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Spatial patterns of mesoplastics and coarse microplastics in floodplain soils as resulting from land use and fluvial processes☆

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A B S T R A C T

Plastic, and especially microplastic, contamination of soils has become a novel research field. After the detection of microplastics in soils, spatial distribution and dynamics are still unknown. However, the potential risks associated with plastic particles in soils cannot be sufficiently assessed without knowledge about the spatial distribution of these anthropogenic materials. Based on a spatial research approach, including soil surveys, this study quantified the mesoplastic (MEP, > 5.0 mm) and coarse microplastics (CMP, 2.0–5.0 mm) content of twelve floodplain soils. At four transects in the catchment area of the Lahn river (Germany), soils down to a depth of 2 m were examined for plastic content for the first time. MEP and CMP were detected through visual examination after sample preprocessing and ATR-FTIR analyses. Average MEP and CMP concentrations range between 2.06 kg^{-1} ($\pm$1.55 kg^{-1}) and 1.88 kg^{-1} ($\pm$1.49 kg^{-1}) with maximal values of 5.37 MEP kg^{-1} to 8.59 CMP kg^{-1}. Plastic particles are heterogeneously distributed in samples. Both plastic size classes occur more frequently in topsoils than in soil layers deeper than 30 cm. The maximal depth of CMP occurrence lies between 75 and 100 cm. Most common CMP polymer type was PE-LD, followed by PP and PA. MEP and CMP particles occur frequently at near channel sides and more often on riparian strips or grassland than on farmland. Vertical distribution of CMP indicates anthropogenic relocation in topsoils and additional deep displacement through natural processes like preferential flow paths or bioturbation. By comparing sedimentation rates of the river with the maximum age of plastic particles, sedimentation as a deposition process of plastic in floodplains becomes probable. From our findings, it can be concluded that an overall widespread but spatial heterogenous contamination occurs in floodplain soils. Additionally, a complex plastic source pattern seems to appear in floodplain areas.

1. Introduction

Worldwide, a large proportion of plastic waste, especially microplastics, is disposed into the environment (Barnes et al., 2009; Karbalaei et al., 2018; Machado et al., 2018a; PlasticsEurope, 2017). Plastic production has rapidly increased since its serial production in the 1950s, and today 348 million tons of plastic are produced each year (Andrady, 2017; PlasticsEurope, 2017). Research on plastic and microplastics has demonstrated the occurrence of plastic particles in different size ranges all around the world and in several ecosystems (Allen et al., 2019; Emmerik and Schwarz, 2020; Peeken et al., 2018). Soils, as terrestrial ecosystems and fundamental resources of the global food security system, are also affected by plastic pollution (He et al., 2018a; Machado et al., 2018b; Rillig, 2012; Wang et al., 2019, 2020; Zhang et al., 2020). Despite the limited number of studies on meso- (MEP) and microplastics (MP) in soils, it could be proven that, in particular, microplastic have various effects on soil biota, soil physical, and chemical properties, as well as plant growth (Engdahl, 2018; Hüffer et al., 2019; Rillig et al., 2017a, 2017b, 2019; Yu et al., 2019). Because one of the hotspots of plastic pollution is located in the oceans (Martin et al., 2017), the transport of plastic by rivers could be seen as a major source of plastic distribution (Alimi et al., 2018; Liu et al., 2019; Siegfried et al., 2017; Tibbetts et al., 2018; Xiong et al., 2018). Immediately beside these main land-to-sea transport paths are floodplains, which are semiterrestrial ecosystem. The first study on microplastics in floodplain topsoils in Swiss nature conservation areas reveals the abundance of microplastic in these ecosystems (Scheurer and Bigalke, 2018). Because floodplains are mainly

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sedimentation and flood retention areas, a systematic accumulation of plastic particles is conceivable in floodplains.

Microplastics as anthropogenic materials are defined with a currently implemented definition of a size spectrum between 1 and 5000 μm (Mausra et al., 2015). This defined size range reaches its limits if studying environmental processes in a soil context (e.g., transport or transfer processes) because, in the case of particles, these processes are also size dependent (Blume et al., 2016; Rillig et al., 2017b; Yu et al., 2019). In soil science, a differentiation is made between different grain sizes and coarse gravel (2–6 mm) or very coarse sand and coarse sand (˃ 2000 μm) against textural sand classes (˂ 2000 μm) (FAO, 2006; IUSS Working Group, 2015). Differentiation between coarse and fine soil/earth has already been used for a long time to describe soils and pedogenesis, and makes a significant distinction for in situ transfer processes (e.g., macropores; Blume et al., 2016). As microplastic particles within the size range of coarse sand fraction can be assumed to be difficult to relocate in situ, soil context research should specify these size classes. The present study distinguishes between macroplastics (˃ 25000 μm, 25 mm; abbreviated as MAP), mesoplastics (˃ 5000 μm; abbreviated as MEP), and coarse microplastics (abbreviated as CMP) with a size range of 5000 to 2000 μm.

For the deeper understanding of spatial plastic dynamics in soils, more systematic studies in contrast to the so far established "explorative studies" are required (Weber et al., 2020). Hence, the present study uses a geospatial approach to investigate the spatial dynamics of MEP and CMP in floodplain soils to answer the following research questions: (1) What are the levels of MEP and CMP pollution in floodplain soils in an entire catchment area? (2) Is the lateral distribution of MEP and CMP a function of river section, land use, or distance to water body? (3) Are MEP and CMP involved in the sedimentation processes and/or vertically displacement processes? This is intended to achieve the objective of an improved understanding of spatial plastic dynamics inside the three-dimensional system of the soil scapes and enable further targeted research about responsible processes in landscapes.

2. Material and methods

2.1. Study area

The investigation was performed in floodplain areas of the Lahn River located in the central German low mountain range (Hesse, Germany; Fig. S1). The Lahn River, with a length of 245.6 km, drains a catchment area of 5924 km^2 (Regional Council Giessen, 2015). The geology of the Lahn River catchment consists principally of different rock types from the Paleozoic age, tertiary basalts and sandstones (Meschede and Warr, 2019). The Lahn Valley reaches a maximum with of ca. 3.0 km (Tichy, 1951). According to hydrological, geological, and landscape properties, the Lahn Valley can be divided into four zones: (A) upper course (Rhenish Slate Mountains, smaller floodplain), (B) middle course (wide valley with wide floodplain), (C) narrow valley (almost without distinctive floodplain), and (D) lower course with individual valley widenings and floodplains.

The floodplains beside the Lahn River and surrounding valleys are built up from organic-rich silt and loams, reaching a thickness of 3.0 m, which was deposited during the late Holocene (Bos and Urz, 2003; Rittweger, 2000). Even if early Holocene depositions occur, the main sedimentation process occurred over the last 3000 years in connection to anthropogenic land-use change (reclamation) (Andres et al., 2001; Bos and Urz, 2003; Delorme and Leuschner, 1983; Kalias et al., 2003; Rittweger, 2000).

These Holocene strata are on top of Pleistocene gravel and sand deposition, which are partially terrain forming in the upper reaches (Bos and Urz, 2003; Mäckel, 1969). The border between Pleistocene and Holocene depositions is marked in the middle reaches between the cities of Marburg and Wetzlar through an incision of the LST during Younger Dryas (Laacher See tephra; Bos and Urz, 2003). Floodplain surface morphology consists of river banks, plain areas with conserved forms (e.g., former loops or meander bends, channels), and back swamps. Different soil types, like Fluvisols, Gleysols, and Stagnisols, have developed in the deposited river sediments (Table 1).

The current land use in the main Lahn River catchment is 7.0% urban and traffic, 1.4% industry, 43.0% agriculture (corp and grassland), and 47.0% forested (Regional Council Giessen, 2015). As of 2010, the Hessian catchment parts had 1.3 million inhabitants (266/km^2; Regional Council Giessen, 2015). The water body of the Lahn River can be named urbanized only in the direct surroundings of the cities Marburg, Giessen, Wetzlar, and Limburg (Martin, 2012). Floodplains are under mainly agricultural land use (crop- or grassland), excluding riparian strips (channel bank) and small amounts of urban areas. As the lower Lahn River reaches were used for shipping, the river is deepened and channelized (Martin, 2012; Regional Council Giessen, 2015).

Similar to other rivers in central Europe, different medieval flood events are documented for the Lahn River (e.g., 1255, 1332, 1552; Gleim and Opp, 2004). Over the past 100 years, flood events with a discharge >400 m^3/s (Station Leun) and water levels > 450 cm (Station Marburg) occur frequently, with a century high-flood event in 1984 (Gleim and Opp, 2004). Different flood water management measures (e.g., flood retention basins, dikes, return of flood retention areas) were established inside the catchment. However, recent flood events have not eroded or deposited high loads of sediments, and local erosion and deposition of fine sands appears (Martin, 2012, 2015, 2019).

2.2. Soil sampling

The spatial distribution of MEP and CMP was investigated on four sites representative for zones A (upper course), B (middle course) and D (lower course) with extended floodplains. In order to investigate the spatial dynamics of MEP and CMP inside the floodplain system, each site was selected according to the following criteria: (1) location inside a natural flood retention area with frequent flood events, (2) sequence of different morphological units and soil differentiation, (3) land use differences in floodplain cross-section, and (4) no direct MP sources (e.g., highways, industrial plants). A soil mapping was carried out on each of the four sites in order to obtain an overview on soil scape properties and set each transect representative for a larger floodplain area with comparable soil properties (Weihrauch, 2019).

Floodplain cross-section transect, containing two or four sampling sites, were established at each site according to the above-mentioned criteria. The distance between the sampling sites depends on the width of the floodplains. Two soil profiles at a distance of 5 m to each other were sampled down to a depth of 2 m through pile core driving (core diameter: 100 mm and 80 mm) and then sampled in sections of 10 cm (0–0.5 m depth), 25 cm (0.5–1.5 m depth), and 50 cm (1.5–2.0m) depth. Sample mass ranged between 388 g and 3225 g (mean 1150 g) of dry soil material including coarse soil fractions and organic material. Samples were transported and stored in corn starch bioplastic bags (Mater-Bi bags, Bio Futura B.V., Rotterdam, Netherlands). A total of 120 samples (10 samples per soil profile) were taken at four sites between August 12, 2019 and August 23, 2019. Additional plastic fragments on topsoil surface were sampled if the number of plastic fragments around the drill hole was noticeable. On sampling site STD (Profile STD-1), visible plastic fragments (macroplastic: > 25000 μm, MAP)

Table 1
Sampling site features.

Sampling Area	Catchment properties			Sampling site	Floodplain properties			Soil properties	
	Catchment zone	River km[a]	Width of flood area (m)		Morphological unit	Land use	Distance to channel (m)	Sampling depth (cm)	Soil type (WRB[b])
ELM 50.865345 8.618088	Upper course	201.9	617.7	ELM-1	Riparian	riparian vegetation	8.8	100	Skeletic Fluvisol (Technic, Arenic)
				ELM-2	Plane	farmland	500.6	100	Endogleyic Skeletic Fluvisol (Anthric, Siltic)
ROT 50.734907 8.733830	Middle cource	168.3	1543.2	ROT-1	Riparian	grassland	15.5	200	Halpic Fluvisol (Arenic)
				ROT-2	Plane	grassland	462.5	200	Stagnic Fluvisol (Anthric, Tephric, Loamic)
STD 50.552517 8.451405	Middle cource	121.0	689.4	STD-1	Riparian	farmland	50.0	200	Fluvisol (Anthric, Arenic)
				STD-2	Plane	grassland	228.9	200	Skeletic Fluvisol (Loamic)
				STD-3	Plane	grassland	433.3	200	Epigleyic Fluvisol (Silitc)
				STD-4	Backswamp	grassland	622.7	200	Fluvic Gleysol (Clayic)
LIM 50.389916 8.039374	Lower cource	57.0	420.4	LIM-1	Riparian	riparian vegetation	12.2	200	Haplic Fluvisol (Arenic)
				LIM-2	Plane	grassland	82.9	200	Endogleyic Fluvisol (Silitc)
				LIM-3	Backswamp	grassland	251.1	200	Fluvic Gleysol (Loamic)
				LIM-4	Lower terrace	farmland	383.2	200	Skeletic Stagnic Fluvisol (Densic, Clayic)

[a] According to WRRL-Viewer Hesse.
[b] World Reference Base for Soil Recourses (2015).

were collected on a 20 m^2 area next to the drill point. This sampling of plastic particles from soil surface was only used on this site because there was a conspicuous amount of visible plastic fragments. MAP surface samples were handled in accordance with sample handling of soil samples.

The stratigraphy and pedogenesis of the sampled floodplain soils was documented according to the German soil classification (Ad-hoc AG Boden, 2005) and the FAO Guidelines for soil description (FAO, 2006) and were classified according to WRB 2015 (IUSS Working Group, 2015). Soil color with Munsell charts and the carbonate content after reaction with a few drops of 3.23 M hydrochloric acid (HCl) according to Ad-hoc AG Boden (2005) was determined in field.

Contamination prevention during soil sampling was performed by the general renouncement of plastic tools. This includes the removal of plastic parts on the pile cores, the sample material removal with stainless steel spatulas, and the final transport of samples in bioplastic bags.

2.3. Laboratory analysis

For each soil sample, a five-step procedure, including sample preprocessing and analyses, was applied (Fig. S2). Samples were short-time stored in corn starch bags and then dried at 45 °C in a dying chamber (step 1) for a maximum of four days. After drying, samples were carefully crashed with pestle and mortar (step 2) to solve soil macroaggregates. Step 2 is important because CMP can be embedded in soil macroplastics (250–2500 μm) and additional microplastic in microaggregates (20–250 μm; Amelung and Zech, 1999; Zhang and Liu, 2018).

According to the size classes of MEP (˃ 5000 μm) and CMP (˃ 2000 μm), soil samples were dry sieved (step 3) through a 5 mm- and 2 mm-wide mesh, stainless-steel sieve (RETSCH GmbH, Haan, Germany). During the sieving process, the sieves were covered and shaken. Each fraction was then weighed and stored again in fresh corn starch bags. A sample proportion ˂ 2000 μm was saved for later analysis of microplastics, which is currently in progress. During sample preprocessing, the exposition time of each sample was reduced as much as possible to avoid air contaminations (e.g., clothing fibres).

Both fractions of each, single sample were then visually determined (step 4) for the MEP and CMP content. Plastic particles found in the sieving fraction ˃ 2000 μm were counted as CMP, whereas particles found in the fraction ˃ 5000 μm were counted as MEP. MAP particles could be detected only from the surface samples of side STD. Therefore, each fraction was transferred to a stainless-steel bowl with imprinted grid (1 × 1 cm grid size), and each grid space was inspected under a stereomicroscope with 40x maximal magnification (SMZ 161 TL, Motic, Hong Kong).

Potential plastic particles were identified under application of criteria for visual determination of microplastics introduced by Norén (2007). A particle was picked if (a) no cellular, organic or undissolved soil aggregate structure was visible, (b) the particle has a homogenous color, and (c) in case of filaments, the particle is equally thick. Subsequently, each identified particle was classified into different categories according to Hidalgo-Ruz et al. (2012), photographed (Moticam 2, Motic, Hong Kong), and stored in glass vessels until further analysis (Hidalgo-Ruz et al., 2012). Even if several authors suggested that visual identification of microplastic is prone to serious errors, this conclusion depends on the particle size (Andrady, 2017; Hidalgo-Ruz et al., 2012; Song et al., 2015). For CMP particles with a size ˃ 2000 μm, identification by microscope seems to be suitable with a low risk of underestimating the total particle number (e.g, in case of transparent or yellow-weathered shapes; Song et al., 2015). Additionally, the MAP samples from site STD were also visual determined, photographed, and stored in glass vessels.

For final identification and to avoid overestimation, all visually identified CMP, MEP, and MAP particles were analyzed with a Tensor 37 FTIR spectrometer (Bruker Optics, Ettlingen, Germany) combined with a Platinum-ATR-unit (Bruker Optics, Ettlingen, Germany). In the case of very dirty particles, these were carefully cleaned at the measuring point using steel tools (tweezers, spatula). The Platinum-ATR-unit, in the case of particles with strong adherent soil material, was cleaned with 2-propanol ($CH_3CHOHCH_3$). The measurement of each particle was performed with 20 background scans, followed by 20 sample scans. Spectral resolution was set to 4 cm^{-1} in a wavenumber range from 4000 cm^{-1} to 400 cm-^{1} following the suggestions of Primpke et al. (2017) and Primpke et al. (2018).

2.4. Statistics and data evaluation

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). Depth variation of particles was illustrated using a "vioplot" package and comparison with sedimentation rates using "sm" package (Adler et al., 2019; Bowman and Azzalini, 2018).

Data processing of FTIR spectra was performed in OPUS 7.0 (Bruker Optics, Ettlingen, Germany). Each spectrum was compared with the entries in OPUS 7.0 internal spectra database. Quality for identifying a polymer was a hit quality of > 300 (Lorenzo-Navarro et al., 2018; Primpke et al., 2017, 2018). Each particle that did not achieve a hit quality above 300 was measured twice after additional carefully cleaning. Additional FTIR spectra analyses and plotting was performed in Spectragryph (Version 1.2.14; Menges, 2020; Oberstdorf, Germany). Previously unidentified particles were additionally compared with spectra database of natural and biogenic materials (Weiner, 2010).

MEP and CMP concentrations are reported in particles (MEP or CMP) per kilogram soil dry weight (MEP kg^{-1}, CMP kg^{-1}). The concentration of surface MAP particles is given in absolute numbers. For comparison of CMP abundance in floodplain soils and sedimentation rates, the following procedure was performed: (a) For each FTIR-identified particle, the age of earliest possible occurrence (e.g., year of first production or patent application) was chosen from "History of Plastics" (British Plastic Federation, 2020) and/or "PLASTIKATLAS 2019" (Caterbow and Speranskaya, 2019). (b) Sedimentation rates and sediment ages (Quartz OSL/14C) from all available regional studies were compared. Finally, the sedimentation rate of 0.07 cm per year (cm/a) calculated by Lang & Nolte (1999) for the Wetter river floodplain was chosen (Lang and Nolte, 1999). This value is closest to the calculated average (0.11 cm/a) of three rivers (Ämoneburg Basin, Wetter River, Dill River) and is most comparable in morphological terms (Lang and Nolte, 1999; Martin, 2015; Rittweger, 2000). (c) Age of earliest possible occurrence was multiplied by sedimentation rate and compared to sample depth.

3. Results and discussion

3.1. Meso- and coarse microplastic abundance in floodplain soil

Inside the floodplain areas of the Lahn River, plastic particles were found at each transect site. Overall, 14 MEP particles (10 significant FTIR identified) and 78 CMP particles (40 significant ATR-FTIR identified) were documented. On site, STD-1 additional 16 MAP particles (13 significant ATR-FTIR identified) were collected on a 20 m^2 area next to the drill hole. Even if this result is only representative for a single area, a value of 6500 particles per hectare can be calculated. Piehl et al. (2018) documented a value of 206 MAP per hectare for an agricultural field (Piehl et al., 2018). In comparison with this study and due to the limited area representativeness, the sampling point STD-1 could be considered a MAP hotspot.

The MEP concentration in soil samples ranges from 0.62 to 5.37 MEP kg^{-1} (median 1.62 MEP kg^{-1}, mean 2.06 MEP kg^{-1}). For visual identified CMP, the concentration lies between 0.31 and 8.59 CMP kg^{-1} (median 2.25 CMP kg^{-1}, mean 2.87 CMP kg^{-1}) and for additional ATR-FTIR-identified CMPs between 0.37 and 6.06 CMP kg^{-1} (median 1.37 CMP kg^{-1}, mean 1.88 CMP kg^{-1}; Fig. 1a). The difference between the clearly visually identified number of CMP and the FTIR identified CMPs could be explained by the strong surface degradation of weathered particles. Additional degraded particle surfaces allow a better adhesion of dirt (e.g., minerals, organic matter), which facilitates cleaning and identification using a spectra database based on fresh particles (Primpke et al., 2018).

Because there are currently only a few quantitative studies on micro-, meso-, and macroplastics in soils, each reporting different quantitative values, interpretation and discussion is limited. In a comparison of the results of this study with the results of Liu et al. (2018) and Zhang and Liu (2018) for agricultural topsoils, their microplastic load range within 78.00 ± 12.91 particles kg^{-1} (respectively 7100 p kg^{-1}) and 62.50 ± 12.97 particles kg^{-1} (respectively 42.96 p kg^{-1}) (Liu et al., 2018). These values clearly exceed the concentration of the recent study. In contrast, the range of mesoplastics reported by Liu et al. (2018) with 6.75 ± 1.51 to 3.25 ± 1.04 p kg^{-1} corresponds to the concentrations documented by this study (Liu et al., 2018). Another microplastic and mesoplastic concentration, reported for Swiss floodplain topsoils in nature reserves by Scheurer and Bigalke (2018), is given with a maximum of 592 p kg^{-1} (55.5 mg kg^{-1}) and an average of 5 mg kg^{-1}. A comparison with the only other study on floodplain soils is complicated by the use of the mg kg^{-1} unit. However, assuming that the maximum is approximately 11 times higher than the average value and that 88% of total microplastic load occurs in a size < 500 μm, the concentration of CMP could be comparable (Scheurer and Bigalke, 2018).

Dominant identified polymer type is PE-LD with 5 MEPs and 16% of total CMP particles (Fig. 2). In CMP fraction PP (6%), PA (5%), PS (4%), POM (4%) and PET (3%) form the majority together with single others. Collected surface samples from Site STD-1 consists of PE-LD, PE-HD, PVC, PP and PS. These findings are clearly comparable to other studies of microplastics and MP in soils. PE and PP seem to represent the majority of plastic particles in soils (Liu et al., 2018; Piehl et al., 2018; Scheurer and Bigalke, 2018; Zhang and Liu, 2018). This finding is not unexpected because PP, PE-LD, and PE-HD are the most frequently used polymer types and show the largest production quantities (Andrady, 2017; Ellen MacArthur Foundation, 2017; PlasticsEurope, 2017).

Strong degradation of particles and poor surface properties (e.g., strongly adherent soil material) leads to a huge amount of not clearly identified particles (49% of total CMP particles). Additional cleaning of larger particles with distilled water is possible but difficult for fragile pieces and must be carried out carefully.

3.2. Characteristics of plastic particles

The detected MEP and CMP occur in different forms. Examples for particle forms are given in supplementary material (Fig. S4). The predominant forms were fragments (32%) and films (32%), followed by filaments (19%; Fig. 3a). A new class was built for "fiber balls" (11%) because fibers were often found in a tangle. This class contains fiber balls that consist of several fibers of the same color and shape, usually in a bunch or bound to a single soil macro aggregate. Foamed or other particle forms occur least. In case of the particle shape, irregular (48%) and regular (32%) shapes are most common (Fig. 3b). This seems to be contrary, but fragments and films occur in both shape forms. Other studies noted fibers (equivalent of filaments) as the most common shape form followed by fragments or films in the microplastic size class (Corradini et al., 2019b; Liu et al., 2018; Zhang and Liu, 2018). In contrast, MEP films and fragments represent the main share of particle shapes (Liu et al., 2018; Piehl et al., 2018). This comparison is limited by the fact that all mentioned studies investigates topsoils or plastic surface accumulations on agricultural land. For MEP and CMP in floodplain soils under agricultural use, films (46%), fragments (31%), and filaments (23%) arise as documented for MEP by other authors. Unlike this, soils under grassland use or in riparian sites fragments (32%) building the majority and filaments are just 17%.

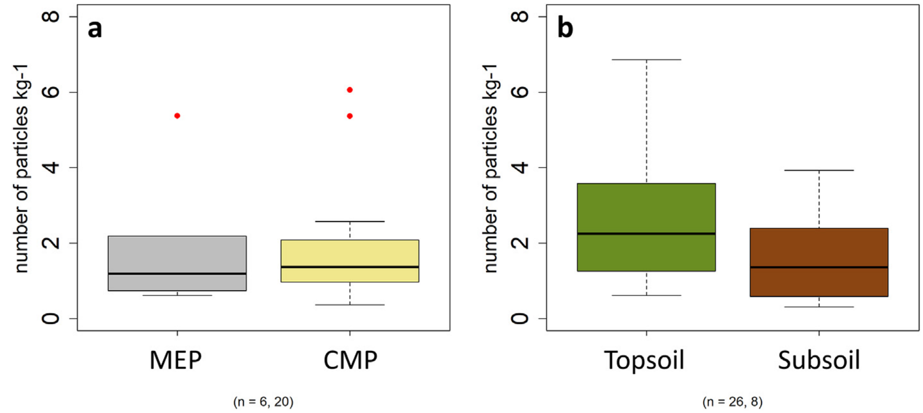

Fig. 1. Overview of number of MEP and CMP particles per kilogram soil dry weight. a) MEP and CMP loads at all sampling sites. b) MEP and CMP loads in topsoil (depth: 0-30 cm) and subsoil (depth: ˃30 cm).

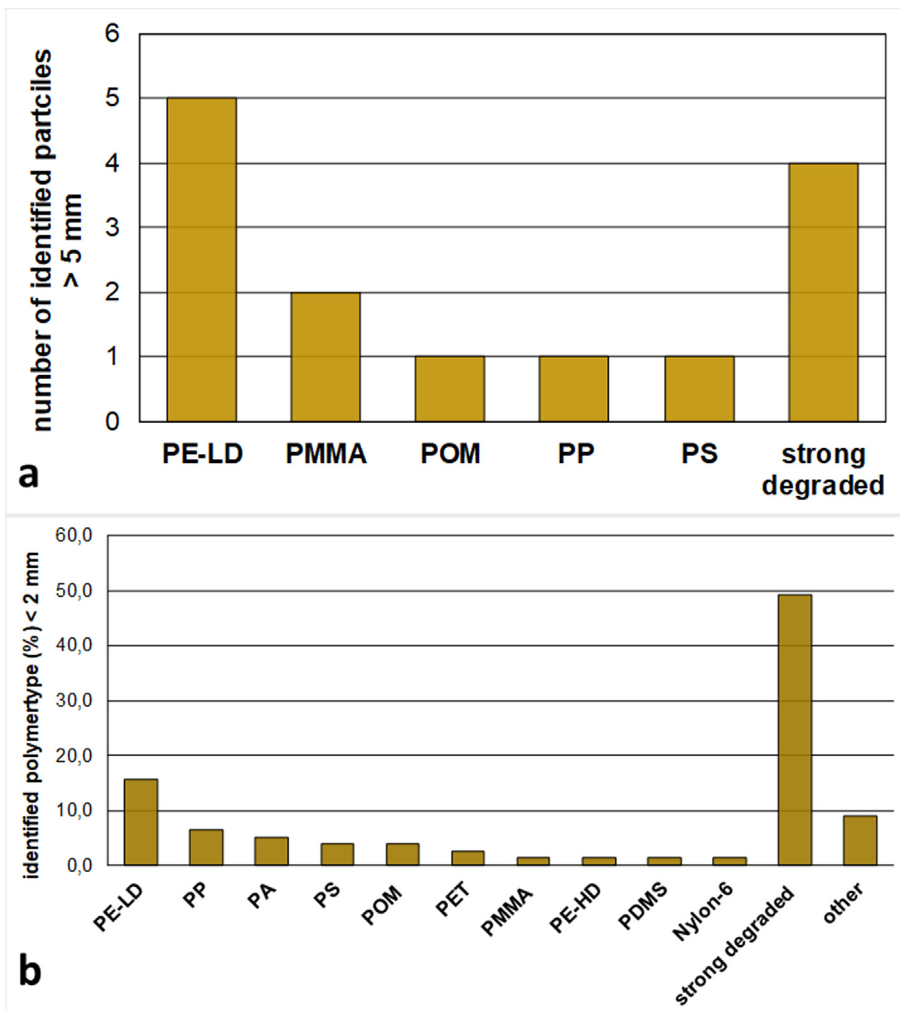

Fig. 2. ATR-FTIR identified polymer types in floodplain soil samples. a) Absolute number of identified MAP particles. b) Polymer type percentage of identified CMP particles (n = 78).

In addition, the condition of particles, forms, and shapes indicates primarily weathered (68%) or very degraded (5%) conditions (Fig. 3c). This could indicate that the majority of particles have been in the environment for a longer time. Both chemical and physical degradation processes, like UV-radiation or mechanical crushing, could lead to the degradation of the particles over time (Song et al., 2015). Out of this indication, these particles could be named as *old*. Unweathered or fresh particles (25% in sum) indicate that fresh and/or young particles are also present. In comparison with land use, fresh and unweathered particles occur more often on

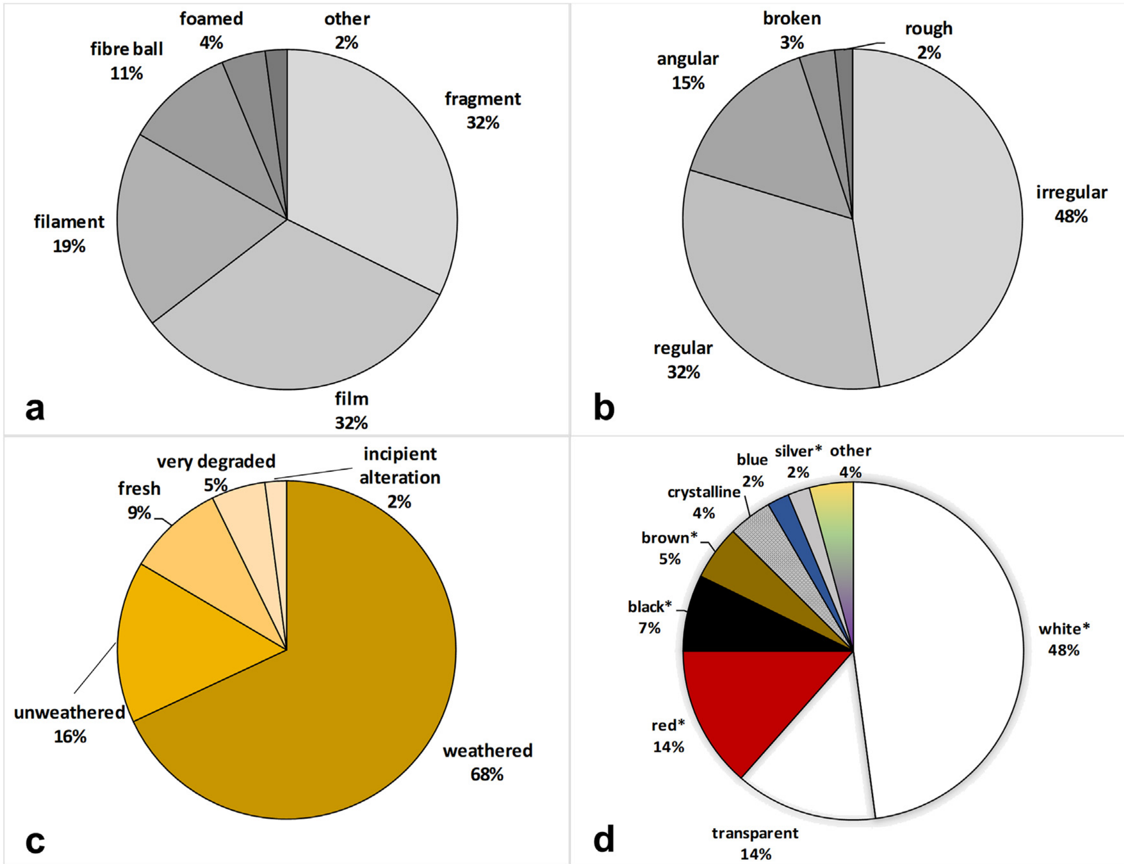

Fig. 3. Shape and surface characteristics of MEP and CMP particles. a) General form of MEP and CMP particles according Hidalgo-Ruz et al. (2012). b) Shape form of MEP and CMP particles according Hidalgo-Ruz et al. (2012). c) State of shape, form and color conservation according Hidalgo-Ruz et al. (2012). d) Surface color of of MEP and CMP particles.

cropland (38%) than on grasslands or riparian (22%). This may suggest a permanent input of fresh MEP and CMP through agricultural practices and a slow degradation (Corradini et al., 2019a; Hurley et al., 2018; Napper and Thompson, 2019).

A comparable finding shows the observation of the particle color. MMP and CMP occur first of all in white (48%), transparent (14%), and red color (14%), followed by minor represented colors (Fig. 3d). The white and transparent color classes also include slight discoloration, often associated with yellowing or bleaching. As this result indicates, strong and fresh colors are minor, whereas bleached colors are widespread and indicate particle weathering (e.g., though UV-radiation; Song et al., 2015). Piehl et al. (2019) documented white and transparent colors as major shares for MEP on agricultural soil surfaces. Other studies note main shares of black or transparent colors in cropland topsoils comparable to the strings or shedding films from agricultural plastics (Liu et al., 2018). In the present study, no difference in particle color shares between land uses could be found. Therefore, the composition particle color share seems to be a question of MMP and CMP source, especially on agricultural soils.

3.3. Lateral particle distribution

The lateral spatial distribution of plastic in floodplain soils can be captured on two levels. Level 1 represents the floodplain catchment area, subdivided in up- and downstream areas, and level 2 represents a detailed differentiation of land use and flood dynamics on transect sides. With increasing flow length and catchment area size, the number of possible plastic sources (e.g., urban areas drainage, agricultural land use) increases (Ballent et al., 2016; Fischer et al., 2016; Klein et al., 2015; Tibbetts et al., 2018). The presence of MEP and CMP at all floodplain transects indicates a widespread occurrence of plastics in soils.

Sampling site ELM, representing the upstream course of the Lahn River catchment, shows an CMP average of 1.51 mg particles kg^{-1}, whereas site LIM located in the downstream course shows a higher CMP average of 2.66 mg particles kg^{-1}. In contrast, sampling sites ROT and STD, representative for the middle course, range between 1.22 and 1.36 particles kg^{-1} (CMP average). A continuous increase relative to the flow length cannot be observed. Nevertheless, significantly higher maximum values occur at the downstream sites (STD: 2.57 particles kg^{-1} CMP; LIM: 6.06 particles kg^{-1} CMP). A higher abundance of MEP and CMP near urbanized areas or behind those in flow direction, found for different MP loads in river waters worldwide, could not be approved for floodplain deposits (Xiong et al., 2018). However, the increase in maximum values suggests that an increase in possible plastic sources leads to an increase in MEP and CMP concentrations, as demonstrated for Swiss floodplain soils by a correlation with population (Scheurer and Bigalke, 2018). It can be concluded that the basic distribution of plastics in floodplains seems to be diffuse, based on the multitude of input paths (Blettler et al., 2017; He et al., 2018; Karbalaei et al., 2018).

The spatial sampling approach in floodplains cross-sections allows a data evaluation on level 2: Considering flood dynamics as a possible transport mechanism of plastics in fluvial systems, each floodplain area could be divided into a proximal area (near to channel, high flood probability) and a distal area (distant to channel, lower flood probability; Bridge, 2003; Fryirs and Brierley, 2013). Superordinately, 75% of MEP and CMP particles occur in near channel samples, whereas 25% occur in a larger distance to the channel (Fig. 4b). As near channel sites including riparian strips (riparian vegetation) and grassland, both land use types contain 87% of MEP and CMP particles in sum (Fig. 4a).

The high loads of MEP and CMP in near channel sites could be explained by the frequent occurrence of flood dynamics at those floodplain parts and the facilitated deposition of plastics through higher vegetation roughness. Only 13% of the total MEP and CMP amount were detected on farmland sites, although this was the second-most frequent land use of sampling sites (Table 1). Therefore, the agricultural land use and cultivation practices, often suggested as a major source of plastic contamination of soils, could not be seen as the primary source (Bläsing and Amelung, 2018; Corradini et al., 2019a; Piehl et al., 2018). However, MEP surface enrichments, like on-site STD-1, may be traced back to sewage sludge application or degradation of microplastic debris (Corradini et al., 2019a; Piehl et al., 2018).

The identified lateral distribution pattern of plastics in floodplain soils leads to the conclusion that flood dynamics could build a link between the aquatic and terrestrial system. Plastic loads in river waters and sediments could be transported or deposited not only in riparian sediments but also in floodplain soils. MAP, MEP, and CMP deposition on soil surfaces was observed in the surroundings of all sampling sites after flood events (Fig. S6). The occurrence of MEP and CMP in distal floodplain areas and on farmlands indicates that several factors interact in the same area. Therefore, a bunch of potential plastic sources could play a role for plastic abundance. Because flood dynamics and deposition, cultivation practices, and the degradation of microplastic debris in direct surroundings could be considered key processes, a complex source pattern has to be assumed (Scheurer and Bigalke, 2018; Wang et al., 2019, 2020; Zhang et al., 2020). This can include both point or diffuse sources and small-scale hotspots (e.g., waste debris) in aquatic and terrestrial systems. Future research should therefore increasingly differentiate the possible sources to develop strategies against a progressive increase of plastic contamination of soils and the environment.

3.4. Vertical dynamics of coarse microplastics

When studying microplastics or CMPs in soil, the vertical distribution is imporirtant because the majority of environmental processes in soils also cover deeper sections than the topsoil. Vertical displacements of MP in soils have already been demonstrated by Rillig et al. (2017a, 2017b) and Yu et al. (2019). Further displacement processes are therefore expected. As far as we know, no other study has dealt with samples from a depth greater than 25 cm (Weber et al., 2020).

In comparing MEP and CMP values in topsoil (0–30 cm) and subsoil (30–200 cm), the topsoils contain an average of 2.94 particles kg^{-1}, whereas the subsoils indicates lower average values (1.62 particles kg^{-1}; Fig. 1b). A decrease in depth is also observed on agricultural soils (Liu et al., 2018; Zhang and Liu, 2018). Liu et al. (2018) report an average number of mesoplastics between 6.75 particles kg^{-1} in shallow soil to 3.25 particles kg^{-1} in deep soils. However, Zhang and Liu (2018) studied agricultural soils, and Huerta Lwanga et al. (2017) studied garden soils, and neither declared significant difference between two soil layers because of cultivation practice. Unfortunately, a comparison is difficult, despite the few studies available, because only depths of 3–6 cm (Liu et al., 2018), 5–10 cm (Zhang and Liu, 2018), and 10–20 cm (Huerta Lwanga et al., 2017) are regarded as deep-soil layers. The comparison is therefore insufficient because these depths are considered topsoils both on agricultural soils and under grassland. The vertical distribution of MEP and CMP is heterogeneously similar to the lateral distribution. As an example, in profile LIM-1 (riparian zone), 10 CMPs occur in the upper 30 cm (0–10 cm: 4 CMP, 10–20 cm: 1 CMP, 20–30 cm: 5 CMP) and a single particle in a depth of 75–100 cm.

The maximum depth at which CMP (single PA particle) was found is 75–100 cm below the surface at the above-mentioned sampling site LIM-1. A difference between visual- and polymer-type identified particles in the maximum depth could not be detected (Fig. 5). The majority of particles found in subsoils is related to proximal floodplain (near channel) sampling sites (Fig. 5). The vertical distribution of MEP and CMP particles for all sampling sites shows that the maximum reached was between 20 and 30 cm depth.

Comparing depth distribution to land use classes, maximum depths are reached under grassland (60.0 cm) and riparian (87.5 cm; Fig. 6). Under farmlands, the maximum depth of plastic abundance is strongly correlated with the frequent working depth of cultivation (approx. 30 cm). However, those differences in depth distribution compared with distance to channel are linked to the land use because farmlands are more frequent in the distal floodplain area. A vertical displacement of the CMP is conceivable only through a few processes, as in situ transfer is particle-size dependent (Blume et al., 2016). The clear depth limit under arable land use indicates that CMP does not penetrate through the frequent compaction under the cultivated topsoil (Blume et al., 2016). In addition to anthropogenic relocation (e.g. by ploughing), further deep displacements by preferential flow paths (e.g., coarse pores,

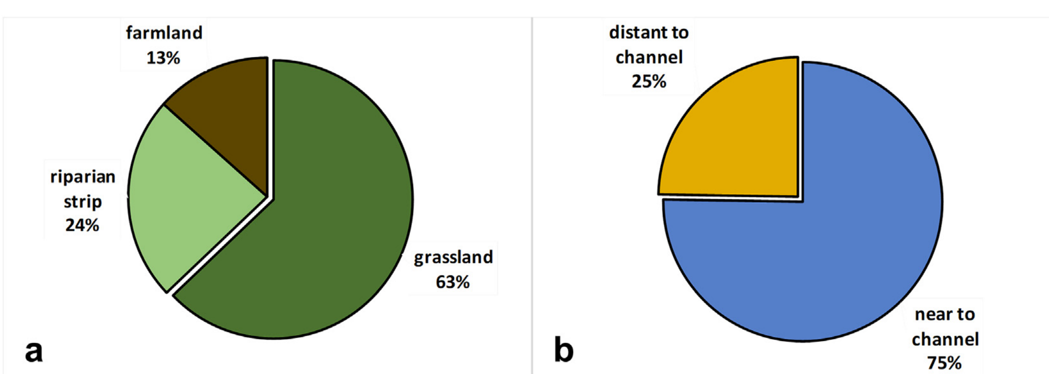

Fig. 4. Lateral MEP and CMP distribution according to sampling sites in floodplain areas. a) According to land use (n = 26). b) According to channel distance (reported in Table 1) (n = 26).

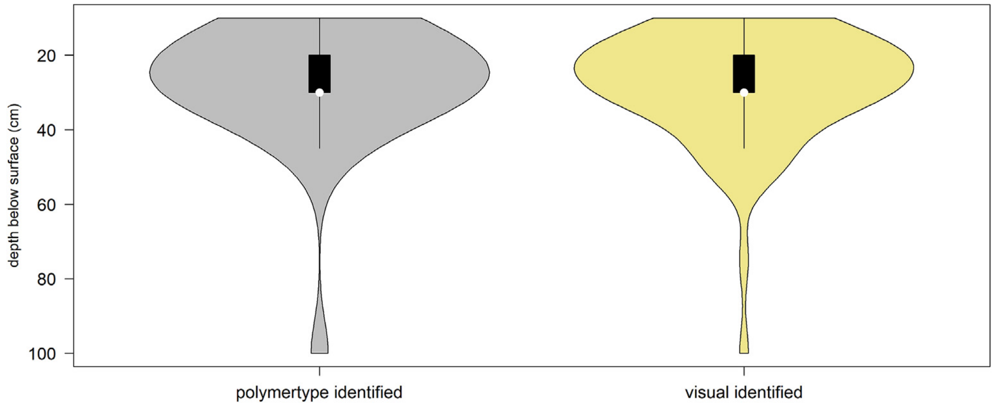

Fig. 5. Depth distribution of MEP and CMP particles for polymer type identified particles (n = 53) and visual identified particles (n = 94).

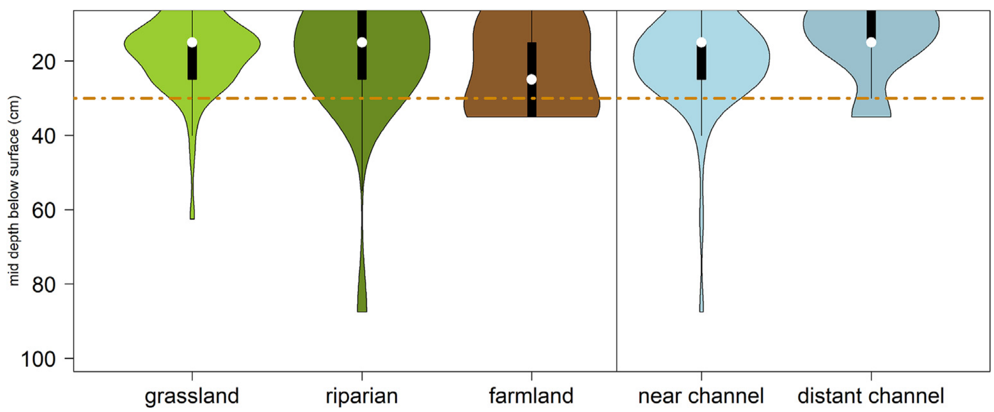

Fig. 6. Depth distribution of visual identified MEP and CMP particles differentiated by land use and channel distance (both parameters reported in Table 1 for each sampling site). Grassland: n = 61, riparian: n = 23, farmland: n = 12, near channel (< 100 m): n = 72, distant channel (> 100 m): n = 24. Maximal soil cultivation depth (approx. -30cm) added with orange dotted line.

earthworm tunnels) or bioturbation are conceivable (Lahive et al., 2019; Rillig et al., 2017b; Selonen et al., 2020; Yu et al., 2019).

On the other hand, the high contents and maximum depth in riparian zones may indicate direct sedimentation or younger relocation of sediments containing plastics (e.g. bank erosion; Klein et al., 2015). Comparing the real CMP depth distribution with the expected CMP depth distribution, calculated by earliest occurrence of identified plastic type and catchment sedimentation rate per year, CMP occurs at much greater depths than could be achieved by sedimentation processes alone (Fig. 7).

The average mid-depth where MEP was detected is 14.0 cm (22.4 cm for CMP). Based on sedimentation rates an average mid-depth of 5.2 cm for both MEP and CMP was calculated. Therefore, CMP occur 4.3 times deeper than assumed by sedimentation rates. As most identified particles have a date of earliest occurrence within the 1930s–1950s, the calculated sedimentation rate can become even smaller because the increase in global plastic production started at the end of the 1950s (Andrady, 2017; Heinrich-Böll-Stiftung and Bund für Umwelt und Naturschutz Deutschland [BUND], 2019). However, this result indicates that sedimentation could not be the main driver for MEP and CMP deposition in deep floodplain soils. To enable CMP to reach such depths, vertical transport processes must take place in the soil. While our results can provide indications merely through the vertical distribution, other authors have examined vertical displacements more closely. Rillig et al. (2017b) have demonstrated vertical displacement by earthworms even for particles of a size from 2360 to 2800 μm (i.e., partly in the CMP size range). For microplastics with a size less than 2000 μm, which are not included in this study, an in situ vertical transfer through soil pore space should be easily conceivable (Engdahl, 2018). Other authors suggest that sedimentation is also not a major source for MP and MEP loads in floodplain soils or riparian sediments based on correlation with grain size data (Scheurer and Bigalke, 2018). However, this comparison is only possible to a limited extent, as the authors examined topsoils in the direct riparian zone which cannot be regarded as representative of the sedimentation dynamics in the entire floodplain. However, the major concentration of MEP and CMP in upper soil layers at riparian zones and local meso- and macroplastic accumulations (Figs. S5a, S6a, and S6b) after floods indicate that this highly dynamic source cannot be excluded here. Flood events can deliver plastic of different sizes. Nevertheless, environmental degradation processes (e.g., chemical or physical alteration) and relocation processes seem to play a key role for the heterogenous distribution of plastic in the floodplain soils. In order to understand vertical processes and dynamics more clearly in future, in addition to the expanded use of laboratory experiments (e.g., column experiments), more field studies with a vertical context are recommended. High-resolution vertical sampling, in connection with the recording of soil parameters such as density, pore fraction or pore size, and also the dating of sediment layers could provide information about these in-situ processes.

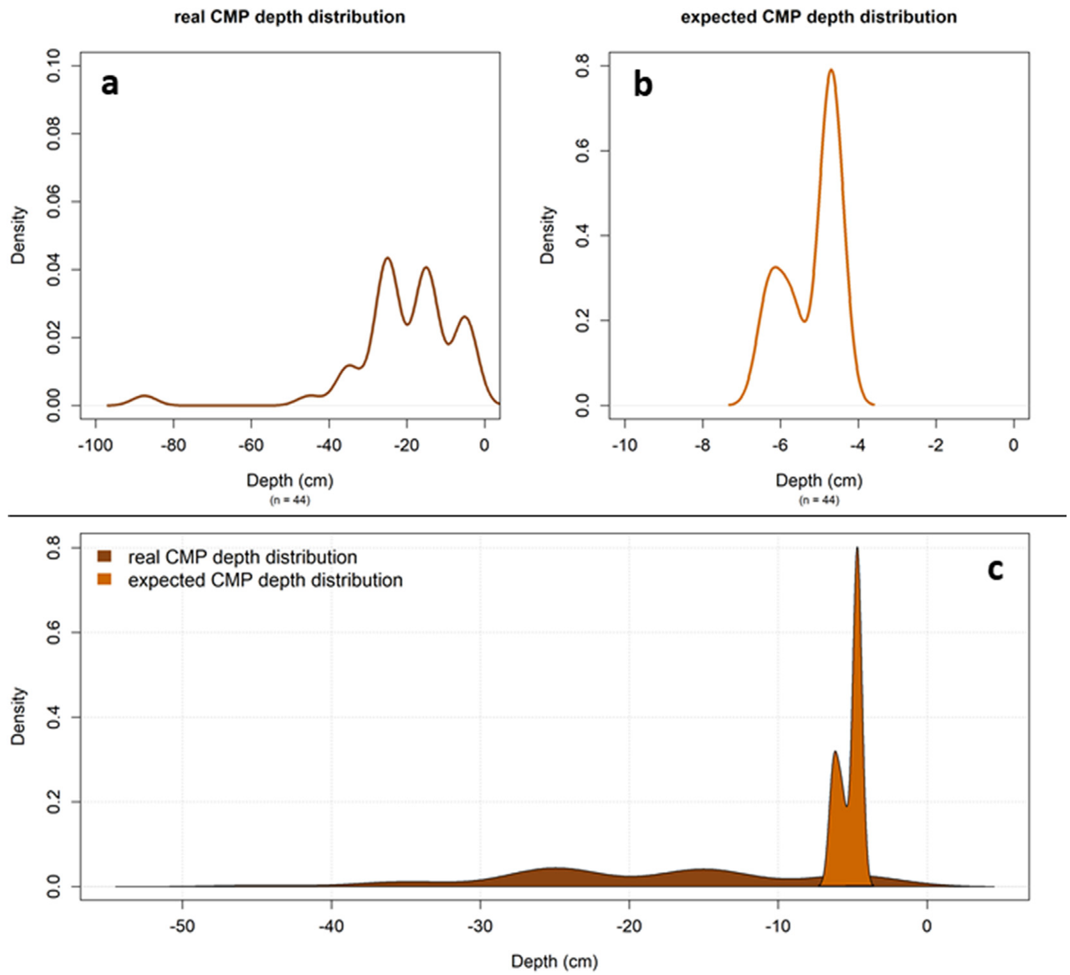

Fig. 7. Depth distribution of CMP in a) real floodplain samples (n = 40) and b) expected depth distribution according plastic age and sedimentation rates (n = 40). c) Density plot of the value distribution for both, real and expected values, by depth.

3.5. *Plastic contaminations in floodplain soils*

If microplastics are considered an environmental pollutant, then it must be assumed that, in contrast to other pollutants (e.g., heavy metals), no natural or geogenic background value exists. It must be anticipated, first, that plastics are purely anthropogenic substances that have been present in the environment for only a short period of time (Andrady, 2017; Zalasiewicz et al., 2016). Second, the variety of occurring particle shapes, the type of plastics reinforced by the variety of additives, make a comprehensive pollution assessment complex (He et al., 2018a; Wang et al., 2020; Zhang et al., 2020). Several studies on the effects of plastics and microplastics in soils show that microplastics pose an emerging threat to soil biota and plants (Rillig et al., 2017a, 2019; Xu et al., 2019). By plant uptake of micro- and nanoplastics, food production and human health could be affected (Rillig et al., 2017a, 2019).

Overall, the abundance of MEP and CMP in floodplain soils of the Lahn River is significantly smaller than in contaminated soils, agricultural soils, river sediments, and river waters (Corradini et al., 2019b; Fuller and Gautam, 2016; Klein et al., 2015; Liu et al., 2018; Piehl et al., 2018; Zhang et al., 2019; Zhang and Liu, 2018). However, the relatively small amount of MEP and CMP is in line with the findings from other soil-related studies, which state that the number of plastic particles in increasing with decreasing size (Corradini et al., 2019b; Huerta Lwanga et al., 2017; Liu et al., 2018; Scheurer and Bigalke, 2018; Zhang et al., 2019; Zhang and Liu, 2018). This could also indicate that the contamination levels of the smaller microplastic particles ($<$2000 μm) inside the floodplain soils of the Lahn River could be considerably higher. Assuming the spatial distribution that proves the occurrence of MEP and CMP partly independent of land use, with different emphases in the entire floodplain of a main river catchment area, a widespread contamination of soils is probable.

In addition, every larger particle will be reduced in size to microplastic and later nanoplastic after a certain time as a result of degradation processes in the environment (Chamas et al., 2020; Napper and Thompson, 2019). However, because this degradation is very slow and with modeled half-lives ranging from 250 years (HDPE bottle) to $>$ 2500 years (PET bottle) in buried conditions, the plastic contamination could be present over a long period (Chamas et al., 2020). The long half-lives and slow degradation are contrary to the larger fraction of strongly weathered and therefore ATR-FTIR unidentified particles in this study. As the number of field studies considering plastics properties in soils (e.g., weathering status, surface degradation) is still very small, an evaluation of the modeled values from field samples is difficult (Andrady, 2017; Xu et al., 2019). A faster degradation of particles through environmental factors, especially of the particles surfaces, which does not represent a half-life, therefore seems, in general, possible. Even if the absolute contents of CMP in this study appear small, they demonstrate that, despite the heterogeneous distribution, plastics (a) widely occur in floodplain soils, (b) occur at greater depths than previously assumed, and (c) have the potential to cause environmental degradation.

Because floodplains are important ecosystems in the transition between terrestrial and fluvial systems, have an important role in food production and act as a connecting space between landscapes, further monitoring in a spatial context of plastic pollution is essential in the future.

4. Conclusion

The spatial-research approach enables statements about plastic occurrence and distribution in the three-dimensional system of the soil scapes. Using the example of floodplain soils, it could be demonstrated that plastic particles are heterogenously distributed but still occur widespread in soils. Even if the comparison between recent studies is limited due to the different spatial conceptions, analytic methods and quantitative values used, we can conclude that the MEP and CMP concentration and particle features (e.g. shape, polymer type) are widely comparable to the findings in other studies. However, the spatial approach allows us to draw first conclusions about the processes which could be relevant for the spatial distribution discovered. From the lateral particle distribution, it can be concluded that a complex source pattern leads to the plastic pollution of floodplain soils. Together with the increase of maximum values with the flow length and the influence of floods as input path from the water bodies, an interaction between terrestrial and fluvial processes can be assumed. The heterogenous vertical MEP and CMP trends, with detections at greater depths than previously assumed, could indicate vertical in situ transfer. Additionally, the comparison with sedimentation rates shows that sedimentation processes alone could not be the major source of plastic pollution. In general, if comparatively large particles already cause widespread pollution in floodplain soils, the contamination through microplastics in soils could be immensely greater. Even if a sufficient data basis is missing so far, this would clearly challenge today's management strategies for handling plastic contamination in soils. As complex sources seem to occur, further research should focus on the identification and quantification of potential sources of plastic contamination. Additionally, the investigation of in situ processes (e.g., vertical transport, particularly for microplastics), risk assessments for plant uptake, and especially more data on spatial dependencies of plastic contamination can improve further management of plastic contamination in soils.

CRediT authorship contribution statement

Collin Joel Weber: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Visualization, Project administration, Funding acquisition. **Christian Opp:** Conceptualization, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115390.

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