition decreases with increasing distance from the watercourse, which is reflected in the grain size differentiation of coarse-grained sandy sediments near the channel and fine-grained (clayey) sediments at the floodplain edge, this can also be assumed for the lateral distribution of plastic particles (Bridge, 2003; Brierly and Fryirs, 2007).

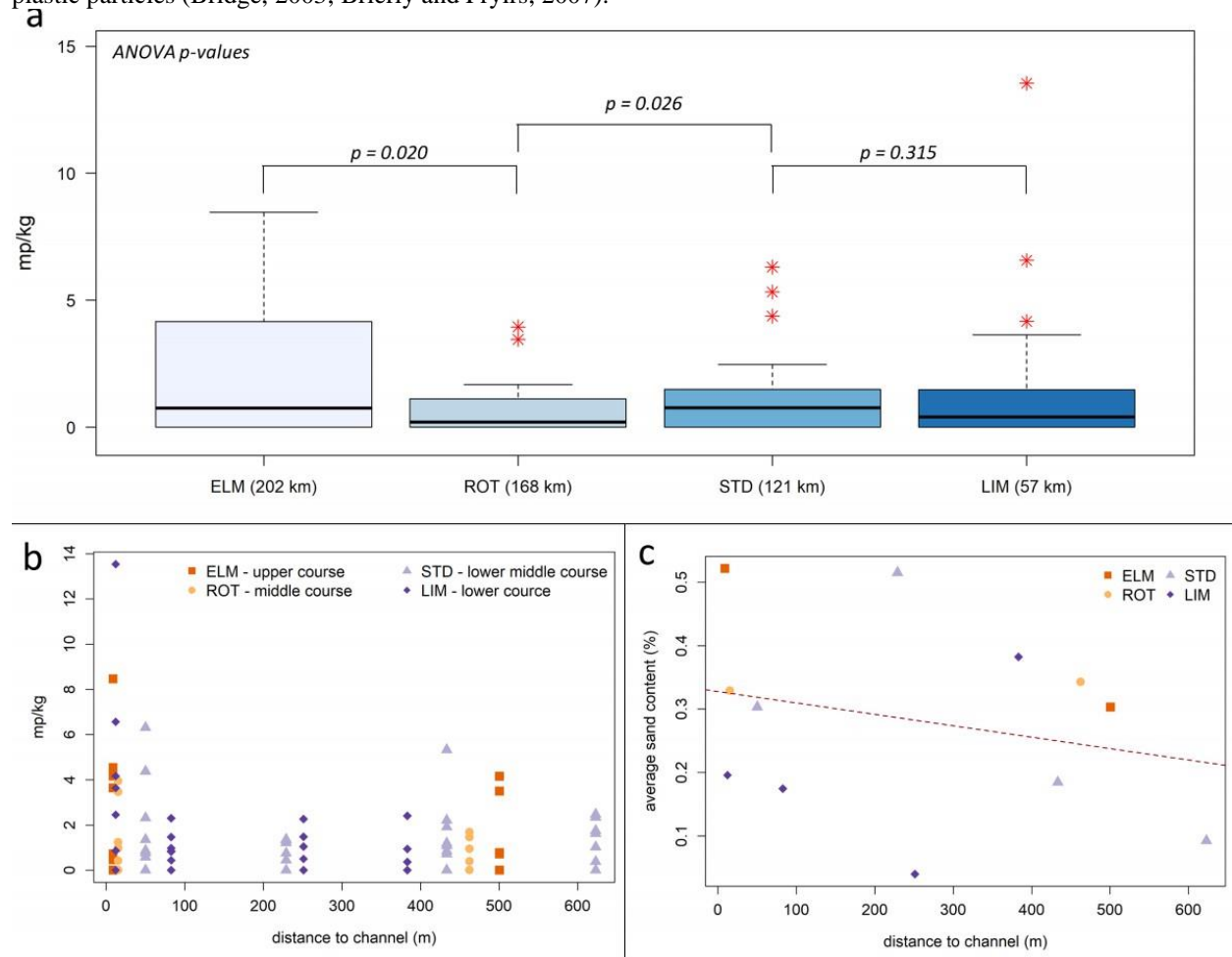


Figure 4. Lateral microplastic distribution. (a) Lateral microplastic distribution on the catchment scale given through total MP concentrations (mp/kg) according to sampling sites (Figure S1, Chapter 2.2) and river km. (b) Total MP concentrations (mp/kg) opposite distance to channel (m) at each sampling point classified according the four sampling sites. (c) Average sand content (%) opposite distance to channel (m) at each sampling point classified according the four sampling sites.

Figure 4c illustrates the average sand concentration within the upper 50 cm soil at each sampling point. Because the upper 50 cm of the floodplain soils contain the most microplastics (see Figure 2 and Chapter 3.3), the average sand content can provide information about the lateral deposition dynamics. With the exception of the ROT transect, the sand content of all transects decreases significantly with distance from the river. Outliers in the STD and LIM transects are due to the location of the points in a flow channel (STD-2, younger sandy flow channel deposits) and the over-deposition by slope erosion at the floodplain edge of the LIM transect (LIM-4). This results in a slight linear relationship between sand contents and distance ($R^2 = 0.126$, $p = 0.002$). The highest contents (maxima) are thus reached where the sand contents at transect level are also highest (near water bodies like river banks). Even

though the sedimentation and erosion properties of plastic particles differ from those of the sediment (Waldschläger & Schüttrumpf, 2019b), the question arises as to how this spatial distribution pattern is formed. In the area close to the watercourse (proximal floodplain), where increased deposition of sands take place, flow velocities should be significantly higher during floods as in the floodplain edge area (distal floodplain), where the reduced velocities (caused by higher terrain roughness) result in the deposition of finer sediments (clay and silt fraction) (Bridge, 2003; Fryirs & Brierley, 2013). However, the deposition of plastic particles also requires a reduced flow velocity because of the properties of the particles, especially their low density (Tibbetts *et al.*, 2018). Another explanation could be that more plastic particles are deposited in areas with increased accumulation of younger sediments take place (frequent flood activity and higher sediment accumulation at river banks). This explanation is supported by the low MP concentrations in active flood channel (STD-2, LIM-2), which might be caused by the erosion of the upper soil material and stored plastic particles inside. An explanation about land use and associated surface roughness (vegetation) also cannot be excluded (Klein *et al.*, 2015; Tibbetts *et al.*, 2018).

In general, the differences between different land use classes and associated vegetation (riparian, cropland, grassland) are not distinct (Figure 5a). Even though the "riparian" class with a median of 2.45 mp/kg⁻¹ is clearly above the concentrations of cropland (median 0.37 mp/kg⁻¹) and grassland (median 0.48 mp/kg⁻¹), this can be attributed to the increased concentrations in the area near the river banks (especially site LIM-1). Comparable to the results for larger plastic particles (MEP, CMP) (Weber & Opp, 2020), an accumulation of higher microplastic concentrations does not seem to occur in the area of intensive agricultural use (cropland). Although agriculture cannot be dismissed as a potential source of microplastics (e.g., from sources such as sewage sludge, compost, fertilizers, hay bale nets) (Corradini *et al.*, 2019; Braun *et al.*, 2021), the spatial position within the floodplain and the surface roughness caused by vegetation appear to be more important factors that can explain the lateral distribution of L-MP and M-MP particles.

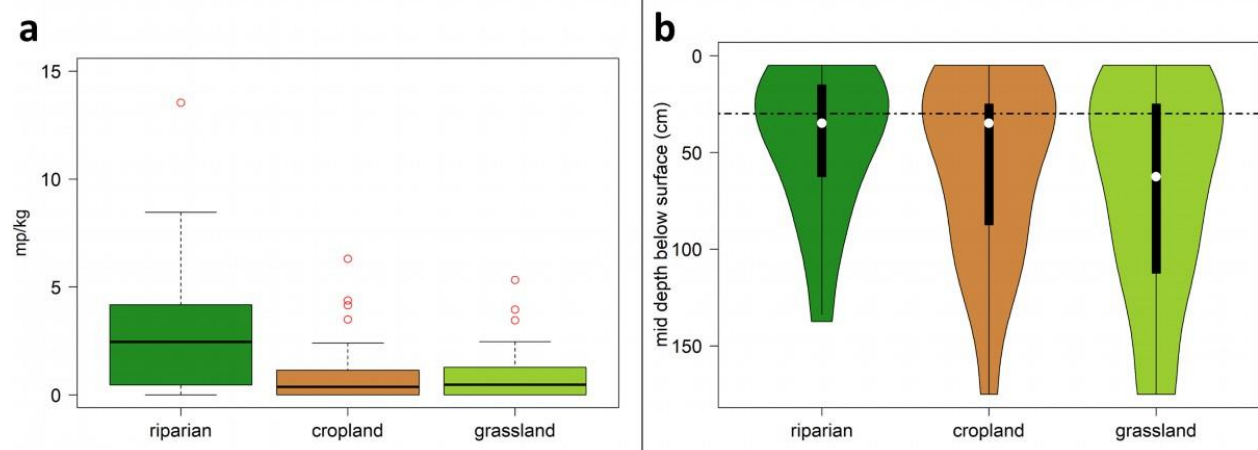


Figure 5: Microplastic distribution under different land use classes. (a) Total microplastic concentration (mp/kg) according to the land use classes. (b) Depth distribution (violinplot: boxplot with kernel density) of plastic concentrations (mean sample depth in cm) with medium tillage depth (plough tillage, for recent and relict soil horizons) by dotted line.

3.3 Vertical microplastic dynamics

The consideration of vertical microplastic distribution within floodplain soils as a three-dimensional system provides important information and enables understanding of vertical microplastic dynamics. With the exception of the study by Lechthaler *et al.* (2021), microplastics have never been searched for at soil depths up to 2 meters until now. If the depth of the floodplain soils is differentiated according to soil horizons, then the higher contents are clearly located in the topsoil horizons (represent A horizons) with average values of 3.33 mp/kg⁻¹ higher than in the subsoil horizons (e.g., B) with 1.34 mp/kg⁻¹ (Figure 1b, Table T1). Independent of the soil stratigraphy, the upper 50 cm (corresponding to 5 samples from one drill core) of the floodplain soils contain 67.65% of the identifiable plastic, whereas the depth range 50 to 200 cm (also 5 samples from one drill core) contains only 32.35% (Figure 2). With the exception of filaments, only particles with a size < 2,000 µm occur in depths greater than 125 cm. The median

particle size within the upper 50 cm lies at 1,421.5 μm with major particle types of fragments, films, and filaments, whereas within the lower 150 cm the median particle size is slightly smaller with 1,301.0 μm (films and fragments, less filaments). Considering the depth distribution in relation to land use (Figure 5b), the depth distribution is basically comparable, even if in the riparian area depths > 150 cm are only reached in a few single cases. Median depth distribution is 35 cm for riparian and cropland and 62.5 for grassland, where it becomes clear that, under grassland, significantly deeper depths are also reached. The vertical distribution at the sampling site level is independent of this. The greatest depths (150–200 cm) are reached at sites ROT (proximal floodplain) and STD (proximal and distal floodplain). The two deepest depths (125–150 cm) are at sites LIM (proximal floodplain) and ROT (proximal floodplain). Particles therefore reach depths of up to a maximum of 2 meters, regardless of the distance to the watercourse, the soil type, or land use. Significant correlations, neither superordinate nor site specific between depth distribution and soil parameters (e.g., grain size, bulk density, OM, root distribution), could not be found (examples given in Figure S8). This and the occurrence of zero-values (empty samples), by unidentified particles but also by no particles found, indicates a clearly heterogeneous depth distribution starting at 50 cm depth, whereas a more homogeneous distribution can be assumed in the upper soil areas (0–50 cm).

This also applies to the vertical distribution of polymer types and associated age of earliest possible occurrence (EPO age). Vertical structuring of floodplain soils is most commonly related to depositional history (sediment deposition), which is related to sedimentation rates (Brigde, 2003). Floodplain chronology can be assessed by different methods, as well as by plastics themselves. Since the global plastic productions started its exponential growth within the 1950s, plastics within floodplain soils could act as a new marker of floodplain chronology (Lechthaler *et al.*, 2021). For each identified polymer type, the EPO age can be added, based on the production starting year or year of patent registration (Weber & Opp, 2020; Lechthaler *et al.*, 2021). Based on the EPO age, each polymer can be used as a specific marker for the time between 1910 and 1990. From Figure 6, it becomes clear that the depth distribution of different polymer types is not equal over the depth. 71% of identified polymers including resins, LDPE, PS, and PA show a peak within the upper 50 cm of floodplain soils. Only HDPE, the third-most frequent polymer, shows an equal distribution over the depth. Assuming that polymers that have been released into the environment for a long time (e.g., rubber, resin, PVC with EPO ages < 1912) are found more frequently at deeper layers than "younger polymers" (EPO age > 1950), this should be reflected in the vertical distribution (Figure 6). In fact, this is only achieved for very young polymers such as chlorosulfonated polyethylene (CSM, EPO age: 1990s). All other polymers do not show a clear superordinate separation but also a heterogeneous distribution over the depth.

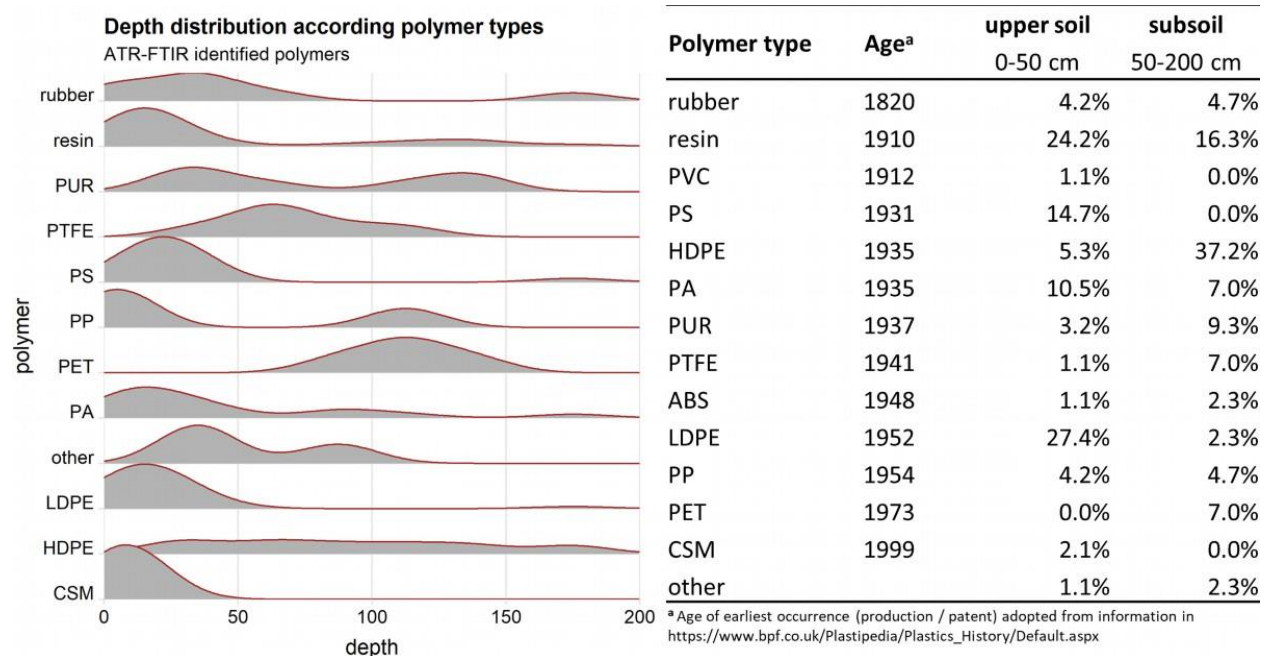


Figure 6. Depth distribution of different identified polymer types and occurrence of different polymer types (sorted by age, production start, or patent submission) in upper and lower soil areas.

The vertical distribution of the MP particles in combination with the evaluation of the EPO age-based depth distribution, underlines the clear separation into two parts: a) The upper floodplain soil area (approx. 0–50 cm) with a clear MP accumulation and the occurrence of very young polymer types as well as the b) lower soil area (approx. 50–200 cm) with partwise MP occurrence, individual hotspots and mixed occurrence of different EPO ages

3.4 Reconstruction of microplastic deposition and translocation

Information on lateral and vertical MP distribution within floodplains soils in combination with derived EPO age and other soil parameters allows a first holistic assessment of the deposition conditions of microplastics in floodplains based on a geospatial sampling approach. Summarizing the spatial distribution, it can be stated that a clear concentration of increased MP occurrences can be found in the area of the proximal floodplain (i.e., the upper 50 cm of the floodplain soil; Figure 7). Areal regression shows a clear decrease in MP content with increasing depth and a decrease with distance from the watercourse (distal floodplain), although individual hotspots occur near the surface.

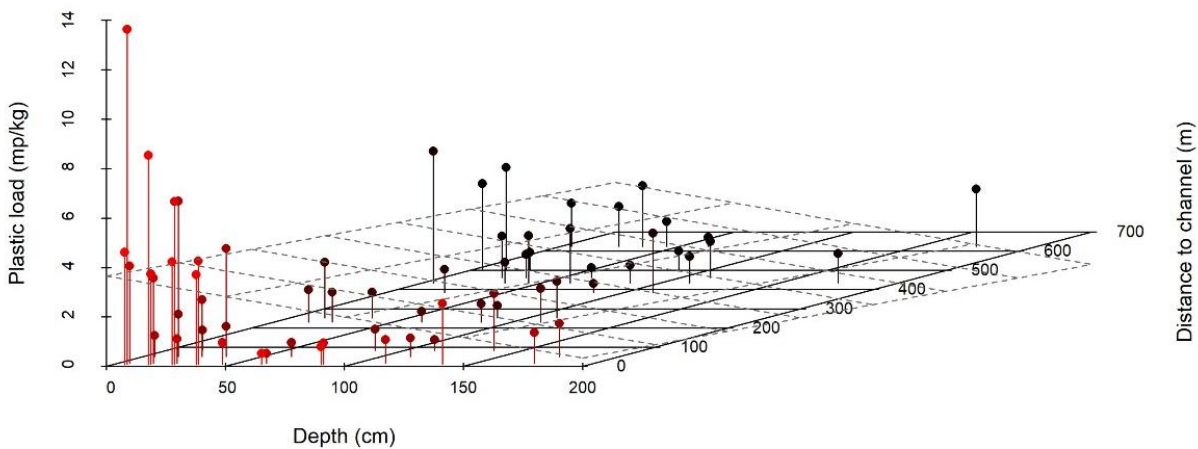


Figure 7. Spatial representation of total microplastic loads (mp/kg) by depth (cm) and distance from the channel (m). Grey area (dotted lines): Regression area of combined concentrations. Color scale of pins ranging from red (proximal floodplain) to black (distal floodplain) based on distance to channel (m).

The areal regression (Figure 7) indicates a dependence on MP inputs on the metric factors (a) depth in the soil profile and (b) distance to the watercourse. Because no dependence on other soil parameters or anthropogenic parameters (e.g., bulk density, land use, proximity to transport routes) could be proven, the MP input seems to be mainly attributable to floods as a transport medium because they reach areas near the banks more frequently than more distant ones. To examine this relationship, the question arises concerning the exact age of the sediments, especially the enrichment layer (0–50 cm). From both dating cores (ELM-D: upper course, LIM-D: lower course), it was possible to achieve sufficient results through the ^{210}Pb and ^{137}Cs dating. In case of the radionuclide ^{210}Pb , both cores show a steady concentration increase the closer to the soil surface (Figure S9). ELM-D shows an average ^{210}Pb concentration of 40.76 Bq kg^{-1} (0–90 cm) with a maximum concentration of 62.36 Bq kg^{-1} in the section 0–2 cm and an increase within the upper 20 cm of sampling core. Comparable concentrations are also found in the LIM-D core with an average ^{210}Pb concentration of 53.33 Bq kg^{-1} (0–90 cm) and maximum concentration of 82.93 Bq kg^{-1} in the section 4–6 cm. The analyses of ^{137}Cs concentrations enables the identification of concentration increases or peaks related to the atomic bomb tests of the 1950s to '60s (increase or peak in 1963) and the entry due to the Chernobyl nuclear disaster in 1986 (peak) (Andersen, 2017). A significant increase in ^{137}Cs concentrations can be observed for core ELM-D from 55 cm depth (first peak: 47 cm with 9.11 Bq kg^{-1} , second peak: 17 cm with 22.50 Bq kg^{-1}) and for core LIM-D from 51 cm depth (first increase: 43 cm with 14.23 Bq kg^{-1} , peak: 31 cm with 24.91 Bq kg^{-1}) (Figure 8a). Based on the dating results, it can be concluded that near channel floodplain sediments were deposited within the 1960s at depths between 40 to 50 cm (Figure 8b). Calculated sedimentation rates related to the period between 1986 and 2020 (34 years) show an average sedimentation rate of 0.5 cm/year for the upper reaches of the Lahn River (ELM-D) and 0.91 cm/a for the lower reaches (LIM-D), significantly higher than the catchment area rates (Lang &

Nolte, 1999; Rittweger, 2000; Martin, 2015; Weber & Opp, 2020). Finally, it can be concluded that the sediment age reaching the 1960s at a depth of 50 cm correspondence clearly with the increase of MP concentrations (Figure 8c).

Combining the findings of lateral and vertical MP distribution with the dating results, it becomes even clearer that MPs are deposited in floodplain soils through sedimentation, with the increase in global production in the 1950s (exponential growth) (Andrady, 2017; PlasticsEurope, 2018, 2020). Because plastics were only used on a small scale before the 1950s, based on the dating results, the underlying plastic particles (> 50 cm) cannot have reached these depths by natural deposition but only by in-situ transport (e.g., preferential flow, bioturbation) (van Schaik *et al.*, 2014; Rillig *et al.*, 2017; Yu *et al.*, 2019).

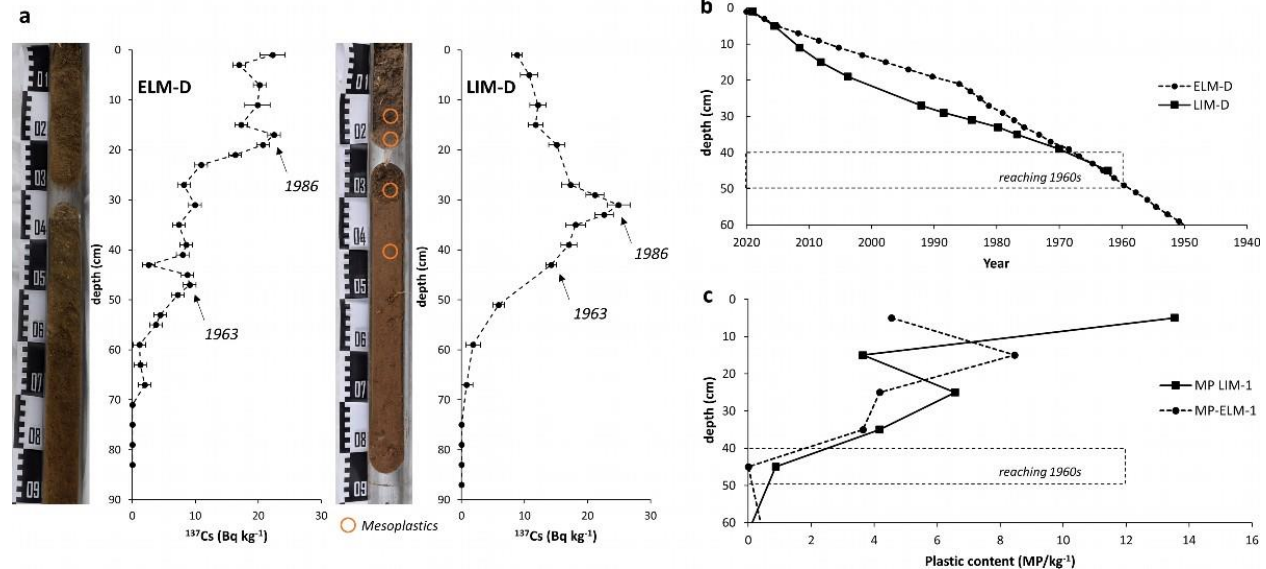


Figure 8. Sediment dating results. (a) ^{137}Cs (Bq kg $^{-1}$) concentrations for dating cores ELM-D and LIM-D with mesoplastic occurrence within core LIM-D (further information on soil density and ^{210}Pb content is given in Figure S9). (b) Calculated sediment ages (years) related to sediment depth (cm) for dating cores ELM-D and LIM-D. (c) Plastic content (mp/kg $^{-1}$) related to sampling depth for sampling sites ELM-1 and LIM-1 (upper 60 cm, plastic content below ranging from ELM-1: 0.72 mp/kg $^{-1}$, LIM-1: 0.85 to 2.45 mp/kg $^{-1}$) corresponding to dating cores.

Finally, it can be concluded that a reconstruction of MP deposition and translocation in floodplain soils, based on the holistic research approach, can become qualitatively feasible (Figure 9). MP particles can reach the floodplain area through different input pathways, whereas the flood water delivery seems to play a major role. Accumulation of MP within young sediments (since 1960s) at near river sections (riparian zone, proximal floodplain) indicates floods as a key environmental driver because only those floodplain areas are reached regularly by floods and sediment deposition (Huggett, 2007; Martin, 2012). The lower and more heterogeneous occurrences of plastic in the remaining part of the floodplain also indicate a strong transport mechanism of floodwater because only flood water reaches the floodplain at times over a wide area (Bridge, 2003). Nevertheless, besides this accumulation since the 1960, however, relocation processes must occur within the soil (Weber & Opp, 2020). Only in-situ relocations can explain the MP occurrences down to depths of 150 to 200 cm (sediments older than increase in global plastic production) because they cannot be pure depositional processes. Although displacement through the pore space of the soil is conceivable, the grain size analyses in this study show the widespread presence of loams (average contents: 19.64% clay, 48.03% silt, 32.22% sand) with a medium pore volume, a displacement of comparatively large particles can take place only through macropores (van Schaik *et al.*, 2014). Further possible processes that could be involved in a relocation and that are limited by the size of the particles (average of 1,171.04 μm) can be flow paths (preferential) or disturbances of the soil structure (corridors, bioturbation) (Rillig *et al.*, 2017; Hüffer *et al.*, 2019; Yu *et al.*, 2019; Hartmann *et al.*, 2020).

In conclusion, MP (a) accumulates in floodplain soils on the one hand and (b) is translocated on the other. Thus, there is also a distinction between a more immobile MP fraction and more mobile MP fraction (particles < 2000 μm), which can reach the deeper soil layers.

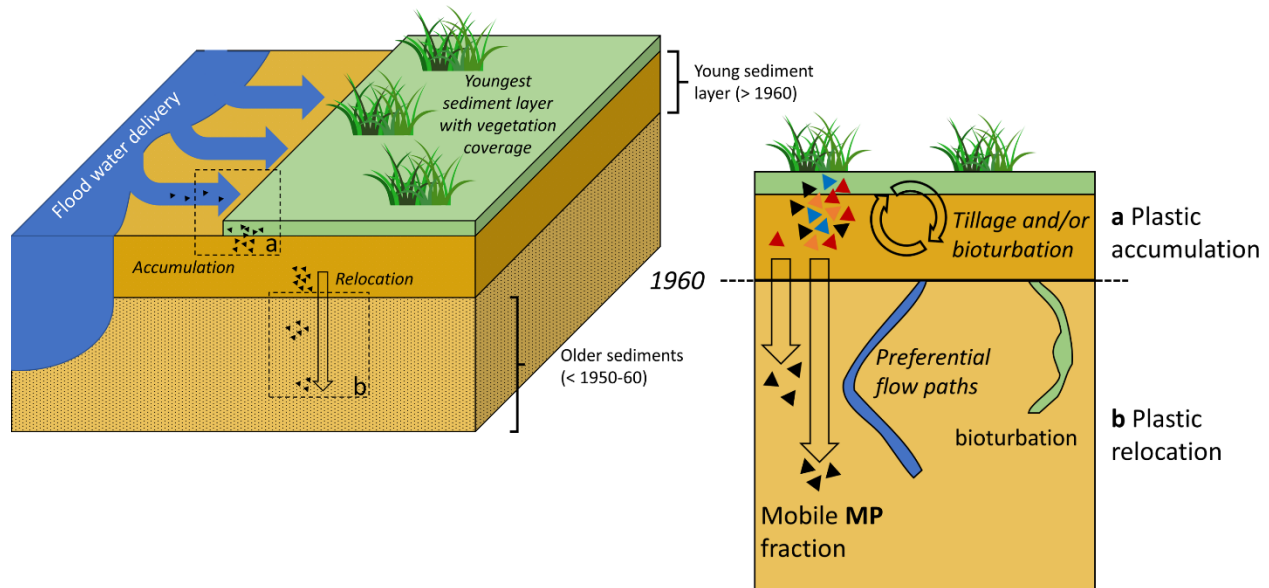


Figure 9. Scheme of MP deposition and redistribution in floodplain soils.

4 Conclusions

The spatial evidence of MP in floodplain soils and thus within the semiterrestrial system (aquatic-terrestrial interface) and its sediments illustrates that MP have become part of the sedimentary transport and thus simultaneously of the geological cycle within the Anthropocene. The heterogeneous but widespread distribution of plastic particles of different size classes, including MEP and CMP (Weber & Opp, 2020), in combination with the evidence from other studies in floodplain soils or freshwater systems in general, underlines this finding. The significant accumulation of MP in young sediments between 0 and 50 cm depth (deposition after 1960), which clearly corresponds to the increase in global plastic production, suggests that floodplains can act as a sink for microplastics. The temporal dimension highlights that plastic can, in principle, act as a stratigraphic marker of sediments in the Anthropocene, as suggested before (Price et al., 2011; Zalasiewicz et al., 2016; De-la-Torre et al., 2021). However, the pure occurrence of plastic cannot represent a specific marker because vertical displacement processes (mobile fraction < 2,000 μm , correspondence to CMP border) are possible.

Even if the consequences of plastic contamination in soils and sediments for different ecosystems are still under investigation and discussion, the widespread occurrence and accumulation of plastic alone should give cause for reflection. Plastic as a purely anthropogenic material without a natural equivalent and long residence times (Zalasiewicz et al., 2016; Chamas et al., 2020) has no place in the environment, especially not in global material cycles such as that of sediments. Humans influence the environment, and thus the future of the Earth, in manifold ways. Plastic should be understood as part of this influence and, against this backdrop, should be researched with a much stronger spatial focus. For further research on plastic and MP in floodplains, and in the semiterrestrial system in general, we recommend the following priorities:

1. **Spatial quantification:** In the context of a global plastic cycle, the representative spatial quantification of plastic and MP amounts in soils and sediments must be expanded. More data is needed to better understand and model plastic fluxes in the environment in the future.
2. **Environmental drivers:** Processes for understanding the mobility and displacement (lateral and vertical) of MP in soils and sediments should be analyzed in more detail. This analysis can be done in laboratory experiments (e.g., pot experiments), as well as in the field (e.g., pore characteristics, tracer experiments with plastic themselves or common tracers). The aim should be to understand the site-dependent key processes for MP mobility and to assess the resulting risks for the environment.
3. **Stratigraphic relevance:** Plastics and microplastics could play an important role in accessing the stratigraphic records and changes within the Anthropocene. The fate of plastics, not only as a stratigraphic

marker in floodplains, but also within different soils (especially anthropogenic soils), should be further investigated. In particular, the mobility of MP particles in soils could limit the use of MP as a marker, whereas larger plastic particles with lower mobility (MEP) could be used. Combinations with or extensions of available dating methods should be proofed.

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Conflicts of Interest: The authors declare no conflict of interest.

Data Availability: Data generated within the framework of this study can be freely accessed at Weber, Collin J.; Andersen, Thorbjørn Joest (2021), “Plastic and microplastics in floodplain soils of the Lahn River catchment (Hesse, Germany)”, Mendeley Data, V1, doi: 10.17632/m864xphyr4.1.

References

- Ad-hoc AG Boden. 2005. *Bodenkundliche Kartieranleitung*, 5th edn. Schweizerbart, Stuttgart.
- Alimi, O.S., Farner Budarz, J., Hernandez, L.M. & Tufenkji, N. 2018. Microplastics and Nanoplastics in Aquatic Environments: Aggregation, Deposition, and Enhanced Contaminant Transport. *Environmental science & technology*, **52**, 1704–1724. doi: 10.1021/acs.est.7b05559.
- Andersen, T.J. 2017. Some Practical Considerations Regarding the Application of ²¹⁰Pb and ¹³⁷Cs Dating to Estuarine Sediments. In *Applications of Paleoenvironmental Techniques in Estuarine Studies* (ed. K. Weckström, K.M. Saunders, P.A. Gell & C.G. Skilbeck), pp. 121–140. Springer, Dodrecht.
- Andrady, A.L. 2017. The plastic in microplastics: A review. *Marine pollution bulletin*, **119**, 12–22. doi: 10.1016/j.marpolbul.2017.01.082.
- Appleby, P.G. 2001. Chronostratigraphic techniques in recent sediments. In *Tracking environmental change using lake sediments. Volume 1: Basin analysis, coring and chronological techniques* (ed. Last, W. M. & J.P. Smol). Kluwer Academic Publishers, Netherlands.
- Barnes, D.K.A., Galgani, F., Thompson, R.C. & Barlaz, M. 2009. Accumulation and fragmentation of plastic debris in global environments. *Philosophical transactions of the Royal Society of London. Series B, Biological sciences*, **364**, 1985–1998. doi: 10.1098/rstb.2008.0205.
- Bläsing, M. & Amelung, W. 2018. Plastics in soil: Analytical methods and possible sources. *The Science of the total environment*, **612**, 422–435. doi: 10.1016/j.scitotenv.2017.08.086.
- Blettler, M.C.M., Ulla, M.A., Rabuffetti, A.P. & Garello, N. 2017. Plastic pollution in freshwater ecosystems: macro-, meso-, and microplastic debris in a floodplain lake. *Environmental monitoring and assessment*, **189**, 581. doi: 10.1007/s10661-017-6305-8.
- Bos, J.A.A. & Urz, R. 2003. Late Glacial and early Holocene environment in the middle Lahn river valley (Hessen, central-west Germany) and the local impact of early Mesolithic people?pollen and macrofossil evidence. *Vegetation History and Archaeobotany*, **12**, 19–36. doi: 10.1007/s00334-003-0006-7.
- Braun, M., Mail, M., Heyse, R. & Amelung, W. 2021. Plastic in compost: Prevalence and potential input into agricultural and horticultural soils. *The Science of the total environment*, **760**, 143335. doi: 10.1016/j.scitotenv.2020.143335.
- Bridge, J.S. 2003. *Rivers and Floodplains: Forms, Processes, and Sedimentary Record*, 1st edn. Blackwell, Malden.
- Brierley, G.J. & Fryirs, K.A. 2007. *Geomorphology and River Management*. Blackwell, Malden.

- Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit. 2009. *Auenzustandsbericht: Flussauen in Deutschland* [accessed on 15 March 2021].
- Carpenter, E.J. & Smith, K.L. 1972. Plastics on the Sargasso Sea Surface. *Science*, 1240–1243.
- Chamas, A., Moon, H., Zheng, J., Qiu, Y., Tabassum, T. & Jang, J.H., et al. 2020. Degradation Rates of Plastics in the Environment. *ACS Sustainable chemistry & engineering*, **8**, 3494–3511. doi: 10.1021/acssuschemeng.9b06635.
- Cole, M., Lindeque, P., Halsband, C. & Galloway, T.S. 2011. Microplastics as contaminants in the marine environment: a review. *Marine pollution bulletin*, **62**, 2588–2597. doi: 10.1016/j.marpolbul.2011.09.025.
- Coppock, R.L., Cole, M., Lindeque, P.K., Queirós, A.M. & Galloway, T.S. 2017. A small-scale, portable method for extracting microplastics from marine sediments. *Environmental pollution (Barking, Essex 1987)*, **230**, 829–837. doi: 10.1016/j.envpol.2017.07.017.
- Corradini, F., Meza, P., Eguiluz, R., Casado, F., Huerta-Lwanga, E. & Geissen, V. 2019. Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. *The Science of the total environment*, **671**, 411–420. doi: 10.1016/j.scitotenv.2019.03.368.
- Cowger, W., Gray, A., Hapich, H., Rochman, C., Lynch, J. & Primpke, S., et al. 2020. *Open Specy*.
- De-la-Torre, G.E., Dioses-Salinas, D.C., Pizarro-Ortega, C.I. & Santillán, L. 2021. New plastic formations in the Anthropocene. *The Science of the total environment*, **754**, 142216. doi: 10.1016/j.scitotenv.2020.142216.
- Durner, W., Iden, S.C. & Unold, G. von. 2017. The integral suspension pressure method (ISP) for precise particle-size analysis by gravitational sedimentation. *Water resources research*, **53**, 33–48.
- Ellen MacArthur Foundation. 2017. *The new plastics economy: Rethinking the future of plastics & catalysing action*. Ellen MacArthur Foundation, Cowes.
- FAO. 2006. *Guidelines for soil description*, 4th ed. Food and Agriculture Organization of the United Nations, Rome.
- Food and Agriculture Organization of the United Nations (FAO). 2015. *Healthy soils are the basis for healthy food production. Fact sheet*.
- Fryirs, K.A. & Brierley, G.J. 2013. *Geomorphic analysis of river systems: An approach to reading the landscape*. Wiley, Chichester West Sussex UK, Hoboken NJ.
- Gleim, W. & Opp, C. 2004. Hochwasser und Hochwasserschutz im Einzugsgebiet der Lahn. *Marburger Geographische Schriften*, 214–229.
- Hartmann, A., Semenova, E., Weiler, M. & Blume, T. 2020. Field observations of soil hydrological flow path evolution over 10 millennia. *Hydrology and earth system sciences*, **24**, 3271–3288. doi: 10.5194/hess-24-3271-2020.
- Hessian Agency for Nature Conservation, Environment and Geology. 2020. *Digital Soil Cover Data 1:50,000 Hesse (BFD50)*, Wiesbaden.
- Hidalgo-Ruz, V., Gutow, L., Thompson, R.C. & Thiel, M. 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. *Environmental science & technology*, **46**, 3060–3075. doi: 10.1021/es2031505.
- Houben, P. 2012. Sediment budget for five millennia of tillage in the Rockenberg catchment (Wetterau loess basin, Germany). *Quaternary Science Reviews*, **52**, 12–23. doi: 10.1016/j.quascirev.2012.07.011.
- Huerta Lwanga, E., Mendoza Vega, J., Ku Quej, V., Chi, J.d.L.A., Sanchez Del Cid, L. & Chi, C., et al. 2017. Field evidence for transfer of plastic debris along a terrestrial food chain. *Scientific reports*, **7**, 14071. doi: 10.1038/s41598-017-14588-2.
- Hüffer, T., Metzelder, F., Sigmund, G., Slawek, S., Schmidt, T.C. & Hofmann, T. 2019. Polyethylene microplastics influence the transport of organic contaminants in soil. *The Science of the total environment*, **657**, 242–247. doi: 10.1016/j.scitotenv.2018.12.047.
- Huggett, R., J. 2007. *Fundamentals of Geomorphology*, 2nd edn. Routledge, New York.
- Hurley, R.R., Lusher, A.L., Olsen, M. & Nizzetto, L. 2018. Validation of a Method for Extracting Microplastics from Complex, Organic-Rich, Environmental Matrices. *Environmental science & technology*, **52**, 7409–7417. doi: 10.1021/acs.est.8b01517.
- Imhof, H.K., Schmid, J., Niessner, R., Ivleva, N.P. & Laforsch, C. 2012. A novel, highly efficient method for the separation and quantification of plastic particles in sediments of aquatic environments. *Limnology and oceanography: Methods*, **10**, 524–537. doi: 10.4319/lom.2012.10.524.
- IUSS Working Group. 2015. World reference base for soil resources 2014, update 2015: International soil classification system for naming soils and creating legends for soil maps. *World Soil Resources Reports, FAO*.
- Jung, M.R., Horgen, F.D., Orski, S.V., Rodriguez C, V., Beers, K.L. & Balazs, G.H., et al. 2018. Validation of ATR FT-IR to identify polymers of plastic marine debris, including those ingested by marine organisms. *Marine pollution bulletin*, **127**, 704–716. doi: 10.1016/j.marpolbul.2017.12.061.

- Kalias, A.J., Merkt, J. & Wunderlich, J. 2003. Environmental changes during the Holocene climatic optimum in central Europe - human impact and natural causes. *Quaternary science reviews*, 33–79. doi: 10.1016/S0277-3791(02)00181-6.
- Karbalaei, S., Hanachi, P., Walker, T.R. & Cole, M. 2018. Occurrence, sources, human health impacts and mitigation of microplastic pollution. *Environmental science and pollution research*, 36046–36063. doi: 10.1007/s11356-018-3508-7.
- Klein, S., Worch, E. & Knepper, T.P. 2015. Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. *Environmental science & technology*, **49**, 6070–6076. doi: 10.1021/acs.est.5b00492.
- Konde, S., Ornik, J., Prume, J.A., Taiber, J. & Koch, M. 2020. Exploring the potential of photoluminescence spectroscopy in combination with Nile Red staining for microplastic detection. *Marine pollution bulletin*, **159**, 111475. doi: 10.1016/j.marpolbul.2020.111475.
- Kooi, M. & Koelmans, A.A. 2019. Simplifying Microplastic via Continuous Probability Distributions for Size, Shape, and Density. *Environmental science & technology letters*, **6**, 551–557. doi: 10.1021/acs.estlett.9b00379.
- Lahive, E., Walton, A., Horton, A.A., Spurgeon, D.J. & Svendsen, C. 2019. Microplastic particles reduce reproduction in the terrestrial worm *Enchytraeus crypticus* in a soil exposure. *Environmental pollution (Barking, Essex 1987)*, **255**, 113174. doi: 10.1016/j.envpol.2019.113174.
- Lang, A. & Nolte, S. 1999. The chronology of Holocene alluvial sediments from the Wetterau, Germany, provided by optical and 14C dating. *The holocene*, 207–214. doi: 10.1191/2F095968399675119300.
- Lechthaler, S., Esser, V., Schüttrumpf, H. & Stauch, G. 2021. Why analysing microplastics in floodplains matters: application in a sedimentary context. *Environmental science: processes & impacts*, **71**, 299. doi: 10.1039/D0EM00431F.
- Lechthaler, S., Waldschläger, K., Stauch, G. & Schüttrumpf, H. 2020. The Way of Macroplastic through the Environment. *Environments*, **7**, 73. doi: 10.3390/environments7100073.
- Liu, H., Tang, L., Liu, Y., Zeng, G., Lu, Y. & Wang, J., et al. 2019. Wetland-a hub for microplastic transmission in the global ecosystem. *Resources, conservation and recycling*, **142**, 153–154. doi: 10.1016/j.resconrec.2018.11.028.
- Liu, M., Lu, S., Song, Y., Lei, L., Hu, J. & Lv, W., et al. 2018. Microplastic and mesoplastic pollution in farmland soils in suburbs of Shanghai, China. *Environmental pollution (Barking, Essex 1987)*, **242**, 855–862. doi: 10.1016/j.envpol.2018.07.051.
- Lorenzo-Navarro, J., Castrillon-Santana, M., Gomez, M., Herrera, A. & Marin-Reyes, P.A. 2018. Automatic Counting and Classification of Microplastic Particles. *Proceedings of the 7th international conference on pattern recognition, applications and methods (ICPRAM 2018)*, 646–652. doi: 10.5220/0006725006460652.
- Machado, Anderson A. de Souza, Kloas, W., Zarfl, C., Hempel, S. & Rillig, M.C. 2018. Microplastics as an emerging threat to terrestrial ecosystems. *Global change biology*, **24**, 1405–1416. doi: 10.1111/gcb.14020.
- Martin, C.W. 2012. Recent changes in heavy metal contamination at near-channel positions of the Lahn River, central Germany. *Geomorphology*, **139-140**, 452–459. doi: 10.1016/j.geomorph.2011.11.010.
- Martin, C.W. 2015. Trace metal storage in recent floodplain sediments along the Dill River, central Germany. *Geomorphology*, **235**, 52–62. doi: 10.1016/j.geomorph.2015.01.032.
- Martin, C.W. 2019. Trace metal concentrations along tributary streams of historically mined areas, Lower Lahn and Dill River basins, central Germany. *Catena*, **174**, 174–183. doi: 10.1016/j.catena.2018.11.008.
- Meschede, M. & Warr, L.N. 2019. *The Geology of Germany: A Process-Oriented Approach*. Springer International Publishing, Cham.
- Möller, J.N., Löder, M.G.J. & Laforsch, C. 2020. Finding Microplastics in Soils: A Review of Analytical Methods. *Environmental science & technology*, **54**, 2078–2090. doi: 10.1021/acs.est.9b04618.
- Napper, I.E. & Thompson, R.C. 2019. Environmental Deterioration of Biodegradable, Oxo-biodegradable, Compostable, and Conventional Plastic Carrier Bags in the Sea, Soil, and Open-Air Over a 3-Year Period. *Environmental science & technology*, 4775–4783. doi: 10.1021/acs.est.8b06984.
- Nardi, F., Annis, A., Di Baldassarre, G., Vivoni, E.R. & Grimaldi, S. 2019. GFPLAIN250m, a global high-resolution dataset of Earth's floodplains. *Scientific data*, **6**, 180309. doi: 10.1038/sdata.2018.309.
- Opp, C., Stach, J. & Hanschmann, G. 1993: Load on differently used soils by heavy metals within the highly contaminated area of Bitterfeld (FRG). *Science of the total environment*, **134**, 141-150. doi: 10.1016/S0048-9697(05)80013-0
- Piehl, S., Leibner, A., Löder, M.G.J., Dris, R., Bogner, C. & Laforsch, C. 2018. Identification and quantification of macro- and microplastics on an agricultural farmland. *Scientific reports*, **8**, 17950. doi: 10.1038/s41598-018-36172-y.

- PlasticsEurope. 2018. Plastics - the facts 2018: An analysis of European plastic production, demand and waste data. *Plastic Europe*.
- PlasticsEurope. 2020. Plastics - the facts 2020: An analysis of European plastic production, demand and waste data.
- Price, S.J., Ford, J.R., Cooper, A.H. & Neal, C. 2011. Humans as major geological and geomorphological agents in the Anthropocene: the significance of artificial ground in Great Britain. *Philosophical transactions. Series A, Mathematical, physical, and engineering sciences*, **369**, 1056–1084. doi: 10.1098/rsta.2010.0296.
- Primpke, S., Lorenz, C., Rascher-Friesenhausen, R. & Gerdt, G. 2017. An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. *Analytical Methods*, **9**, 1499–1511. doi: 10.1039/C6AY02476A.
- Primpke, S., Wirth, M., Lorenz, C. & Gerdt, G. 2018. Reference database design for the automated analysis of microplastic samples based on Fourier transform infrared (FTIR) spectroscopy. *Analytical and bioanalytical chemistry*, **410**, 5131–5141. doi: 10.1007/s00216-018-1156-x.
- Ragusa, A., Svelato, A., Santacroce, C., Catalano, P., Notarstefano, V. & Carnevali, O., et al. 2021. Plasticenta: First evidence of microplastics in human placenta. *Environment international*, **146**, 106274. doi: 10.1016/j.envint.2020.106274.
- Regional Council Giessen. 2015. *Hochwasserrisikomanagementplan für das hessische Einzugsgebiet der Lahn (Flood risk management plan)*. Regional Council Giessen, Giessen.
- Rillig, M.C., Lehmann, A., Souza Machado, A.A. de & Yang, G. 2019. Microplastic effects on plants. *The New phytologist*. doi: 10.1111/nph.15794.
- Rillig, M.C., Ziersch, L. & Hempel, S. 2017. Microplastic transport in soil by earthworms. *Scientific reports*, **7**, 1362. doi: 10.1038/s41598-017-01594-7.
- Rittweger, H. 2000. The “Black Floodplain Soil” in the Amöneburger Becken, Germany: a lower Holocene marker horizon and indicator of an upper Atlantic to Subboreal dry period in Central Europe? *CATENA*, 143–164. doi: 10.1016/S0341-8162(00)00113-2.
- Ruggero, F., Gori, R. & Lubello, C. 2020. Methodologies for Microplastics Recovery and Identification in Heterogeneous Solid Matrices: A Review. *Journal of Polymers and the Environment*, **28**, 739–748. doi: 10.1007/s10924-019-01644-3.
- Scheurer, M. & Bigalke, M. 2018. Microplastics in Swiss Floodplain Soils. *Environmental science & technology*, **52**, 3591–3598. doi: 10.1021/acs.est.7b06003.
- Selonen, S., Dolar, A., Jemec Kokalj, A., Skalar, T., Parramon Dolcet, L. & Hurley, R., et al. 2020. Exploring the impacts of plastics in soil - The effects of polyester textile fibers on soil invertebrates. *The Science of the total environment*, **700**, 134451. doi: 10.1016/j.scitotenv.2019.134451.
- Siegfried, M., Koelmans, A.A., Besseling, E. & Kroeze, C. 2017. Export of microplastics from land to sea. A modelling approach. *Water research*, **127**, 249–257. doi: 10.1016/j.watres.2017.10.011.
- Stock, F., Kochleus, C., Baensch-Baltruschat, B., Brennholt, N. & Reifferscheid, G. 2019. Sampling techniques and preparation methods for microplastic analyses in the aquatic environment - A review. *TrAC Trends in Analytical Chemistry*, **113**, 84–92. doi: 10.1016/j.trac.2019.01.014.
- Thomas, D., Schütze, B., Heinze, W.M. & Steinmetz, Z. 2020. Sample Preparation Techniques for the Analysis of Microplastics in Soil—A Review. *Sustainability*, **12**, 9074. doi: 10.3390/su12219074.
- Maes, T., Jessop, R., Wellner, N., Haupt, K. & Mayes, A. G. 2017. A rapid-screening approach to detect and quantify microplastics based in fluorescent tagging with Nile Red. *Scientific reports*. doi: 10.1038/srep44501.
- Tibbetts, J., Krause, S., Lynch, I. & Sambrook Smith, G. 2018. Abundance, Distribution, and Drivers of Microplastic Contamination in Urban River Environments. *Water*, **10**, 1597. doi: 10.3390/w10111597.
- van Schaik, L., Palm, J., Klaus, J., Zehe, E. & Schröder, B. 2014. Linking spatial earthworm distribution to macropore numbers and hydrological effectiveness. *Ecohydrology*, **7**, 401–408. doi: 10.1002/eco.1358.
- Waldschläger, K. & Schüttrumpf, H. 2019a. Effects of Particle Properties on the Settling and Rise Velocities of Microplastics in Freshwater under Laboratory Conditions. *Environmental science & technology*, **53**, 1958–1966. doi: 10.1021/acs.est.8b06794.
- Waldschläger, K. & Schüttrumpf, H. 2019b. Erosion Behavior of Different Microplastic Particles in Comparison to Natural Sediments. *Environmental Science & Technology*, **53**, 13219–13227. doi: 10.1021/acs.est.9b05394.
- Weber, C.J. & Opp, C. 2020. Spatial patterns of mesoplastics and coarse microplastics in floodplain soils as resulting from land use and fluvial processes. *Environmental pollution*, **267**, 115390. doi: 10.1016/j.envpol.2020.115390.
- Weber, C.J., Weihrauch, C., Opp, C. & Chiffard, P. 2020. Investigating microplastic dynamics in soils: Orientation for sampling strategies and sample pre-processing. *Land degradation & development*, 270–284. doi: 10.1002/ldr.3676.

- 888 Weihrauch, C. 2019. Dynamics need space – A geospatial approach to soil phosphorus' reactions and migration.
889 *Geoderma*, **354**, 113775. doi: 10.1016/j.geoderma.2019.05.025.
- 890 Willgoose, G. 2018. *Principles of Soilscape and Landscape Evolution*. Cambridge University Press, Cambridge.
- 891 Wright, S.L., Thompson, R.C. & Galloway, T.S. 2013. The physical impacts of microplastics on marine organisms:
892 a review. *Environmental pollution*, **178**, 483–492. doi: 10.1016/j.envpol.2013.02.031.
- 893 Xiong, X., Wu, C., Elser, J.J., Mei, Z. & Hao, Y. 2018. Occurrence and fate of microplastic debris in middle and
894 lower reaches of the Yangtze River - From inland to the sea. *The Science of the total environment*, **659**, 66–73.
895 doi: 10.1016/j.scitotenv.2018.12.313.
- 896 Yu, M., van der Ploeg, M., Lwanga, E.H., Yang, X., Zhang, S. & Ma, X., et al. 2019. Leaching of microplastics by
897 preferential flow in earthworm (*Lumbricus terrestris*) burrows. *Environmental chemistry*, **16**, 31. doi:
898 10.1071/EN18161.
- 899 Zalasiewicz, J., Waters, C.N., Ivar do Sul, J.A., Corcoran, P.L., Barnosky, A.D. & Cearreta, A., et al. 2016. The
900 geological cycle of plastics and their use as a stratigraphic indicator of the Anthropocene. *Anthropocene*, **13**, 4–
901 17. doi: 10.1016/j.ancene.2016.01.002.
- 902 Zhang, B., Yang, X., Chen, L., Chao, J., Teng, J. & Wang, Q. 2020. Microplastics in soils: a review of possible
903 sources, analytical methods and ecological impacts. *Journal of chemical technology & biotechnology*, **37**, 1045.
904 doi: 10.1002/jctb.6334.
- 905 Zhang, G.S. & Liu, Y.F. 2018. The distribution of microplastics in soil aggregate fractions in southwestern China.
906 *The Science of the total environment*, **642**, 12–20. doi: 10.1016/j.scitotenv.2018.06.004.
- 907 Zhang, G.S., Zhang, F.X. & Li, X.T. 2019. Effects of polyester microfibers on soil physical properties: Perception
908 from a field and a pot experiment. *The Science of the total environment*, **670**, 1–7. doi:
909 10.1016/j.scitotenv.2019.03.149.
- 910 Zink, T., Geyer, R. & Startz, R. 2018. Toward Estimating Displaced Primary Production from Recycling: A Case
911 Study of U.S. Aluminum. *Journal of industrial ecology*, **22**, 314–326. doi: 10.1111/jiec.12557.
- 912