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Meso- and microplastic distribution and spatial connections to metal contaminations in highly cultivated and urbanised floodplain soilscapes – a case study from the Nidda River (Germany)

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Abstract

Floodplain soilscapes act as temporary sinks in the environment and are nowadays affected by multiple contaminant accumulations and exposures, including different trace metals and plastics. Despite increasing knowledge about the occurrence and behaviour of plastics at the interface between aquatic and terrestrial systems, there are still major uncertainties about the spatial distribution of plastics, their sources and deposition, as well as spatial relationships with other contaminants. Our recent case study addresses these questions, using the example of a river system ranging from rural to urban areas. Based on a geospatial sampling approach we obtained data about soil properties, metal contents via ICP-MS analyses, and particle-based (171 μ m – 52 mm) plastic contents, analysed using sodium chloride density separation, visual fluorescence identification and ATR-FTIR analysis. We found plastic contents of 0.00–35.82 p kg⁻¹ and zero to moderate metal enrichments. Levels of both contaminations occur in the lower range of known concentrations in floodplain soils and show a different spatial distribution along the river course and in the floodplain cross-section. Furthermore, we found that plastic enrichment occurs in the uppermost soil layers, while trace metal enrichment is equally distributed over depth, indicating different sources like flood dynamics and agricultural practice during different deposition periods. Finally, direct short to long-term anthropogenic impacts, like floodplain soilscapes.

Keywords: Plastic, Heavy metal pollution, Wetland, Soil, Fluvial, Density separation

Introduction

The global contamination of the environment through plastics has led to worldwide detections of plastic in almost any environmental system, including marine, freshwater and terrestrial environments among others [1]. After an initial half-decade of investigation plastic

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contamination also in terrestrial systems and their soils, it has become clear that soils of different soil landscapes (soilscapes) contain far more plastics than perhaps previously assumed [2, 3]. These initial findings include the evidence of plastics from highly cultivated to semi-natural soilscapes [2, 4], opening questions about the role of soil systems as a potential plastic reservoir within the global plastic cycle. Additionally, it was already proven that plastics can affect a wide variety of soil functions and related ecosystem services [5, 6], like influences on soil



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structure, material balance, the release of pollutants or the uptake of plastics into the food chain [6].

Investigating plastics in the environment has led to various definitions and size classification resulting from the different research disciplines dealing with plastic contaminations [7]. Plastics within the size of microplastics (MP) defined by size as particles with a size between 1 μ m and 1000 μ m [8] or 1 μ m and 5000 μ m as well as nanoplastics (NP, < 1 μ m) are currently receiving the most attention in environmental research [7, 9]. Additionally, particle size-based distinctions have been made between coarse microplastics (2–5 mm) [10], meso- (>5 mm) [9] and macroplastics (>5 or > 25 mm) [7, 8].

Regarding the role of soil systems as a reservoir within the global plastic cycle, floodplains and their soils as floodplain soilscapes have a special role. Even if floodplains cover only 0.5–1% of Earths land area [11], they act as transitional landscapes between aquatic and terrestrial systems and therefore as temporary transfer and deposition areas of plastics as a semi-terrestrial part of fluvial transport corridors [12-14]. Due to the spatial location of floodplain soilscapes within an aquatic-terrestrial interface, they are entangled with the river through flood dynamics and show a widespread genesis within a human-natural entanglement by the deposited legacy sediments [15]. This has also led to the circumstance that floodplain soilscapes have been subject to a constant impact of different contaminants (e.g., potential toxic metals like lead or copper, PAHs) from e.g., mining or industry sources in the past and nowadays also for plastics [16-18].

Until now six studies investigating five floodplains were able to show that plastics enter floodplain soilscapes mainly through area-wide flood processes (flood deposits) and local point sources like agriculture [10, 18-22]. Additionally, further sources like littering and surface runoff from slopes are probable [23]. Reported particlebased plastic loads include different ranges like 4.94-252.70 p kg⁻¹ (0.1–5.0 mm, NaCl separation) down to 80 cm soil depth in intensively utilised agricultural soils of the lower Yangtze River floodplain (CHN) by Cao et al. [19] or 23–330 p kg⁻¹ in proximal floodplain topsoils (0-4 cm) of three rivers in Virginia (US) with average sizes of 290–1160 µm (NaCl separation) by Christensen et al. [20]. Within Central Europe, Lechthaler et al. [18] documented average loads between 47.9 p kg⁻¹ (depth profiles) and 25.4 p kg⁻¹ (topsoils) for microplastics within the Inde River floodplain (Germany) with a size between 500 and 5000 µm (canola oil extraction) and Weber et al. [22] reported loads of 0.36-30.46 p kg⁻¹ with a size of 219.0-8321.0µm (NaCl separation) for the Lahn River floodplain (Germany). Previous research identified that the lateral distribution along the river

and potential plastic inputs has been linked to population density in the catchment [21], land use (direct input agriculture) [19] and vegetation (trapping) [10] as well as floods (sediment deposition) [18, 20]. Regarding vertical distributions within floodplain soilscapes, incorporated plastics can accumulate in the youngest and uppermost floodplain soil layers due to sedimentation since the 1960s, or agricultural utilisation, but also reach deeper soil layers through in-situ displacement [10, 19]. Plastic abundance in floodplain soils seems to have a clear drop with the soil depth [19, 22]; Christensen et al. [20] found in some parts higher plastic loads in the floodplain than in the stream channel, which may relate to remobilization of deposited plastics, and exports during floods at the river channel [24, 25].

Regarding spatial patterns of MP distribution within floodplain soilscapes it can be summarized that first, the global data availability and number of studied floodplains remains small, second, an increase of potential plastic sources along the river leads to higher plastic loads within floodplain soilscapes (source-to-sink) [21] and third, that plastics are deposited within the youngest and upper floodplain soils and are subject to vertical insitu displacements. When combining these summarized points to the well-known contamination of floodplain soilscapes with metals or potential toxic metals, different processes between plastics and metals, like the dissolution of additive metals from plastics [26, 27], adsorption of different metals (e.g., Cd, Zn) from the soil matrix [28, 29] or the influence of MPs on metal behavior in soils (e.g., reduction of the exchangeable, carbonate-bound and Fe-Mn-oxide bound fraction) [30], seem also plausible in floodplain soilscapes. With regard to the abovementioned interactions between plastics and metals, it is therefore important to determine whether both contaminants are equally distributed within floodplain soilscapes. Finally, future risk assessments of MP contaminations or the consideration of plastic-metal interactions requires basic knowledge about the spatial distribution, the identification of potential floodplain soilscapes hotspots, and the natural and anthropogenic processes responsible for them.

Against the background of current research on MP in floodplain soils and initial findings about the deposition, spatial distribution as well as interrelationships to other pollutants, there is still a lack of spatial MP data with regard to strongly different floodplain properties (e.g., flood frequency and extension, sediment origin and budget, soil stratigraphy or land use compositions). Furthermore, questions arise about whether previous findings from other catchments are transferable and whether concrete spatial contamination patters within floodplain soilscapes are identifiable. Based on these general

questions, we conducted a case study within the floodplains of the Nidda River and its catchment area focusing on the analysis of macroplastics (>5mm), coarse microplastics (2-5 mm) and microplastics $(>300 \mu \text{m}, \text{ lower})$ detection limit) in combination with different metals. The Nidda River can be considered very representative of Central European river basins due to the gradient from the rural upper reaches over agricultural heartland to the highly urbanised lower reaches [31]. Furthermore, the associated land use, possible point and diffuse plastic and metal sources and the conflicts arising around the floodplain and watercourse management are also comparable to many intensively utilised floodplain areas. Regarding the predominant soilscape properties, we aim to investigate the following aspects and related issues at the aquatic-terrestrial transitional zone of the Nidda River:

- 1. Providing spatial MP and metal data combined with basic information about plastic abundance and their spatial distribution within the floodplain soilscapes of the Nidda River to enable comparisons with other catchments.
- 2. Study the lateral spatial distribution of plastics and metals along the river and within floodplain crosssection, testing the hypothesis that contaminant loads increase with the river course with potential contamination sources (hypothesis 1).
- 3. Investigate the vertical spatial distribution of plastics and metals within floodplain soils, testing the hypothesis of comparable deposition conditions and times for both contaminants (hypothesis 2).
- 4. Compare lateral and vertical spatial patterns of both contaminants to soil stratigraphy and local land use changes, testing the hypothesis of uniform, area-wide deposition patterns (hypothesis 3).

Methods

Study area

The Nidda River, with a length of 89 km, drains large parts of the Wetterau basin (1942 km²) into the River Main, located in the in central Germany (Fig. 1) [32]. The Wetterau basin is an important cultural landscape, which has been inhabited and cultivated almost continuously since the early Neolithic period [33, 34]. This long-lasting cultivation and deforestation influence the soilscapes of the catchment area. While the upper reaches and upland positions are dominated by Cambisols developed in periglacial solifluction layers (basalt), the remaining catchment area is dominated by soils developed on loess (Luvisols, Regosols, relictic Chernozems, Stagnosols) [34]. The floodplains of the Nidda River first appear after the Vogelsberg (headwaters) and progressively expand with the river course; they consist mainly of Fluvisols (fine-grained Holocene flood loam and colluvial deposits) and partly of Gleysols and Stagnosols [32]. The Nidda floodplains, except for the direct riparian zones, are often cultivated as cropland, meadows and pastures. Large parts of the Nidda floodplains are part of a landscape conservation area for the protection of near-natural floodplain areas. However, the amount of land used for residential development and infrastructure facilities is also increasing, reaching a high level in the Frankfurt metropolitan area where the Nidda River joints the River Main, and restricts near-natural or just cultivated floodplain areas severely.

The Nidda River can be classified as a characteristic medium-sized stream system with six major tributaries (Fig. 1). The stream is influenced by intense industrial and agricultural activities, including six industrial and municipal wastewater treatment plants in its course, and river engineering for flood protection [31, 35] (Fig. 1). Today's land use along the Nidda River changes from a rather rural environment in the upper reaches (population density: 72 people km², settlement and traffic area: 10.6%) to a heavily cultivated agricultural heartland in the middle reaches (population density: 282 people km^2 , settlement and traffic area: 16.1%), to the highly urbanised lower reaches in the Frankfurt metropolitan region (population density: 3077 people per km², settlement and traffic area: 58.6%) [36], resulting in multiple possible point and diffuse (micro-) plastic as well as metal sources along the river course. Furthermore, agricultural practice includes mainly conventional crop production (mainly wheat, barley, maize) and grassland cultivation for livestock farming [36], resulting in a additional direct source for plastics and metals on crop- and grassland.

Flood events under the hundred-year flood level (<HQ100) occur frequently between December and February in flood retention areas along the river course, with a total area of 44.8 km² [37]. Flood protection measures, such as dams, widening of the cross-section, or flood retention basins in the catchment area, have been constructed since the 1920s and have been continuously expanded [37]. Nevertheless, ten high flood events with a discharge $> 25.7 \text{ m}^3 \text{s}^{-1}$ for the gauge Nieder-Florstadt (ID: 24830050, between sampling sites OKA and FRA) occurred between 1967 and 2011, showing an average discharge of 28.2 m³s⁻¹ (max: 37.6 m³s⁻¹ in 1981), average discharge rate of 53.41 (s km^2)⁻¹ and an average water level of 308 cm (max: 343 cm in 1981) [37]. The average discharge during flood events exceeds the longterm middle discharge (MQ) of $3.0 \text{ m}^3 \text{s}^{-1}$ by 9.4 times.



Soil sampling

The selection of sampling sites was carried out with the aim of implementing a geospatial approach [10] and identifying sites representative of the floodplain land-scape and its different soilscapes as already introduced by Weihrauch [38] as well as Weber and Opp [10]. The

selection was carried out with the help of a preliminary evaluation of geodata (aerial photos, geological map, soil map, morphology) and the following conditions: each sampling site a) must be representative of a stretch of watercourse with typical soil formations and landscape characteristics [38], b) must be located in the designated floodplain (10–100 year flood events) (Fig. S1), c) should not be located in close proximity to potential MP point sources (e.g., garbage dump, sewage plant), and d) should be free of interruptions (infrastructure, dams) in the floodplain cross-section [10]. Furthermore, each site should consist of a clear structure of floodplain morphology including levee, inactive flood channels and backswamp [10].

In contrast to other catchments, the anthropogenic utilisation and land use in the Nidda River catchment was the major restriction for the identification of suitable sampling sites. Except for the headwaters, where no floodplain can be found due to narrow valley morphology, there are still four areas in the middle and lower reaches of the Nidda River that show a floodplain width of 400-900 m (on each side) and have not been dammed excessively. In these areas, which are part of the protected landscape area "Floodplain Association Wetterau", four transect locations were selected after a preliminary soil survey, in order to represent the floodplain cross-section (Fig. 1). Each transect location can be reached by annual floods, and would be flooded by 50-200 cm (sites NID and MOK) or 1-100 cm (sites OKA and FRA) during a hundred-year flood [37]. Furthermore, the transect locations are partly affected by river and floodplain renaturation measures. All sites are under agricultural land use, mainly pastoral and only at site OKA also arable (Table S1). Land use comparison based on satellite images from 1933 and 2020 show that arable land changed to grassland at the distal floodplain of site OKA, and floodplain renaturation is taking place at sites NID, MOK and FRA (Figs. S2 and S3).

Finally, soil sampling was conducted during summer 2019 and 2020 at the river transect locations, using pile core driving with stainless-steel cores (diameters of 100 mm and 80 mm) down to a depth of 2 m. At each site, two (NID, FRA) or three (MOK, OKA) points were sampled, each localised in the proximal or distal flood-plain area, with an additional third point in the central floodplain at sites MOK and OKA as the floodplains are wider there. Sampling points were numbered as 1 (distal), 2 (central) and 3 (proximal) at sites MOK and OKA, 1 (distal) and 2 (proximal) at site NID and 1 (proximal) and 2 (distal) at site FRA. Sampling points are furthermore available as .kml file within a data repository.

At each sampling point of the transect, two complete cores at a distance of 5 m from each other were extracted, resulting in 20 cores. Soil stratigraphy and pedogenesis were documented according to the FAO Guidelines for soil description [39], and classified according to WRB 2015 [40] and German soil classification [41]. Samples were collected from the two cores with stainless-steel spatulas and pooled in the field according to fixed

depth levels (10 cm sections in 0–0.5 m, 25 cm sections in 0.5–1.5 m and 50 cm section from 1.5–2.0 m), resulting in 10 composite samples per sampling point (total: 100 samples, 385.5–3704.6 g dry fine earth per sample), and stored bioplastic bags, made from corn starch (biological origin) and biodegradable (Mater-Bi bags, BioFutura B.V., Rotterdam, Netherlands).

Additionally, plastics fragments on topsoil surfaces were sampled if a conspicuous amount of plastics could be found around the drill points [42]. Visible plastic fragments were collected in a 20 m² area around the drill points, by two people walking straight lines in parallel (four-eyes-principle), according to Piehl et al. [42]. Surface sampling area was prepared by means of a tape measure and measuring rods, while a rectangle with the extension of 4×5 m was measured and marked around the centre (drill point). This additional procedure was conducted at the OKA sampling site (points OKA-2 and OKA-3).

Laboratory analysis

Field fresh soil samples were immediately dried at 45 °C within opened bioplastic bags for 4 days in a closed drying chamber, without fan to avoid cross-contamination of samples by airflow. Subsequently the sample material was carefully mortared (ceramic mortar) to break down soil macro-aggregates, and dry-sieved through stainless-steel sieves (Retsch, Haan, Germany), covered with a stainless-steel plate, to the size fractions >5 mm (mesoplastics), >2 mm (coarse microplastics and rock fragments) and <2 mm (large microplastics and fine-earth fraction). The fine-earth fraction was afterwards homogenised in a stainless-steel bowl and divided via a rotary sampler (Retsch, Haan, Germany), to obtain representative subsamples for soil parameter and metal analysis. Each fraction was stored in corn starch bags.

Soil parameter and metal analysis

The residual moisture content of representative sub-samples (fine-soil < 2 mm, average sub-sample mass: 99.6 g) taken after drying at 45 °C, was determined by drying (105 °C), and the content of organic matter (OM) was determined via loss of ignition at 550 °C (DIN 19684–3:2000–08) and both were recorded as percentage by weight (wt%). Additionally, the pH was measured with a pH 91 electrode (WTW, Weilheim, Germany) in a 0.01 M CaCl₂ solution (m:V 1:2.5). Carbonate content was determined after reaction with a few drops of 3.23 M hydrochloric acid (HCl) according to Ad-hoc AG Boden [41] and soil texture was analysed via the Integral Suspension Pressure Method [43] after the samples had been prepared according to DIN ISO 11277:2002–08. Soil textures were reported according to Guidelines for soil

description [39] and individual shares of clay, silt and sand, in percentage by weight (wt%).

Pseudo-total concentrations of the metal Fe, the metalloid As and the trace metals V, Cr, Co, Ni, Cu, Zn, Cd, Hg, Pb were determined from digests of 1 g prepared subsample with 20 ml aqua regia (12.1 M HCl and 14.4 M HNO₃; ratio 1:3; DIN ISO 11466:2006-12). Metal concentrations were quantified using inductively coupled plasma-mass spectrometry (ICP-MS; XSERIES 2; Thermo Fisher Scientific, Bremen, Germany) and system calibration with a certified multi-element standard solution (ROTI®STAR; Carl Roth GmbH, Karlsruhe, Germany). Each digest was measured three times and averaged, resulting in converted results given in mgkg⁻¹. Relative standard deviation (RSD) after threefold measurements, and detection limits resulting from the multiplication of the mean standard deviation of 10 repeated blank measurements by factor 3, were used for correction [44, 45].

Plastics and microplastics analysis

Plastics and microplastics analyses were carried out according to the method and application first published by Weber & Opp [10] and Weber et al. [22]. Visual identification with naked eye, or the help of a magnifying glass, was conducted for macro- and mesoplastics (>5 mm) and with the help of a stereomicroscope (SMZ 161 TL, Motic, Hong Kong) for coarse microplastics (>2 mm) after dry-sieving. Potential plastic particles were cleaned (deionised water) [46], dried (45 °C), photographed and stored in rim jars made of glass with PE-caps for polymer identification.

The total fine-earth fraction (<2 mm) with a sample mass between 94.0 and 3552.1g (mean: 1295.3g, excluding coarse soil fragments) and a related average volume of 1053 ml (250-2750 ml), was used to separate out microplastic particles (Fig. S4). For this purpose, a density separation with the "MicroPlastic Sediment Separator" (MPSS) (Hydro-Bios Apparatebau GmbH, Kiel-Altenholz, Germany) under the application of a saturated and>300µm filtered NaCl-solution (density adjusted to $1.2 \,\mathrm{g/cm^3}$ and controlled by balance and aerometer) was performed [47]. Density control, before and after separation, shows a range between 1.195 to $1.218 \,\mathrm{g \, cm^{-3}}$ with an average of $1.203 \,\mathrm{g \, cm^{-3}}$ (measured by gravity), at an average solution temperature of 19.47 °C (Fig. S4). The sample solution was stirred for 60 minutes and then allowed to settle for 19 hours. At the end of the separation time, the integrated ball valve was closed, and separated material was rinsed into glass beakers using filtered NaCl solution.

Afterwards, the remaining sample material, consisting of organic material and potential plastic particles, was separated into the following size classes using stainless-steel sieves (Ø 75 mm, Atechnik, Leinburg, Germany), and filtered (> 50 μ m) deionised water: > 1000 μ m, > 500 μ m and > 300 μ m. After sieving, the sieve residues were filtered via vacuum-filtration on cellulose filters (Ø 47 mm, LLG-Labware, Meckenheim, Germany), and then transferred to glass petri dishes (Ø 90 mm or 200 mm) by rinsing with deionised water and drying at 50 °C for 2 days, according to Prume et al. [48].

To differentiate between organic material and potential plastic particles, a Nile Red staining procedure $(20 \,\mu g \,m L^{-1}$ Nile Red ethanol-acetone (1:1) solution, Sigma-Aldrich, Taufkirchen, Germany) was applied [49, 50]. Nile Red solution was applied with the help of a glass pipette and plastic (PE) spray bottle and stained for 10 minutes at 50 °C in a drying chamber [49]. Stained petri dishes were afterwards visually inspected systematically under a stereomicroscope (SMZ 161 TL, Motic, Hong Kong), with fluorescence setup (Excitation: 465 nm LED; Emissions 530nm colour long pass filter: Thorlabs, Bergkirchen, Germany) and transmitted light [48]. This approach allows the visual identification of plastic particles, but is disrupted by the fluorescence of natural organic components in the red fluorescence range (e.g., chitin shell ants, freshwater mussel fragments), so the exclusion of natural organic particles must be based on surface structure (e.g., cell structures) or, in case of uncertainty, by spectroscopic analysis. Each fluorescent or other potential plastic particle that shows no cellular or biologic structure and a clear and homogenic colour [51] was collected and individually stored in microplates (Brand, Wertheim, Germany). Each particle collected was then classified according to surface characteristics (particle type, shape, surface degradation, colour), photographed (Moticam 2, Motic, Hong Kong) and size-measured (longest diagonal, Motic Images Plus 3.0, Motic, Hong Kong) [51, 52]. Surface degradation status has been characterized into fresh, incipient alteration and weathered particles, according the presence of visible (40x magnification) of cracks, abrasion and colour bleaching.

Polymer type identification for a) previously visually determined mesoplastic to coarse microplastic particles and b) microplastic particles identified via staining-fluorescence procedure was performed using the Tensor 37 FTIR spectrometer (Bruker Optics, Ettlingen, Germany) combined with a Platinum-ATR-unit (Bruker Optics, Ettlingen, Germany). Measurement was carried out using 20 background scans followed by 20 sample scans for each sample, with a resolution of 4 cm^{-1} in a wave-number range from 4000 cm^{-1} to 400 cm^{-1} [46, 53, 54]. The ATR-unit used is the limiting factor for the lower size limit of the particles determined in this study, since particles with a size < 300 µm have insufficient contact area and are difficult to handle [22]. Quality assurance

and contamination control follows the procedures introduced within Weber et al. [22] and include the avoidance of plastic materials during field and lab work, contamination control via blank samples during all analysis steps and contamination prevention by wearing cotton lab coats, filtration (<50 μ m) of all solutions used as well as cleaning of all laboratory materials via filtered water. Blank samples have shown a contamination with five synthetic particles (4 fragments and one fibre) with an median size < 300 μ m (except a single fibre) and therefore below our detection limit (Fig. S5).

Statistics and data evaluation

Data processing, basic statistical operations and data visualisation operations were conducted using Microsoft Excel (version 1808, Microsoft, Redmond, USA) or RStudio (version 1.3.1093, RStudio, PBC, 2020) within an R environment (version 4.0.3, R Core Team, 2020). Data processing of FTIR spectra was performed in OPUS 7.0, including atmospheric compensation and baseline correction (concave rubber band method) (Bruker Optics, Ettlingen, Germany), and in Spectragryph (Version 1.2.14; Menges, 2020; Oberstdorf, Germany). Spectra identification of pre-processed spectra was done via the OpenSpecy database using full spectra and Pearson's r with $r^2 > 0.6$ as match quality indicator [55]. Spatial data was processed in ArcGIS (ArcMap version 10.8, Esri 2019, West Redlands, CA, United States).

In order to allow an effective assessment of spatial contamination differences between plastics and potentially toxic metals of interest, we calculated the metal "Pollution Load Index" (PLI) based on "Single Pollution Index" (PI) according to Kowalska et al. [56] following Eq. (1) and Eq. (2):

$$PI = \frac{Cn}{GB} \tag{1}$$

$$PLI = \sqrt[n]{PI_1 x PI_2 x PI_3 x PI_n}$$
(2)

where Cn is the concentration (mgkg⁻¹) of individual metals of environmental interest (V, Cr, Co, Ni, Cu, Zn, Cd, Hg, Pb) and the metalloid As, and GB is the geochemical background concentration, calculated for the individual metals (V: 38.25 mgkg⁻¹, Cr: 25.0 mgkg⁻¹, Co: 8.75 mgkg⁻¹, Ni: 24.0 mgkg⁻¹, Cu: 13.25 mgkg⁻¹, Zn: 58.5 mgkg⁻¹, Cd: 0.14 mgkg⁻¹, Hg: 0.04 mgkg⁻¹, Pb: 27.0 mgkg⁻¹, As: 8.0 mgkg⁻¹) based on the average values from 341 soil samples (64 floodplain silt substrates, 277 floodplain sand substrates) from Hessian floodplain soils [57]. As a second measure to assess a potential anthropogenic impact on metal concentrations, the "Enrichment factor" (EF) was calculated in order to measure the potential impact of anthropogenic metal contamination against geogenic background contents [56] following Eq. (3):

$$EF = \frac{\left\lfloor \frac{Cn}{LV} \right\rfloor sample}{\left\lfloor \frac{Cn}{LV} \right\rfloor GB}$$
(3)

where Cn is the concentration of individual metal and LV the reference content of Fe concentration $(mgkg^{-1})$ [57]. Index values were also evaluated according to Kowalska et al. [56] for PLI levels <1 as "denote perfection", levels around 1 as "only baseline levels of pollution" and levels >1 with "deterioration of soil quality", and for EF with "deficiency to minimal enrichment" at values <2 and "moderate enrichment" at values between 2 and 5.

Plastic loads were documented as particles per kg soil dry weight (p kg^{-1}), including the total number of sufficient ATR-FTIR identified particles per sample (sample mass 94.0 to 3552.1g). From 263 particles, previously identified visually (>2mm, coarse soil fraction) or via staining-fluorescence procedure, 35 particles (13.31% of all collected particles) were identified as natural organic matter with an r^2 between 0.77 and 0.96 for the spectra correlation by the OpenSpecy database. These organic particles were excluded from the entire data evaluation, which means that 228 particles are counted and evaluated as sufficiently $(r^2>0.6)$ identified as plastics. Ages of possible earliest occurrence (EPO age) were assigned to the particles identified based on the year of polymer development or start of production, according to Weber and Lechthaler [58]. In the case of identified rubber particles, EPO age were differentiated between natural (EPO: 1820) and synthetic (EPO: 1910) rubbers. Plastics identified as polymers grouped as rubbers or resins were summarized within the respective group following the OpenSpecy grouping. Macro- and mesoplastic loads on soil surfaces from sampling at site OKA were reported as particles per square meter ($p m^2$).

All data collected do not display a normal distribution (Shapiro-Wilk test) and some data show significant differences in variance by group. Normality and homoscedasticity of the residuals was checked graphically within R, showing no significant variance differences. Comparison of means was carried out using the Wilcoxon test or Kruskal-Wallis test, using R standard functions. Data visualisation was conducted with R standard functions (R Core Team, 2020), "ggplot2" (Wickham, [59]: https://ggplot2.tidyverse.org) and "corrplot" (Wei and Simko, [60]: https://github.com/taiyun/corrplot). Spearman correlation was performed with a significance level of $p \le 0.05$, and correlation coefficients were interpreted as: weak (r_{SP} 0.4 - <0.6), clear (r_{SP} 0.6 - <0.8), and strong (r_{SP}>0.8): https://CRAN.R-project.org/package=plot3D).

Results

Plastic loads and features

In the floodplain soils along the Nidda River, plastic particles were found at each transect site and sampling point, resulting in a positive rate of 73% of all samples (n = 100) which contain plastics. Based on our methodological approach we found a false-positive rate (fluorescent but no synthetic polymer according ATR-FTIR) of 12.5% for Nile Red fluorescent particles and could not measure 117 particles (28.8%) via ATR-FTIR because of their size $< 300 \,\mu\text{m}$. In each sample, 0 to 20 particles (average: 2.64 particles, SD: 3.49) were found; while overall concentrations ranged from 0 p kg⁻¹ up to a maximum of 35.82 p kg^{-1} , with an average of 3.23 p kg^{-1} (± 1.75 p kg^{-1} RSD, n=100). Samples containing no detectable plastic particles occur mainly at depths >75 cm, whereas higher concentrations can be found in the upper 30 cm of each soil column (Table 1).

The extracted and identified plastic particles appeared as films (45.8%) and fragments (38.3%), or as filaments, pellets and foams (Fig. 2b), with weathered (49.0%), fresh (28%) or incipient alteration (23.0%) surface structures. The shape composition consists of irregular (71.0%), regular (25.0%) or rounded (4.0%) shapes; particle colour is often transparent or white (49.0%), or bright red (16.0%), blue (9.0%) or pink (8.0%), followed by different colours with an individual share \leq 6.0% (e.g., black, orange, yellow).

Polymer type composition is dominated by low- and high-density polyethylene (LDPE and HDPE) making up 46.0%, followed by polypropylene (PP, 10.0%), rubbers (9.0%), chlorinated or chlorosulfonated polyethylene (CPE, CSM, 7.0%) and polyethylene terephthalate (PET, 6.0%) (Fig. 2a). All identified polymers show a density of \leq 1.2 g cm³ in pure form, except PET with 1.37 g cm³ [61], which was thus only recorded semi-quantitatively based on the density separation method with NaCl.

Plastic particles in the coarse soil fraction (>2 mm) occur in a size range between 2.1 mm and 52.0 mm, with an average of 20.68 mm (Fig. 2c), while in the fine soil fraction (<2 mm) particle size ranges between $171.0 \,\mu$ m and $1680 \,\mu$ m with an average of $598.6 \,\mu$ m and a clear accumulation of outliers over $1000 \,\mu$ m (Fig. 2c).

Additionally, sampling of plastics on the soil surface (site OKA) shows an occurrence of 1 p m² (OKA-3) to 1.05 p m² (OKA-2). The macroplastics collected occur as films (54.8%), fragments (38.7%) or styrofoam (6.5%), with mainly irregular and weathered surfaces and an average size of 66.3 mm (Table S3). The plastics consist of HDPE (35.5%), LDPE (16.1%), PP (12.9%) and other polymers like PET, polyvinyl chloride (PVC), polystyrene (PS) or polymethyl methacrylate (PMMA). In some cases, the original function of plastic items is still identifiable, for example, DIY store shed for flowers, plastic fork, bottle cap or food wraps (Fig. S6).

Metal concentrations and soil properties

All of the 10 trace metals analysed, as well as the metalloid As, were detectable by ICP-MS measthe urement with contents above detection limit. In relation to the mean values (n=100), the content decreases from Fe $(21,026.3 \,\mathrm{mg \, kg^{-1}}) > Zn$ (55.4 mgkg^{-1}) > Ni $(31.17 \text{ mgkg}^{-1})$ > Cr $(29.82 \text{ mgkg}^{-1})$ > Pb $(23.78 \text{ mg kg}^{-1})$ >Cu $(16.58 \text{ mg kg}^{-1})$ >V $(16.30 \text{ mg kg}^{-1})$ >Co $(10.93 \text{ mgkg}^{-1})$ > As (6.63 mgkg^{-1}) > Cd (0.28 mgkg^{-1}) to Hg $(0.11 \,\mathrm{mg \, kg^{-1}})$. A comparison with geogenic background and legislation values is possible for Cr, Ni, Cu, Zn, Cd, Hg

Table 1 Plastic loads (p kg⁻¹) in soil samples (for plastics size range 171 μ m – 52 mm)

Sampling depth (cm)	Plastic loads (p kg ⁻¹)												
	Transect NID		Transect MOK			Transec	t OKA	Transect FRA					
	1	2	1	2	3	1	2	3	2	1			
0-10	2.92	15.14	11.78	0.00	1.10	4.77	35.82	11.44	7.76	5.80			
10–20	4.58	3.61	13.86	0.00	0.00	2.48	19.39	4.93	1.40	4.45			
20–30	27.25	4.18	1.26	0.00	2.61	5.16	2.76	2.07	3.98	5.43			
30–40	2.22	0.71	3.47	2.27	1.94	2.18	2.57	1.81	2.20	21.27			
40-50	0.00	12.00	2.32	1.65	1.87	0.00	1.49	1.92	5.83	0.00			
50–75	0.55	0.43	0.14	1.04	1.34	0.44	0.00	0.00	1.71	2.36			
75–100	1.01	1.57	2.27	0.00	0.00	0.48	0.77	0.00	2.04	1.40			
100-125	0.00	0.00	0.00	5.60	5.18	0.00	1.66	0.00	2.09	0.63			
125-150	0.70	0.00	5.11	0.00	3.76	1.80	0.00	0.00	1.45	0.59			
150-200	0.00	1.59	1.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00			



example of a blue PP film and pink PET fragment (Polymer type abbreviations explained in Table S4); **b** Particle type composition (n = 228); **c** Particle size composition for particles in coarse soil fraction (> 2 mm) (upper boxplot, n = 35) and fine soil fraction (< 2 mm) (lower boxplot, n = 193) with first and third quartile represented by outer box border

and Pb. Mean and median values fall below the worldwide average contents of surface horizons [62], but exceed the geogenic background values for Hessian floodplain soils in the case of Cr, Ni, Cu, Cd and Hg. Legislative precautionary values are exceeded by all metals except Hg by the respective maxima (Table 2); these levels occur in individual samples at sites FRA-1, MOK-1 to MOK-3, NID-2 and OKA-3 at varying depths between 20 and 200 cm.

With regard to the calculated pollution indices, the PLI ranges between 0.4 and 3.2, with a mean of 1.1, just above the limit value of 1 at which baseline levels of pollution begin. The PLI of maximum concentration values of 3.2 indicates a partial deterioration of soil quality [56]. The

values of EF are also comparable, with a mean just above the limit where a moderate enrichment can be assumed, and maximum values indicating a significant contaminant enrichment.

Soil conditions for trace metal behaviour in the floodplain soils investigated consist of predominantly silty to clayey soils (soil textures: SiCL, SiC, C), OM contents range from 1.57 wt% up to 24.91 wt% with an average of 8.33 wt% and pH values indicate a very weak acidic environment with a total average of 6.33 (range 4.90–7.69, moderate acid to weak alkaline) (Table S1). Furthermore, in groundwater affected Fluvisols or Gleysols, pedogenic oxides (Fe, Mn) in Bl-horizons as well as reductive **Table 2** Summary of trace metal contents compared to geogenic background levels and legislation values with calculated pollution indices and their thresholds

		Elemental concentrations							Indices		
		Cr	Ni	Cu	Zn	Cd	Hg	Pb		PLI ^a	EF ^b
		mg kg ⁻¹							(—)	(—)	
Mean		29.8	31.2	16.6	55.4	0.28	0.11	23.8		1.1	2.2
Median		26.3	27.6	13.5	40.0	0.16	0.07	17.7		0.9	1.8
Min.		9.8	8.7	3.5	17.0	0.05	0.01	8.3		0.4	1.1
Max.		75.4	100.7	55.8	194.5	1.39	0.46	77.1		3.2	5.9
Mean upper soil	0–50 cm	28.6	29.9	15.1	50.1	0.25	0.09	22.9		1.0	2.1
Mean lower soil	50–200 cm	31.1	32.5	18.1	60.8	0.31	0.13	24.6		1.2	2.4
Wilcox <i>p</i> -value	upper vs lower	0.52	0.55	0.21	0.34	0.54	0.22	0.73		0.45	0.37
SHW c	Average content surface horizons worldwide	60.0	29.0	38.9		0.41		27.0	perfection low ^f	< 1	<2
GBHF ^d	Geochemical background in Hessian flood- plain soils (0–2 m)	25.0	24.0	13.3	58.5	0.14	0.04	27.0	baseline moderate ^f	1	2–5
PV ^e	Precautionary values	60.0	50.0	40.0	150.0	1.00	0.50	70.0	deterioration significant ^f	>1	>5

^a Pollution Load Index; ^b Enrichment factor; ^c Kabata-Pendias [62]; ^d Friedrich & Lügger [57]; ^e German Federal Soil Protection Ordinance - BBodSchV [63]; ^f Pollution assessment according to Kowalska et al. [56] with "denote perfection" (PLI) or "deficiency to minimal enrichment" (EF), "baseline level of pollution" (PLI) or "moderate enrichment" (EF) and "deterioration of soil quality" (PLI) or "significant enrichment" (EF)

conditions occur (Table S1). Deviations in soil texture occur only in subsoils (>100 cm), where the sand content increases locally, and at the middle and lower reaches, with average values up to 22.0 wt% (OKA-1) or 30.5 wt% (FRA-2). Similarly, organic enrichment occurs through deep peat bands or layers with OM contents of 16.2 wt% (NID-2, >125 cm) or 24.9 wt% (MOK-1, >100 cm).

Spatial distribution of plastics and metals Contaminants along the river course and floodplain-cross transects

Both contaminants under consideration are present within all floodplain soils along the Nidda River course. Plastic loads occur with average values between 2.34-3.92 p kg⁻¹ and a variation of ± 1.58 p kg⁻¹ with no significant (p = 0.5106) mean differences along the river course (Fig. 3). Highest average values occur at site NID (upper middle reaches) with 3.92 p kg^{-1} and upper middle reaches and lower reaches contain the highest plastic contamination levels with plastic sums per sampling site around 39.24 p kg-1 (NID), 41.87 p kg-1 (MOK) and 41.92 p kg-1 (FRA). Regarding the metal loads, we found a slight increase of average EF values per site from NID (upper middle reaches) at 2.06, MOK (middle reaches) at 2.42 and OKA (upper lower reaches) at 2.44, and then decrease to 1.97 at site FRA (lower reaches), with no significant mean differences between sites (p = 0.2208). PLI shows average PLI values per site <1 at sites MOK or FRA, and PLI > 1 at sites NID or OKA (Table S2).

Plastics within the river cross-transects show no significant (p = 0.6916) mean differences between proximal,

central and distal floodplains. Slightly higher median values occur within distal (remote-channel) floodplains along the course, while higher values at central site OKA are dominated by accumulation of plastics in the arable topsoils (Fig. 3a) (Tables S1 and S2). While macro- and mesoplastic particles only occur isolated within topsoils (plough horizons) of those OKA sites, a general lateral sorting of plastic sizes, depending on the distance to the channel, could not be determined (Fig. S8). Also, for metals we found slightly higher median values within central and distal floodplains, but comparable to plastic loads without significant differences between sites (Fig. 3b).

Vertical contaminant patterns

In contrast to lateral contaminant distributions, contaminant loads and soil properties show clearer vertical patterns across the vertical spatial scale. First, the floodplain soils investigated show typical texture and SOM variations along the sampled depth gradient (Fig. 4). Accumulation of SOM is thereby related to topsoils (A-horizons) as well as thin peat bands in deeper soil layers. Soil texture differences are mainly expressed through low (<25%) sand contents in topsoils and upper soil layers (>50 cm) with alternating clay contents in the upper floodplain loams (Fig. 4). While in upper soil layers clays (C), silty clays (SiC) and silty clay loams (SiCL) prevail, deeper soil layers show primarily silty loams (SiL), loams (L) or sandy loams (SL), and loamy sands (LS) (Fig. S10).

Plastics could be determined over the entire depth sampled, but with a clear increase in upper (<50 cm) soil



layers, while metal concentrations expressed through the PLI show stronger variations across the depth gradient with different maxima on different depth positions (Fig. 4). The individual metals analysed show a variable distribution over depth, but with insignificant mean differences between deeper soil (50-200 cm) and upper soil (0-50 cm) layers (Table 2). Slightly higher mean concentrations occur for Cr, Ni, Cu, Zn, Cd, Hg and Pb within lower soil layers (>50 cm) related to the respective maxima (Table 2). A combined consideration of the metals using the PLI shows a baseline level of contamination within the entire soil column with individual peaks at proximal floodplains at sites NID (upper middle reach), OKA (middle reaches) and FRA (lower reaches) in depths below 50 cm (Fig. 4). Vertical mean differences of EF and PLI are not significant (p > 0.05).

Clearly significant differences occur in the vertical plastic load distribution (Figs. 4 and 5). While the upper soil layers (0–50 cm), consisting of topsoil A-horizons as well as upper B-horizons, show an average plastic load of 6.36 p kg $^{-1}$, the subsoil layers, consisting of B-horizons (50– 200 cm), have a significantly lower mean value of 0.99 p kg^{-1} (*p* < 0.01). The vertical distribution of plastic loads shows a clear maximum in the uppermost sampling layer (0-10 cm), mostly consistent with A-horizon boundaries, with an average of 9.65 p kg^{-1} continuously decreasing to a depth of 40–50 cm, with an average of 2.71 p kg⁻ and even lower average values ranging between 1.52 p kg^{-1} and 0.33 p kg^{-1} below that (Fig. 5d). This vertical decrease is comparable to the decrease in mean organic matter content (with the exception of layers deeper than 100 cm which contain peat), and the increase in mean sand content from a depth of 50 cm (Fig. 4). Conspicuous accumulation in the vertical distribution occur at sampling site NID-1 (27.25 p kg $^{-1}$ at 20–30 cm), MOK-1 (11.78 p kg⁻¹ to 13.86 p kg⁻¹ at 0–20 cm), OKA-2 (35.82

(See figure on next page.)

Fig. 4 Vertical distribution of multi-proxy parameters within investigated floodplains soils including plastic loads (p kg⁻¹), PLI, sand and clay content (%) as well as soil organic matter (SOM) content (%) grouped according floodplain positions (proximal, central, distal). **a** Sampling site NID (upper middle reaches) at river km 62; **b** Sampling site MOK (middle reaches) at river km 55; **c** Sampling site OKA (lower middle reaches) at river km 35; **d** Sampling site FRA (lower reaches) at river km 2





depth gradient (n = 10 per depth section), e rice

p kg⁻¹ to 19.39 p kg⁻¹ at 0–20 cm) and FRA-1 (21.27 p kg⁻¹ at 30–40 cm) (Fig. S9). Summarized, the vertical spatial patterns of the contaminants considered, can be summarized with a clear accumulation of plastics within upper (< 50 cm) soil layers and metal contamination peaks within 30–40 cm, 75–100 cm and only for EF within 150–200 cm depth sections (Fig. 5d–f).

Comparing the vertical spatial patterns of the contaminants with soil properties, a statistical correlation of plastics or individual metals with soil textures could not be proven (p>0.63 for each texture fraction), although the sand content increases from the upper to the lower course while the clay content decreases (Table S2). Regarding potential plastic adsorption processes on clay minerals or micro- to macroaggregates, a comparison of mean plastic contents with individual clay shares shows no significant (p=0.3845) correlations. However, the influence of soil depth on plastic loads is supported by a significantly ($p^{<}$ 0.05) weak negative correlation (r_{SP} =-0.56). Furthermore, plastic loads show slight negative correlations with EF (r_{SP} =-0.19) and PLI values (r_{SP} =-0.12). Slight to weak positive correlations (r_{SP} 0.2-0.6) occur between the river course (km) and distance to channel (m) with clay and OM content, as well as slight negative correlations with sand content. Notable correlations between clay or OM and metals, indicating a strong absorbance to clay minerals or humic substances, could not be found. Inter-element correlations show clear to high positive correlations (r_{SP} 0.6–1.0), except for lower

correlation coefficients (r_{SP} ⁶0.4) for Cd, Hg, Pb with V, Cr, Fe, Co, Ni and partly As (Fig. S11).

Furthermore, a vertical differentiation could be found for plastic particle characteristics. Figure 6a indicates that particles with a size >2000 μ m (coarse microplastic border) consist mainly of fragments and films, whereas smaller particles show a heterogenous distribution related to particle type, particle size and soil depth (Fig. 6).

Deepest soil layers are reached by filaments, fragments and films: the share of particle types shows comparable values of films (46.43%) and higher values of fragments (41.04%) in lower soil layers than in upper layers (films: 45.40%, fragments 34.48%). Overall, the particle size ranged from 171 µm to 52,000 µm (average: 4566.49 µm) in upper soil layers (0-50 cm), and smaller average sizes of 512.15 µm (242-2700 µm) in lower (50-200 cm) layers. The gap of plastics between the 2–10 mm range could be traced back to the release of microplastic particles into topsoils of two sampled agricultural fields (Fig. 6). The share of particle surface characteristics shows an increase for weathered particles with depth, from 45.98% in upper (0-50 cm) to 62.5% in lower (50-200 cm) soil layers, at the cost of fresh and incipient alteration particles. EPO ages range in upper and lower layers from 1820 to 1990, with an average occurrence of 1938 in upper (0-50 cm), and 1941 in lower (50-200 cm) layers, indicating no significant polymer age differentiation. The vertical distribution of EPO ages (Fig. 6b) shows no clustering of polymers of the same age at certain soil depths. There is no dominant polymer type in the deep soil layers, and only two young (>1990) chlorosulfonated or chlorinated polyethylene (CSM/CPE) polymers occur. In general, the depth distribution of the youngest CSM/CPE polymer group shows an average depth of 23.5 cm and an enrichment in soil layers between 0 and 35 cm (third quartile of CSM/CPE depths, n = 24).

Discussion

Plastic abundance

The observed plastic content, ranging from 0 p kg⁻¹ up to a maximum of 35.82 p kg⁻¹, is in the lower range of previously determined plastic contents in floodplain soils. Even if plastic particle abundances recorded for different locations with different methods are difficult to compare, our results can be viewed in relation to other results using comparable methods and soils (floodplain soils). For example, Christensen et al. [20] found plastic loads of 23.0-330.0 p kg⁻¹ for particle sizes of 280-1160 µm (NaCl solution) in three river floodplains in Virginia (US), whereas Lechthaler et al. [18] document average loads of 25.4–47.9 p kg⁻¹ for plastics with a size of 500-5000 µm (canola oil separation) in the Inde River floodplain (Germany). Both studies investigated near-channel depositions (bank profiles, levee situations), with a focus on topsoils and single depth profiles. Further investigations, based on the same geospatial sampling approach as in the present study, and



conducted in the more rural river system of the Lahn River (Hesse, Germany), found loads of 0.62–5.37 p kg⁻¹ for mesoplastics (>5 mm) and 0.31–8.59 p kg⁻¹ for large microplastics (2–5 mm) based on sieving [10], as well as 0.36–30.46 p kg⁻¹ for microplastics sized 219.0–8321.0 μ m based on NaCl separation of the fine soil fraction (<2 mm) [22]. The average value of all samples from the Nidda River catchment at 3.23 p kg⁻¹ is comparable to the average values of 2.06 p kg⁻¹ (mesoplastics), 1.88 p kg⁻¹ (coarse microplastics) or 2.75 p kg⁻¹ for microplastics from the Lahn catchment [10, 22].

With regard to intensive agriculturally utilised floodplain soils of the lower Yangtze River floodplain and the lower particle size range investigated, the average of 37.32 p kg⁻¹ (100–500 μ m) in 0–80 cm soil depth clearly exceeds the Nidda River catchment average [19]. In comparison to further studies which examine plastic contents in agricultural soils, it becomes clear that these clearly exceed the values from the Nidda River floodplain [64–66]. This could indicate the role of intensive agriculture in the contribution of plastic inputs. In the Nidda catchment, plastic enrichment through agriculture is also probable, as for example only 5.5% of farms work with organic agriculture. The maximum plastic load (35.83 p kg^{-1}) and higher values, especially for meso- and macroplastic contents, occur where plastic accumulation is also visible at the surface (site OKA, agricultural field). Plastic particles collected on soil surfaces at site OKA (Fig. S6; Table S3) could be partially identified as parts of consumer articles. The identification of consumer articles may also indicate local littering as a potential source. At this sampling site, the occurrence of plastics on soil surfaces at 1.0-1.05 p m⁻² is clearly above the value of 0.021 p m^{-2} (206 p ha⁻¹) reported by Piehl et al. [42] for microplastic particles on an agricultural farmland in Germany. Nevertheless, the comparability against different studies conducted on agricultural soils is limited, especially because different separation solutions are used, and different particle sizes are considered. Higher plastic contents in agricultural soils could therefore also be caused by the consideration of particles < 300 µm in other studies.

With a view to the entire aquatic-terrestrial interface, it seems until now, that floodplain soils could contain lower plastic loads than river sediments in the active channel (riverbed, shore). For example, shore sediments of the Main River contain plastic loads of 786.0–1368.0 p kg⁻¹ (63–5000 µm) and values >50 p kg⁻¹ for particles > 200 µm directly before and after the inflow of the Nidda River [67]. Even in more rural areas, such as the Tisza River (eastern central Europe) contents reach values of 3808 ± 1605 p kg⁻¹ (90–5000 µm, zinc chloride solution) already in the upper reaches [12]. Both examples exceed

the plastic loads in the Nidda River floodplain by a multiple factor which, however, may be traceable in part to the examination of smaller particles and different spatial representativeness in the comparative studies. Up to now, only the results of Christensen et al. [20], based on a cross-transect sampling from river sediments to floodplains, suggest equal or slightly increased levels in floodplain samples instead of channel samples, while larger plastic particles occur in floodplain deposits.

Plastic particle characteristics found in floodplain soils of the Nidda River are comparable to other findings from floodplain soils, as well as river sediments. Films and fragments prevail, followed by filaments and pellets, with a typical distribution for soils, with the exception that filaments are sometimes dominant in other studies [20, 64]. Most of the particles show a weathered or incipient alteration of surface structure, indicating prolonged exposure to degradation factors (e.g., physical break, UVlight) [52, 68, 69]. The increase in the number of particles with smaller particle size also is a typical distribution independent of the environmental media studied [70]. Due to the method used, particles $< 300 \,\mu m$ could only be detected semi-quantitatively here if their size and degradation state allowed a manual ATR-FTIR measurement. Therefore, it can be assumed that plastic loads would still increase using a lower detection limit, as in studies quantifying particles $< 300 \,\mu m$ [19].

The dominant polymer types found correspond to those of commonly produced and used polymers in Europe (top 10 ranking), like polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET) or rubbers [71]. This polymer composition, resulting from the frequency of use in everyday life, industry, agriculture and infrastructure (e.g., rubber car tyre), is also found in most soil studies, with fluctuations around the most dominant polymer type [4]. Furthermore, a composition following the frequency of use is also found in channel bed sediments and seems to overlap in different river systems [4]. For river shore sediments of the Main River, Klein et al. [67] found a composition of PE, PP, PVC and dominant polystyrene (PS) which occupies only a small share <6% in Nidda River floodplain samples. Based on the density separation fluid used here, polymers with a density > 1.2 g cm³ can only be detected semi-quantitatively (e.g., PET). However, except for PET, and without taking additives into account, the common polymer types show a density < 1.2 g cm³. Considering the binding of plastic particles to and in soil aggregates, the question arises whether all particles could be separated [23, 66]. Although manual mortaring allows a gentle sample preparation, it does not dissolve soil microaggregates in which plastic might still be retained [72].

Besides the methodological limitations described, the comparison of plastic contents with other investigations results frequently in restrictions based on methodical differences. These differences arise from different sampling concepts, the sample quantity examined, size classes, and the different separation methods. It is therefore difficult to evaluate the plastic content against the background of a contamination assessment, as is usual for other pollutants. In the previous discussion, mainly studies on floodplain soils (mostly near bank and topsoil) which work by means of NaCl- or canola oil-based separation were consulted. Against the background of the comparisons made, it can be cautiously assumed that the contamination level of plastics over the entire soil depth of 2m is in the lower range of known contamination, also with regard to smaller particles.

Metal abundance

Trace metals and the metalloid As are present in the floodplain soils of the Nidda River catchment. Even if the average concentrations of Cr, Ni, Cu, Zn, Cd, Hg and Pb fall below the average contents of surface (topsoil) horizons worldwide [62], the exceeding of local geochemical background values of Cr, Ni, Cd and Hg indicates a contamination enrichment of those metals compared to other regional floodplains in Hesse (Germany) (Table 2) [57]. From a legal perspective, only individual breaches of the precautionary values require legal measures (e.g., further investigations, risk designation), traceable to the absolute maximum values and therefore single enriched samples [63]. A pronounced contamination from a single element was not detected.

Pollution indices, like the Enrichment factor (EF), or Pollution Load Index (PLI), enable an assessment of the possible anthropogenic impact on trace metal concentrations [56]. Both indices show average values just above the threshold values for moderate enrichment (EF; > 2) and baseline pollution (PLI; >1), with a significant enrichment for single samples with EF > 5. As both indices require a geochemical background value for calculation, the exceeding of the thresholds indicates a deviation of metal loads from the theoretical natural background variation [56, 73]. Elemental concentrations and pollution indices show a homogeneous distribution over depth with maxima in lower soil layers (>50 cm) (Table 1). This pattern could indicate older contamination in deeper floodplain sediments, or mobilised trace metals that reach deeper soil horizons through relocation processes [16, 73]. However, against the background of possible metal displacements, the soil properties indicate adsorption tendencies. Dominant silty to clayey, organic rich Fluvisols and Gleysols, provide good adsorption potentials on clay minerals, humic-substances and the formation of metal-humus-complexes [73, 74]. Furthermore, adsorbent pedogenic-oxides and reductive conditions in groundwater effected layers, with single peat layers in deeper soil sections, can increase the retention of metals [73–75]. Additionally, the very weak acid environment falls below the pH values for incipient mobilisation of Cd, Zn, Ni, but not for Cu, As, Cr, Pb and Hg, even in the minima [74, 75].

Due to spatially widespread moderate enrichment and baseline contamination by different metals under strong adsorption tendencies, anthropogenic impacts on metal enrichment can be assumed. Because of the slight vertical differences observed, this vertical pattern could also be due to an historical metal input to deeper soil layers and a more recent one in upper soil layers. Possible older sources of trace metals may include mining in the headwaters of the Nidda River (Vogelsberg mountains: iron ore and basalt mining), as well as early industrial metal processing throughout the river catchment [16]. Mining and metal industry represent one of the main sources of historical metal enrichment in river floodplains before, and especially during the Industrial Revolution from the 1850s onwards [76]. In contrast, recent sources could be related to wastewater treatment plants, industrial and traffic discharges as point sources, or uptake of polluted legacy sediments, as well as erosion on agricultural land [73, 77-79]. Former studies, assessing the ecological quality and ecotoxicological effects from channel sediments along the Nidda River, concluded that, other than the headwaters, the whole Nidda River is affected by anthropogenic chemical contamination (e.g., PAH, PCB, metals) [31, 35]. A relationship between anthropogenic point sources and ecotoxicological effects could not be proved, which leads to the assumption of diffuse sources for chemicals and consequently also for metal contamination [31, 35].

Spatial differences between plastic and metal contamination

Within our case study, we found that spatial contamination patterns of plastics and metals differ only along the vertical spatial extension. Along the lateral spatial extension, we found no significant contaminant levels along the river course (i) or the floodplain cross-sections (ii) (Figs. 3 and 4). However, both contaminants show significant differences along vertical depth profiles (iii) (Fig. 5).

Regarding contaminant levels along the river course (i) showing no enrichment of plastic loads or heavy metal enrichments along the river course (Table S2), the hypothesis that plastics accumulate with the river course cannot be supported. This finding is supports similar findings from other floodplains in Germany [18, 22]. Further catchment specific correlations, like the relation of plastic loads to the population densities in the respective river catchments [20, 21] or the higher abundance of plastics in rivers near urbanized areas [80] do not appear to be applicable on the Nidda River. In contrast we found already higher levels of plastics within the upper reaches of the Nidda River, which rises in rural landscapes. This finding is supported by the study of Kiss et al. [12], which found plastic enrichment in tributaries of the Tisza River, indicating a higher emission of plastics also in suburban and rural areas. Therefore, suburban to rural areas also seem to provide potential plastic and metal sources.

Spatial patterns on the floodplain cross-transect scale (ii) seem to result in no significant spatial differentiation of plastic loads, characteristics or relationships to soil properties (e.g., soil textures). Comparisons to other scientific studies are limited in this case, as until now only the work of of Weber and Opp [10], for coarse microplastics and mesoplastics, and the study of Weber et al. [22], for medium and large microplastics, have examined plastics across floodplain transversal sections. Both studies found a clear enrichment of plastic loads at near-channel (proximal) floodplain sites, interpreted as a consequence of frequent flood occurrence at levee situations, and easier plastic retention during floods, due to higher vegetation density. In contrast, plastic loads in the Nidda River floodplain seem to be much more homogeneously distributed over the floodplain area, and no differentiation based on flood dynamics and related sediment deposition occur. The slight but not significant increase of metal enrichment could be traced back to the clear association of metal loads with sediment particles, reaching the floodplain when flood water overflows the riverbank [81]. Following the diffusion mixing model, and water-flow velocity slowing with increasing distance from the channel, metals associated with finer sediment fractions show higher concentrations in floodplain zones behind the levee (central, distal) [81]. Assuming flood delivery to be the dominant source of plastics as well as metal contamination, it can be stated that transport and deposition of both contaminants by floodwater is conceivable. The spatial patterns of the metal distribution correspond partly to well-known distribution patterns, whereas the plastic distribution shows differences to previous findings.

Vertical spatial patterns (iii) of both contaminants, which are so far underrepresented especially within plastic case studies, can be considered as the clearest patterns with differences between the contaminants. Whereas plastic loads showing a clear distinction between upper (0-50 cm) and lower (50-200 cm) soil layers (Figs. 4 and 5), metal loads or pollution indices showing different enrichments within two or three depth positions below 40 cm. A plastic accumulation within uppermost topsoils, and an overall decrease in concentrations with

increasing depth, was also found by the few studies that have investigated different soil depths [10, 65], while the work of Cao et al. [19] documents renewed increases of concentration below 40 cm depth in agricultural soils. Metal contamination tendencies between 40 and 120 cm depth for PLI levels, show no significant mean differences between different soil sections caused by the high data variability. Assuming a more or less low mobility of metals, due to good adsorption conditions in the floodplain soils examined, this pattern can probably be attributed to deposition processes of metals bound to sediments [17, 81]. With regard to the assumption that fluvial processes lead to the deposition and accumulation of plastics as well as metals in floodplain soils, no further indicator, like a relationship to soil texture [18, 19], or a clear stratigraphic distinction, could be found. Nevertheless, other surface discharge pathways for microplastics, such as surface runoff on slopes, can be excluded for the studied floodplains, as all study sites have no direct connection to slopes (e.g., through roads). Beside agricultural activities contributing to plastic deposits, plastic can originate from the river and deposited via flood water since further sources, except for atmospheric deposition, are excludable.

The vertical spatial contaminant patterns could also reflect a temporal sequence of dynamic sedimentation and erosion processes, based on the vertical formation of floodplains. From this background, the question arises whether different time periods for the input of the two contaminants could be relevant? Although no clear temporal differentiation could be established by EPO ages, the increase in plastic loads between 50 and 30 cm, depending on the sampling point, suggests that deposition of plastic started at these depths, beginning in the 1950s at the earliest. Assuming plastics load increases as a general marker for sediment dating, the upper soil layers containing significant enriched plastic loads could have been deposited after the 1950s [58, 82]. The application as a general marker also works when plastic is shifted vertically (e.g., by bioturbation), as significant content increases can be seen in the upper soil section [58]. In contrast, and although the metal inputs have not stopped in recent times, deeper metal accumulations may indicate earlier impacts (e.g., mining, industry), with maximum accumulation since the 1850s, and before widespread environmental protection laws in the 1960s [73]. This assumption could be supported by the, in some cases, strong inter-element correlations (Fig. S11), indicating a combined metal contamination from similar long-term sources with the same origin and controlling factors [83, 84]. Even though the concentrations in deeper soil layers decrease significantly, the results from the Nidda River indicate that plastic particles can shift

vertically, as was also shown for larger particles by Weber and Opp [10], and smaller particles by Cao et al. [19] and Weber et al. [22]. The size distribution of plastic particles found here, with the occurrence of coarse microplastics (>2000 μ m) only in upper soil layers and a considerably smaller particle size average in deep layers, suggests that smaller particles can more easily reach deeper soil sections. This result is supported by the findings of Rehm et al. [23], showing that microplastic particles with a size of 53–100 μ m tend to be carried vertically in soils more than larger particles. Possible transport paths through the soil, depending on the particle size, are assumed to be the pore space (macropores) or preferential flow paths, but also transport by soil organisms (earthworms) [23, 85–87].

In general, it can be summarized that a spatial homogenous distribution, mainly independent from local site and soil features, exists for each contaminant. Despite spatial comparisons, possible interactions between plastics and metals could not be identified in this study. Although the spatial differences of both contaminants were studied, no correlations between plastic content and individual elements exist.

Anthropogenic activities might directly impact distribution of plastics

Despite the general spatial distribution of plastic loads in floodplain soils of the Nidda River, the question arises whether there are high accumulations of plastics per site (Fehler! Verweisquelle konnte nicht gefunden werden.), exclusively related to the vertical distribution. Trace metal concentrations and pollution indices show only minor outliers, which can usually be associated with individual deep-lying soil layers and single-element enrichments (Figs. 4 and 5). In contrast, four significant, vertical outliers of plastic loads could be found in the cores NID-1, MOK-1, OKA-2 and FRA-1. Thus, at each sampling site one soil column shows a concentration of 13.86-27.25 p kg⁻¹, exceeding the average value of the respective profile by 3.3 to 6.9 times (Fig. S9, Table S2). The first assumption that these enrichments are attributed to flood processes could not be confirmed, because none of the four sites has a special micro-morphology (e.g., flood channel, depression) (Fig. S1) [17, 88]. Also, an influence from outside the floodplain, such as slope erosion and surface flow [23] can be excluded, since either no lower slopes are present (sites: FRA, OKA) or these are separated by roads (sites: NID, MOK).

In contrast, a relationship is evident at the OKA site between macro- and mesoplastic accumulations on the soil surface of the agricultural fields at OKA-2 (distal) as well as OKA-3 (proximal) and the enrichment of microplastic loads ranging between $35.82-19.39 \text{ p kg}^{-1}$

(0-20 cm, OKA-2) or $11.44-4.93 \text{ p } \text{kg}^{-1}$ (10-20 cm, cm)OKA-3) in ploughed topsoil. The marked decrease of concentrations with depth could be due to compaction by tillage below the Ap horizons [10]. Even if it is obvious to seek the source for this enrichment in agriculture, local littering cannot be ruled out. As no plasticulture was carried out on the field, the findings of macro- and mesoplastic may indicate an application of a compost or sewage sludge (origin of some plastic pieces still recognisable) [89, 90]. However, according to information provided by the land tenant (anonymous for data protection reasons), only fertilization with manure has been carried out in recent years. Because this information is not verifiable, the entire range of possible agricultural plastic input, from fertilizer application to machine abrasion, or local littering cannot be retraced.

For the remaining enriched sites, the soil stratigraphy shows slight layer differences for the sites MOK-1 and FRA-1, but not in NID-1, and likewise without changes in soil texture. However, with consideration of land use changes (Figs. S2 and S3), it becomes clear that these sites were immediately adjacent to earthworks for construction purposes (bridge construction, site FRA-1) and floodplain and river restoration (sites NID-1 and MOK-1) during the 2000s and 2010s. The influence of restoration on microplastic loads in near-channel deposits, through a remobilisation of deposited plastics and a high and young sedimentary activity, was also found in the lower reaches of the Inde River (Germany) [18].

Therefore, direct anthropogenic impacts like tillage and fertilization, as well as earthworks for building or restoration purposes, might have an influence on the microplastic abundances found in this study.

Conclusion

With the results of our case study, we were able to demonstrate that there are detectable plastic and metal contaminations within the lower range of known MP loads and metal enrichments in a further typical floodplain soilscape in Central Europe. Regarding the hypothesis of this case study, we were able to identify increased contaminant presence already within upper stream sections within rural areas (hypothesis 1). Furthermore, vertical contaminant pattern indicates a sequence of younger plastic deposits and older metal deposits within the studied floodplain soilscapes, which allows to assume different time periods of deposition and maybe different mobilities within floodplain soilscapes (hypothesis 2). Additionally, direct short to long-term anthropogenic impacts (e.g., agriculture, tillage, earthworks) seems to impact the contaminants distribution patterns on local scales (hypothesis 3).

In summary, our case study illustrates the role of floodplain soilscapes as temporal sinks for plastics and metals as well as the role of catchment-specific or site-specific properties like land use and site history on plastic and metal distribution. Based on our results we can conclude that within floodplain soilscapes, agriculture and flood processes could be interpreted as emitting and spreading agents for plastics and metals, while earthworks and renaturation could be seen as redistributing agents especially for plastics. The alongside presence but different spatial patterns of both contaminants considered within this study allow the following recommendations for future research on plastics and metals within floodplains:

- 1. We recommend more efforts on the investigation of MPs and metals within river headwaters, their small-scale floodplain soilscapes and freshwater systems for a better understanding of contaminant discharge, recent contaminant sources and transports in those "remote" areas.
- 2. We recommend the detailed consideration of geobiochemical interactions between plastics, metals and the soil matrix within floodplain soilscapes, in order to understand possible risks from multiple contamination frameworks within floodplain ecosystems in future.
- 3. We recommend an enhanced practice of spatial sampling approaches including the analysis of subsoils and multiple contaminant sources or anthropogenic redistribution agents, to quantify different impacts on MP and metal distributions on different spatial scales.

Abbreviations

ATR: Attenuated Total Reflection; EF: Enrichment Factor; FRA : Frankfurt (location name); FTIR: Fourier-transform infrared spectroscopy; HQ: Flood; ICP-MS: Inductively Coupled Plasma - Mass Spectrometry; MOK: Mockstadt (location name); MP: Microplastic; MQ: Mean discharge; NID: Nidda (location name); NP: Nanoplastic; OKA : Okarben (location name); OM: Organic matter; PAHs: Polycyclic Aromatic Hydrocarbons; PLI: Pollution Load Index; Polymer abbreviations (e.g., PE): Listed in Table S4; WWTP: Waste Water Treatment Plant.

Supplementary Information

The online version contains supplementary material available at https://doi. org/10.1186/s43591-022-00044-0.

Additional file 1.

Acknowledgements

We acknowledge the support by all landowners who granted access to their land as well as the Wetteraukreis for a landscape protection area permit. Furthermore, we thank Alexander Santowski for his assistance during fieldwork. Finally, we thank the anonymous reviewers for their comments and improvements on the manuscript.

Authors' contributions

Conceptualisation, C.J.W. and C.O.; Methodology, C.J.W. and J.A.P.; Validation, C.J.W.; Formal Analyses, C.J.W.; Investigation, C.J.W.; Resources, C.J.W., C.O., M.K. and P.C.; Data Curation, C.J.W., Writing – Original Draft, C.J.W., Writing – Review & Editing, C.J.W., C.O., J.A.P. and M.K., Visualisation, C.J.W.; Supervision, C.O. and M.K.; Project Administration, C.J.W.; Funding Acquisition, C.J.W. and C.O. The author(s) read and approved the final manuscript.

Funding

Open Access funding enabled and organized by Projekt DEAL. The authors acknowledge the funding of this work by the Hessian Agency of Nature Conservation, Environment and Geology (Hesse, Germany), and PhD Scholarship from the Marburg University Research Academy (MARA) for C.J.W.

Availability of data and materials

The dataset generated and/or analysed during the current study as well as the exact sampling point positions are available in: Meso- and microplastic distribution and heavy metal contaminations in floodplains of the Nidda River (Germany). figshare. Dataset. https://doi.org/10.6084/m9.figshare.17714909.v2

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare that they have no competing interests.

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Received: 2 July 2022 Accepted: 12 October 2022 Published online: 16 November 2022

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