RESEARCH

Evaluating the effectiveness of the MicroPlastic Sediment Separator (MPSS)

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Abstract

Effective laboratory methods are a requirement to obtain accurate data on the contamination of the environment with microplastics. However, current methods often lack specification and validation of performance. The aim of this work was to provide the first evaluation of the effectiveness of the commercially available MicroPlastic Sediment Separator (MPSS) operated with sodium chloride. We performed density separation experiments with pristine plastic particles spiked into both commercial sand (experiment I) and natural river sediments (experiment II) and with environmental microplastics contained in natural river sediments (experiment III). The natural sediments were taken from the Lahn River in Germany, a medium-scale central European tributary. The pristine test particles were ground polypropylene fragments, sieved in three different size classes: larger than 1 mm, 0.5–1 mm, and 0.3–0.5 mm. In experiment I, the mean recovery rate was 97 % for particles of the largest size class (standard deviation $s = 6$ %; $n = 30$ per size class), but dropped to 75.33 % ($s = 21.29$ %) in the medium and to 54 % ($s = 25$ %) in the smallest size class. After density separation, 87 % of all unsuccessfully separated test particles were found at the inner walls of the MPSS. In experiment II, the recovery rate was not correlated with the particle size distribution of the natural river sediments. In experiment III, a subsequent second density separation step contributed on average 38 % ($s = 18$ %; $n = 5$) to the total number of extracted environmental microplastics. This study illuminates central aspects of a density separation and aims to contribute to quality improvements of recovery rate experiments and field studies for the generation of reliable data on microplastics in the environment.

Keywords Microplastics, River sediments, Density separation, Sodium chloride, Size fractionation, QA/QC, Method validation, Positive control, Recovery rate, Stokes' law

Introduction

Since the first report by Carpenter and Smith in 1972 [1], an interdisciplinary research community has detected microplastics, defined as plastic particles between 1 µm and 1000 µm [2, 3] or smaller than 5000 µm [4] (with other lower size limits being 20 µm [5] or 100 µm [6]), in waters [7], sediments [8], soils [9], the atmosphere [10, 11], on continents [12], and in oceans [13] all over the world. Numerous laboratory and field studies demonstrated the ingestion of microplastics by biota [14, 15] and humans [16–19]; however, potential impacts on organismic health and ecosystem functions are still subject to intense research [20]. Given that plastic production and associated emissions

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into the environment will likely continue to rise in the future [21], global management efforts are required.

Measures for the reduction of plastic emissions presuppose an accurate quantitative knowledge of particle numbers, characteristics, sources, sinks, and spatio-temporal distribution patterns. The quantification of microplastic particles consists of three main steps – sampling, sample preparation, and analysis [3]. For each step, different techniques have been proposed [22–24], depending on the type of environmental medium to be investigated [25]. Among different environmental media, sediments and soils tend to stand out as a temporary or final sink for microplastics [26, 27]. In samples from sediments, soils, and also sludge ('complex solid matrices' [28]), preparation is challenging given the high content of organic and inorganic natural particles microplastics have to be separated from. Before digestion of natural organic matter, the inorganic fraction, which accounts for the largest share of the sample matrix, has to be removed. It is this latter step, we focus on in our work.

Non-destructive approaches use physical differences between microplastics and minerals regarding density [29, 30], lipo- or oleophilicity [31, 32], hydrophobicity [33], and electrostatics [34]. Most studies use the so-called density separation [23, 28, 35, 36], which is 'gravitation-based' [37]. While the density of minerals ranges from 2.5 g cm^{-3} to 2.8 g cm^{-3} [38], the density of pure synthetic polymers ranges from 0.05 g cm^{-3} [39] to 2.3 g cm^{-3} [40]. However, in environmental microplastics, the density can be altered compared to pure polymers because of chemical additives, biofouling [41, 42], and aggregation with natural and synthetic particles [43]. When a solid sample is placed in a static fluid, both gravity and buoyancy force act on every particle. It depends on the density difference between a particle and the fluid, whether a particle sinks or ascends. Thus, the density of the separation fluid determines the spectrum of separable particles.

Numerous saline solutions were tested as separation fluid [36, 44, 45]; however, researchers choose a zinc chloride (density $\rho \approx 1.7$ g cm^{-3}) or a sodium chloride solution ($\rho \approx 1.2$ g cm^{-3}) most often [36]. Due to the relatively low density, the latter fails to effectively separate denser polymers like polyvinyl chloride (PVC; density of pure polymer: 1.16–1.58 g cm^{-3}), polyethylene terephthalate (PET; density of pure polymer: 1.37–1.45 g cm^{-3}) [46], or polyoxymethylene (POM; density of pure polymer: 1.41 g cm^{-3}), but is cheaper, readily usable, non-toxic, and more environmentally friendly than zinc chloride, and therefore frequently used [47] (more detailed reasoning in the Supplementary Information, 'Sodium chloride as separation fluid' section).

Besides the choice of the separation fluid, studies vary by several aspects, such as the separation vessel, the agitation of the matrix, the duration of the separation process, and the way of removing floating microplastics from the vessel [48]. This methodological diversity is further evolving as long as there is no standardization of methodology and operation protocols with quality assurance / quality control (QA/QC) methods, and hampers the comparability of data between studies. At the same time, standardization efforts are even more complicated when methods are ineffective or not sufficiently validated. A recent review finds that most studies investigating microplastics in soils are, among other critical points, deficient in 'positive control and method validation' [49]. As a result, meta-studies, which are necessary to compile and critically analyse the current knowledge about microplastics in the soils and sediments, are scarce.

The first commercially available device to perform a density separation of microplastics from sediment or soil samples is the 'MicroPlastic Sediment Separator' (MPSS; Hydro-Bios Apparatebau GmbH, Altenholz, Germany; published by Imhof et al. in 2012 as 'Munich Plastic Sediment Separator' [29]). The large multi-component steel vessel processes several litres of sediments per run, but requires 30 litres of separation fluid, boosting the costs of operation and – depending on the type of salt – hampering the environmental friendliness. An advantage of the MPSS, and of the Sediment–Microplastic Isolation (SMI) unit [50], is the ball valve technology to reliably collect the floating particles from the fluid's surface, while the classical way of decanting floating particles in other setups is associated with problems of sediment turbulence and particle adhesion [29, 50–52]. While Imhof et al. demonstrated the MPSS to be highly effective [29], a later re-evaluation by Zobkov and Esiukova (2017) reproduced the high recovery rates only partially [53]. This contradiction between the two studies about the effectiveness of the MPSS can be explained by the lack of systematic validation experiments in terms of test particle size, separation fluid and duration, as well as the number of replications. For instance, the number of replications was only three in [29], which challenges the precision of their results. In [53], the test particle size was only about 1 mm. Furthermore, the separation effectiveness of the MPSS operated with sodium chloride to reduce costs and environmental hazard and improve operation convenience, which is in line with principles of green analytical chemistry [54], has not yet been tested.

The aim of this study was thus to validate the commercially available MPSS operated with sodium chloride as separation fluid, taking into account *precision* and *trueness* according to ISO 5725-1:2023 [55]. We performed recovery rate experiments with test particles of different

sizes larger than 0.3 mm in artificial (experiment I) and environmental samples (experiment II), tested whether a repeated density separation would improve the separation of environmental microplastics from natural fluvial sediments (experiment III), and explored the impact of sediment grain size distribution on the separation effectiveness of the MPSS (experiments II and III). Our results can help to improve techniques of sample preparation and method standardization, contributing to more accurate data on microplastics in the environment.

Methods

MicroPlastic Sediment Separator (MPSS)

The MPSS consists of a sediment container, two standpipes (bottom and top), and a sample chamber (Fig. 1). In the sediment container, the sample is stirred with a flat blade agitator to expose buoyant particles to the separation fluid. Particles less dense than the fluid are released from the matrix and ascend within the standpipes to the fluid surface in the sample chamber. The conical shape of the standpipes, whose diameters are continuously narrowed from the bottom to the top, enables a reduction of the sample volume of up to six litres in the sediment container to 95 ml in the sample chamber. The ball valve closes the sample chamber, encapsulating the floating particles. Further details on valves, materials, and dimensions of the MPSS are in the Supplementary Information ('MPSS' section).

Separation fluid

To prepare a sodium chloride solution, crystalline sodium chloride (about 19 kg; Poolsalz Alposal, Südwestdeutsche Salzwerke AG, Heilbronn, Germany) and tap

Fig. 1 Components of the MPSS. Photo by Hydro-Bios Apparatebau GmbH, modified

water (about 50 l) were mixed in a 60 l-barrel (blue, high-density polyethylene [PE-HD]; Mauser Packaging Solutions) with an electronic mixer designated for colours and mortar (Einhell Germany AG, Landau/Isar, Germany). The density of the solution was calculated as the quotient of mass (analytical balance ABJ 120-4NM, reading precision: 0.1 mg; Kern & Sohn GmbH, Balingen-Frommern, Germany) and volume of 5 ml (volumetric pipette; Brand GmbH & Co KG, Wertheim, Germany), and averaged over two subsamples. As soon as a density of 1.19 g cm^{-3} or higher was reached, the solution was pumped over a sieve to remove potential contamination (300 µm; funnel with inset filter, made by the mechanical workshop, Philipps-Universität Marburg) into the MPSS. In experiment III, a second sieve (100 µm) was added as additional prevention of contamination (lower particle size: 0.3 mm). After the separation, the solution was drained into the barrels for reuse.

Separation procedure

At the beginning of each run, the separation fluid was filled in the bottom standpipe to its medium fill height. The motor was started (9 to 10 rpm) and the sample added slowly through the opening of the bottom standpipe ('wet sediment loading' [53]). Afterwards, the top standpipe and the sample chamber were mounted. The sample chamber's dead volume in the ball valve had been primed before by filling it with separation fluid and closing the lever, to remove air inside the ball valve. In experiment I, it was closed twice; in experiments II and III only once, to reduce the contamination arising from shutting the ball valve (cf. Results, "Contamination and technical difficulties" section). The separation fluid was poured through the open sample chamber with a beaker to fill the MPSS completely. The chamber's opening was then closed with the top cannula. After a run time of one hour, the motor was turned off, and the MPSS left standing overnight for separation.

In experiment I, the separations lasted 15 hours. However, for a more convenient time schedule and a longer separation, we extended the duration to 20 hours in experiments II and III (cf. Table 1). This extra time potentially allowed more particles to arrive in the sample chamber. Thus, 15 or 20 hours after the motor had been started, the separation process was stopped by closing the ball valve. Both the side and aeration valve were opened to drain the separation fluid into the storage barrels. The sample chamber with the encapsulated microplastics was removed, turned by 180 °, hung in a custom-made holding (mechanical workshop, Philipps-Universität Marburg), opened to drain the sample into a glass flask with glass plug or a beaker, and carefully rinsed to transfer any particles sticking to
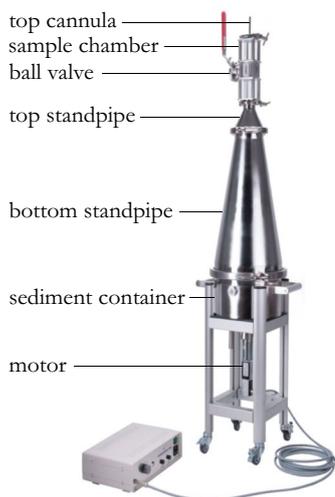
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Table 1 Parameters in the study design of the three experiments

	I	II	III
Matrix	commercial sand	fluvial sediments	fluvial sediments
Sampling location	–	Lahn River, Marburg	Lahn River, Gießen
Particle type	PP test particles	PP test particles	environ. microplastics
Particle size [larger than; mm]	0.3, 0.5, 1	0.3, 0.5	0.3, 0.5, 1, 5
Total particle no.	900	370	525
Particle no. per sample and size	10	5	–
Replicates	30	37	5
Separation duration [h]	15	20	20
Density separations per sample	one	one	two

the inner walls into the beaker. The beaker was covered with a glass dish or aluminium foil to safely store the sample until the next step. After the sample was taken, the MPSS was disassembled and the sediment removed from the sediment container. All components were cleaned with tap water and dried at room temperature.

Experimental design

In this study, we present three different experiments on the separation effectiveness of the MPSS (cf. Table 1). In the first two experiments, the separation effectiveness is measured by the *recovery rate*, defined as the quotient of successfully separated and originally added test particles. In experiment I, the impact of particle size on recovery rate was investigated by separating three different size classes (or *fractions*; lower limits: 1 mm, 0.5 mm, 0.3 mm; labelled *large*, *medium*, and *smallest* size class, respectively) from commercial sand. Commercial sand was chosen for unhindered particle release from the matrix into the inner separator. In experiment II, the separation of small and medium test particles from an environmental matrix was explored by using samples from the surface of near-shore sediments of the Lahn River in Marburg (Hesse, Germany). Finally, in experiment III, we tested to which extent the separation of real microplastics could be improved by a repeated density separation, using sediments from the Lahn River in Gießen.

A special feature in experiment I is the participation of members of the *Citizen Lab for Microplastics* in Marburg. The *Citizen Lab* was a citizen science project from 2018–2020, where interested citizens participated in the quantification of microplastics in environmental samples. In the present study, a team of four previously trained citizen scientists was successfully involved in running the first recovery rate experiment with the medium and large test particles.

Experiment I: recovery by size class
Test particles and density separation
Test particles were *signal blue* polypropylene (PP) fragments, incurring as post-consumer waste in the automotive industry (obtained from General-Industries Deutschland GmbH, Kassel, Germany; photos and FTIR spectrum in Supplementary Fig. S1). The thickness and length of the fragments were diverse; the colour was chosen to spot the particles easily and to distinguish them from potential contamination of the black and white plastic materials of the MPSS. The polymer type PP is not only theoretically separable in a saturated sodium chloride solution due to its lower density (density of pure polymer: 0.85 g cm^{-3} [56]; the least dense unfoamed commodity plastic), but also one of the most frequently produced polymer types worldwide [57], often found in natural habitats, and thus environmentally relevant.

The test particles were classified in three size classes by dry-sieving in a stainless-steel sieve cascade (diameter: 150 mm; VWR International GmbH, Darmstadt, Germany), namely larger than 1000 μm, 500–1000 μm, and 300–500 μm. From each fraction, ten particles were randomly picked with tweezers and mixed in a 2 l-glass beaker with about two litres of commercially obtained sand (Spielsand 25 kg; toom Baumarkt GmbH, Cologne, Germany; grain size distribution was analysed as described in the Supplementary Information, 'Grain size distribution analysis' section, data in Supplementary Table S1). In the case of the smallest size class, 30 MPSS runs with ten particles each were performed. The two larger size classes were combined: in each of the 30 MPSS runs, 20 particles were tested. Thus, in experiment I, the number of MPSS runs amounted to 60 and the number of values for *particle counts* and *recovery rate* to 90.

Our laboratory is equipped with three MPSS devices, which allowed us to perform three runs in parallel (control for a potential impact by device in Supplementary Fig. S4). The commercial sand was reused

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after each run. In the first six runs of the smallest size class, we checked if the sand still contained test particles from a previous run. To this end, the sand was left in the sediment container after the standpipes had been removed, and the motor started at 9 rpm for 60 minutes. After two hours of settling, the container and the fluid surface were visually inspected for test particles. Since never any particles were found, this test was not continued, and we concluded that all test particles were removed from the sand during density separation and no further treatment was necessary before the next run. Nevertheless, to account for a potential cross-contamination of particles between the runs, a further control was performed as detailed in 'Localization of test particles' section.

Detection and quantification

After density separation, the recovered test particles were counted. In the case of the large and medium size classes, the suspensions were wet-sieved and the particles counted by eye. They were clearly recognizable on the sieve meshes (cf. photo in the Supplementary Fig. S1). In the case of the smallest size class, the samples were thoroughly rinsed from the glass flask into a vacuum-filtration set-up (47/50 mm; Sartorius AG, Göttingen, Deutschland) and filtered on quartz fibre filters (pore size: 0.5 μm, MN QF-10; Macherey-Nagel GmbH & Co. KG, Düren, Deutschland). In the first runs, the filters were stained with Nile Red according to the protocol of [58] (50 mg cm^{-3}, N3013-100MG; Merck KGaA, Darmstadt, Deutschland; solved in ethanol and methanol [1:1]) to prevent overlooking of particles. The stained filters were excited with blue LED light (465 nm, LED465E; Thorlabs GmbH, Dachau, Deutschland) and observed through a long pass filter (532 nm) under a stereo microscope (WILD Heerbrugg AG). As the sediment matrix was reused, the samples became cleaner, probably because some finer material was washed out. Thus, staining became unnecessary, and the particles could be counted directly under the stereo microscope.

Localization of test particles

Since the measurand was determined by counting particles under a microscope, observation bias needed to be accounted for. Therefore, a separate step of quality control was implemented (in 49 of 60 samples; after the first eleven samples of the size fractions larger than 0.5 mm and 1 mm), where, after a separation run, the inner MPSS was carefully inspected for any unsuccessfully separated particles. More details can be found in the Supporting Information, 'Localization of test particles' section.

Experiment II: recovery from fluvial sediments
River sediment sampling

The aim of the second experiment was to explore the recovery of test particles in a real environmental matrix. Therefore, 37 Lahn River sediment samples were spiked with pristine test particles. The sediments originated from the Lahn River in the city of Marburg (Hesse, Germany). Samples were taken in 0.5 km intervals, alternately from both river sides, along an 18 km long river section, in May 2020 (first sampling location [upstream]: N 50°51.0', E 8°47.585'). Several litres of sediment per sample were taken from the surface of near-shore sediments with a Van Veen grab sampler (volume: 2 l, testable surface: 260 cm^2; Royal Eijkelkamp, Giesbeek, Netherlands), placed in pre-cleaned metal buckets with lids (5 l, tin-plate; Eimer-Welt.de, Hamburg, Germany), transported to the laboratory, and stored at room temperature until further processing. The analysis of the grain size distribution is described in the Supplementary Information, 'Grain size distribution analysis' section, while total organic carbon (TOC) analysis was out of scope here.

Sample preparation and density separation

Before density separation, the supernatant river water was decanted over a stainless-steel sieve (mesh size: 300 μm, diameter: 150 mm; VWR International GmbH, Darmstadt, Germany), and the sieve residue rinsed with saturated ($\rho \approx 1.2$ g cm^{-3}) and filtered (300 μm) sodium chloride solution into a 1 l-glass beaker. The sieved supernatant was later returned to the main sample through the bottom standpipe at the beginning of the separation process. The remaining sediment sample was placed in a metal bowl, covered with aluminium foil, and oven-dried for 24 h at 65 °C (BM 700; Memmert GmbH + Co. KG, Schwabach, Germany) to further remove river water from the sample. Since this short oven-drying did not appear to substantially remove any water, it was not conducted with the samples of experiment III. The dry mass of the sediment was determined gravimetrically as described in the Supporting Information, 'Dry sediment mass' section.

Every sample was spiked with five test particles of the size classes 0.5–1 mm and 0.3–0.5 mm, which were randomly chosen and picked with forceps. The test particles were from the same batch as in experiment I, excluding the size class larger than 1 mm because of its high recovery rate in experiment I. The separation was performed as described above; the separation time was 20 hours. When taking the encapsulated particles, the top standpipe was remounted onto the sample chamber hanging in the custom-made holding (Supplementary Fig. S2) to include the particles sticking to the top standpipe's wall

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into the beaker by thorough rinsing. Before disassembling the MPSS, the density of the separation fluid was determined at the top of the bottom standpipe to check a possible fluctuation after the separation.

Size fractionation and detection

After the density separation, the samples were fractionated in different size classes by wet-sieving over 1000 µm, 500 µm, and 300 µm (stainless-steel sieves, diameter: 7.5 cm; Atechnik GmbH, Leinburg, Germany). In the case of the first samples, the sieves were flipped by 180 ° and laid on a glass funnel with a small glass beaker (250 ml) below and rinsed thoroughly. The sample in the beaker was then vacuum-filtrated (50 mm filtration device with glass frit and polytetrafluoroethylene [PTFE] ring; Sartorius AG, Göttingen, Germany) onto a cellulose filter (LLG-Filter circles, qualitative, medium/fast, diameter: 47 mm; Lab Logistics Group GmbH, Meckenheim, Germany) and analysed under a stereo microscope (SMZ-171T; Motic Deutschland GmbH, Wetzlar, Germany). However, cellulose filters turned out to be unsuitable for microscopy since they are opaque and do not allow for transmitted light, which is necessary to reveal cellular structures and identify biogenic material (important in experiment III). Further, they tend to warp during drying. Searching for a better approach to transfer a sample fraction from the sieve to the microscope, we developed a two-step procedure [59]. The subsequent samples were processed according to this approach, where, in short, the sieve residue was concentrated via vacuum-filtration onto a cellulose filter and directly rinsed from the wet filter with a small amount of water into a glass petri dish. The water was evaporated in an oven at about 60 °C, and the dried sample in the petri dish examined microscopically with both reflected and transmitted light. During microscopy, test particles were differentiated from environmental microplastics contained in the river samples based on colour, shape, and environmental impurities (more details on this procedure and continuative thoughts on spiking environmental samples with test particles in the Supplementary Information, 'Spiking of environmental samples' section).

Experiment III: repeated density separation
Repeated density separation

To explore a way to improve the separation effectiveness of the MPSS, a series of two subsequent density separations was performed with real environmental samples (without the addition of test particles). For this purpose, both the top standpipe and the sample chamber were remounted on the bottom standpipe after the first density separation and re-filled with filtered sodium chloride solution. The motor was started again for one hour and

the floating particles taken after 20 hours. This test was conducted with five environmental samples (not previously analysed), where the amount of microplastics from the first and second density separation was compared. The samples were taken as described in 'River sediment sampling' section from two adjacent near-shore sediment bars of the Lahn River in the urban centre of Gießen (sampling location: N 50°35.183′, E 8°39.83′) in October 2021.

The sediment grain size distribution was measured as described in the Supplementary Information, 'Grain size distribution analysis' section, while TOC analysis was out of scope here. Before density separation, the supernatant river water was removed by oven-drying the samples at about 60 °C for several weeks. Up to an hour after filling the separation fluid into the bottom standpipe, large matrix components were removed with forceps from the fluid surface, followed by mounting the remaining MPSS components. In between, the opening of the bottom standpipe was covered with a clean metal lid to prevent airborne contamination. The size fractionation of the environmental samples was conducted as described in 'Size fractionation and detection' section, but with an additional 5 mm-sieve to eliminate possible environmental macroplastics.

Detection of microplastics

Putative microplastic particles from the first ($n = 308$) and second ($n = 217$) density separation were selected under a stereo microscope (SMZ-171T, equipped with six continuous zoom steps from 0.75x to 5x and an 1.5x objective; Motic Deutschland GmbH, Wetzlar, Germany) according to common visual sorting criteria [9, 51, 60], fixed on glass microscopic slides with pieces of double-sided adhesive tape (product ID: 57912-00000 and 64621-00000; tesa SE, Norderstedt, Germany), counted, and photographed (software: Motic Images Plus 3.0). The system was calibrated for size measurements with the calibration slide provided by Motic. Particle size was measured on the photos as the largest Euclidean distance between two points on the 2D area of the particle (maximum Feret diameter). Particle morphology was classified as fragment, foam, fibre, foil, or sphere.

FTIR measurements

To identify the polymer types of the putative microplastic particles, measurements were performed with attenuated total reflection (ATR) Fourier transform infrared (FTIR) spectroscopy. The overall 'spectroscopy assessment rate' [61] was 97.9 % (514 of 525 putative microplastics were measured). From the first density separations, 11 particles of the fraction 300–500 µm were not measured because they broke upon pressure with the forceps

($n = 2$) or were so thin ($n = 9$) that fragmentation upon contact with the ATR crystal and subsequent measurement of the underlying adhesive tape was likely. Nevertheless, they were classified as 'plastic' because their appearance was obviously synthetic and included in the calculation of particle concentrations. The rest of the particles were measured with the μATR FTIR spectrometer LUMOS II (Bruker Optics GmbH, Ettlingen, Germany), operated by the software OPUS (version 8.5.29). The spectral range was 4000–680 cm^{-1}, the resolution 4 cm^{-1}. The measuring conditions were thermo-electrically cooled mercury-cadmium-telluride (TE-MCT) single-element detector, low crystal pressure (material: germanium), open aperture, and 50 scans. The background was recorded with the same number of scans and repeatedly measured after usually not more than five sample measuring points. Each particle was measured at one or two points free of impurities. The resulting spectra were subjected to an atmospheric compensation which slightly smoothes the signal in the wave number regions of water (4000–3600 cm^{-1} and 1900–1800 cm^{-1}) and CO_2 (2400–2300 cm^{-1}). Apart from that, no further processing was conducted.

Identification
All spectra were visually assessed and an additional spectral library search was conducted in most cases. On the one hand, commercial libraries from Bruker were used (BPAD-Bruker Polymer ATR Library, ATR-FTIR LIBRARY KIMW, and BIBL ATR-FTIR FORENSICS Library), on the other hand, five own libraries were set up. These libraries contained spectra of (i) fresh reference standards, obtained from the *Polymer Kit 1.0* of the Center for Marine Debris Research (Hawaii Pacific University, USA), the Federal Institute for Materials Research and Testing (BAM, Berlin, Germany), and two German companies operating in the field of plastics' manufacturing and processing (Reinhard Bretthauer GmbH, Dillenburg, Germany, and Günther Heißkanaltechnik GmbH, Frankenberg [Eder], Germany); (ii) artificially weathered polymers obtained from the University of Bayreuth (details of the weathering procedure are provided in [62]); (iii) materials occurring in the two laboratories where this work was conducted, representing potential contamination; (iv) reliably identified environmental microplastics, obtained from Lahn River sediments in Marburg in a previous sampling; (v) natural biogenic matter (plants and invertebrates) also obtained from Lahn River sediments. In total, the libraries contained 54 entries of 28 different materials. The library spectra were recorded under the same conditions as the sample spectra. Spectral library search conditions were a standard algorithm, a maximum number of results of 30, and a minimum hit

quality index (HQI) of 1 (full scale: 1–1000). No minimum match score was applied for positive identification because, depending on the weathering state of the particle, the HQI calculated in OPUS turned out to be too variable.

Prevention of microplastic contamination
To prevent potential airborne contamination, white cotton laboratory coats were worn at all times and synthetic clothes avoided as much as possible. Samples were always covered. All components containing or contacting the samples were cleaned with filtered tap water (mesh size: 50 μm, diameter: 7.5 cm; Atechnik GmbH, Leinburg, Germany) or treated with pressurized air (e.g., the MPSS components, which were too large to wash them with filtered water). A limited number of plastic tools was used, and the two laboratories, which were exclusively utilized for the analysis of microplastics, were kept as clean and plastic-free as possible. More information on the plastic tools used and the blank runs applied can be found in the Supplementary Information, 'Prevention of contamination'.

Data analysis
The data were processed, analysed, and visualized in *R* [63]. For data processing and visualization, the packages *readxl* [64], *tidyverse* [65], and *scales* [66], and for descriptive statistics, *skimr* [67] were used. Spearman rank correlations and corresponding significance levels were calculated with *hmisc* [68], correlation matrices with *PerformanceAnalytics* [69]. The correlation coefficients (r_s) were interpreted according to [70].

Recovery rates were calculated as the quotient of the number of recovered and spiked particles, and expressed as percentages. Precision, assessed as random error (cf. [71]), was expressed as standard deviation s and coefficient of variation v (quotient of standard deviation and arithmetic mean $\bar{x}$: $v = \frac{s}{\bar{x}}$). Mean and standard deviation were rounded according to DIN 1333 1992-02 [72]. Trueness, assessed as systematic error (cf. [71]), was expressed as mean difference in recovery rates, called effect size, and estimated with *dabestr* [73], where the sampling distribution and the 95 % confidence interval (CI) are calculated based on 5000 bootstrap re-samples [74]. In experiment I, the large size class was chosen as reference group due to its generally high recovery rate [28, 29, 45]; in experiment II, the recovery rate from commercial sand of experiment I was used as reference for the recovery from fluvial sediments.

The microplastic content of the environmental samples (hereafter: concentration) was expressed as the number of particles per kilogram of dry sediment (p kg^{-1}). The mean sediment dry mass of the samples equalled

$\bar{x} = 0.73$ kg ($s = 0.16$ kg; $n = 5$). The sum of the concentrations resulting from the first and second density separation was considered as the total concentrations extracted from the samples.

To control for a potential impact by the reuse of sediments or any unknown systematic errors in experiment I, the recovery rate was plotted against the separation run number (Supplementary Fig. S3). The correlation between both variables was negligible and not significant for any size classes ($r_s = 0.13$ and $p = 0.505$, $r_s = 0.12$ and $p = 0.543$, $r_s = 0.25$ and $p = 0.191$; large, medium, and smallest size class, respectively). Further, a potential impact of the individual MPSS devices was analysed using the effect size estimation as outlined above. Although the recovery rates of MPSS 3 were higher than those of MPSS 1 and 2, the size of the effect was negligible (5.54 % [95 % CI: -6.56 and 19 %]; Supplementary Fig. S4).

In experiment II, a correlation matrix was calculated for the recovery rate, the fluvial grain size distribution (Marburg), and control variables of the density separation (fluid density before and after the separation, sample mass). No correlation between recovery rate and density before the separation ($r_s = 0.04$ and $p = 0.835$), density after the separation ($r_s = -0.24$ and $p = 0.157$), or sediment dry mass ($r_s = -0.02$ and $p = 0.922$) was found. In

experiment III, a correlation matrix was calculated for the microplastic concentrations (first run, second run, total) and the fluvial grain size distribution in Gießen (sand, clay, silt) (Supplementary Fig. S10).

Results

Experiment I: recovery by size class

Overall, 679 of 900 test particles were successfully recovered in the sample chamber (mean recovery rate $\bar{x} = 75$ %, standard deviation $s = 26$ %, coefficient of variance $v = 0.34$, $n = 30$). The mean recovery rate decreased with size class, along with the measurement precision (Supplementary Fig. S5 and Table S2) and trueness: The effect size between the medium and large size class was -21.7 % (95 % CI: -30.3 and -14.6 %) and -43.0 % (95 % CI: -52.0 and -34.3 %) between the smallest and large size class (Fig. 2). In the smallest size class, the distribution of the recovery rate exhibited a slight bimodal pattern (Supplementary Fig. S6).

To account for observation bias and understand the fate of the test particles, we analysed their location ($n = 871$) in the MPSS after the separation. Altogether, 97.2 % particles were found throughout the MPSS, mostly in the sample chamber (78 %). Many particles were attached to the inner wall of the bottom and top standpipe (10.1 % and 9.0 %, respectively; Supplementary Figs. S7 and S8),
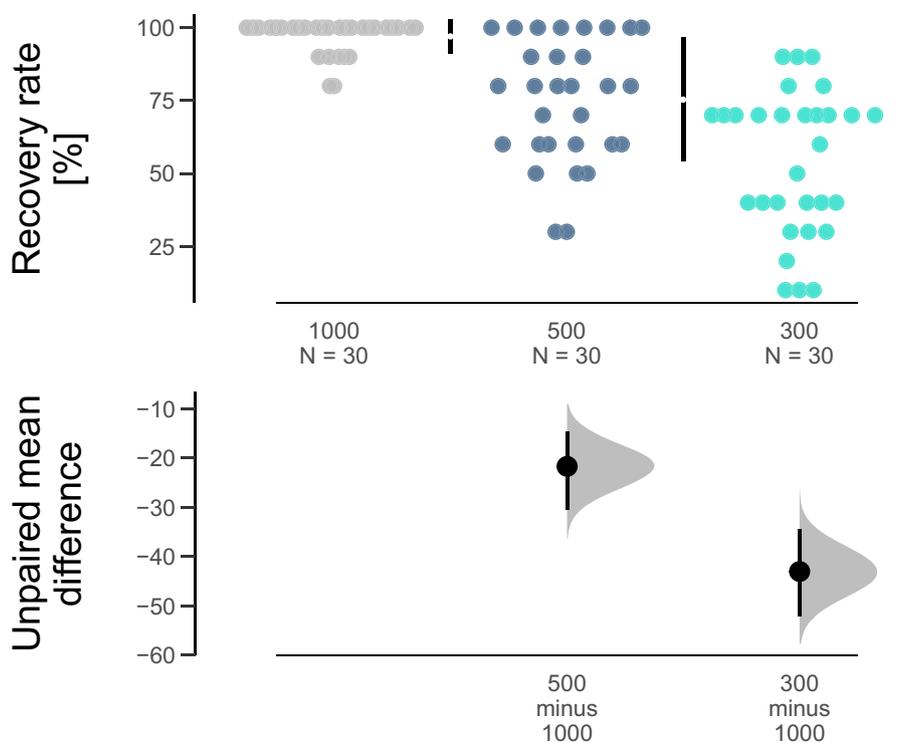

Fig. 2 Recovery of test particles (sample size: 10 particles, number of replications: 30) from commercial sand (experiment I)

equalling to 87 % of all unsuccessfully recovered particles. Each one particle of the smallest size fraction was found in the sediment container and the separation fluid. Among 24 missing particles, 6 were found in other samples, yielding a cross-contamination rate of 0.69 %. Only 18 particles (2 %) were overlooked or lost.

Experiment II: recovery from fluvial sediments

Overall, 308 of 370 test particles were recovered ($\bar{x}$ = 83.24 %, s = 22.24 %, v = 0.27, n = 37). The recovery rate did not correlate with the samples' grain size distribution, i.e., the contents of sand, silt, or clay (Supplementary Figs. S9 and S10). A comparison of the findings with the summarized recovery rate of the medium and smallest size class of experiment I (Supplementary Fig. S11) shows that the recovery from commercial sand was lower in average than from river sediment, but of similar variability (experiment I: $\bar{x}$ = 65 %, s = 16 %, v = 0.25). The effect size equalled 18.6 % (95 % CI: 8.94 and 27 %; Fig. 3).

Regarding particle size, as in experiment I, the recovery of the *medium* was higher than of the *smallest* size class (Supplementary Fig. S12). Although no test particles originated from the largest size fraction, 66 of 370 particles (17.8 %) were found here. Apparently, during size fractionation, some particles were caught in a sieve with a larger mesh size than in the preparation of the

spiked samples. Because of this, an analysis of size was not pursued.

Experiment III: repeated density separation

In all five river samples from the urban centre of Gießen, microplastics in a size range of 0.3–5 mm were found in both separation runs (absolute numbers in Supplementary Table S3, concentrations in Table 2). The most frequent polymer and morphological type were 'PE' and 'fragment', respectively (cf. Supplementary Information, 'Polymeric and morphological particle characteristics' section). The total concentration ranged from 95.9–213.5 particles kg^{-1}, to which the second separation contributed with 14.6–60.4 % ($\bar{x}$ = 38 %, s = 18 %, v = 0.46, n = 5) (Table 2). The mean microplastic concentration increased with decreasing size class in both runs. In all size classes, we found the largest share of particles in the first density separation. (Supplementary Fig. S13). Excluding the size classes larger than 1 and 5 mm reduces the mean share of the first run in total concentrations from 62 % to 50 % (s = 20 %, v = 0.39, n = 5), which is a much lower separation effectiveness than expected from the recovery rates of the small and medium size class in experiment I and II ($\bar{x}$ = 65 % and $\bar{x}$ = 83.24 %, respectively).

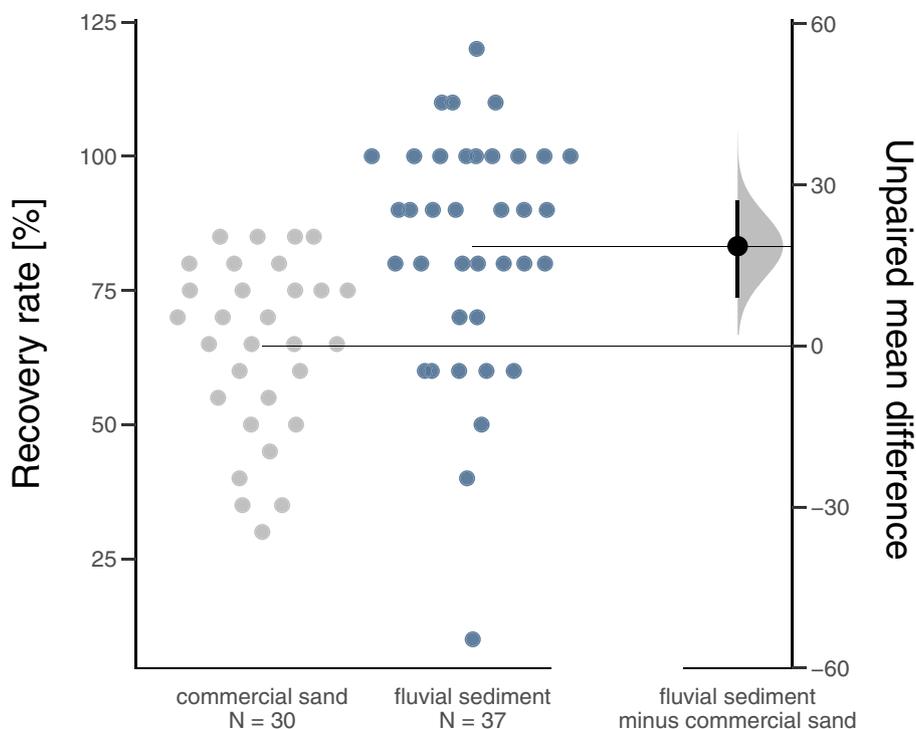The sediment grain size distribution (Supplementary Fig. S14) and the total concentration of microplastics

Fig. 3 Recovery of small and medium test particles from commercial sand (experiment I) and fluvial sediments (experiment II)

Table 2 Concentrations of microplastics (particle number per kilogram of dry sediment) obtained from two density separation runs of five Lahn River sediment samples each. Sdm. = Dry sediment mass

Sample ID	Sdm. [g]	Number of particles kg^{-1}			Percentage of particles	
		1st run	2nd run	Total	1st run	2nd run
1	795.6	94.3	52.8	147.1	64.1	35.9
3	487.2	147.8	65.7	213.5	69.2	30.8
7	928.0	48.5	47.4	95.9	50.6	49.4
8	732.0	95.6	16.4	112.0	85.4	14.6
9	690.7	57.9	88.3	146.2	39.6	60.4
rounded mean	730	89	54	140	62	38
rounded sd	160	40	26	50	18	18

were strongly correlated: sand ($r_s = -1.0$) was negatively correlated with the total concentration, silt ($r_s = 0.9$) and clay ($r_s = 0.82$) positively. However, only the correlation between the total concentration and content of sand ($p < 0.0001$) was significant, which may be attributed to the small sample size ($n = 5$). The correlation coefficients between the first and second density separation and the contents of sand, silt, and clay were low to moderate and not significant (Supplementary Fig. S15).

Contamination and technical difficulties

Numerous blank samples analysed in our laboratory over an MPSS operation life of four years consistently contained a specific type of plastic contamination, arising from the PTFE components in the sample chamber (ball valve, spacer disc, and sealing ring). Especially the PTFE parts in the ball valve are likely to cause thin PTFE particles with an abrasive shape (Supplementary Fig. S17), since the ball valve is closed and opened before the separation for priming, and closed at the end of the separation. By contrast, no contamination has ever been observed from the O-rings (EPDM).

Apart from the distinguishable PTFE contamination, the blank samples did not contain other plastic contamination in the particle range investigated (larger than 300 μm). Regularly, thin transparent fibres and, less frequently, blue fibres, were found, which probably originated from lab coats and blue jeans. Therefore, in the environmental samples of experiment III, such fibres were excluded from the analysis.

Finally, some minor technical difficulties are described in the Supplementary Information, 'Technical problems' section, and in Figs. S16 and S18.

Discussion

Comparison with MPSS literature

Our data confirm the high recovery rates of the first MPSS validation study by Imhof et al. [29] only partly.

Their methodology differed from ours regarding the MPSS (self-built prototype), the separation fluid (zinc chloride, $\rho = 1.7$ g cm^{-3}), the matrix (river sediment purified with zinc chloride and deionized water), the separation duration (1–2 h), the filling of the MPSS with separation fluid from the bottom valve after the sample has been entered, as well as the determination of recovery rates (large fraction: count-based, small fraction: gravimetric; commented in the Supplementary Information, 'Calculation of recovery rate in the literature' section). Their recovery rates were 100 % for particles from 2–5 mm and 95.5 % for particles from 40–309 μm. Although these size categories differ from ours, we can confirm the high recovery rates for test particles larger than 1 mm, but not for small particles. Due to the methodological differences mentioned, the comparison is limited.

Nevertheless, our challenge of the high effectiveness of the MPSS is in line with findings of the validation study by Zobkov and Esiukova [53] (self-built MPSS; separation fluid: equal amounts of zinc and calcium chloride, $\rho = 1.5$ g cm^{-3}; dry-loading; 30 min stirring at 120 rpm; about 20 h of separation). In their recovery experiment with PET sheets of about 1 mm edge length, the mean extraction efficiency was 97.1 % (CI: 2.6 %) – as in our large size class. However, in their test with environmental microplastics in marine sediments, Zobkov and Esiukova (2017) obtained a low recovery rate of 13–39 %, which they attributed to microplastic particle retention in the matrix. This is even lower than our extracted amount of environmental microplastics from fluvial sediments ($\bar{x} = 62$ %, $s = 18$ %, $n = 5$; share of the first run in total concentrations), which may be due to differences in methodology (e.g., the shorter stirring time in [53]), the sizes and shapes of the extracted microplastics, or the natural matrices. In sum, it can be assumed that the MPSS-based density separation

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caused an underestimation of microplastic loads in the existing field studies (Supplementary Table S4).

Effect of particle size on recovery

The key finding of this work is the effect of particle size on separation effectiveness (measurement precision and trueness). The recovery rate was excellent for particles larger than 1 mm, but dropped for smaller particles. As Stokes' law describes, the vertical velocity of a sphere in a fluid decreases with the diameter of the sphere. However, this phenomenon alone cannot explain our finding, since a separation time of 15 or 20 hours should be excess time for a vertical distance of one metre. According to Stokes' law, in 15 hours, a distance of one metre can be covered by a spherical PP particle with a diameter of 46 µm. Localizing the particles after the separations showed they were successfully released from the matrix, but stuck to the standpipes. Further, as the recovery from complex fluvial sediment was not impaired compared to that from commercial sand, a retention of particles in the matrix can be ruled out. Instead, we attribute the decline in recovery rate to particle–separator surface interaction in the MPSS.

Due to the strong reduction of the cross-section, particles rising in the MPSS likely encounter the inner vessel surface. The surface is manufactured to be especially smooth to avoid mechanical adhesion. However, this does not prevent hydrophobic or electrostatic attraction. Moreover, the flange connections and valves indeed represent physical traps for ascending particles. When particles meet the wall, the hydrophobic plastic–metal contact might be maintained because it is energetically favourable in an aqueous solution. Particles attaching to the inner MPSS surface might continue to rise, but due to adhesion and friction force, at a reduced speed. Consequently, the separation time of 15 or 20 hours might be insufficient.

While the probability for a particle to hit the wall of the MPSS is a question of coincidence, the tendency to adhere to the metal surface likely depends on particle characteristics. In particular, the surface-to-volume ratio, which increases with decreasing size [75] and elongation [76] of a particle, could influence the adhesion positively. Therefore, the small and medium size fractions have a higher tendency to adhere to the wall than the larger one, potentially causing the smaller recovery rates. Moreover, both the higher variability and the slight bimodal distribution of the recovery rate in the smallest size class might have been caused by the diversity in particle morphology, which is less influential in larger particles. Nevertheless, adhesion can also occur in larger and more regular particles (e.g., expanded polystyrene [EPS] spheres of 1 mm

[77], rectangles of 0.25 mm² [78], or particles larger than 1 mm [79]) or in vessels without inclination [29].

In interpreting our results, however, we need to consider several limitations of this study. First, particle number per size class was relatively low (cf. Table 1). Second, the commercial sand matrix was reused despite a potential impact on the recovery rate which, however, did not turn out to be the case (Supplementary Fig. S3). Third, the test particles were of pristine nature, but their behaviour can be different when exposed to environmental conditions, which alter their surface characteristics. Therefore, future work should investigate the factors affecting particle adhesion, such as particle size, shape, surface properties (e.g., roughness, charge, zeta-potential), and weathering state. It is important to find solutions to the common problem of particle adhesion in the MPSS and other separation setups, for example, by filling in the fluid from the bottom valve of the MPSS, using oil which concentrates the particles swimming at the surface [78], or agitating the fluid [80].

Despite the limitations, the key finding of our study appears to be in line with the literature. A topical meta-analysis of 144 studies about the extraction and analysis of microplastics in solid matrices found a decreasing recovery rate with particle size [28]. Recovery rates decreased from 92.8 % (1–5 mm), over 90.1 % (0.5–1 mm), to 86.6 % (0.1–0.5 mm). A later meta-analysis including 71 recovery studies on all media used in the field of microplastics found the same trend: the recovery decreased from 100 % (larger than 5 mm), over 84.8 % (1–5 mm), to 84.5 % (1 µm–1 mm) [45]. In the size class larger than 1 mm, our recovery (97 %) was higher than the mean reported in both reviews. This comparison should be treated with caution because of the large diversity of methodological approaches reflected by the two meta-analyses, including different separation fluids. A comparison with the smaller size categories is even more difficult because of the different size ranges. However, based on our data, a continuing decreasing trend in the separation effectiveness of the MPSS for particles smaller than 0.3 mm can be expected, indicating a performance worse than the average calculated in both meta-analyses. Indeed, in [81], a recovery of 80 % was achieved for the size class 0.3–0.5 mm with river sediments, sodium chloride, beakers, and the overflow technique, potentially due to chemical purification, small sample volumes, and five repeated extractions.

We set the lower size class limit to 0.3 mm, as particles of this size can still be reliably detected by eye (as shown in the localization step), manually handled by tweezers, and measured by ATR-FTIR spectroscopy. The size limit of 0.3 mm was also advocated in [82] due to its correspondence with a common mesh size of sampling nets for

water samples. However, in an investigation on the extent of underreporting data on microplastics extracted from different environmental media (i.e., water, sediment, soil, biota, air) and methodological procedures for extraction and characterization, microplastics sized 1–300 µm were found to be affected most [83]. Thus, future work should investigate the recovery of particles smaller than 300 µm, also due to their higher abundance in environmental samples and a potentially higher ecological relevance. However, this requires a more elaborated methodological approach regarding sample preparation and detection and contamination management.

In the literature, a decreasing recovery rate with size was not always shown. Although several studies observed this trend [80, 84, 85], in a setup with sodium chloride, beaker, and overflow, there was no difference in the recovery between small (0.25–0.5 mm) and large (0.5–1 mm) particles [46]. One study even reported an increased recovery with decreased size [44]. Although it is already known that smaller particles are more difficult to separate [86], this is not yet sufficiently considered in recovery rate experiments – particularly, when the test particle sizes in the recovery rate experiment do not correspond with those of the microplastics investigated. For example, in [80], the test particle size range was 100–3000 µm while the environmental microplastics investigated were larger than 20 µm.

Extraction from environmental matrices

It was suggested that the separation of particles may be influenced by the type [87] or characteristics of the matrix [46, 79], entailing the call for matrix-specific purification methods [88]. For instance, a higher content of clay could be associated with a stronger retention of buoyant particles in the matrix [89]. Other studies found no impact of the matrix on the recovery. In [53], neither a difference in recovery from three different sediment types ranging from fine to coarse marine sediment was found, nor from matrix with different contents of organic matter up to 2.6 % (test particle size around 1 mm). Similarly, Constant et al. (2021) found no significant difference in recovery from fine and coarse sediments, a result they attributed to the pristine test particles (size larger than 1 mm) [79]. Radford et al. (2021) found no significant correlation between matrix grain size (clay and fine sand) and plastic particle recovery (test particle size larger than 250 µm), whereas the presence of organic matter decreased the recovery rate [46].

We found neither evidence for a test particle recovery lower in fluvial sediment than in commercial sand (experiments II and I), nor for an impact of river sediment grain size distribution on test particle recovery (experiment II) or on microplastic extraction in the second density separation (experiment III). However, this absence of evidence could simply be due to methodological reasons, i.e., the way of spiking the matrix with test particles, namely only some millimetres deep and without any agitation or incubation (experiments I and II), or the small sample size (experiment III), and must not necessarily hold true for microplastics, which may be embedded in a matrix more tightly than test particles. Therefore, a stronger interaction of microplastics with the fluvial matrix could still be a reason for the lower separation of microplastics extracted in experiment III compared to the test particles. Another reason could be that the test particles did not represent the spectrum of microplastics regarding polymeric and morphological characteristics as well as surface properties, which change in the environment. Therefore, we refrained from using the recovery rates obtained in experiment I as 'correction factors' [90] in experiment III, to adjust environmental concentrations in the Lahn River sediments in Gießen. Finally, more research is necessary to clarify to which extent a sample matrix impairs the recovery and if 'matrix-specific calibration' [46] is appropriate.

In our study, the recovery from environmental sediment was *higher* than from commercial sand. Apart from the matrix, the experiments differed in several aspects, which is why this comparison should be interpreted with caution. The separation duration was 15 and 20 hours, the number of test particles 20 and 10, the two size fractions were separated and combined, and the number of replicates was 30 and 37, in experiment I and II, respectively (cf. Table 1). If the three latter aspects are negligible, the difference in recovery rate may be attributed to the longer separation duration in experiment II – in line with Stokes' law. In contrast, a recent work using sodium chloride and beakers found an optimum separation duration of one to six hours and a decreasing recovery rate with a longer duration (sieved particle sizes of 2.5 µm to 5 mm, ground PE-HD bottle caps) [89]. The reason for this observation is unclear. Apparently, the relationship between separation duration and effectiveness requires further research. Nonetheless, given that particle separation is a function of time, it is important to keep the separation duration constant within a study for comparability.

Repeated density separation

Subsequent separation runs of the same sample were shown to improve the recovery of test particles and are therefore commonly performed (e.g., in [81, 85, 89, 91, 92]). Our study is the first to test the effect of a second separation on the effectiveness of the MPSS, which was also devised with the intention to avoid repeated density separation runs [29]. We demonstrated that a second separation provided considerable numbers of microplastics.

Still, the average proportion of the first run in total concentrations was higher than that of the second run. In both runs, the number of separated particles increased with decreasing size class, which can be explained by a higher proportion of smaller particles in environmental samples.

Microplastics in Lahn River sediments

Here, we provide first evidence for microplastics in sediments of the Lahn River, a medium-scale tributary of the Rhine River. Total concentrations were related positively to clay and negatively to sand, indicating that microplastics and finer sediment grains are deposited at higher rates in areas of lower water flow velocity. Due to the small number of samples ($n = 5$), this correlation requires further testing. Compared to Lahn River floodplains, which contained a mean of 2.75 and a maximum of 30 particles per kilogram of dry mass (size range: 0.3–2 mm) [93], river bed concentrations were an order of magnitude higher. The comparability of the two sampling campaigns is high, since similar methods were applied, partly in the same laboratories.

Conclusions

In the density separation with the MPSS operated with sodium chloride, the separation effectiveness is high for pristine test particles larger than 1 mm, but declines with particle size. Whether the levels of trueness and precision achieved by single-run separations in the size range of 0.3–1 mm are acceptable depends on the intended use or research question. Due to the systematic underestimation of particles smaller than 1 mm, resulting concentrations of microplastics should be considered as minimum levels.

Further work is recommended to validate existing and develop new separation technology: Firstly, to prevent particle adhesion, separation vessels should avoid inclined walls, reduce the ratio of vessel surface to fluid volume, or agitate the whole volume of separation fluid. Secondly, density separation procedures must be designed in such a way that the smallest target particles have enough time to ascend. Thirdly, future recovery studies should use weathered test particles of different polymer types, sizes, and shapes as well as environmental microplastics to obtain more environmentally relevant data.

This study highlights the interplay of separation vessel, particle size, and separation time, and stresses the importance of testing recovery rates by particle size. Understanding and improving methods is a requirement to obtain accurate data, which are needed to reduce plastic emissions.

Abbreviations

ATR	Attenuated total reflection
BAM	Bundesanstalt für Materialforschung und -prüfung
EPDM	Ethylene propylene diene monomer rubber
EPS	Expanded polystyrene
EVA	Ethylene-vinyl acetate
FTIR	Fourier transform infrared
HQI	Hit quality index
MPSS	MicroPlastic Sediment Separator
PE-HD	High-density polyethylene
PE-LD	Low-density polyethylene
PET	Polyethylene terephthalate
PEST	Polyester
POM	Polyoxymethylene
PP	Polypropylene
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PVC	Polyvinyl chloride
QA/QC	Quality assurance/quality control
TOC	Total organic carbon

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s43591-023-00073-3.

> **Additional file 1.** Supplementary Information. Appendix (.pdf) including additional text, figures, and tables for a more detailed understanding.

Authors' contributions

J.A.P.: Conceptualization, Methodology, Software, Validation, Formal Analysis, Investigation, Data Curation, Writing – Original Draft, Writing – Review & Editing, Visualization. H.L.: Investigation, Writing – Review & Editing. M.G.J.L.: Writing – Review & Editing, Supervision. C.L.: Writing – Review & Editing, Supervision. C.B.: Formal Analysis, Writing – Review & Editing, Visualization, Supervision. M.K.: Resources, Writing – Review & Editing, Supervision, Project administration, Funding acquisition.

Availability of data and materials
The data generated and analysed during the current study are available in the Zenodo repository [DOI: 10.5281/zenodo.10104525].

Declarations

Ethics approval and consent to participate
Not applicable.

Consent for publication
All authors agree to the publication.

Competing interests
The authors declare no competing interests.

References

1. Carpenter EJ, Smith K Jr. Plastics on the Sargasso Sea surface. Science. 1972;175(4027):1240–1.
2. Hartmann NB, Huffer T, Thompson RC, Hassellöv M, Verschoor A, Daugaard AE, et al. Are we speaking the same language? Recommendations for a definition and categorization framework for plastic debris: ACS Publications; 2019.
3. ISO/TR 21960:2020. Plastics - Environmental aspects - State of knowledge and methodologies. 2020. https://www.iso.org/obp/ui/#iso:std:iso:tr: 21960:ed-1:v1:en. Accessed 9 Apr 2022.
4. Arthur C, Baker J, Bamfort H, editors. Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris. Technical Memorandum NOS-OR &R-30: NOAA; 2009.
5. Wagner M, Scherer C, Alvarez-Muñoz D, Brennholt N, Bourrain X, Buchinger S, et al. Microplastics in freshwater ecosystems: what we know and what we need to know. Environ Sci Eur. 2014;26(1):1–9.
6. Koelmans AA, Besseling E, Shim WJ. Nanoplastics in the aquatic environment. Critical review. In: Bergmann M, Gutov L, Klages M, editors. Marine Anthropogenic Litter. Springer Open; 2015. p. 325–340.
7. Vivekanand AC, Mohapatra S, Tyagi VK. Microplastics in aquatic environment: challenges and perspectives. Chemosphere. 2021;282:131151.
8. Van Cauwenberghe L, Devriese L, Galgani F, Robbens J, Janssen CR. Microplastics in sediments: a review of techniques, occurrence and effects. Mar Environ Res. 2015;111:5–17.
9. Yang L, Zhang Y, Kang S, Wang Z, Wu C. Microplastics in soil: a review on methods, occurrence, sources, and potential risk. Sci Total Environ. 2021;780:146546.
10. Zhang Y, Kang S, Allen S, Allen D, Gao T, Sillanpää M. Atmospheric microplastics: a review on current status and perspectives. Earth Sci Rev. 2020;203:103118.
11. Kernchen S, Löder MG, Fischer F, Fischer D, Moses SR, Georgi C, et al. Airborne microplastic concentrations and deposition across the Weser River catchment. Sci Total Environ. 2022;818:151812.
12. Koutnik VS, Leonard J, Alkidim S, DePrima FJ, Ravi S, Hoek EM, et al. Distribution of microplastics in soil and freshwater environments: global analysis and framework for transport modeling. Environ Pollut. 2021;274:116552.
13. Ajith N, Arumugam S, Parthasarathy S, Manupoori S, Janakiraman S. Global distribution of microplastics and its impact on marine environment-a review. Environ Sci Pollut Res. 2020;27(21):25970–86.
14. Egbeocha CO, Malek S, Emenike CU, Milow P. Feasting on microplastics: ingestion by and effects on marine organisms. Aquat Biol. 2018;27:93–106.
15. Kukkola A, Krause S, Lynch I, Smith GHS, Nel H. Nano and microplastic interactions with freshwater biota-Current knowledge, challenges and future solutions. Environ Int. 2021;152:106504.
16. Cox KD, Covernton GA, Davies HL, Dower JF, Juanes F, Dudas SE. Human consumption of microplastics. Environ Sci Technol. 2019;53(12):7068–74.
17. Schwabl P, Köppel S, Königshofer P, Bucsics T, Trauner M, Reiberger T, et al. Detection of various microplastics in human stool: a prospective case series. Ann Intern Med. 2019;171(7):453–7.
18. Ragusa A, Svelato A, Santacroce C, Catalano P, Notarstefano V, Carnevali O, et al. Plasticenta: first evidence of microplastics in human placenta. Environ Int. 2021;146:106274.
19. Leslie HA, Van Velzen MJ, Brandsma SH, Vethaak AD, Garcia-Vallejo JJ, Lamoree MH. Discovery and quantification of plastic particle pollution in human blood. Environ Int. 2022;163:107199.
20. Catarino AI, Kramm J, Voelker C, Henry TB, Everaert G. Risk posed by microplastics: scientific evidence and public perception. Curr Opin Green Sustain Chem. 2021;29:100467.
21. Lebreton L, Andrady A. Future scenarios of global plastic waste generation and disposal. Palgrave Commun. 2019;5(1):1–11.
22. Prata JC, da Costa JP, Duarte AC, Rocha-Santos T. Methods for sampling and detection of microplastics in water and sediment: a critical review. TrAC Trends Anal Chem. 2019;110:150–9.
23. Möller JN, Heisel I, Satzger A, Vizsolyi EC, Oster J, Agarwal S, et al. Tackling the challenge of extracting microplastics from soils: a protocol to purify soil samples for spectroscopic analysis. Environ Toxicol Chem. 2021;41:844–57.
24. Beaurepaire M, Dris R, Gasperi J, Tassin B. Microplastics in the atmospheric compartment: a comprehensive review on methods, results on their occurrence and determining factors. Curr Opin Food Sci. 2021;41:159–68.
25. Brander SM, Renick VC, Foley MM, Steele C, Woo M, Lusher A, et al. Sampling and quality assurance and quality control: a guide for scientists investigating the occurrence of microplastics across matrices. Appl Spectrosc. 2020;74(9):1099–125.
26. Bläsing M, Amelung W. Plastics in Soil: Analytical Methods and Possible Sources. Sci Total Environ. 2018;612:422–35.
27. Kawecki D, Nowack B. Polymer-Specific Modeling of the Environmental Emissions of Seven Commodity Plastics As Macro- and Microplastics. Environ Sci Technol. 2019Aug;53(16):9664–76.
28. He D, Zhang X, Hu J. Methods for separating microplastics from complex solid matrices: comparative analysis. J Hazard Mater. 2021;409:124640.
29. Imhof HK, Schmid J, Niessner R, Ivleva NP, Laforsch C. A novel, highly efficient method for the separation and quantification of plastic particles in sediments of aquatic environments. Limnol Oceanogr Methods. 2012;10(7):524–37.
30. Claessens M, Van Cauwenberghe L, Vandegehuchte MB, Janssen CR. New techniques for the detection of microplastics in sediments and field collected organisms. Mar Pollut Bull. 2013;70(1–2):227–33.
31. Crichton EM, Noël M, Gies EA, Ross PS. A novel, density-independent and FTIR-compatible approach for the rapid extraction of microplastics from aquatic sediments. Anal Methods. 2017;9(9):1419–28.
32. Lechthaler S, Hildebrandt L, Stauch G, Schüttrumpf H. Canola oil extraction in conjunction with a plastic free separation unit optimises microplastics monitoring in water and sediment. Anal Methods. 2020;12(42):5128–39.
33. Grbic J, Nguyen B, Guo E, You JB, Sinton D, Rochman CM. Magnetic extraction of microplastics from environmental samples. Environ Sci Technol Lett. 2019;6(2):68–72.
34. Felsing S, Kochleus C, Buchinger S, Brennholt N, Stock F, Reifferscheid G. A new approach in separating microplastics from environmental samples based on their electrostatic behavior. Environ Pollut. 2018;234:20–8.
35. Hanvey JS, Lewis PJ, Lavers JL, Crosbie ND, Pozo K, Clarke BO. A review of analytical techniques for quantifying microplastics in sediments. Anal Methods. 2017;9(9):1369–83.
36. Bellasi A, Binda G, Pozzi A, Boldrocchi G, Bettinetti R. The extraction of microplastics from sediments: an overview of existing methods and the proposal of a new and green alternative. Chemosphere. 2021;278:130357.
37. Jakobs A, Gürkal E, Möller JN, Löder MG, Laforsch C, Lueders T. A novel approach to extract, purify, and fractionate microplastics from environmental matrices by isopycnic ultracentrifugation. Sci Total Environ. 2023;857:159610.
38. Skopp J. Physical properties of primary particles. In: PM Huang MS Y Li, editor. Handbook of soil science. 2nd ed. New York: CRC Press; 2012.
39. Ye S, Andrady AL. Fouling of floating plastic debris under Biscayne Bay exposure conditions. Mar Pollut Bull. 1991;22(12):608–13.

40. Chubarenko I, Bagaev A, Zobkov M, Esiukova E. On some physical and dynamical properties of microplastic particles in marine environment. Mar Pollut Bull. 2016;108(1–2):105–12.

41. Kowalski N, Reichardt AM, Waniek JJ. Sinking rates of microplastics and potential implications of their alteration by physical, biological, and chemical factors. Mar Pollut Bull. 2016;109(1):310–9.

42. Kaiser D, Kowalski N, Waniek JJ. Effects of biofouling on the sinking behavior of microplastics. Environ Res Lett. 2017;12(12):124003.

43. Besseling E, Quik JT, Sun M, Koelmans AA. Fate of nano-and microplastic in freshwater systems: a modeling study. Environ Pollut. 2017;220:540–8.

44. Quinn B, Murphy F, Ewins C. Validation of density separation for the rapid recovery of microplastics from sediment. Anal Methods. 2017;9(9):1491–8.

45. Way C, Hudson MD, Williams ID, Langley GJ. Evidence of underestimation in microplastic research: a meta-analysis of recovery rate studies. Sci Total Environ. 2022;805:150227.

46. Radford F, Zapata-Restrepo LM, Horton AA, Hudson MD, Shaw PJ, Williams ID. Developing a systematic method for extraction of microplastics in soils. Anal Methods. 2021;13(14):1695–705.

47. Cutroneo L, Reboa A, Geneselli I, Capello M. Considerations on salts used for density separation in the extraction of microplastics from sediments. Mar Pollut Bull. 2021;166:112216.

48. Möller JN, Löder MGJ, Laforsch C. Finding Microplastics in Soils: a Review of Analytical Methods. Environ Sci Technol. 2020;54(4):2078–90.

49. Ziajahromi S, Leusch FD. Systematic assessment of data quality and quality assurance/quality control (QA/QC) of current research on microplastics in biosolids and agricultural soils. Environ Pollut. 2022;294:118629.

50. Coppock RL, Cole M, Lindeque PK, Queirós AM, Galloway TS. A small-scale, portable method for extracting microplastics from marine sediments. Environ Pollut. 2017;230:829–37.

51. Hidalgo-Ruz V, Gutow L, Thompson RC, Thiel M. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ Sci Technol. 2012;46(6):3060–75.

52. Klein S, Worch E, Knepper TP. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. Environ Sci Technol. 2015;49(10):6070–6.

53. Zobkov MB, Esiukova EE. Evaluation of the Munich Plastic Sediment Separator efficiency in extraction of microplastics from natural marine bottom sediments. Limnology and Oceanography: Methods. 2017;15(11):967–78.

54. Picó Y, Barceló D. Micro (Nano) plastic analysis: a green and sustainable perspective. J Hazard Mater Adv. 2022;6:100058.

55. ISO 5725-1:2023. Accuracy (trueness and precision) of measurement methods and results - Part 1: General principles and definitions. 2023. https://www.iso.org/obp/ui/en/#iso:std:iso:5725:-1:ed-2:v1:en:term:3.1. Accessed 9 Oct 2023.

56. Andrady AL. Microplastics in the marine environment. Mar Pollut Bull. 2011;62(8):1596–605.

57. Europe Plastics. Plastics-The Facts 2022. Plast Facts. 2022;2022:1–81.

58. Konde S, Ornik J, Prume JA, Taiber J, Koch M. Exploring the potential of photoluminescence spectroscopy in combination with Nile Red staining for microplastic detection. Mar Pollut Bull. 2020;159:111475.

59. Prume JA, Gorka F, Löder MG. From sieve to microscope: an efficient technique for sample transfer in the process of microplastics' quantification. MethodsX. 2021;8:101341.

60. Norén F. Small plastic particles in coastal Swedish waters. Kimo Sweden. 2007;11:1–11.

61. Thiele CJ, Hudson MD, Russell AE, Saluveer M, Sidaoui-Haddad G. Microplastics in fish and fishmeal: an emerging environmental challenge? Sci Rep. 2021;11(1):2045.

62. Sommer C, Nguyen J, Menzel T, Prume JA, Ruckdäschel H, Koch M. Weathering-induced oxidation: An investigation of artificially aged polystyrene samples using Laser-induced Breakdown Spectroscopy. Polym Test. 2022:107623.

63. R Core Team. R: A Language and Environment for Statistical Computing. Vienna, Austria; 2021. https://www.R-project.org/. Accessed 18 Aug 2022.

64. Wickham H, Bryan J. readxl: Read Excel Files. 2022. https://readxl.tidyverse.org, https://github.com/tidyverse/readxl. Accessed 18 Aug 2022.

65. Wickham H, Averick M, Bryan J, Chang W, McGowan LD, François R, et al. Welcome to the Tidyverse. J Open Source Softw. 2019;4(43):1686.

66. Wickham H, Seidel D. scales: Scale Functions for Visualization. 2022. https://scales.r-lib.org, https://github.com/r-lib/scales. Accessed 11 May 2023.

67. Waring E, Quinn M, McNamara A, Arino de la Rubia E, Zhu H, Ellis S. skimr: compact and flexible summaries of data. 2023. https://docs.ropensci.org/skimr/, https://github.com/ropensci/skimr/. Accessed 11 May 2023.

68. Harrell Jr FE, Harrell Jr MFE. Package 'hmisc'. CRAN2018. 2019;2019:235–6.

69. Peterson BG, Carl P, Boudt K, Bennett R, Ulrich J, Zivot E, et al.. Package 'performanceanalytics'. 2006. https://cran.r-project.org/web/packages/PerformanceAnalytics/PerformanceAnalytics.pdf. Accessed 13 Jan 2023.

70. Mukaka MM. A guide to appropriate use of correlation coefficient in medical research. Malawi Med J. 2012;24(3):69–71.

71. Squara P, Scheeren TW, Aya HD, Bakker J, Cecconi M, Einav S, et al. Metrology part 1: definition of quality criteria. J Clin Monit Comput. 2021;35(1):17–25.

72. DIN 1333:1992-02. Presentation of numerical data. 1992.

73. Ho J, Tumkaya T, Aryal S, Choi H, Claridge-Chang A. Moving beyond P values: everyday data analysis with estimation plots. 2019.

74. Ho J, Tumkaya T, Aryal S, Choi H, Claridge-Chang A. Moving beyond P values: data analysis with estimation graphics. Nat Methods. 2019;16(7):565–6.

75. Ji ZG. Hydrodynamics and water quality: modeling rivers, lakes, and estuaries. Hoboken: John Wiley & Sons; 2017.

76. Sumich JL, Morrissey JF. Introduction to the biology of marine life. Sudbury: Jones & Bartlett Learning; 2004.

77. Nuelle MT, Dekiff JH, Remy D, Fries E. A new analytical approach for monitoring microplastics in marine sediments. Environ Pollut. 2014;184:161–9.

78. Karlsson TM, Vethaak AD, Almroth BC, Ariese F, van Velzen M, Hassellöv M, et al. Screening for microplastics in sediment, water, marine invertebrates and fish: method development and microplastic accumulation. Mar Pollut Bull. 2017;122(1–2):403–8.

79. Constant M, Billon G, Breton N, Alary C. Extraction of microplastics from sediment matrices: experimental comparative analysis. J Hazard Mater. 2021;420:126571.

80. Liu M, Song Y, Lu S, Qiu R, Hu J, Li X, et al. A method for extracting soil microplastics through circulation of sodium bromide solutions. Sci Total Environ. 2019;691:341–7.

81. Duong TT, Le PT, Nguyen TNH, Hoang TQ, Ngo HM, Doan TO, et al. Selection of a density separation solution to study microplastics in tropical riverine sediment. Environ Monit Assess. 2022;194:1–17.

82. Waldschläger K, Brückner MZ, Almroth BC, Hackney CR, Adyel TM, Alimi OS, et al. Learning from natural sediments to tackle microplastics challenges: a multidisciplinary perspective. Earth Sci Rev. 2022;228:104021.

83. Pérez-Guevara F, Roy PD, Kutralam-Muniasamy G, Shruti V. Coverage of microplastic data underreporting and progress toward standardization. Sci Total Environ. 2022;829:154727.

84. Konechnaya O, Lüchtrath S, Dsikowitzky L, Schwarzbauer J. Optimized microplastic analysis based on size fractionation, density separation and μ-FTIR. Water Sci Technol. 2020;81(4):834–44.

85. Nava V, Leoni B. Comparison of different procedures for separating microplastics from sediments. Water. 2021;13(20):2854.

86. Löder MG, Gerdts G. Methodology used for the detection and identification of microplastics—a critical appraisal. In: Bergmann M, Gutov L, Klages M, editors. Marine Anthropogenic Litter. Springer Open; 2015. p. 201–227.

87. Schütze B, Thomas D, Kraft M, Brunotte J, Kreuzig R. Comparison of different salt solutions for density separation of conventional and biodegradable microplastic from solid sample matrices. Environ Sci Pollut Res. 2022;29(54):81452–67.

88. He D, Luo Y, Lu S, Liu M, Song Y, Lei L. Microplastics in soils: analytical methods, pollution characteristics and ecological risks. TrAC Trends Anal Chem. 2018;109:163–72.

89. Maisto M, Ranauda MA, Zuzolo D, Tartaglia M, Postiglione A, Prigioniero A, et al. Influence of sediment texture on HDPE microplastics recovery by density separation. J Environ Manag. 2022;317:115363.

90. Claessens M, De Meester S, Van Landuyt L, De Clerck K, Janssen CR. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar Pollut Bull. 2011;62(10):2199–204.

91. Browne MA, Crump P, Niven SJ, Teuten E, Tonkin A, Galloway T, et al. Accumulation of microplastic on shorelines woldwide: sources and sinks. Environ Sci Technol. 2011;45(21):9175–9.

92. Morgado V, Gomes L, da Silva RJB, Palma C. Validated spreadsheet for the identification of PE, PET, PP and PS microplastics by micro-ATR-FTIR spectra with known uncertainty. Talanta. 2021;234:122624.
93. Weber CJ, Opp C, Prume JA, Koch M, Andersen TJ, Chifflard P. Deposition and in-situ translocation of microplastics in floodplain soils. Sci Total Environ. 2022;819:152039.

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