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# Probabilistic environmental risk assessment of microplastics in soils

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#### ABSTRACT

Despite the convenience that plastics provide to society, there have long been concerns regarding the risks they present to the environment. Microplastics (MPs, plastic particles smaller than 5 mm) have been found in every environmental compartment—air, freshwater, oceans, but also soils—yet the emphasis to date has been on the risks they represent to aquatic environments. The present study, therefore, aimed to perform a comprehensive risk assessment of MPs in soils. A probabilistic approach was used to account for the variability and uncertainties in the available data. Measured exposure concentrations and ecotoxicity thresholds were extracted from pertinent peer-reviewed literature. Risk characterization ratios (RCRs) were then calculated for different land uses and geographical regions by dividing the predicted no-effect concentration (PNEC) distribution by the respective measured environmental concentration distribution. Using particle-number concentrations, the mean PNEC was calculated to be 82,000 part.kg<sup>-1</sup>. The measured concentrations of MPs in soils ranged from 0 to 410,000 part.  $kg^{-1} \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; with \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; influence \; was \; clearly \; demonstrated, \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. \\ kg^{-1}). \; A \; human \; higher \; concentrations \; (median; \; 930 \; part. )$ dian: 3,600 part.kg<sup>-1</sup>) in soils directly influenced by human activity than in natural or agricultural soils. 4.8 % of RCR calculations (median: 0.013) of the probability distribution were above 1, which is 40 and 240,000 times greater than that predicted for freshwater and marine habitats, respectively. Urban and industrial soils had the highest RCR, followed by agricultural and natural soils. The comparability of exposure and hazard datasets could be improved if the MPs tested for ecotoxicity were more representative of those found in environmental samples. There is a need for more ecotoxicity data on fibers, films, polyethylene, and weathered or aged MPs so that comparisons with real-world, observed exposure data can be built on more solid foundations.

## 1. Introduction

Plastics present various interesting properties, such as high malleability, low mass, and high corrosion resistance. These make them the materials of choice for a large range of applications. Despite their convenience, evermore concerns are being raised regarding the risks they pose to the environment (Priya et al., 2022; Parker, 2019). Microplastics (MPs) are one class of pollutants on which research has increased tremendously in the past few years (Klingelhöfer et al., 2020). Their common point is a size smaller than 5 mm in diameter (GESAMP, 2015), but they are also diverse in size, shape, and how and where they were produced. Primary MPs are produced deliberately for use as beads or pellets, e.g., in hygiene and abrasive products. Secondary MPs result from weathering, e.g., by ultraviolet radiation or the thermal oxidation of larger plastic products, such as packaging and textiles, and their subsequent fragmentation (Nasseri and Azizi, 2022; Horton et al., 2017; Andrady, 2011). Secondary MPs are mainly fragments and fibers.

The main release pathway for plastics is to the terrestrial environment. Worldwide, it is estimated that plastic pollutants are 4 to 23 times more abundant in soils than in oceans (Horton et al., 2017). In Switzerland alone, emissions to soils were estimated to be 40 times higher than to surface water (Kawecki and Nowack, 2019). After use, plastics and MPs find their way to the terrestrial environment via different routes (Rillig, 2012). Littering is the main route, but construction and agriculture are also significant sources (Kawecki and Nowack, 2019). In agriculture, MPs find their way into soils, e.g., through sewage sludge application (Corradini et al., 2019; Nizzetto et al., 2016; Zubris and Richards, 2005), irrigation with wastewater (Bläsing and Amelung, 2018; Zhang and Liu, 2018), and the application of plastic mulching film (Huang et al., 2020). Tire-wear particles are another large source of the MPs that enter soils and rivers via runoff from the streets where they are produced in the first place (Campanale et al., 2022; Bläsing and Amelung, 2018; Sieber et al., 2020; Chen et al., 2020).

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Microplastics can alter soil properties (e.g., their water-holding capacity or soil bulk density), microbial activity (de Souza Machado et al., 2018b), enzyme activity (Zhao et al., 2021), and the composition of microbial communities (Seeley et al., 2020). Moreover, their leachates can negatively impact soil biota (Kim et al., 2020). Microplastics are also directly ingested by organisms present in soils, such as earthworms (Huerta Lwanga et al., 2017), snails (Panebianco et al., 2019), or collembolans (Maass et al., 2017), most likely because these animals mistake these particles for food (Cole et al., 2013). After ingestion, MPs can cause false satiation and deleterious effects on the digestive system, such as damage to the esophagus and intestinal obstruction (Ju et al., 2019; Song et al., 2019; B.-K. Zhu et al., 2018; D. Zhu et al., 2018). Eventually, MPs affect organisms' reproduction, growth, and survival (Cao et al., 2017; Lahive et al., 2019). Jiang et al. (2019) showed growth inhibition in Vicia faba in the presence of polystyrene MPs, and Bosker et al. (2019) observed reduced germination rates and decreased root length in Lepidium sativum after exposure to MPs. Smaller particles can also be taken up by plants, including edible crops (Li et al., 2020a; Li

Given the adverse effects of MPs on soil organisms, toxic doses of MPs should be compared to the real-world environmental concentrations to which they are exposed to understand their potential impacts fully. Risk assessment methodologies compare exposure concentrations and toxicity doses. Microplastics risks have been assessed in marine waters using modeled environmental concentrations and ecotoxicity data extracted from the literature (Everaert et al., 2018). These authors concluded that MPs in surface marine environment would probably not create a risk before the year 2100.

Adam et al. (2018, 2020) used a probabilistic approach to consider the uncertainties in their risk assessments of MPs in freshwaters and marine waters, respectively. Risks were shown to be unlikely based on the available data. Burns and Boxall (2018) aggregated both freshwater and marine water in their assessment, also concluding that MPs' risks to aquatic organisms were unlikely, although they highlighted the limited data available. Mehinto et al. (2022) proposed a framework for categorizing the risks of MPs in aquatic systems by the level of concern. More recently, Redondo-Hasselerharm et al. (2023) also assessed the risks of MPs in freshwater sediments, reporting that there was a potential risk to benthic communities.

However, to date, risk assessments of MPs in soil systems have been very limited. To the best of our knowledge, only one study has conducted a complete environmental risk assessment of MPs in soils (Jacques and Prosser, 2021). These authors built environmental exposure distributions and species sensitivity distributions (SSDs) based either on no-observed effect concentrations (NOECs) or lowest observed effect concentrations (LOECs), and risks were calculated considering the overlap between the distributions. Their results showed that the current MP levels in soils might affect 7 %–28 % of species 5 % of the time.

Given the need for an updated risk assessment of MPs in soil, the present study's aim was to use a probabilistic species sensitivity distribution (pSSD) model to shed further light on the possible risks of MPs on soil organisms. The probabilistic species sensitivity distribution was built by using uncertainty factors to consider different endpoints (NOEC, LOEC, and the highest observed no-effect concentration or HONEC) and to transfer acute toxicity to chronic toxicity endpoints. As per Adam et al. (2018), this approach was used to include the uncertainties and variabilities associated with the experimental data available in the peerreviewed literature. The risk assessment also considered the percentage of overlap between the two distributions and suggested RCRs that considered different land uses and locations for the first time. The adequacy of these datasets for risk assessment needs is discussed in order to identify potential research gaps that need to be addressed to obtain the most accurate risk assessment possible.

#### 2. Materials and methods

We performed a terrestrial risk assessment of MPs at the global scale, incorporating types of land usage and locations that were then compared to identify risk hot spots. Using the Web of Science search engine, we analyzed the peer-reviewed literature published up to August 2021 to extract exposure concentrations and ecotoxicity doses. Effects due solely to the leaching of additives or sorbed pollutants were excluded from this assessment as it was focused on the direct risks of MPs. All the data on plastic particles smaller than 5 mm in diameter or length was examined, but data on nanoplastics (particles smaller than 1  $\mu$ m) were excluded since their toxicity mechanisms in terrestrial systems are different (de Souza Machado et al., 2018a).

After data collection, probability distributions associated with exposure and effect concentrations were built. These were then compared to assess their potential risks. All calculations were made in R software (R Core Team, 2019) using the probabilistic risk assessment method developed by Gottschalk and Nowack (2013) and modified by Wigger et al. (2019) (pSSD<sup>+</sup>). The "trapezoid" (Hetzel, 2022), "mc2d" (Pouillot and Delignette-Muller, 2010), "mvtnorm" (Genz et al., 2021), "psych" (Revelle, 2022), "xlxs" (Dragulescu and Arendt, 2020), "msm" (Jackson, 2011), "stringr" (Wickham, 2022), "ggplot2" (Wickham, 2016), "car" (Fox and Weisberg, 2019), "tidyverse" (Wickham et al., 2019), "rstatix" (Kassambara, 2021), "ggpubr" (Kassambara, 2020), and "FSA" (Ogle et al., 2022) packages were all used. The general methodology used for this risk assessment was similar to the method used for MP risk assessment in freshwater and marine waters by Adam et al. (2018, 2020). The details of this method are described in the following sections.

#### 2.1. Hazard assessment

The terms "microplastic", "soil", "terrestrial", "ecotoxicity", "toxicity", and combinations thereof were used to search the internet for hazard data. The regulatory guidelines for ecotoxicity on terrestrial organisms define three soil organism groups that should be covered by hazard data for a comprehensive assessment. These include plants, invertebrates such as earthworms, springtails and mites, and microorganisms like bacteria, protozoa, and fungi. This ensures that all the relevant ecological levels and exposure pathways to stressors in a terrestrial environment are considered (ECHA, 2017). The preferred endpoints considered for inclusion were growth, survival, and reproduction; thus, data for soil, animals, and plants were collected. To ensure the quality of the dataset used for risk assessment, several criteria (e.g., lack of information on polymer type or shape, differentiation of soil and organism matrix, focus of the study) were used to exclude studies considered irrelevant (Table S1).

The hazard dataset was then used to build pSSDs following the pSSD<sup>+</sup> methodology developed by Wigger et al. (2019) that uses uncertainty factors (UFs). First, an uncertainty factor-dose descriptor (UF<sub>D</sub>) was used. NOEC data points were derived from the literature since this is the preferred dose descriptor in REACH regulations (ECHA, 2008). When NOEC data were not available, LOEC and HONEC data points were derived and then converted to NOECeq using the UFD. To derive NOECeq from LOECs (ECHA, 2008) and HONECs, UFDs of 2 and 1 were used, respectively. HONEC values were only included if no other descriptors were reported, and only if they were higher than 0.1 g.kg<sup>-1</sup> thus ensuring that the lowest part of the pSSD would not be skewed by highly uncertain data. Secondly, an uncertainty factor-time (UF<sub>T</sub>) was used when chronic data—which is preferable to acute data (ECHA, 2008)—were unavailable. A threshold duration for each species was considered for the UF<sub>T</sub>. If the experimental duration was higher than the threshold duration derived from the guidelines (Table S2), UF<sub>T</sub> was taken as 1, whereas it was taken as 10 when the experimental duration was below the threshold value. Endpoints were also considered when determining the UF<sub>T</sub>. For example, if the guideline specified the threshold duration according to an endpoint, such as mortality or reproduction, then the study's endpoint was considered accordingly to determine  $UF_T$ . Considering these uncertainty factors, the  $NOEC_{eq}$  was derived by using Equation (1).

No observed effect concentration equivalent

$$= \frac{\text{Dose descriptor data point}}{\text{Uncertainy factor\_time*Uncertainty factor\_dose descriptor}}$$
 (1)

Second, probability distributions were calculated for each species in the dataset by considering inter-laboratory variations and uncertainties due to UFs. From these distributions for each species, multiple pSSDs were formed using the Monte Carlo simulation. Finally, the probability distribution associated with the predicted no-effect concentration (PNEC) was calculated by combining the fifth percentiles of each pSSD into one vector (ECHA, 2008), which results in a probabilistic PNEC distribution instead of a single value. Both mass-based and particle-based MP levels were considered for the pSSD distributions.

Both metrics are needed because hazard studies often report MP concentrations as mass-based, and exposure studies report them as particle-based. To make the results comparable, mass-based data was transformed into particle-based data by considering the density and volume of the MPs used. If the density of the MP used in the study was not reported, the density values were derived from Plastics Technology (n.d.). For volume determination, spheres, fragments, and films were all modeled as spherical, and fibers were modeled as cylindrical (considering both length and diameter). If the research papers did not define shape, but scanning electron microscopy images (if available) showed an irregular shape, the volume was modeled as spherical. If the size of the particle was not reported, but the size distribution was, the mean value was considered. For instance, if the size was reported as "< 250  $\mu m$ ", it was considered to be 125  $\mu m,$  or if it was reported as "20 %  $<100~\mu m$ and 80 % 100–200  $\mu m$ ", then the value was calculated as 130  $\mu m$  based on an appropriate weighting of these ranges.

The effect of particle size on mass-based and particle-based toxicity was also checked. The relationship between the size and the particle number was also assessed for particle-based toxicity to see if any of the noted effects were due to the increase in particle number as particle size decreased. To test this, the particle-based toxicity of the largest-sized MP was taken as a reference point and the particle number was set to 1. Particle numbers for the same mass of smaller-sized MPs were calculated by considering each particle's volume. Toxicity values were then adjusted by dividing the reference point by the calculated particle number for each size of MP.

## 2.2. Exposure assessment

For our exposure assessment, the peer-reviewed literature was searched using the terms "microplastic", "soil", and "terrestrial". Most of the studies retained reported measured environmental concentrations (MECs) as the particle number per soil mass. Since only a few MECs on a mass-per-mass basis were provided, we preferred the particle-based unit of measurement and used this to conduct the exposure and subsequent risk assessments.

To ensure the quality of the final exposure assessment dataset, we defined exclusion criteria, data extraction and harmonization methodologies, the calculation of proportions of shapes, and polymer compositions (Tables S3, S4, and S5). The uncertainty and variability of the measured concentrations reported in the literature were used to build probability distributions. Normal distributions were built when mean and standard deviations were provided, whereas triangular distributions were calculated when minimum, maximum, and mean concentrations were given for a single location. In most cases, however, only single values were reported, in which case no probability distribution was calculated. Monte Carlo simulations were used to calculate cumulative exposures by using all the data points and their associated probability distributions. Data points produced by iteration that were higher than

real, measured concentrations (plus their standard deviation, if available) were removed from the distributions.

The present study considered three types of land use: agricultural soils, natural soils, and urban and industrial soils. All soils used for agricultural activities involving crops, including orchards and greenhouses, were listed under agricultural soils, regardless of the sampling area's location. Natural soils grouped samples taken from locations with no reported human impact, mostly consisting of forest and floodplain soils. However, when a human impact was reported close to the sampling point's location, or if a human influence was clear in photos of the sampling site, data points were excluded from this category and included in the urban and industrial soils category. It should also be noted that if no information was available regarding the sampling point's surroundings, and in case it was reported as forest or woodland, data points were included in the natural soil category. Samples of urban and industrial soils came from residential areas, recreational spaces, roadsides, industrial zones, wastelands, and areas where there was soil management (e.g., managed pine plantations) or animal activity (e.g., pastures). The continental locations of the samples were also considered to be used in the geographical comparison.

Probability distributions and single values associated with land use and location were then plotted as cumulative curves and combined into overall probability distributions representing the exposure probabilities in various types of land use, and in different locations.

#### 2.3. Risk assessment

The potential risks of MPs in soils were characterized using two approaches. The first consisted in plotting the exposure and hazard probability distributions on the same graph. If the two curves overlapped, meaning that the maximum MEC was higher than the minimum PNEC, a risk was expected. If there was no overlap, risks were considered unlikely, given current knowledge. The second means of characterizing a risk was to calculate the risk characterization ratio (RCR) by dividing the MEC probability distribution by the PNEC probability distribution, as shown in Equation (2) (ECHA, 2016). A risk was expected if the RCR was greater than or equal to 1 (RCR  $\geq$  1), meaning there was a chance that the MEC exceeded the PNEC.

$$RCR = \frac{Measured environmental concentration distribution}{Predicted no - effect concentration distribution}$$
 (2)

# 2.4. Dataset comparability

The shape, polymer type, and size of MPs were also collected from the hazard and exposure studies to compare their datasets. After considering the main shapes used in ecotoxicity studies and found in exposure assessments, we formed five different groups of shapes for comparison, representing fibers, foams, films, spheres, and fragments. Sheets and foils were combined under films; granules, beads, virgin MPs, powder, pellets, particles, and balls were considered under spheres; columns, flakes, and platelets were considered under fragments. For examining hazard studies, if MPs had been ground, milled, cut, or shredded, they were assumed to be fragments. If scanning electron microscopy or transmission electron microscopy images were available, but the shapes were unclear, these data points were excluded from the shape-toxicity assessment to prevent uncertainty, even though they were included in the hazard assessment, as described in Section 2.2. Regarding the shares of MP shapes, if there was no information about a specific type of shape, it was assumed not to be present since all the main shapes defined in the literature were covered in the datasets formed. Regarding the types of MPs, we formed seven different groups: polystyrene (PS), polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polyamide (PA), polyethylene terephthalate (PET), and polyester together (as PES), and "others". When a value was not reported for a specific type of polymer (e.g., PP), it was not included in the calculation,

averaging, or weighting.

#### 3. Results

#### 3.1. Hazard assessment

The species included in the present dataset for pSSD calculations generally agreed with the REACH guidance, covering the major taxonomic groups of plants and invertebrates and providing suitable endpoints, including 63 toxicity values from 16 species. Most of these data points were HONECs (n: 36), followed by LOECs (n: 19), and NOECs (n: 8). The whole dataset is available in Supplementary Information 1 (Table S6). Probabilistic species sensitivity distributions and PNEC distributions calculated in mass-based and particle-based concentrations are shown in Fig. 1A and 1B.

When using mass-based concentrations, Zea mays L. was the least sensitive species, with a HONEC:  $100~\rm g.kg^{-1}$  (NOEC<sub>eq</sub>:  $100~\rm g.kg^{-1}$ ), while L. sativum was the most sensitive (Fig. 1A), with LOEC: 0.2 and a HONEC:  $0.2~\rm g.kg^{-1}$  (NOEC<sub>eq</sub>:  $0.1~\rm and~0.2~\rm g.kg^{-1}$ , respectively). When using particle-based concentrations, however, Allium fistulosum became the least sensitive species (LOEC:  $6.8\cdot10^9~\rm part.kg^{-1}$ , NOEC<sub>eq</sub>:  $3.4\cdot10^9~\rm part.kg^{-1}$ ), and Eisenia andrei was the most sensitive species (HONEC/NOEC<sub>eq</sub>:  $3.900~\rm part.kg^{-1}$ , Fig. 1B).

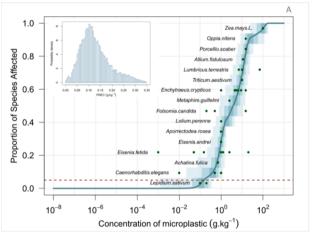
Since HONECs are unreliable dose descriptors, calculations were

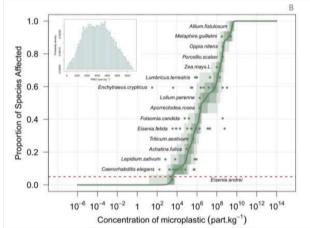
repeated excluding them (Fig. 1C and 1D), thus decreasing the total number of species from 16 to 7. Many of the species were lost in the cluster of least sensitive species because they only had HONEC values. Lumbricus terrestris (NOEC<sub>eq</sub>: 70 g.kg<sup>-1</sup>) became less sensitive than A. fistulosum, and this shift was due to the removal of five lower NOEC<sub>eq</sub>. There was also a shift among the most sensitive species because Eisenia fetida had relatively high HONEC data points. When using particle number concentrations, the least sensitive species did not change, whereas Caenorhabditis elegans became the most sensitive species, as E. andrei had no data points except HONECs.

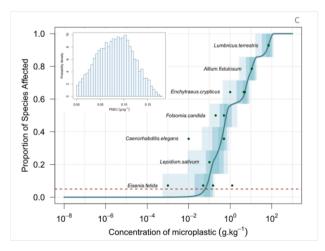
The predicted no-effect concentration were extracted from the pSSDs presented in Fig. 1. When all the data were included, the PNEC distribution calculated based on particle number concentrations presented a mean: 3,300 part.kg $^{-1}$  (Table 1). This value shifted slightly higher when HONECs were excluded (82,000 part.kg $^{-1}$ ). When using mass-based concentrations, the mean values of the PNEC distributions were found to be 0.13 and 0.08 g.kg $^{-1}$  when including and excluding HONEC data, respectively. The particle-based PNEC excluding HONEC was used for risk assessment, with a mean value: 82,000 part.kg $^{-1}$ . The MP size range used in the hazard dataset was 10–4000  $\mu m$ .

#### 4. Hazard data analysis

No single species had enough data available for us to perform







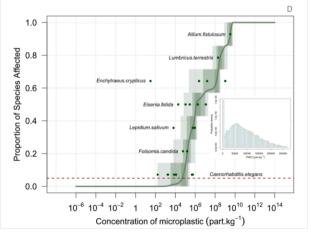


Fig. 1. Probabilistic species sensitivity distributions (pSSDs) for terrestrial species exposed to MPs in mass-based (A) and particle-based (B) concentrations, including all available data, and excluding HONECs from the dataset (C and D, respectively). Mean pSSD curves are shown by the blue/green lines; surrounding bars indicate the range of uncertainty (lightest shade. area between minimum and maximum, mid shade: area between Q5 and Q95, darkest shade: area between Q25 and Q75). Points indicate single NOEC values. Common names for species can be found in Table S7. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Statistical analyses of the probability distributions associated with predicted noeffect concentrations (PNECs) in soil for particle-based and mass-based evaluation. HONEC: Highest observed no-effect concentration. All values are rounded to 2 significant figures.

Dataset	Unit	25 <sup>th</sup> quantile	Mean	Median	75 <sup>th</sup> quantile
Including HONECs	part. kg <sup>-1</sup>	2,100	3,300	3,200	4,400
Including HONECs	g.kg <sup>-1</sup>	0.082	0.13	0.11	0.16
Excluding HONECs	part. kg <sup>-1</sup>	38,000	82,000	72,000	120,000
Excluding HONECs	$g.kg^{-1}$	0.06	0.08	0.08	0.11

statistical tests on how varying MP composition, particle shape, and size had a potential influence on toxicity values. Thus, for all organisms, these relationships were analyzed using a combination of particle-based and mass-based toxicity.

Regarding polymer type, the effects of PES and PA were found to be higher than those of other polymers when examining mass-based toxicity (Figure S4), but the only statistically significant difference was observed between PES and PET (p: 0.006) when HONEC values were included. When particle-based toxicity was considered, in addition to PES and PET, statistically significant differences were also observed between PES and PE, PA, and PET. The p values can be seen in Table S8. When HONEC values were excluded, there were no data points left for PES.

When it came to shapes, fibers seemed to have higher toxicity than spheres and fragments for mass-based toxicity (Figure S5), whereas a significant difference was only observed between fibers and fragments (p: 0.032 and 0.013 for mass-based and particle-based toxicity, respectively) when HONEC values were included. There were no data points left for fibers when HONEC values were excluded.

It should be noted that some polymer types (e.g., PVC, PA, PES) and shapes (e.g., films) were under-represented, and some of the data points were completely or mostly lost when HONEC values were excluded. Differences are likely to occur as more data is published in the literature. Another factor adding uncertainty is the transformation of mass-based toxicity into particle-based toxicity, as described above.

As Fig. 2A shows, there was no clear relationship between particle size and toxicity when particle mass was used as the metric. This means that, overall, the same mass of smaller particles does not result in greater toxicity (and thus smaller NOEC values). When particle number is used

as the metric (Fig. 2B), a clear relationship between particle number and NOEC can be seen, with larger NOECs (and thus less toxicity) at smaller particle sizes. Smaller-sized particles have higher particle numbers for the same mass. The line in Fig. 2b shows the relationship between particle size and particle number, assuming that the toxicity of the largest particle in the dataset remains the same and only the particle number increases. Particle numbers were calculated by assuming that the relative total volume of different-sized MPs was the same as that of the largest-sized particle. There was visibly no size effect in the dataset, and the trend in the data was caused solely by the increasing number of particles.

#### 4.1. Exposure assessment

The final dataset for terrestrial exposure assessment included 713 MECs (Supplementary Information 2, Table SI2). Most of the data (59 %) were composed of single measurements with no associated uncertainty, and the variability reported was mainly in the form of replicates. Measured concentrations ranged between 0 and 410,000 part.kg<sup>-1</sup> (Fig. 3), with a mean concentration: 9,300 part.kg<sup>-1</sup> (Table 2).

Globally, urban and industrial soils showed the highest MP

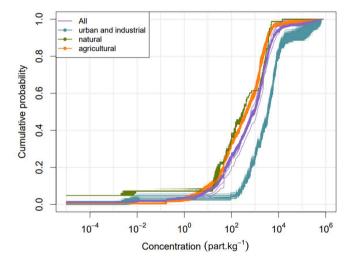


Fig. 3. Cumulative probability curves of microplastic concentrations in soils. Samples with no detected microplastics are reported as  $10^{-5}$  part.kg $^{-1}$  to keep them visible on the logarithmic scale.

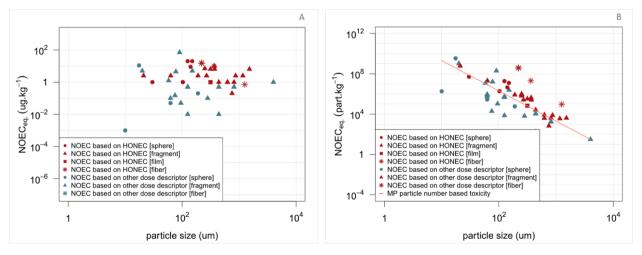


Fig. 2. Relationships between the particle sizes used in terrestrial toxicity studies and the resulting no-effect concentrations for mass-based (A) and particle-based units (B). The line in Fig. 2B is calculated assuming that toxicity remains the same and that just the particle number increases at smaller sizes, keeping the total mass the same. Dose descriptors other than HONECs include NOECs and LOECs. Undefined shapes were excluded from the chart.

**Table 2** Key figures for measured environmental concentrations in part. $kg^{-1}$  reported in soils. All values are rounded to 2 significant figures.

Land use	25 <sup>th</sup> quantile	Median	Mean	75 <sup>th</sup> quantile
All	98	930	9,300	2,600
Urban and industrial soils	1,000	3,600	28,000	8,000
Natural	60	300	1,400	2,400
Agricultural	58	440	4,400	1,700

concentrations, with a mean value:  $28,000 \, \text{part.kg}^{-1}$ , followed by agricultural and natural soils with mean values:  $4,400 \, \text{part.kg}^{-1}$  and  $1,400 \, \text{part.kg}^{-1}$ , respectively (Table 2).

The breakdown of urban and industrial soils according to different categories can be seen in Figure S6. The results showed that there were some variances between sub-categories. The highest data points were measured in woodland in an industrial zone in Asia that received discharge from many different factories (Zhou et al. 2019). The MP levels found in the sub-categories were: industrial zones: 2,000–410,000 part.kg<sup>-1</sup>; construction land: 570–14,000 part.kg<sup>-1</sup>; recreational spaces: 0.004–12,000 part.kg<sup>-1</sup>; residential areas: 280–19,000 part.kg<sup>-1</sup>; wasteland: 1,800–6,400 part.kg<sup>-1</sup>; and others (vacant land, savanna, pastures, and a site where untreated sewage waste was dumped) 0.004–120,000 part.kg<sup>-1</sup>. Overall, savanna, pasture, and managed pine plantations had considerably lower MP concentrations, whereas the greatest concentrations were in the woodland in the industrial zone mentioned above.

The amounts of MPs according to different land uses and regions, and the cumulative probability curves of MP concentrations in different geographical regions, can be seen in Figures 4 and S1, respectively. Most of the data points were for Asia and Europe (631 and 63 values, respectively). Asian data came from China, India, South Korea, and Pakistan. European concentrations came from Switzerland, Greece, Germany, Spain, and the Netherlands. A mere 15 data points came from three studies in Latin America (Chile and Mexico), whereas just one study with 4 data points was available from Canada, representing all of North America. This overall geographical coverage is quite limited, with a lack of data from different land use types for different locations, as shown in Fig. 4.

### 4.2. Risk assessment

MEC and PNEC distributions were compared for overlap (Fig. 5A).

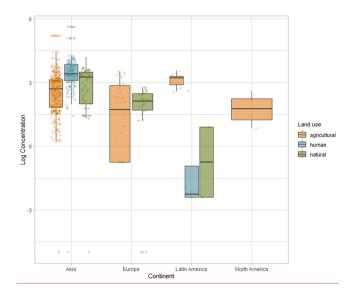
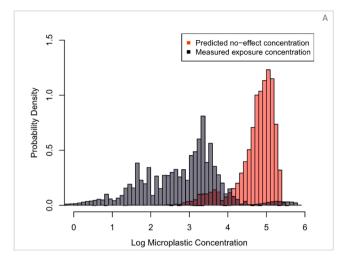
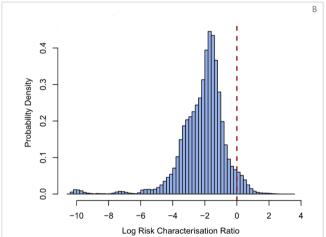


Fig. 4. Microplastic concentrations according to land uses and continents. Null concentrations were represented as  $10^{-5}$  part.kg $^{-3}$  for visibility on the log scale.





**Fig. 5.** Terrestrial microplastics risk characterization. A: Worldwide measured environmental concentration (MEC) and predicted no-effect concentration (PNEC) distributions for terrestrial habitats. B: Probability distribution associated with the risk characterization ratio.

The whole PNEC distribution (from minimum PNEC to maximum PNEC) overlapped the MEC distribution by 69.9~%.

Secondly, RCRs were calculated for all soils together (Fig. 5B) and for each type of land use and location separately (Table 3, Figure S2, Figure S3). Globally, 4.8 % of calculations resulted in an RCR  $\geq 1$ , which means a risk existed for a maximum of 4.8 % of cases when all the uncertainties and locations were taken into account. For the different land uses, 12.7 % of the calculations resulted in an RCR  $\geq 1$  for urban and industrial soils, followed by agricultural and natural soils at 2.8 % and 1.8 %, respectively. Regarding comparisons based on locations, the highest expected risk was 5.8 % for the cases in Asia, followed by 0.96 % and 0.4 % for Latin America and Europe, respectively. It should be noted that the number of data points for Latin America was quite limited, and statistical assessment was impossible for North America due to a lack of

**Table 3**Key figures of the risk characterization ratio (RCR) distributions in the terrestrial compartment.

Location	25 <sup>th</sup> quantile	Mean	Median	75 <sup>th</sup> quantile	% RCR $\geq$ 1
World	0.002	0.61	0.013	0.051	4.8
Urban and industrial	0.014	1.8	0.053	0.19	13
Natural	< 0.001	0.088	0.006	0.034	1.8
Agricultural	< 0.001	0.28	0.008	0.030	2.8

data.

#### 5. Discussion

#### 5.1. Hazard assessment

Most of the hazard data points were HONEC values, which means that in most studies, no toxicity due to MPs was observed, even at the highest tested concentration. This shows that soil organisms had an overall low sensitivity to MPs, although some specific species may be at risk. The inclusion of HONEC values, therefore, constitutes a precautionary assessment as the true NOEC values observed will be higher. This is also visible in the PNEC values, which are lower when HONECs are included. Removing HONEC values from the SSD reduced the number of species affected from 16 to 7. ECHA (2008) recommends the inclusion of at least 10 NOEC values from 8 taxonomic groups for freshwater organisms. There is as yet no clear requirement or recommended number of species of soil organisms that should be included in an SSD; however, a dataset for calculating the PNEC should preferably be comprised of primary producers, consumers, and decomposers (ECHA, 2008). Our dataset included species from each group, even when the HONECs were excluded.

The mass-based PNEC distributions remained quite similar whether HONECs were included or not, with mean values: 0.13 g.kg<sup>-1</sup>, and 0.08 g.kg<sup>-1</sup>, respectively. Kim and Rillig (2022) found a PNEC value: 0.52 g. kg<sup>-1</sup> for soil biota. However, there were many methodological differences between the studies examined (e.g., number of data points considered per study, factors used to convert LOECs into NOECs, the inclusion of acute data, choices of endpoints). There were also differences between the sensitivities used in the present paper and those used for some of the species in the SSD-like curves in Kim and Rillig's paper (2022). For instance, this study found the most sensitive species to be E. fetida, whereas it was at the top of the SSD-like curve in the Kim and Rillig study (2022), representing their least sensitive species. Furthermore, the two least sensitive species found in the present study (L. terrestris and A. fistulosum) were positioned around the middle of the SSD-like curve by Kim and Rillig (2022). These differences could be due to the available dataset, choices authors made on which studies to include in their datasets as well as the methodological differences listed above.

Toxicity concentrations extracted from the literature were mostly mass-based, whereas exposure concentrations were normally reported in particle numbers. To enable a comparison of these two datasets and the calculation of risk characterization ratios, toxicity concentrations were converted into particle-number concentrations. When mass-based toxicity values were transformed into particle-based toxicity values, a change in the least sensitive species was observed. This was due to the small size of the MPs used in the study involving A. fistulosum, i.e., 17.5  $\mu m$  (de Souza Machado et al. 2019). Small-sized MPs resulted in higher particle-based toxicity values since each particle's mass was smaller. In other words, dividing mass-based NOECeq by volume and density to obtain particle-based NOECeq (as stated in Section 2.1) produces higher NOECeq values. Bigger particle sizes would result in higher total volumes of MPs, which would then be used to divide mass-based toxicity values, eventually producing lower particle-based toxicity.

The generalizations required to compare our data on the sizes and shapes of the particles tested led to uncertainties that could not be quantified. Many assumptions had to be made as almost none of the studies included provided the complete size distributions that would be needed for accurate conversions. Using an average particle size or a size range can result in inaccuracies in particle numbers, as this metric is highly dependent on the actual size distribution. This shows how critical the current situation is concerning the availability of a high-quality dataset, especially for the particle-based PNEC. One possible approach to overcoming this limitation is using probability density functions to describe MP size and shape, as proposed by Kooi and Koelmans (2019).

The debate on which metric best characterizes MP toxicity is currently ongoing in the MP-related literature. For marine data, one meta-analysis showed the incompatibility of the measured concentration units used in the real world and those used in effect studies (Cunningham and Sigwart, 2019). This is similar to the situation with engineered nanomaterials, where there remains a gap between predicted environmental concentrations and those used in laboratory studies (Holden et al., 2014). Because of engineered nanomaterials' minuscule size, high surface reactivity, and potential for aggregation, some studies have indicated that particle number concentrations might be more relevant than mass concentrations for describing biological effects (Petersen et al., 2015). These characteristics are the same for MPs, resulting in some of the same dosimetry challenges previously found for nanomaterials. The size distribution of MPs that tested organisms encounter appears to be an important characteristic that should be described in ecotoxicity assays, especially considering the very wide range of sizes over which they are defined. Moreover, because plastic particles have a low density, their weight might be difficult to measure in some environmental samples (Weber et al., 2020). Therefore, it seems that particle number concentrations might be more appropriate than mass-based concentrations when describing MP hazards. Specific tests need to be performed to define which metric correlates best with the biological effects observed. In the meantime, it is important that toxicity assay researchers provide both mass and particle number concentrations, as they are more familiar with the materials and can make more accurate calculations. Efforts are under way to propose guidelines that will increase dataset comparability, including dose metrics (Cowger et al., 2020).

Current data suggest that overall particle type, shape, and size are not always the main parameters affecting MP toxicity in soils. This is especially true for particle size, where the data do not show any overall effects of particle size on toxicity—they notably do not show that small sizes are more toxic. Fibers and PA and PES particles may result in higher toxicity depending on the dataset considered, as stated earlier. It should be noted that there were very few data points for some types or shapes (e.g., PA or films) of MPs. Kim and Rillig (2022) used an effect distribution dependent on the type and shape of MP, and they considered the proportion of significant effects in soil by evaluating it with an SSD-like curve. Even though there were differences between the methodological approaches and the data sets, as described above, the results are partially similar. They found that MP fibers and films had larger effects than other shapes and suggested that toxicity decreased from PVC (highest) to LDPE, PES, PE, PA, PS, HDPE, PAN, and finally, PP. Kim and Rillig (2022) also assessed whether there was an effect based on the MP size tested, and they stated that significant effects mainly occurred with particles ranging from  $10^2$ – $10^5$  µm.

The PNEC values reported here could serve as the starting point for a risk assessment, but they may very likely need to be adapted in the near future as more data are published. However, these values could then shift lower if very sensitive organisms were tested, or perhaps higher should chronic EC10 or NOEC values be published, because these will require only a small assessment factor to obtain the PNEC, which will therefore be higher than when obtained using acute tests.

#### 5.2. Exposure assessment

Urban and industrial soils had the highest MP concentrations (median: 3,600 part.kg<sup>-1</sup>), ahead of agricultural (median: 440 part.kg<sup>-1</sup>) and natural soils (median: 300 part.kg<sup>-1</sup>). The exposure assessment clearly showed the influence of anthropogenic activities on MP levels in soils. Agricultural soils showed higher values than natural soils, and it is well known that many agricultural applications result in MP contamination, such as sewage sludge application or the use of plastic mulch (Huang et al., 2020). However, there were only a limited number of studies providing complete information on agricultural applications; thus, no comparisons could be made based on specific application types.

Natural soils showed the lowest MP concentrations. Even though the sampling points furthest away from any human impacts were grouped in this category, it was hard to find information regarding their use histories or descriptions of their surroundings. For instance, a data point may be considered natural, but the sample may have been taken from natural soil next to a road or a recreational space, which might actually represent more urban or industrial-type soils. Samples taken from riparian forests formerly used as cropland showed very high levels of MPs (Zhang and Liu, 2018). The lowest levels were found in a forest sample from South Korea (Choi et al. 2021) and floodplain soil samples from Switzerland (Scheurer & Bigalke 2018), followed by a tropical rainforest and an unmanaged pine plantation in Mexico (Álvarez-Lopeztello et al., 2021).

Another issue is that the geographical distribution of the studies included was far from ideal, with large differences in the types of soil studied in different regions. Not all land-use types were reported for every different location, so a robust geographical comparison was impossible. For instance, urban and industrial soil data were limited to Asia and Latin America, whereas there were no data points reported for urban and industrial soils in Europe, and only agricultural soil data was available for North America.

Another point deserving consideration was the extraction process used to remove MPs from soils in the different studies. Before the identification and quantification of MPs in soils, samples must undergo 1) a digestion step to remove organic matter, 2) a flotation step to extract MPs from the soil matrix, and 3) a filtration step to isolate MPs from the rest of the sample. Since there are no standardized methodologies for the quantification of MPs in soil samples, the different methods for extracting MPs from soil samples in the literature could cause uncertainties about the levels of MPs reported. Different methods have different limits of detection, which could considerably affect the concentrations described.

### 5.3. Risk comparison

The risks to terrestrial systems found in our study were all higher than those for marine systems (Adam et al., 2018) and freshwaters (Adam et al., 2020). The comparison of mean values for PNEC and MEC, as well as RCR percentages higher than 1, are all shown in Table 4.

These results suggest that MPs in soil pose a higher risk than MPs in freshwater and marine water. The mean PNEC calculated for soil is much higher than those for marine water and freshwater, however, MEC values were also much higher in soil ecosystems. Even though soil organisms may seem more resistant to MPs, based on PNEC values, they are much more likely to be exposed to greater amounts of MPs, which results in higher risks, as shown in Table 4. The proportion of RCRs greater than one is 40 times higher for soil ecosystems than for freshwater ones, and 240,000 times higher than for marine ecosystems. It should be noted, however, that potential methodological differences in MP determination in aquatic and soil ecosystem exposure studies and the limited data may result in some uncertainties. Another factor affecting risk assessment is the dataset's distribution of maximum MEC values. Differences in RCR calculations may occur depending on whether the maximum MEC is reported as a single value or a normally distributed value since iterations are made to build up an exposure

distribution.

Jacques and Prosser (2021) also showed that MPs might pose a risk to terrestrial ecosystems. The 5th percentiles of their SSDs ranged between 162–4824 part.kg $^{-1}$ , depending on whether NOEC or LOEC values were used and whether the lowest or the geometrical mean of the NOEC and LOEC values were used. These methodological differences preclude very robust comparisons. Their results showed that from 7 % to 28 % of species might be impacted by MPs 5 % of the time, representing a risk to soil ecosystems in line with the present study's results. By comparing the PNEC and exposure concentration values calculated in selected literature, Kim and Rillig (2022) also concluded that MPs could be expected to affect some agricultural and industrial zones.

## 5.4. Dataset comparability

The characterizations of the MPs used in hazard studies were compared with the MPs found in environmental samples in Fig. 6. Regarding MP shapes, fragments comprised 71 % of the ecotoxicity data points in hazard tests. This was generally due to plastics being milled in liquid nitrogen to obtain smaller MP particles for hazard studies. However, only 39 % of the samples reported fragments in their exposure assessments. On the contrary, fibers were under-represented in ecotoxicity assays, constituting 11 % of this dataset, whereas they represented 30 % of the MPs observed in soils. Similarly, films constituted 24 % of the MPs characterized in samples, whereas they were only used in one ecotoxicity study. This evaluation is hampered by different definitions of what constitutes a *fragment* in toxicity studies and monitoring studies.

Most of the ecotoxicity data points considered for risk calculations represent the effects of polyethylene (PE), followed by polypropylene (PP), polyester (PES), and polystyrene (PS). The share of PS and PES in ecotoxicity studies is comparably high, whereas PE's share is lower by 7%. None of the hazard data points included the effects of weathered or aged MPs, actions that could enhance their toxicity (Luo et al., 2022). To ensure greater representativity of real-world environmental scenarios, scientists are encouraged to use fibers, films, and PE MPs more frequently, as well as consider the effects of additives and effects.

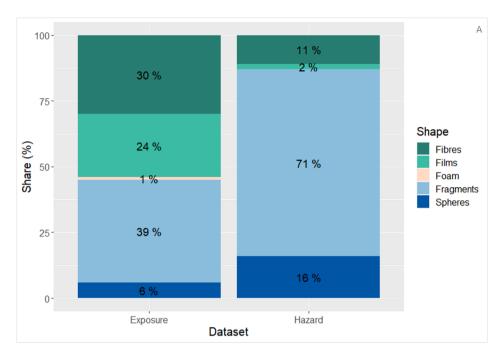
Still, the hazard dataset in this study was much more representative of the actual MP exposure in soils compared to the datasets for the marine environment (Adam et al., 2020); with gaps calculated at 49 %, 14 %, and 27 % for PS, PE, and PP usage, respectively. The same applied for the MP shapes used in the marine risk assessment: 78 % of the hazard studies used spheres, contradicting the 56 % of shapes found to be fibers in the exposure dataset (Adam et al., 2020).

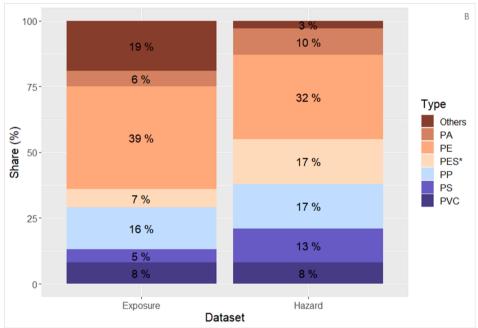
A detailed evaluation comparing MP particle sizes in hazard and monitoring studies can be seen in Table S9: 45 % of the data points used in our hazard assessment were based on MP particles from 150 to 500  $\mu m$ , followed by 27 % for 50–150  $\mu m$ , 13 % for < 50  $\mu m$ , and 11 % for 500–1000  $\mu m$ , respectively. < 4 % of data points were based on particles greater than 1 mm in size. Most of the studies reported in the exposure dataset examined particles smaller than 1000  $\mu m$ . For the studies that reported particle sizes smaller than 500  $\mu m$ , most of them reported higher MP abundances for this range than particle sizes higher than 500  $\mu m$ . Also, studies reporting MP particle sizes smaller than 50  $\mu m$  generally had higher shares (between 13 and 49 %) than those in hazard studies (7 %). Exposure and hazard datasets had some similarities,

Table 4
Comparison of risk in different ecosystems. Soil and marine water data exclude HONEC values, whereas they are included in the freshwater assessment. The PNEC and MEC values for freshwater and marine water have been transformed into part.kg $^{-1}$  to make their results comparable with soil data. Calculations use a density of 1036 kg/m $^{3}$  for marine water (Pawlowicz, 2013) and 1000 kg/m $^{3}$  for freshwater. The MEC data for freshwater represents mean values for the regions.

	PNEC	MEC	Risk characterization Ratios $\geq 1$	Reference
Soil	82,000 part.kg <sup>-1</sup>	9,300 part.kg $^{-1}$ 0.014 $^{-1}$ 9 part.kg $^{-1}$ 1.4 part.kg $^{-1}$	4.8 %	This study
Freshwater	950 part.kg <sup>-1</sup>		0.12 %	Adam et al. (2018)
Marine	3,700 part.kg <sup>-1</sup>		0.00002 %	Adam et al. (2020)

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**Fig. 6.** Properties of the microplastics observed in soils and tested in ecotoxicity assays. A: Comparison of shapes. B: Comparison of polymer types. PA, Polyamide; PE, Polyethylene; \*PES (PET + polyester); PP, Polypropylene; PS, Polystyrene; PVC, Polyvinylchloride.

nevertheless (e.g., share of MPs greater than 1 mm), but showed more discrepancies when examinations became more detailed (e.g., share of MPs < 50 um).

## 6. Conclusions

With the current state of knowledge, the probabilistic risk assessment performed in this study showed that we could not exclude the probability that microplastics (MPs) in soils pose a risk to these environments, especially urban and industrial soils, where measured concentrations were the highest. This assessment could have been more accurate if the MPs tested in biological assays were more representative of those characterized in real-world environmental samples, especially regarding their shape and polymer composition. Moreover, uncertainties could be

minimized if toxicity studies also reported particle-based concentrations or gave more detailed information (e.g., density, volume) on the MPs used in the experiments.

It should be noted that this work's risk assessments represented a first step towards a full environmental risk assessment for MPs in soils. As discussed above, both our exposure and hazard datasets had limitations, and a new risk assessment should be performed when more data is available. However, as answers about the potential risks of MPs in the environment are urgently needed, it is of paramount importance to assess the available data in the light of requirements for robust environmental risk assessment procedures.

#### **Declaration of Competing Interest**

The authors declare that they have no potential competing interests.

#### Data availability

All data are provided in the SI

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geoderma.2022.116315.

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