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Microplastic in long-term soil monitoring: first spatial and temporal data on plastics in agricultural topsoils

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Abstract

Purpose Within the emerging field of soil related microplastic (MP) research, little is known about spatio-temporal variations of MP concentrations in soils. To overcome this current knowledge gap, we suggest the exploitation of long-term soil monitoring programmes (LTSM) and its archived soil samples, to gain first data on spatio-temporal variations within soil MP contamination.

Methods We analysed 32 archived topsoil samples of 13 cropland and 3 control sites, including grassland and forest sites, which are part of the LTSM programme of the German federal state of Hesse. Analysed samples cover a time span of 10–16 years. MP (>300 μm) have been extracted via sodium chloride (NaCl, 1.2 g cm^{-3}) density separation, Nile Red staining and fluorescent optical identification followed by ATR-FTIR analysis of individual MP.

Results We found MP contamination (mean 34.66 ± 24.08 p kg^{-1}) from mainly PE and synthetic rubber polymers. Furthermore, we were able to detect spatial MP contamination hotspots, with unexpected high MP concentrations in control grassland and forest LTSM sites. In case of temporal trends, we cannot conclude a general increase of MP concentrations over time. However, as we found MP in older and newer samples, MP contamination is likely to be ubiquitous in topsoils and has already been present in the environment for several decades.

Conclusion Our findings underline the role of cropland topsoils as MP reservoirs, but raise questions on the MP storage in grassland and forest soils. Applying MP analytical tools to archived LTSM samples enables first insights within non-targeted spatial source analysis and temporal trends of soils MP pollution. If enhanced analytical methods will be applied to LTSM samples, it will be possible to gain more detailed insights within spatio-temporal MP dynamics in soils.

Keywords Microplastic sources · Temporal data · Temporal trends · Cropland soil · Forest soil · Archived soil samples

1 Introduction

Within the last 70 years, around 8.3 million tons of plastics have been produced and consumed worldwide, while 79% from this quantity could have been accumulated in the

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environment (Geyer et al. 2017). In recent years, it becomes obvious that terrestrial systems and their soils act as a main reservoir for plastics, as plastic emissions to soils are 3–24 times higher than to oceans (Horton et al. 2017). Plastic items and especially microplastics (MP) with a size between 5000 and 1 μm (Andrady 2017) reach soils via several emission pathways (Bläsing and Amelung 2018; Duis and Coors 2016). Well-studied are the direct emissions for agricultural soils, where agricultural practice and management lead to plastic inputs from fertilizers (e.g., sewage sludge, compost) (Scopetani et al. 2022; Tagg et al. 2022), plastic mulching (Huang et al. 2020) and application of other plastic materials (Steinmetz et al. 2022) as well as inputs from adjacent environmental systems due to irrigation practice (Pérez-Reverón et al. 2022). Diffuse sources like littering (Braun et al. 2023; Cowger et al. 2022), atmospheric (Aeschlimann et al. 2022) or flood deposition (Lechthaler et al. 2021)

occur both for agricultural soils and others (e.g., forest soils; natural grasslands).

Globally, detected MP concentrations in soils, given in MP particles (p) per kilogram soil dry weight (kg^{-1}), vary strongly from amounts < 100 p kg^{-1} for non-intensively used soils up to $> 10,000$ p kg^{-1} in soils with intensive, conventional agricultural practice (Büks and Kaupenjohann 2020). In contrast, MP characteristics like chemical composition or particle size distribution, show a more comparable picture, with recovery of commonly produced polymers and significant increase in particle numbers with decreasing particle size (Lwanga et al. 2022). However, data on MP in soils is difficult to compare between studies, because of the widely different MP extraction and analytical protocols applied, especially with respect to extraction recovery and lower particle size detection limit (Möller et al. 2020).

MP in soils lead to different effects on soil and the soil environment. Briefly, those effects include changes in soil structure (e.g., aggregate formation) and therefore physical soil properties (e.g., water-holding capacity) (Rillig et al. 2021; Zhang and Liu 2018). But also influences on soil organisms (de Souza Machado et al. 2018; Zhang et al. 2019) or impacts within soil–plant systems including negative consequences for plant growth or the uptake of MPs into food chains (Ren et al. 2022). Depending on MP concentrations, surface and chemical properties as well as exposure time, MP contamination can therefore turn to MP pollution, with significant soil- and ecosystem consequences (de Souza Machado et al. 2018, 2019; Ng et al. 2021).

Generally, recent data on MP in terrestrial environments must be considered as a snapshot of soils MP contamination in a spatial and temporal context. This comes up because all previous studies only quantify and chemo-physically describe MP in a limited spatial scope and at one point in time (Büks and Kaupenjohann 2020; Weber and Bigalke 2022). Currently, however, there is only limited information about MP contamination across different spatial units and soil formations and basically no information about the temporal MP variations in soils. Yet it is especially temporal data that is important when considering effects of MP in soils. Regarding MP sources, input pathways for plastics in agriculture and beyond have been increasingly regulated at different levels of legislation (e.g., sewage sludge; ban of single use plastics) (Da Costa et al. 2020). Furthermore, social awareness on plastic pollution rises, leading to other consumption patterns and plastic production (European Commission. Directorate General for Environment 2021) with differences regarding urban or rural environments. Once in the environment, plastics are supposed to be aged by UV-radiation (Ali et al. 2021; Ren et al. 2021) and biogeochemical processes (Canopoli et al. 2020; Ren et al. 2021), leading to chemo-physical surface changes and subsequent changing effects on soil environments. Since

socio-economic, environmental as well as physico-chemical material changes itself, underlying temporal and partwise also spatial dynamics, a clear knowledge gap is given in the absence of spatio-temporal MP data.

In order to address this significant knowledge gap, we propose the use of long-term soil monitoring programmes (LTSM) and its archived soil samples, to access spatio-temporal data on soil MP contamination. Against this background, we analysed soil samples of different ages from 13 LTSM cropland sites together with three control sites located in the German federal state of Hesse to answer the following questions:

1. Are archived soil samples from LTSM programmes suitable for MP analysis?
2. Can significant spatio-temporal differences in the MP contamination of soils be detected?

Based on our MP extraction and analytical approach, we were able to analyse large (> 1000 μm) and medium sized (> 300 μm) MP made from polymers with a density < 1.2 g cm^{-3} (e.g., PE, PP, PA), covering the most common polymers, in a simple and easily transferable analytical implementation.

2 Material and methods

2.1 Long-term soil monitoring programmes

Long-term soil monitoring (LTSM) programmes investigate representative soils and their status as well as their further development and are a central part of an integrated environmental monitoring (European Commission 2021). The aims of these programmes are to describe the current status of soils, to monitor their long-term changes and to predict future developments. LTSM programmes or surveys are carried out worldwide on different scales and covering different time periods (Drewry et al. 2021; Gubler et al. 2020; Michel et al. 2018; Orgiazzi et al. 2018). Specific examples can be found within the supplementary information of this publication.

In Germany, the federal states are responsible for the individual LTSM programmes, with the first LTSM sites established in the mid-1980s in some federal states. Meanwhile, approximately 800 LTSM sites are operated nationwide (Umweltbundesamt 2015). Due to the very long duration of the programmes, the LTSM sites are among the best studied soils in Germany and serve as important national reference areas for various research studies. Within the Federal State of Hesse, where this study was conducted, the official authority currently operates 67 basic LTSM sites and one intensive LTSM site (with path-related material flow measurements). These sites were established in the period from

1992 to 1999 and have been repeatedly sampled at regular intervals since then. Depending on the site, the investigation intervals cover 5 to 10 years. The Hessian LTSM sites comprise plots of different land use types, like cropland, grassland and forests, vineyard and one subhydric soil (Hessian State Agency for Nature Protection, Environment and Geology 2022). The agriculturally used LTSM sites in the state of Hesse remain in the normal use by the farmers who cultivate the areas. As previous agreements and contracts with the farmers unfortunately did not include the provision of detailed management information (e.g., crop rotation, application of sewage sludge, biowaste composts, pesticides), this important data basis for further interpretation of microplastic investigations is missing. At present, a survey and the corresponding documentation of the management data have been started on the basis of new contracts with farmers who participate in the programme.

The investigation of the samples includes the determination of a large number of chemical and physical parameters. In addition to standard parameters such as pH-value, TOC or grain size, nutrients and trace elements as well as different organic pollutants and certain radionuclides are analysed by the state authority. Relevant data for the selected LTSM sites are given in the supplementary information (Fig. S3, Table S1). Residual material from all samples gets air-dried and archived in a soil sample bank, where it is available for later investigations.

2.2 Sampling and sample pre-processing

To access the feasibility of MP analysis from LTSM samples, we used archived soil samples from the LTSM programme of the federal state of Hesse (Germany) managed by the responsible official authority, the Hessian State Agency for Nature Conservation, Environment and Geology (HLNUG). Out of the whole soil sample archive, we decided to analyse sample material from 13 cropland LTSM sites, together with two grassland and one forest LTSM site as a non-cropland control group. The selected LTSM cropland sites cover the three major agricultural production areas across the federal state. In addition, we added three control sites within rural areas of Hesse. Spatial locations of all analysed LTSM sites are given with coordinates in the supplementary information (Table S2). The selected LTSM sites contain mostly Luvisols and Cambisols, followed by Vertisols and Planosols (classified according World reference base for soil resources, 2015) with silt loam to silty clay textures, a total organic carbon (TOC) content between 0.7–6.1% and a pH range of 4.3–7.4, both depending on soil type, location and land-use (Table S1).

From each selected LTSM site, archived samples taken by staff of the HLNUG in the years 2003–2004 (19–20 years old), as well as 2014–2015 and 2019 (4–5 and 9 years old)

have been analysed. For the control forest site, fresh topsoil samples and one subsoil sample from spring 2022 have been used, as we wanted to control the sampling procedure with regard to sample contamination. The different time intervals as well as the new sampling of the forest soils in 2022, resulted from the availability of sufficient (> 100 g) reserve material in comparable time spans in the archive of the LTSM programme. Each LTSM site covers 900 m^2 (30 × 30 m). Topsoil samples have been taken as a composite sample according to soil horizons from 18 individual boreholes across the diagonals of the full LTSM site and pooled in the field (Fig. S1). Sample extraction was performed with stainless-steel augers and transport within amber glass bottles. Sample material has been air-dried, representatively divided and a sub-sample stored as archived sample in amber glass bottles with PE-caps separated by aluminium foil from the sample material. For MP analysis, we received aliquots of >100 g dry soil from the HLNUG. We used one topsoil sample of each agricultural site with a sampling depth of 0–25 to 0–30 cm under cropland and 0–10 cm under grassland (Table S1). Only from the forest control site SCHOT2, we analysed several samples including two topsoil A-horizons and one subsoil B-horizon of the sampled Cambisol (Table S1 and Fig. S2).

2.3 Microplastic extraction

Our MP extraction protocol was adapted from Crawford and Quinn (2017) as well as Weber et al. (2022) and Weber and Opp (2020) including four major steps. Initially, LTSM sample material was extracted from the storage amber glass bottles and dry-sieved via stainless-steel sieves (Atechnik, Leinburg, Germany) to the size fractions of >5000 μm (macroplastics), >2000 μm (coarse MPs, corresponding to coarse soil fraction) and <2000 μm (corresponding to fine soil fraction) (Weber and Opp 2020). Large MP particles within the size fraction >2000 μm have been identified visually. The remaining sample material of the size range < 2000 μm was homogenized in a stainless-steel bowl and a sub-sample of 100 g has been representatively extracted via rotary sampler for further analysis.

Density separation of each sub-sample was performed using a saturated sodium chloride (NaCl) solution after density adjustment to 1.20 g cm^{-3} at constant room temperature (19 °C) and after solution filtration (<50 μm) (Weber et al. 2022). Solution density was controlled via areometer (1.900 to 2.000 g cm^3, Greiner-Glasinstrumente, Lemgo, Germany). 100 g sub-sample has therefore been mixed with 300 ml NaCl solution in a glass baker (Crawford and Quinn 2017). The sample solution has been stirred for 1 min at 800 rpm, whereafter the beaker glass was rinsed with NaCl solution. The sample solution was left standing for 10 min and subsequently, the separation supernatant was sucked

off into a Woulfe bottle (Carl Roth, Karlsruhe, Germany) via vacuum. After each suction process, the tube made of PE was rinsed with filtered (<50 µm) deionized water. Stirring, sedimentation and suction were repeated three times per sample (Crawford and Quinn 2017).

The remaining supernatant solution was sieved via stainless-steel sieves (Atechnik, Leinburg, Germany) to the size classes of >1000 µm (large MP) and >300 µm (MP). Sample material remaining in the sieves was afterwards rinsed with filtered (<50 µm) deionized water to a vacuum filtration unit (Prume et al. 2021) and stored on cellulose filters (Ø 47 mm, ROTILABO, Carl Roth, Karlsruhe, Germany) within closed glass petri dishes (Ø 150 mm, ROTILABO, Carl Roth, Karlsruhe, Germany).

Finally, a Nile Red staining procedure (20 µg ml^{-1} Nile Red ethanol-acetone (1:1) solution, Sigma-Aldrich, Taufkirchen, Germany) was applied to differentiate between the remaining particulate organic matter (POM) and potential MP particles (Maes et al. 2017). Nile Red solution was applied to each cellulose filter via a glass pipette and a spray bottle (PE) and stained at 50 °C for 10 min within a drying chamber (Konde et al. 2020). Stained filters within petri dishes have been visually inspected systematically under a stereomicroscope (SMZ 161 TL, Motic, Hong Kong), with fluorescence 276 setup (Excitation: 465 nm LED; Emissions 530 nm colour long pass filter: Thorlabs, Bergkirchen, Germany (Konde et al. 2020; Weber et al. 2022). Each fluorescent or other potential plastic particle that shows no cellular or biologic structure (Hidalgo-Ruz et al. 2012) was collected and individually stored in microplates (Brand, Wertheim, Germany). Each plastic particle collected was size-measured (longest diagonal, Motic Images Plus 3.0, Motic, Hong Kong), photographed (Moticam 2, 284 Motic, Hong Kong) and characterised according to particle shape, surface degradation and colour.

2.4 Microplastic analysis

MP particle analysis has been performed with an ATR-FTIR device setting the lower detection limit of 300 µm, as smaller particles show an insufficient contact area to the ATR crystal (Hurley et al. 2018). We used a Tensor 37 FTIR spectrometer (Bruker Optics, Ettlingen, Germany) including a Platinum-ATR-unit (Bruker Optics, Ettlingen, Germany). Single particle measurement has been carried out after 20 background scans and with 20 sample scans for the wavenumber range between 4000 and 400 cm^{-1} with a resolution of 4 cm^{-1} (Primpke et al. 2017). Spectral data processing of each individual FTIR spectra was performed with OPUS 7.0 (Bruker Optics, Ettlingen, Germany), including atmospheric decompensation and baseline correction (concave rubber band method). Pre-processes spectra have been identified using the FTIR database of OpenSpecy (https://openanalysis.org/openspecy/) (Cowger

et al. 2021). Spectra identifications have been performed with the pre-processed spectra. Particles with a polymeric match and a $R^2 > 0.5$ have been identified as MPs. The matching rate found was on average 82% ($R^2 = 0.82$).

2.5 Quality assurance and contamination control

Spike experiments have been performed to control the quality of our MP extraction protocol (Brander et al. 2020; Möller et al. 2020). Therefore, MP particles within a size range between 2000–1000 µm and 1000–300 µm have been produced by cutting from environmental macroplastics (Weber et al. 2022) made of HDPE, LDPE, PP, PS and PMMA. We focussed on those polymers as they represent the most common produced (Geyer et al. 2017) but also the most commonly identified polymers in soil samples so far (Büks and Kaupenjohann 2020). The macroplastics have been sampled on cropland soil surfaces after a probable environmental presence of over 30 years and identified via the same analysis procedure stated above (Weber et al. 2022). For each MP size class and polymer type, we have added five particles mixed with 100 g dried forest topsoil (Cambisol) material and performed three replicate extractions following the extraction protocol. During 30 spike experiments, we found an average recovery rate of 100% for particles > 1000 µm and 92% for particles > 300 µm, comparable to the recovery rate reported by the original method developers (Crawford and Quinn 2017). Therefore, we had an underestimation of 8% within 1000–300 µm particles.

Next to general measures to avoid MP contamination during MP extraction and analysis, including the avoidance of plastic materials, the filtering (<50 µm) of all solutions used, the wearing of cotton lab coats and the constant covering of all open vessels to avoid air contaminations, we have performed blank controls during MP extraction. A blank sample has been set up after every five sample extractions, resulting in a total number of six blank samples analysed. In those blank samples, we found a contamination with single filaments (filament length: 900–1100 µm) in 50% of our blank samples. Therefore, we had a false positive rate of 0.5 particles per sample.

2.6 Statistics and data evaluation

In total, 103 MP (>300 µm) and four macroplastic (>5000 µm) particles have been extracted and analysed via ATR-FTIR. Detected MP concentrations are given in particles per kg soil dry weight. Based on the underestimation of 8% for 1000–300 µm particles and the false positive error of 0.5 p kg^{-1} for filament bearing samples, we corrected our MP concentrations according to the following equations (Eqs. (1) and (2)):

$$MP_{total=} \frac{MP_{det}}{100} \times MP_{rec} \qquad (1)$$

with MP_{total} (p kg^{-1}): total number of MPs in 1 kg dry soil corrected with recovery rate, MP_{det} (p kg^{-1}): total number of MPs detected in 1 kg dry soil and MP_{rec} (%): recovery rate difference of 8% for 1000–300 μm particles.

$$MP_{corr=}MP_{total} - MP_{error} \qquad (2)$$

with MP_{corr} (p kg^{-1}): total number of MPs recovery and blank-value corrected in 1 kg dry soil, MP_{total} (p kg^{-1}) total number of MPs detected in 1 kg dry soil, and MP_{error} (p kg^{-1}): total number of MPs detected as blank contamination. Within the result section, only MP_{corr} values are presented (Brander et al. 2020).

Statistical data analysis was performed in R (Version 4.0.3, https://www.r-project.org/) and in RStudio (Version 3.4.1; RStudio Inc.; Boston, MA, USA). Data visualization, tests for normal distribution (Shapiro–Wilk), linear regression analyses, Spearman correlation analyses, and variance analyses (ANOVA) were conducted with the standard R-packages and in "ggplot2" and "ggpubr". We interpreted statistical analysis results as significant with a p-value <0.05. Geostatistical analysis has been performed with QGIS (A Coruna, Version 3.10.11, http://www.qgis.org/) using data of the hessian municipal statistical office (Hessian State Statistical Office 2022) and administrative

area data of the Federal Agency for Cartography and Geodesy (2018).

3 Results

3.1 Microplastic abundance and composition

We found MP in all analysed LTSM archived samples independent of site location and sample age, except one single sample of LTSM site HAI1 from the year 2003. The MP concentrations within the 13 cropland topsoils range from 0 to 75.59 p kg^{-1} with an average of 29.04 ± 18.85 p kg^{-1} (Table 1). Mean relative standard deviation (RSD) from all cropland topsoil samples was 5.48%, indicating an equal data variability. In contrast, control LTSM sites show significant ($p = 0.0171$) higher MP amounts of 56.19 ± 24.42 p kg^{-1} (RSD: 13.73%). For the control forest site, we were able to receive samples of three different soil depths. Here we found the highest concentrations with 65.22 p kg^{-1} within the top Ah horizon (0–5 cm) and lower concentrations (10.87 and 32.61 p kg^{-1}) for the underlying mineral soil horizons (5–15 and 15–30 cm) (Fig. S2). The MP distribution of all samples shows a strong increase with decreasing particle size. The sizes range from 300 to 6000 μm with a mean of 1166.6 ± 1234.5 μm and a median of 650.5 μm. 38.8% of the MP particles have a size between 300 and 500 μm and

Table 1 Microplastic concentration (p kg^{-1}) in soil samples of LTSM sites and sample information

LTSM ID	Land use	Soil type	Sampled horizon[a]	Sampling depth (cm)	MP concentration (p kg^{-1}) per sampling year		
					2003–2004	2014–2015, 2019	2022
ALL	Cropland	Vertisol	Sw-Ap	30	11.44	43.48	
GUMS1	Cropland	Luvisol	Ap	30	32.11	43.48	
REI	Cropland	Luvisol	Ap	30	46.55	10.87	
RI	Cropland	Luvisol	Ap	30	10.87	64.72	
OE	Cropland	Luvisol	Ap	30	32.11	75.59	
WEI2	Cropland	Cambisol	Ap	30	57.51	21.78	
GRE1	Cropland	Luvisol	Ap	28	28.01	21.82	
UD	Cropland	Planosol	Ap	28	32.61	54.01	
ALS1	Cropland	Cambisol	Ap	25	10.87	10.87	
HAI1	Cropland	Planosol	Ap	30	0.00	32.11	
ERB	Cropland	Luvisol	Ap	27	10.87	10.98	
HÜT	Cropland	Luvisol	Ap	27	32.11	21.78	
BUBO	Cropland	Planosol	Ap	25	27.05	11.59	
SCHOT3[b]	Grassland	Cambisol	Ah	10	78.43	64.72	
ALT[b]	Grassland	Cambisol	Ah	10	86.46	55.04	
SCHOT2[b]	Forest	Cambisol	Ah	5			65.22
			Ah-Bv	5–15			10.87
			Bv	15–30			32.61

[a] According to German soil systematics (Ad-hoc AG Boden 2005)

[b] Control group sampling sites

22.3% between 500 and 900 μm (Fig. S4). Only 18.4% of the identified MP particles show a size > 2000 μm and are thus within the coarse soil fraction. The average MP size did not change significantly between older (18–19 years) and younger (3–8 years) samples (Fig. 1a). For the forest control site, average particle sizes differ with soil depth, with the largest average particle sizes in the lowest mineral soil horizon (Fig. S2).

Within cropland and control sample set, polyethylene (PE) with 38.7% including high-density and low-density PE, and synthetic rubbers including chloroprene rubber (CR) with 19.4% or ethylene propylene diene monomer rubber (EPDM) with 6.5% are the most common polymers analysed. In addition, we found resins (7.5%), polyamide (5.3%) and polyvinyl alcohol (3.2%) among different other thermoplastic polymers (16.1%). Polymeric composition was not equal for older and younger samples (Fig. 1b). While the share of CR polymers remained the same over time, we were able to detect an increase in EPDM, resin, TPR, PA and PVA polymers. On the other hand, the share of PE decreased slightly. MP particles are present as films (46.6%), fragments (31.1%) and filaments (22.3%) with white and transparent colours (54.3%) in cropland and control sample sets. Significant differences in particle properties or sizes, could not be found for cropland and control sample set.

3.2 Spatial microplastic distribution

With regard to potential spatial hotspots of MP contamination, we found clear spatial maxima of MP concentrations of the cropland sites at site OE (2014) with 75.59 p kg^{-1}, while minima include the sample of site HAI1 with a zero-detection value in 2003 and the site ERB with concentrations of 10.87–10.98 p kg^{-1} for both sampling dates (Fig. 2 and Table 1). MP size composition did not show significant differences between the LTSM sites. On a regional scale, we found highest average MP concentrations with 40.43 ± 18.34 p kg^{-1} in northern Hesse, followed by southern Hesse (32.95 ± 18.89 p kg^{-1}) and central Hesse (16.82 ± 10.23 p kg^{-1}), but no significant ($p > 0.20$) regional average differences. Unexpectedly, we found the absolute maximum concentration of 86.46 p kg^{-1} (site ALT, 2003) and average maximum concentration from both sampling dates of 78.4 p kg^{-1} (site SCHOT3) in the grassland control group (Fig. 2). Generally, control samples show a significant ($p < 0.05$) higher MP abundance with average of 56.19 ± 12.13 p kg^{-1} within the control group containing two grassland and one forest LTSM site, versus the cropland group with an average of 29.05 ± 18.85 p kg^{-1} (Fig. 3). Regarding potential spatial correlations or dependencies between MP abundance at different LTSM sites and, e.g., MP sources or transport pathways, we found no significant correlation between MP concentrations and spatial proximity to roads ($p > 0.18$), but a slight linear relationship between MP concentrations and the number of farms ($R^2 = 0.32$, $p < 0.05$) or inhabitant number on community scale ($R^2 = 0.13$, $p < 0.05$) (Hessian State Statistical Office 2022). As MP emissions can also be considered in the context of proximity to urban areas, we tested the dependencies between MP concentration and distance to urban areas with > 50,000 inhabitants (Fig. 4). With respect to soil types or soil properties (TOC, pH), no significant difference ($p > 0.08$) occurs. A strong differentiation between the cropland and control group is obviously by data density distribution, with respect to distance measures and MP concentration, respectively. Focusing on the distance to urban areas relationship, we found a slight MP
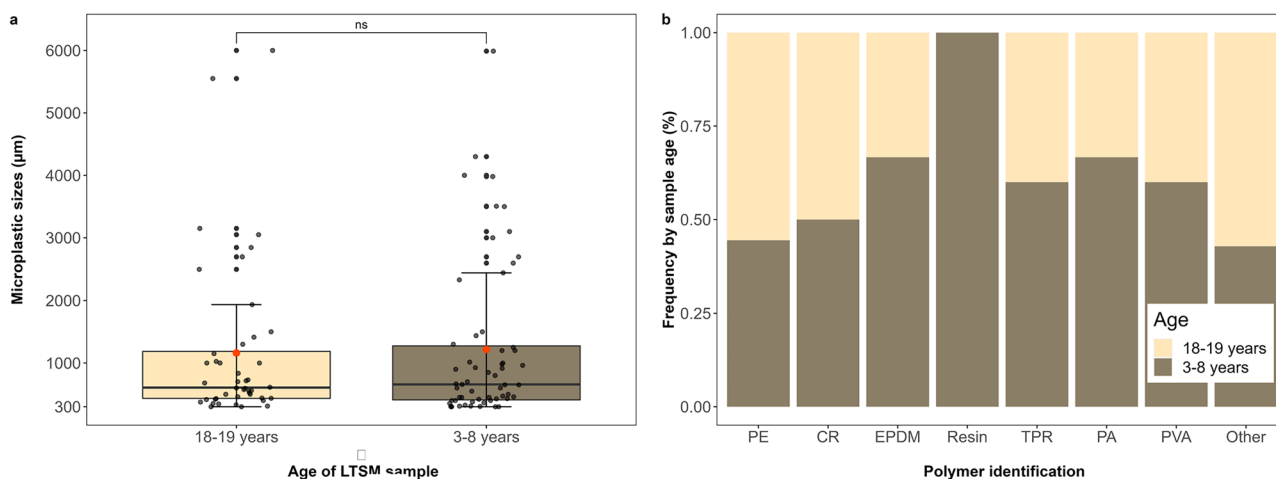

Fig. 1 Temporal trends in microplastic particle features from cropland and control sites. **a** Microplastic particle sizes (>300 μm) within "older" (118–19 year stored, $n = 44$) and "newer" (3–8 year stored, $n = 49$) LTSM samples. **b** Frequency of identified polymers according to OpenSpecy database ($n = 103$) grouped by polymer type and age of samples. ns = not significant
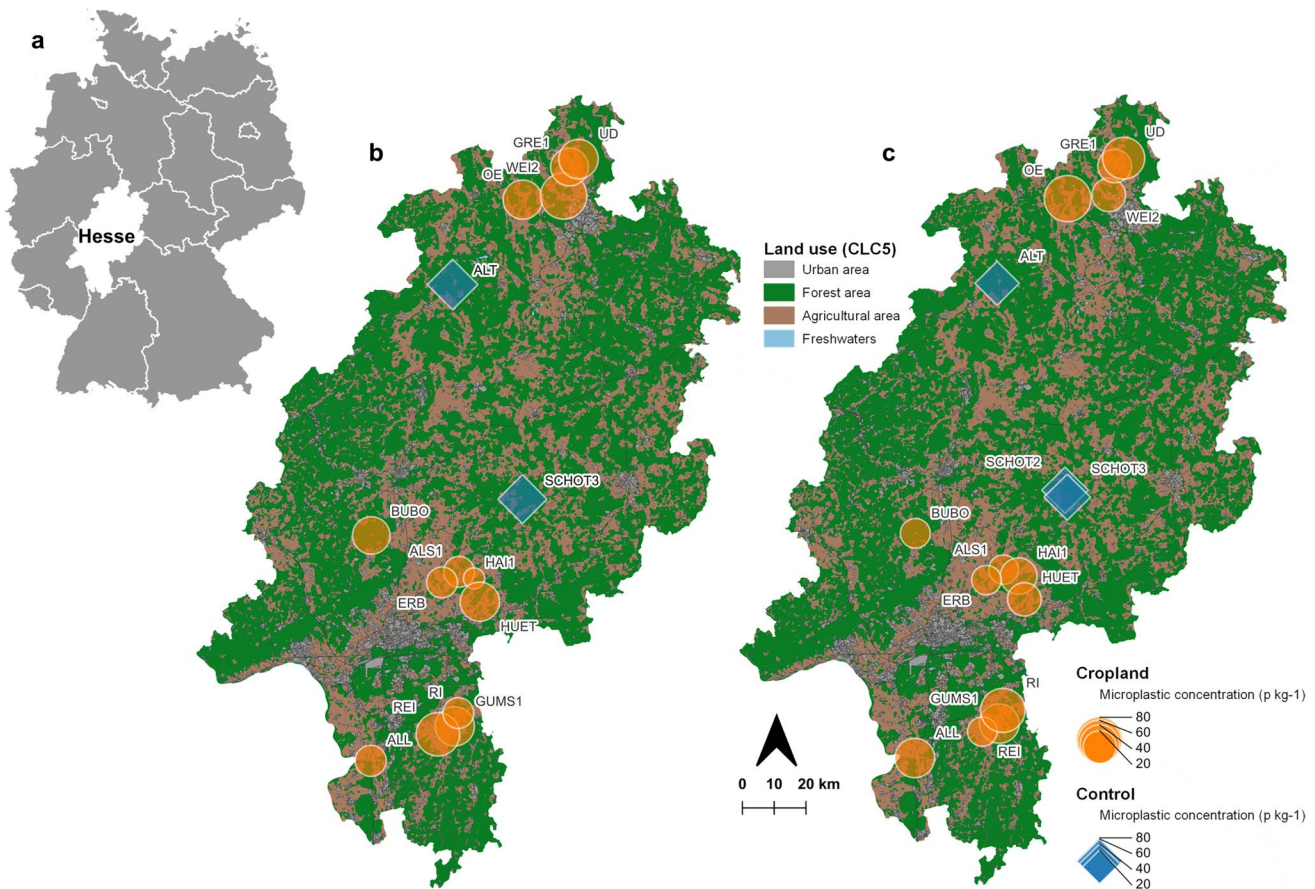
Fig. 2 Location of LTSM sites in the federal state of Hesse (Germany) and microplastic abundance. **a** Location of the federal state of Hesse in Germany. **b** Microplastic concentration in topsoil samples (p kg^{-1}) in sampling years 2004–2005 for cropland and control group. **c** Microplastic concentration in topsoil samples (p kg^{-1}) in sampling years 2014–2015, 2019 and 2022 for cropland and control group. Land use according to CORINE Land Cover 5 ha (CLC5), data received from Federal Agency for Cartography and Geodesy (2018) https://daten.gdz.bkg.bund.de/produkte/dlm/clc5_2018/aktuell/clc5_2018.gk3.shape.zi

concentration decrease with increasing distance, but no significant ($p = 0.17$) linear correlation ($R^2 = -0.28$) (Fig. 4).

3.3 Temporal microplastic trends

Regarding the temporal differences of MP contamination, we found unsteady MP concentrations within the archived samples of different ages. Considering only the cropland topsoil group, older soil samples (sampled in 2003–2004) have an average MP concentration of 25.54 p kg^{-1} (± 3.98 RSD), while younger soil samples (sampled in 2014–2015, 2019) show an average of 32.54 p kg^{-1} (± 6.86 RSD). Figure 5a shows concentration increases at seven LTSM sites and decreases at five sites, where one site (ALS1) shows constant MP concentrations. Overall, we can therefore observe an increase in MP concentrations for 53.8% of the LTSM cropland soils during a time period of 10–16 years ($n = 26$). In contrast, MP concentrations at control LTSM sites under grassland show an average decrease of 22.57 p kg^{-1} for 12 years (2003–2015).

Nevertheless, we are not able to detect a significant overreaching temporal trend of MP abundance increase or decrease (Fig. 5b). However, younger LTSM samples show an increase of 27.4% compared to the initial or past MP concentration level. Additional soil properties which were already part of analysis during the LTSM programme like pH, cation exchange capacity and total organic carbon as well as total carbon remained stable between sampling periods (Fig. S3).

4 Discussion

4.1 Spatio-temporal variability of MP in soils

MP concentrations in soils reported in the current literature depend strongly on the applied extraction protocols and MP sizes analysed (Lwanga et al. 2022). Compared to global data, the MP concentrations of LTSM sites are in the lower range of known concentrations (Büks and Kaupenjohann
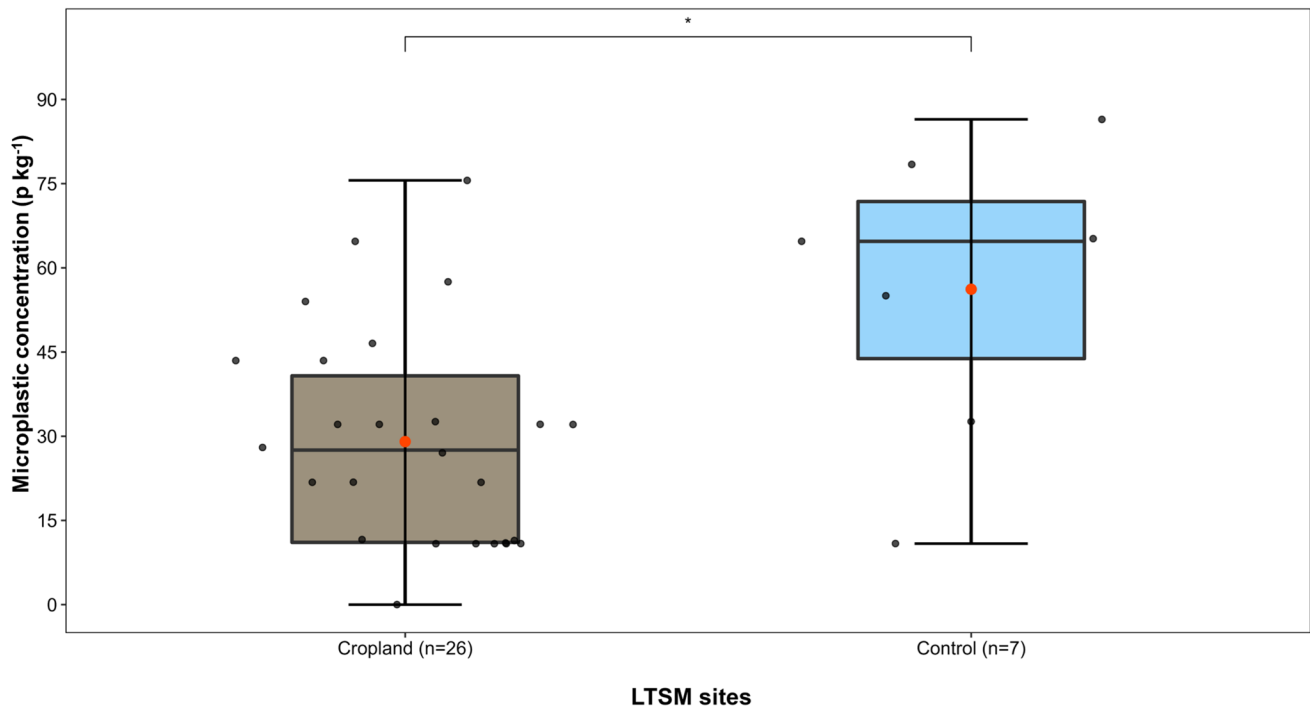
Fig. 3 Microplastic concentration (p kg^{-1}) differentiated for cropland and control LTSM sites. Control LTSM samples include topsoil samples from grassland sites (two sampling times) and three forest soil horizons (one sampling time). Significance levels: **** < 0.001; *** 0.001–0.01; ** 0.01; * < 0.05; ns = not significant. Red dot indicates mean value position

2020), mainly due to the lower detection limit of 300 μm, as MP concentrations are supposed to increase with decreasing particle sizes in the environment (International Organization for Standardization 2020; Zhang et al. 2020). Taking three regional examples from cropland sites in Germany, all using different MP extraction protocols, Tagg et al. (2022) report 845–4367 p kg^{-1} related to sewage sludge applications on study sites, while Braun et al. (2023) report 0–64 p kg^{-1} for soils with compost application and Harms et al. (2021) report 0–217.8 p kg^{-1} (average: 3.7 p kg^{-1}) for large MP (>1 mm) at different arable lands in northern Germany. Comparisons of our results with studies applying similar extraction protocols, based on NaCl density separation, for example, Cao et al. (2021) reported an average of 37.32 p kg^{-1} (>100 μm MPs) or Zhou et al. (2020) an average of 503.3 p kg^{-1} (>50 μm MPs), both for cropland soils in China partly with mulching application. Overall, it becomes obvious that the measured LTSM MP concentrations are in a comparable range within a regional level of known concentrations.

MP composition in LTSM samples follows known particle and polymeric composition in cropland topsoils (Cao et al. 2021; Kumar et al. 2020) especially for the observed increasing particle number with decreasing particle size (Fig. S4). This seems to be one of the most common MP data features regardless of environmental compartment

studied, including soils, sediments or water (Cole et al. 2011; Zhang et al. 2020). The large PE shares within our samples are comparable to European plastic production data (2010–2019), where PE is the most produced polymer (PlasticEurope 2020). Again, methodological limitations such as the applied density separation using NaCl-solution (1.2 g cm^{-3}), lead to an underestimation of high-density polymers such as PET or PVC (Cutroneo et al. 2021). The comparable high abundance of synthetic rubbers (e.g., CR, EPDM and resins) could be traced back to a couple of items used for agricultural purposes like seals, hoses, or abrasion from machine paints (Domininghaus 2012; Lwanga et al. 2022).

With regard to the spatial distribution patterns, we were able to identify spatial hotspots of MP contamination within the federal state of Hesse. Here, it should be highlighted that those hotspots are located partly in rural regions at a greater distance to municipal areas and follow no clear identifiable spatial pattern, based on the limited LTSM site number analysed. This contradicts previous findings assuming higher MP concentrations in proximity to municipal areas (Brandes et al. 2021; Choi et al. 2021). Overall, we could not determine a clear spatial dependency between MP concentrations and potential MP sources (e.g., streets, rivers) or possible indicators such as the number of inhabitants. This result underlines the spatial heterogeneity of MPs in soils (Zhang
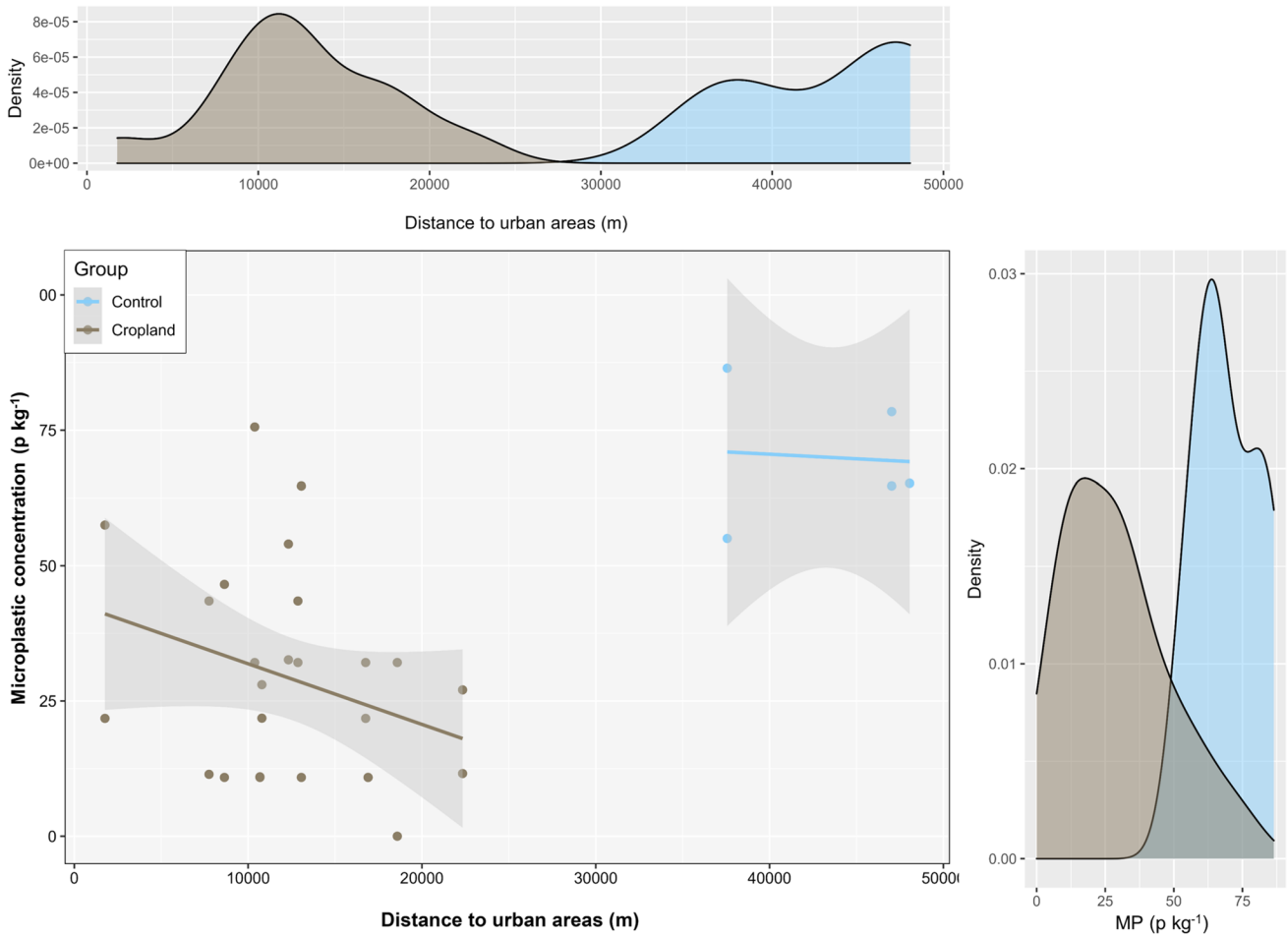
Fig. 4 Microplastic concentration (p kg^{-1}) for cropland and control LTSM sites at an increasing distance from urban areas (cities >50,000 inhabitants) with marginal data distribution as density plots for x- and y-axis

et al. 2020), but also the limited spatial representativeness of the selected LTSM sites for a whole federal state. However, this finding could emphasize the role of local point sources

as well as management practice for MP entries to arable soils (Duis and Coors 2016; Lwanga et al. 2022). The decrease of MP concentrations with increasing distance from urban
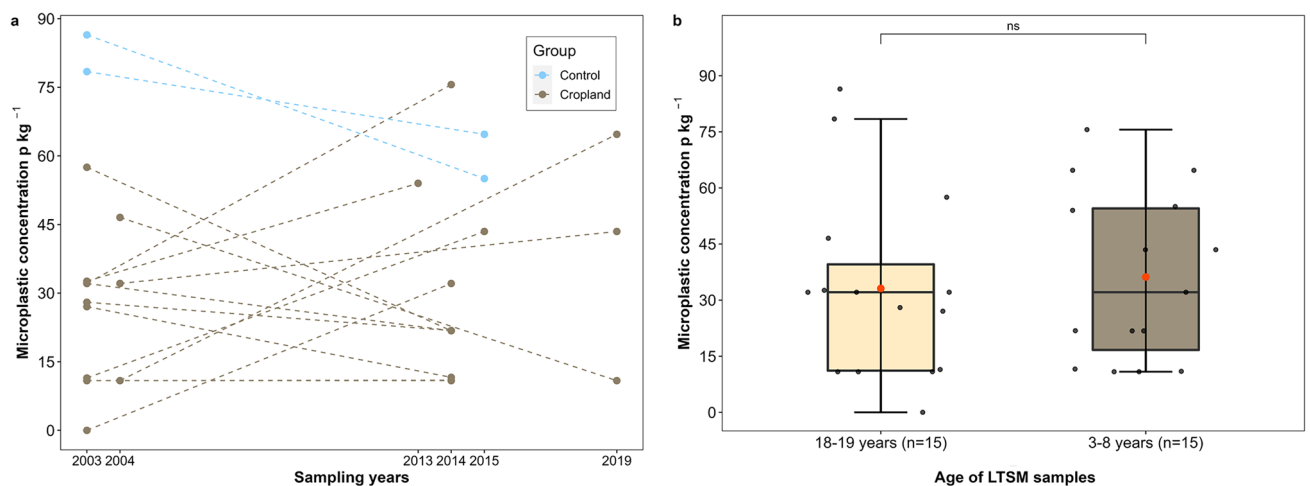
Fig. 5 Temporal trends within LTSM microplastic abundance with **a** Absolute microplastic concentrations (p kg^{-1}) at each LTSM site at the respective sampling years, without forest control site SCHOT2;

b microplastic concentrations (p kg^{-1}) of "older" (18–19 year stored) and "newer" (3–8 year stored) LTSM samples. ns = not significant

centres, could furthermore indicate the underlying input of MPs through littering, depending on the regional inhabitant density (Choi et al. 2021; Cowger et al. 2022), but also shows the role of rural areas as MP reservoir whose sources might be currently undetermined.

As the majority of MP soil studies has been conducted on single arable fields or fields under the same management practice (Weber and Bigalke 2022), we can derive implications from MP concentration differences according to land use practice. Even if data about management practice on LTSM sites is missing, other studies reported differences in MP abundance primarily between intensive and non-intensive agricultural practice including dependence on crop rotation, use of fertilisers or plastic materials (e.g., mulch films) (Corradini et al. 2019; Huang et al. 2020; Yang et al. 2021). Unexpected was the finding that our control group containing grassland and especially forest soils with particles >500 µm (Fig. S2), show partly higher MP concentrations as the regular sample set from cropland soils. Contrary to these results, Feng et al. (2021) found clearly lower concentrations in grassland soils (<30 p kg^{-1} on average) than in cropland soils (>30 p kg^{-1} on average) of the Qinghai-Tibet plateau. Even if the significance of our findings for grassland and forest soil is strongly limited, due to the small sample number analysed here within the control group ($n=4$ for grassland soil; $n=1$ including 3 soil horizons for forest soil), this circumstance should be taken notice. For forest soils, Choi et al. (2021) reported an abundance of 20 p kg^{-1} (<1000 µm) for a single forest soil within the Republic of Korea, while Xu et al. (2022) report average concentrations of 637.5 ± 181.6 p kg^{-1} for primary tropical forests within China. For European forests, there are currently no known evidence of MP in soils (Büks and Kaupenjohann 2020; Weber and Bigalke 2022), except of the SCHOT2 LTSM site MP abundance, presented here. The decrease in MP concentration and increase in particle size along soil depth within the forest control site, could be traced back to the high MP inputs potentially by atmospheric deposition already found for forest systems (Klein and Fischer 2019) and the adhesion to soil organic matter in forest Ah-horizons (Hurley et al. 2018). Since, in contrast to arable land, no tillage takes place here, the MP evidence in deeper horizons must be due to displacement processes of MP particles (e.g., bioturbation, leaching) (Bigalke et al. 2022; Ren et al. 2021).

Similarly, temporal trends of MP abundance in soils have been underestimated in soil-related studies so far (Dahl et al. 2021), in contrast to other environmental systems like marine or limnic systems (Dong et al. 2020; Li et al. 2020). Based on data from these systems, we assumed an increase of MP concentrations over time, in line with the global plastic production increase (Geyer et al. 2017; Martin et al. 2022). For our best knowledge, the temporal results presented above are the first temporal assessments of MP contamination in soils. Within the LTSM MP dataset, we can trace an increase of MP concentrations over time for 53.8% of all samples for a maximum time span of 16 years. Even if increases are not significant in the mean comparison, our result stays comparable to the initial assumption of increasing MP concentrations over time. Based on regional market data, the increase in European plastic production between 2010 and 2019 is small, but a significant increase over 32% can be seen in the collected plastic waste for Germany between 2006 and 2018 (PlasticEurope 2020). This indirectly reflects the increased consumption of plastic materials on a regional scale, which could be one of the responsible factors for the MP concentration increase in parts of the LTSM samples. However, the also observed decrease over time, especially at grassland control sites, also indicates the high variability of MP abundance in soils. This could be traced back on the one hand to spatial heterogeneities (Möller et al. 2020) of MPs in the sampled LTSM sites, which would indicate a more or less constant MP abundance over time. On the other hand, we could assume an overall dynamic MP presence in topsoil, which is subject to dynamic MP inputs (Lwanga et al. 2022), biogeochemical MP degradation (Kale et al. 2020) and leaching to deeper soil layers (Bigalke et al. 2022). Regarding the degradation or ageing of MP in soils, it could be assumed that microplastic particle sizes should decrease over time, based on disaggregation (Ren et al. 2021). However, we found no significant change in particle size over time (Fig. 1), which may be due to the large share of PE polymers and their particularly slow disaggregation within soils (Binda et al. 2023). Therefore, we conclude that cropland topsoils act as MP filters and can partwise accumulate MP over time, which nevertheless underlies dynamic processes (Ren et al. 2021) that should be investigated in more detail under field conditions, to reveal open questions on temporal MP trends in soils.

4.2 Implications for MP in long-term soil monitoring programmes

Here, by our best knowledge, we present the first analysis of MP within archived soil samples from a LTSM programme. As stated above, the harnessing of those special soil sample databases makes it possible to investigate both spatial and temporal occurrence as well as changes within MP abundance and characteristics over time. Major advantages of LTSM programmes for MP analysis are (i) the availability of soil samples mostly spatial representative across different administrative units (e.g., federal states) or soil formations and soilscapes (Toschki et al. 2015); (ii) the availability of a sample time series with consistent sampling and treatment protocol (Umwelt Bundesamt, 2022) and (iii) the availability of pedogenetic and soil property data along for each LTSM

site, which could support the so far underrepresented documentation of soil properties within MP research (Weber and Bigalke 2022). Moreover, the applied sampling protocol within the German LTSM programme, including an area-based composite sampling (cross wise within defined area) seems to be well suited for heterogenous distributed MP from heterogenous sample matrices (Yu and Flury 2021).

Despite these advantages, it must be clear that LTSM programmes have not been developed for the investigation of MP, which means that sampling and sample treatment protocols do not intend the minimization or avoidance of sample contamination by MP (Brander et al. 2020). This leads to the disadvantage of possible sample contamination during field sampling and later sample preparation, as

common precaution measures (e.g., cotton clothing, laminar flow working places) are not intended (Pérez-Reverón et al. 2022; Silva et al. 2018). However, at least the usage of stainless-steel and glass equipment for sample extraction and transport promotes contamination prevention (Möller et al. 2020; Prata et al. 2019). To overcome these circumstances, each potential MP contamination should be traced as far as possible, by e.g., field and laboratory blank control during future LTSM sampling, if MP analyses are planned. Additionally, LTSM sample archives only contain a certain amount of sample material. In our case, the applied extraction protocol has required a total amount of 100 g dry-soil material, leading to the fact that only archived samples with enough soil material could be analysed, resulting in breaks

Table 2 Checklist for microplastic analysis from archived soil samples.

Points to be checked		Further recommendations
Sample and data access Do I get access to sample material and data?	✔	• Make clear agreements and, if necessary, contracts with the LTSM operator about sample analysis and data usage
	✘	• Go for a personal discussion and explain the benefits that your research offers for the LTSM programme itself (e.g., public visibility)
Structure of the LTSM programme Does the LTSM programme have a high spatial resolution for its target area?	✔	• Research question-guided selection of LTSM sites
	✘	• If necessary, take additional own samples in unrepresented areas
Temporal sample availability Are all samples from all intended years available?	✔	• Ensure that equal time intervals are available for all selected samples
	✘	• Check for alternative or modified extraction protocols (e.g., with lower sample mass)
LTSM sampling procedure Has a minimum of contamination control (e.g., stainless-steel tools) been applied?	✔	• Check for further MP contamination sources and try to include these in QA/QC[a] assumptions
	✘	• Check carefully whether samples are still suitable • Strictly control possible MP contamination during QA/QC[a]
LTSM archive characteristics Were the samples divided representatively and stored in glass jars?	✔	• Check integrity of stored samples for QA/QC[a]
	✘	• Check carefully whether samples are still suitable • Strictly control possible MP contamination for QA/QC[a]
Available LTSM datasets Are additional data such as soil characterisations, nutrient contents, pollutant contents available?	✔	• Examine which datasets can be analysed together with the MP data
	✘	• Collect own data if necessary (strongly dependent on research question)

[a] Quality assurance and quality control

in the temporal continuity of the samples analysed. We therefore recommend the application of different state-of-the-art extraction protocols, which require a lower sample volume (Möller et al. 2020; Zhang et al. 2020). Finally, we found a clear drawback in unavailable or limited data about land management. In general, MP concentration variances within a soil but also changes over time could be traced back to changing land management with regard to MP emissions (e.g., fertilization practice or the use of mulch foils) (Kumar et al. 2020). Here, as in our case, only insufficient conclusions can be drawn due to the lack of management data.

Going further, the dynamics of MP emissions and behaviour in soils over time is so far totally unknown. Referring to the global plastic production development, with a clear exponential growth since the early 1950s, a time span covering the last 70 years is of special interest for MP research (Geyer et al. 2017; Porta 2021). If we try to understand the role of soils as reservoirs and/or interfaces within global MP material cycles, several questions could be asked about the starting time of MP deposition in soils and for the enrichment over time. Since MP is a "newly" discovered environmental contaminant (Andrady 2017), only archived soil samples can offer the opportunity to investigate past MP depositions. Globally, comparable programs or soil sample archives are available. Exemplarily, we could name the Australian National Soil Archive (Australian National Soil Archive—CSIRO) with over 40,000 archived samples back to 1924, the National Soil Archive of Scotland (National Soils Archive | Facilities | The James Hutton Institute) with over 48,000 archived samples back to 1934 or the Rothamsted Sample Archive (The Rothamsted sample archive | Rothamsted Research) with about 300,000 samples back to 1843, among others. Those examples illustrate the underlying potential of archived soil samples to understand the "historical" development of MP contamination in soils and its spatial distribution globally.

Based on our own experiences presented above, we suggest the following checklist if planning MP analysis and the usage of data from archived soil samples out of LTSM programmes or other sample archives in future (Table 2).

5 Conclusion

Our results show for the first time the presence of MP in LTSM archived samples and imply a ubiquitous MP contamination of topsoils independent of spatial location, land use and time in the millennium. Polymeric composition and the increasing particle number with decreasing size are clearly consistent with previous data for MP in arable soils. Slightly increasing MP concentrations over time underline the role of soils as MP reservoirs, but also indicate the dynamic changes in MP abundance within soils.

Unexpected high MP concentrations in previously not contemplated control group of grassland and forest soils, suggest that also non-agricultural soils in rural areas could have a major role in MP storage over space and time within terrestrial systems. Basically, we were able to demonstrate that MP analysis from archived samples of LTSM programmes is possible and suitable to gain spatio-temporal MP data. Nevertheless, both MP analytical method regarding lower particle size detection limit and polymer recovery as well as the sample quantity investigated can be significantly expanded. However, applying MP analytical tools to LTSM samples in the future could enable first conclusions about soils MP pollution sources based on non-targeted spatial source analysis which goes beyond cropland areas. Additionally, it will be possible to monitor temporal trends of MP pollution in soils. Furthermore, legislative measures for precautionary soil protection against MP contamination need information on soil background levels of MP in addition to ecotoxicological metrics, to enable appropriate risk assessments in the future. Comparable MP extraction protocols and analytical frameworks, together with the use of LTSM programmes worldwide, can be a powerful tool for further soil protection measures.

Declarations

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