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Microplastics and nanoplastics barely enhance contaminant mobility in agricultural soils

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Farmland soils are prone to contamination with micro- and nanoplastics through a variety of agricultural practices. Concerns are recurrently raised that micro- and nanoplastics act as vector for organic contaminants to deeper soil layers and endanger groundwater resources. Whether and to what extent micro- and nanoplastics facilitate the transport of organic contaminants in soil remains unclear. Here we calculated the ratio between transport and desorption time scales using two diffusion models for micro- and nanoplastics between 100 nm and 1 mm. To identify micro- and nanoplastics bound contaminant transport we evaluated diffusion and partitioning coefficients of prominent agrochemicals and additives and of frequently used polymers e.g., polyethylene and tire material. Our findings suggest that the desorption of most organic contaminants is too fast for micro- and nanoplastics to act as transport facilitators in soil. Contaminant transport enabled by microplastics was found to be relevant only for very hydrophobic contaminants ($\log K_{\rm ow} > 5$) under preferential flow conditions. While micro- and nanoplastics might be a source of potentially harmful contaminants in farmland soils this study suggests that they do not considerably enhance contaminant mobility.

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lobally, 26,000 Mt of plastic waste is projected to be generated until 2050, of which at least 45% are expected not to be recycled or incinerated¹. The large public attention and unprecedented individual efforts have triggered political endeavors to regulate and mitigate plastic waste². And vet, enormous amounts of plastics escape from regulated waste streams along their life cycle and, in combination with slow degradation rates, contribute to the ubiquitous accumulation of plastics in environmental systems³. Plastic particles and abraded tire materials are re-emitted into the air with sea spray aerosols and resuspended road dust⁴. Atmospheric transport and wind drift contribute to their distribution and add to the plastic load deposited in soils⁴. Farmland soils in particular receive an additional input of micro- and nanoplastics (MNP)⁵ through common agricultural practices (Fig. 1)^{6,7}. MNP that are effectively removed in waste water treatment plants enter farmland soils through the application of biosolids. Recent mass balance approaches estimate these biosolids to contain at least 300,000 plastic particles kg⁻¹ introducing up to 70 kilotons of MNP annually to farmland soils in the U.S. alone⁸. Additional MNP input originates from compost applications⁷ or plastic mulching films⁹, over 40% of which are not recovered from the soil and can break down into a continuum of smaller fragments¹⁰. In intensively used farmland soils, plastic loads of up to 43,000 particles kg-1 encompassing a multitude of different polymers have been reported¹¹.

MNP can contain up to several percent of intentionally added substances, such as functional additives and fillers. Additionally, MNP can take up organic contaminants before entering farmland soils, including, e.g., pharmaceuticals, polyaromatic hydrocarbons (PAH), or agrochemicals. This makes MNP a potential source for organic contaminants in farmland soils where they can have detrimental effects on soil organisms and the microbiome^{12,13}. Unlike immobile contaminants, which interact strongly with the soil matrix, mobile contaminants can be carried with the pore water flow to deeper soil layers or groundwater¹⁴. However, concerns are recurrently voiced that MNP are a vector facilitating the relocation of immobile organic contaminants additionally endangering groundwater resources^{15–18}. As studies thoroughly examining this issue are lacking, further investigation is required.

For MNP-facilitated transport of organic contaminants to be environmentally relevant, four conditions have to apply: particles must be present in sufficiently high concentrations, the contaminant must be of concern, particles must be more mobile than the (non-sorbed) contaminant, and the desorption of the contaminant during the travel time of the MNP must be low^{19–21}.

To assess MNP mobility in soil, existing knowledge on the mobility of natural and engineered nanoparticles and colloids can be transferred to the mobility of nanoplastics²⁰ with some concepts equally applying to the mobility of microplastics²². The particle mobility in soil depends on flow conditions, solution chemistry, and physicochemical properties of the soil and the

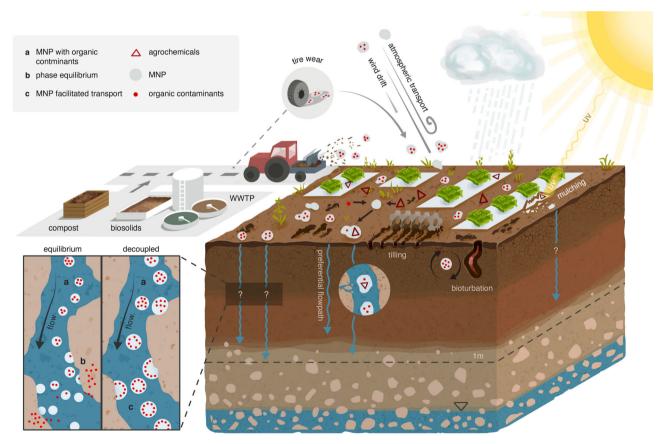


Fig. 1 Pathways and sources of micro- and nanoplastics (MNP) and organic contaminants in farmland soils. Atmospheric input, tire wear, and common agricultural practices like the application of compost, biosolids from waste water treatment plants (WWTP), or mulching introduce MNP and organic contaminants to the soil. Physical and chemical degradation of plastics, e.g., by UV irradiation causes their fragmentation to MNP. Bioturbation, tilling or preferential flow paths enhance vertical transport of MNP. **a** MNP containing organic contaminants are suspected to vertically relocate them towards the groundwater table. **b** If contaminants are in equilibrium, MNP do not enhance contaminant relocation; **c** if the transport is decoupled, no significant desorption of contaminants occurs during transport and MNP facilitate as vector their relocation. Establishing equilibrium of contaminants with plastics and all soil phases is faster for smaller MNP than for larger MNP.

particle²³. Maximum nanoplastics transport can be expected for an attachment efficiency to the soil matrix of zero and negligible physical removal by sedimentation, sieving, interception, or diffusion. These settings may be found for short travel distances and time scales^{24,25}. Particles with a density of 1 g cm⁻³, approximately corresponding to many commonly used polymers, are most mobile at a diameter of 1 μ m²⁶. Transport of larger microplastics and fibers is restricted to larger soil pore diameters or preferential flow paths, while the transport of nanoplastics might be limited by diffusion to the collector surface²⁷.

If MNP particle concentration and mobility are high, and the potentially transported contaminants are of concern, the contaminant desorption rate remains the determining parameter to assess the relevance of MNP-facilitated transport. The desorption of the contaminants in the soil environment is triggered if their fugacity in the plastic phase is higher than in the surrounding soil phases²⁸. Desorption rates can be controlled by intra-particle diffusion (IPD), i.e., the migration of the contaminant through the particle, and/or by the diffusion through the aqueous boundary layer (ABLD) between the particle and the surrounding aqueous phase²⁹. Under field conditions, ABLD usually is the rate-limiting mass transfer process^{29,30}. The Damköhler number, a useful parameter derived from fluid mechanics, describes the ratio between transport and desorption time scales to assess the relevance of particle facilitated contaminant transport^{21,31}. Damköhler numbers < 0.01 indicate fully decoupled transport, i.e., contaminant desorption during the MNP travel time is negligible and MNP are expected to facilitate contaminant transport. In contrast, Damköhler numbers >100 indicate full equilibrium conditions, where MNP will not facilitate contaminant relocation (Fig. 1) 32 . In between, kinetic models are required to fully describe the MNP-contaminant transport process. Narrower thresholds (0.1-10) can be assumed for dominantly decoupled or equilibrium transport conditions³¹.

This study identifies environmental settings in which MNP may play a part in the relocation of organic contaminants in farmland soils and aims to clearly discriminate cases in which MNP will not add to contaminant mobility. We show that in most cases nanoplastics will not contribute to contaminant relocation as smaller particles establish equilibrium quickly due to short diffusion distances. Microplastics may contribute to the relocation of contaminants in farmland soils for a few specific scenarios, i.e., very fast soil pore water flow, high MNP mobility, and very slow desorption rates.

Results

Environmental setting: slow and fast flow conditions. Our first set of calculations is based on an average soil water flow velocity of 1 m a^{-1} . We determined the prevalence of decoupled (relevant) or equilibrium (irrelevant) MNP bound contaminant transport through a homogeneous soil of 1 m thickness. The first meter of farmland soils constitutes the rooting zone of the most commonly cultivated crops³³ and it is crucial to assess if MNP derived contaminants remain within this soil layer, or reach the groundwater table³⁴. This upper soil layer is influenced by biological and physical processes such as bioturbation and tilling³⁵ and is particularly susceptible to preferential flow paths³⁶. Our second set of calculations, therefore, includes the presence of preferential flow paths with fast flow velocities of 1 m h^{-1} . Irrespective of the soil water flow velocity, we assumed a very high particle mobility and no interactions between the particles and the soil matrix. The influence of these factors will be further elaborated in the discussion section. Damköhler numbers were calculated in dependence of the diameter of a spherical MNP particle and the apparent diffusion coefficient of a contaminant

through the polymer ($\log D_{\rm app}$) for IPD, or the partitioning coefficient of a contaminant between the MNP particle and water ($\log K_{\rm PW}$) for ABLD (Fig. 2a, c). We then assessed if the combination of certain polymers and contaminants are likely to reach MNP-facilitated transport by comparing to what extent $\log D_{\rm app}$ or $\log K_{\rm PW}$ values reported in the literature (Supplementary Tables 1 and 2) correspond to the calculated values necessary for decoupled transport (Fig. 2b, d). Since experimental data available in the literature is not exhaustive, cases, where $\log D_{\rm app}$ or $\log K_{\rm PW}$ values exceed those presented in Figs. 2 and 3, cannot be excluded.

Under slow flow conditions MNP do not enhance contaminant mobility in farmland soils. Our assessment elucidates that for IPD-limited desorption MNP particles ≥1 mm can facilitate contaminant relocation if $\log D_{\rm app}$ values are <-17. As $\log D_{\rm app}$ of all polymer-contaminant combinations are >-16, equilibrium conditions occur for MNP <100 µm within a transport time of one year (Fig. 2a, points I and II, Fig. 2b). Here, contaminant desorption is too fast to sustain particle-bound transport conditions. Only for larger microplastics (100 µm-1 mm) the calculations indicate that equilibrium may not be reached within the assumed transport time. Under such conditions, the transport of a few contaminant—polymer combinations with $log D_{app} < -14$ to -16 is in the kinetic range, where particle-bound contaminant relocation could be of relevance for shorter time scales or faster pore water flow. These polymer-contaminant combinations include additives in polyethylene and polystyrene as well as PCB in polyvinylchloride. Due to the high $log D_{app}$ of more rubber-like polymers such as polydimethylsiloxane and styrene butadiene rubber, major composite of tire materials, their role for contaminant relocation in the soil is negligible. In case that IPD is the rate-limiting mass transfer process, MNP will not be able to enhance contaminant relocation beyond shallow soil layers.

For ABLD-limited mass transfer, microplastics <100 μ m do not facilitate the transport of contaminants with $\log K_{\rm PW}$ <5.5, covering most contaminants (Fig. 2c, point II and Fig. 2d). The transport of PAH, PCB, and few pesticides by polyethylene and polydimethylsiloxane microplastics <100 μ m is in the kinetic region, while the transport for none of the contaminants by microplastics <100 μ m is decoupled. Large microplastics >1 mm do not facilitate the transport of contaminants with $\log K_{\rm PW}$ <4.5, covering most of the contaminants. Only the transport of highly hydrophobic PCB by larger polyethylene-microplastics >1 mm with $\log K_{\rm PW}$ >6.5 is decoupled. If ABLD is the rate-limiting mass transfer process, MNP do not contribute to the relocation of contaminants in farmland soils with few exceptions of microplastics >100 μ m.

Under fast flow conditions MNP may add to the mobility of highly hydrophobic contaminants. Fast flow conditions do not affect the diffusion rate for IPD limited desorption, but Damköhler numbers decrease due to a shorter transport time. Even under preferential flow conditions, nanoplastics <1 µm do not enhance contaminant mobility in the soil as $log D_{app}$ of all polymer-contaminant combinations are >-16 (Fig. 3a, point I and 3b). Microplastics <100 µm do not facilitate the transport of contaminants with $log D_{app} > -12$ (Fig. 3a, point II) covering contaminants in tire materials and polydimethylsiloxane. Transport of contaminants with $log D_{app}$ –15 to –12 is in the kinetic range, including the combination of polyethylene, polystyrene, and polyvinylchloride and most PAH, PCB, pesticides, and additives (Fig. 3b). Decoupled transport is only observed for larger polyethylene, polystyrene, and polyvinylchloride

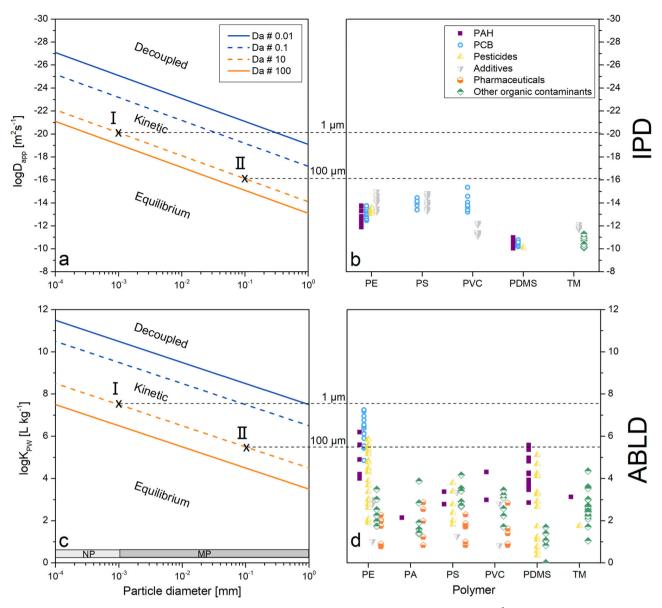


Fig. 2 Identification of MNP-facilitated transport of contaminants in farmland soils for a travel time of 1 m a^{-1} using Damköhler numbers. Lines of equal Damköhler numbers indicate equilibrium conditions (>10-100) and decoupled transport (<0.1-0.01) depending on the particle size (x-axis) and a the $logD_{app}$ for IPD or c $logK_{PW}$ for ABLD (y-axis). b $logD_{app}$ 30.39,71-77 and d $logK_{PW}$ 30.37-40.55,59,78-82 for contaminant groups and polymers reported in the literature: polyaromatic hydrocarbons (PAH) (\blacksquare), pesticides (\bigwedge), polychlorinated biphenyls (PCB) (\bigcirc), additives (\bigvee), pharmaceuticals (\bigcirc) and other organic contaminants (\bigcirc) in polymers such as polyethylene (PE), polyamide (PA), polystyrene (PS), polyvinyl chloride (PVC), polydimethylsiloxane (PDMS) and tire materials (TM).

microplastics >100 μm in combination with PAH, PCB, and additives (log D_{app} <-13).

In cases of ABLD as rate-limiting mass transfer, the Damköhler numbers decrease under fast flow conditions, which is only slightly counteracted by a faster diffusion. Nanoplastics <1 μ m can facilitate transport of contaminants with $\log K_{\rm PW}$ values >7, which have only been reported for PCB³⁷. The majority of contaminants have $\log K_{\rm PW}$ values <4 and is within the range of equilibrium transport for nanoplastics <1 μ m (Fig. 3c, point I), confirming that nanoplastics with few exceptions do not contribute to contaminant relocation in soil. Microplastics can enhance the relocation of contaminants in soil for very specific cases. Smaller microplastics <10 μ m reach equilibrium within one hour if $\log K_{\rm PW}$ is <3.5, which includes a range of contaminants such as most pharmaceuticals, many pesticides, additives and other organic contaminants (Fig. 3d). Larger

microplastics >100 µm can facilitate the transport of contaminants with $log K_{PW}$ >4. However, such $log K_{PW}$ values were reported only for the more hydrophobic PAH with octanol -water partition coefficient ($logK_{OW}$) >5 such as pyrene, chrysene, or benzo[b]fluoranthene, as well as the highly hydrophobic PCB and certain pesticides (Fig. 3b, d)³⁸. The latter include mostly organochlorine pesticides such as hexachlorobenzene, DDT and its metabolites, aldrin, endrin, and transnonachlor^{30,39,40}. The linear relationship between the $log K_{PW}$ and logK_{OW} value and thus transferability of both concepts is described elsewhere³⁰. For microplastics >1 mm, decoupled transport can be observed for $log K_{PW} > 3$, covering combinations of all polymers and most contaminant groups except for the less hydrophobic pharmaceuticals. Under preferential flow conditions microplastics >10 μm can add to a relocation of contaminants to deeper soil layers for some polymer-contaminant combinations.

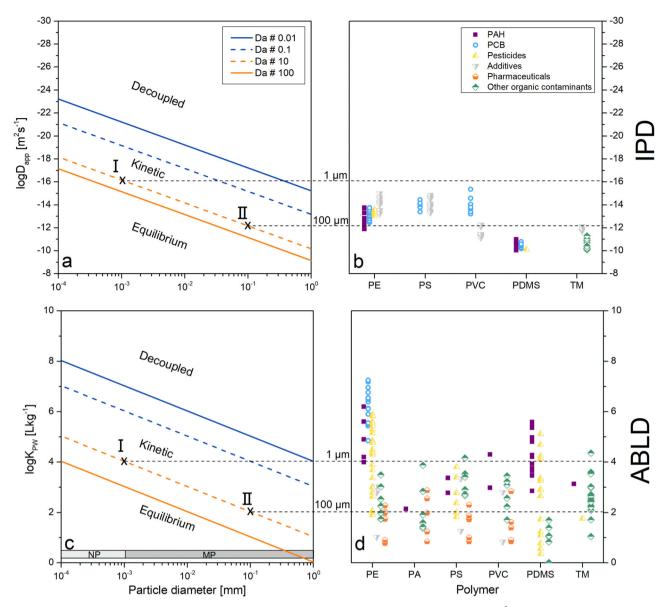


Fig. 3 Identification of MNP-facilitated transport of contaminants in farmland soils for a travel time of 1 m h⁻¹ using Damköhler numbers. Lines of equal Damköhler numbers indicate equilibrium conditions (>10-100) and decoupled transport (<0.1-0.01) depending on the particle size (x-axis) and a the logD_{app} for IPD or (c) log K_{PW} for ABLD (y-axis). b LogD_{app} $\frac{30.39,71-77}{300}$ and (d) log K_{PW} $\frac{30.37-40.55,59,78-82}{300}$ for contaminant groups and polymers reported in the literature: polyaromatic hydrocarbons (PAH) (\blacksquare), pesticides (\bigwedge), polychlorinated biphenyls (PCB) (\bigcirc), additives (\bigvee), pharmaceuticals (\bigcirc) and other organic contaminants (\bigcirc) in polymers such as polyethylene (PE), polyamide (PA), polystyrene (PS), polyvinyl chloride (PVC), polydimethylsiloxane (PDMS) and tire materials (TM).

Our analysis shows that under few conditions MNP facilitated contaminant transport may be possible. However, a more comprehensive assessment of the model limitations is called for to estimate the applicability of the calculations under more complex environmental conditions. The model does not include particle—soil interaction and neglects the biological and physicochemical effects influencing the fate of MNP in soils. It further simplifies the polymer morphotypes, i.e., using spherical particle shapes for the calculations. The influence of these factors on the presented results will be discussed in the following section.

Discussion

Short travel times and slow diffusion rates can result in fully decoupled transport (Damköhler number <0.01) and the possibility of MNP to act as vector for contaminants; hence, factors influencing both parameters need to be discussed.

Soil water flow velocities can vary drastically from a few cm per year to several meters per day41. At slow soil water flow conditions of 1 m a-1, organic contaminants desorb from small particles before reaching deeper soil layers, limiting the relevance of MNP-facilitated transport (Fig. 2). In contrast, very fast transport regimes can provide pathways for MNP-bound contaminant transport. Most prominent examples include exposed fractured rocks, karst⁴², or soils exposed to long draughts followed by heavy rain events, which can provide larger transport channels and cracks⁴³. In such cases, MNP may be relocated within very short time scales during which the contaminants will barely desorb and will, as a result, be carried over longer distances with the MNP particles. Systems with high infiltration rates like karstic rocks are, however, vulnerable to multiple kinds of pollution to which the contribution of MNP-facilitated contaminant transport is rather negligible. Furthermore, soils overlying these systems are less

likely to be used for substantial farming activities. Other factors affecting MNP mobility include the biogenic activity of earthworms or moles or tilling of farmland soils, which can transport larger plastic fragments to a depth of approximately 30-50 cm^{16,44}, where they can interact with the roots and soil microbiome^{35,45}. The impact of these factors, however, decreases with soil depth³⁵. In our transport scenarios (Figs. 2 and 3), particle attachment to the soil was neglected. This may be valid for fast transport in cracks or bioturbation structures, but in most cases, soils are excellent filters with a high potential to retain particles^{25,26,46-48}. While microplastics may transport organic contaminants due to longer desorption times compared to nanoplastics, they are not as mobile and will be retained in soil by natural filtration mechanisms²⁶. Although pore size exclusion effects may lead to an apparently faster transport of particles compared to a soluble tracer⁴³, they lead to a higher overall particle retention in soil. As MNP in the natural environment have mostly irregular shapes further decreasing their mobility, MNP transport can be overestimated assuming spherical particles. Plastic fibers, for example, have less favorable hydrodynamic properties and are subject to enhanced straining²⁷. Extracellular polymeric substances (EPS) can triple the attachment efficiency of MNP and enhance the formation of hetero-aggregates between MNP, EPS, and soil minerals and organic matter^{49,50}. Our analysis shows that preferential flow paths in cracks and macropores provide the only relevant condition where MNP may be a contributing factor to contaminant relocation in farmland soils²⁰.

Even under preferential flow conditions, for which we assumed fast flow velocities of 1 m h^{-1} , nanoplastics of almost all contaminant-polymer combinations establish equilibrium before reaching deeper soil layers (Fig. 3). For microplastics, decoupled transport might be possible for slow diffusion rates, which depend on the polymer and contaminant properties. Assuming IPDlimited diffusion, glass-like polymers are more likely to transport contaminants (Fig. 3a, b) than more rubber-like polymers⁵¹. Furthermore, UV-induced embrittlement of glass-like polymers can enhance polymer porosity and thereby the retention of hydrophobic contaminants⁵². Accordingly, polystyrene or polyvinylchloride may facilitate the relocation of contaminants in farmland soils which is not expected for silicone and tire materials. Calculations, which are based on spherical particles, may also underestimate MNP-facilitated contaminant transport. Diffusion through a sphere is faster than through a cube, regardless of the lower surface-to-volume ratio, since the distance for a contaminant to diffuse from the center to the edge of the cube is longer than the radius of a sphere⁵³. On the other hand, IPD is faster for sheets or fibers compared to spheres, leading to an overestimation of MNP-facilitated contaminant transport³⁰. To evaluate transport processes of particles with different geometries than spheres, equations to calculate the diffusion rates need to be adapted according to the particle geometry. Overall, only if preferential flow paths are available for MNP transport, glass-like polymers may contribute to the relocation of hydrophobic, slowly diffusing high molecular weight contaminants (Eqs. 2 and 3) in farmland soils, assuming that mass transfer is IPD-limited.

The importance of IPD in contaminant mass transfer increases with particle size and smaller apparent diffusion coefficients^{29,53}. However, mass transfer of highly hydrophobic contaminants is usually expected to be ABLD limited²⁹. Damköhler numbers indicated decoupled transport primarily for hydrophobic contaminants with $\log K_{\rm OW}$ >5 (Fig. 3), where ABLD limited desorption will be the rate-limiting mass transfer process⁵⁴.

ABLD-limited mass transfer depends on $\log K_{\rm PW}$ (Eq. 4) and thus on the interaction between the organic contaminants and MNP. Hydrophobic partitioning is one of the driving mechanisms for sorption^{55–57}. Accordingly, decoupled transport is

mainly possible for hydrophobic contaminants ($\log K_{OW} > 5$) with a slow diffusion rate. Apart from contaminant characteristics, the interactions depend on the polymer type. Figure 3d may imply that polyethylene most efficiently retains contaminants as most sorption studies have focused on polyethylene as sorbent, but some hydrophobic contaminants sorb more strongly to polystyrene due to the additional formation of $\pi - \pi$ electron—donor -acceptor interactions between delocalized π -electrons⁵⁵. Also tire materials can strongly sorb hydrophobic contaminants through hydrophobic partitioning to its main components styrene—butadiene rubber and carbon black⁵⁸. Sorption of less hydrophobic and polar contaminants to polyamide can be enhanced by hydrogen-bonding⁵⁵. In spite of these interactions, the $log K_{PW}$ of these less hydrophobic contaminants is too small for decoupled transport and their MNP-facilitated relocation⁵⁹. Therefore, decoupled transport only occurs for more hydrophobic contaminants, assuming that microplastics are transported along preferential flow paths and are mobile therein.

Desorption models predict that environmental factors such as biofilm formation could triple the desorption time of a contaminant from polyethylene⁶⁰. The presence of biofilm would, however, not substantially change our conclusions on the role of MNP as a vector for contaminants, since even at a third of the desorption rate, the log*K*_{PW} threshold for decoupled transport remains within the same order of magnitude. UV-induced aging can introduce oxygen-containing functional groups such as carbonyl and hydroxyl groups onto the polymer surface⁵¹, increasing sorption and retention of polar contaminants but decreasing sorption of hydrophobic contaminants^{52,61}. For ABLD-limited desorption, UV-induced aging may therefore further decrease the relevance of MNP for transport of hydrophobic contaminants.

Contaminant transport in the soil can be facilitated by mobile natural colloids, such as macromolecular soil organic matter⁶², clay minerals, precipitates, or weathering products⁴³. MNP containing contaminants before entering the soil may also transport these to deeper soil layers. Conversely, if the contaminants do not come into contact with MNPs until they reach the farmlands, it is questionable if MNP are of any relevance at all in terms of facilitating contaminant transport due to the overabundance of mobile natural colloids and organic matter. In aquatic systems, a minor fraction (<1%) of contaminants sorb to plastics due to the low abundance of plastics compared to all other phases present (water, air, and organic matter). Although the relative abundance of plastics in soil differs compared to aquatic systems, it is still too low to considerably contribute to the uptake of contaminants in soil^{63,64}. Furthermore, the sorption coefficient of soil organic matter and bulk soil is higher than that of most plastics^{32,65}, which limits the uptake of organic contaminants by MNP in soil and thereby their function as vector for contaminants. As the uptake of contaminants by MNP prior to their input to soils is slow, which leads to lower concentrations in MNP in the first place²⁸, the input and vertical transport of non-intentionally added substances will play a minor role in overall contaminant concentration. Thus, plastic additives, which are intentionally incorporated into the plastics during manufacturing, are more prone to being transported into the soils and relocated to deeper soil layers. This input pathway is especially important as MNP are the only source to farmland soils for some of these additives.

The use of plastic mulch or biosolids offers great benefits for agricultural producers in their effort to sustain a growing global population. To minimize the impact of agricultural practices on the environment and the consumers, the complete removal of plastic foils after use or their replacement by fully biodegradable materials should be advocated. The amount of plastics unintentionally entering farmland soils via biosolids and compost needs to be reduced and should be regulated. Good governance

and targeted regulations are required to improve waste management and to promote a sustainable, effective circular economy². Simultaneously, a thorough understanding of the risk associated with the presence of MNP in farmland soils is crucial to reduce the impact on the consumers' health. There is no doubt that the input of plastics contributes to the overall flux of potentially harmful organic contaminants to farmland soils. Although MNP carrying these contaminants may be physically worked into topsoil layers by physical and biological activities, the scenarios under which MNP play a relevant role for contaminant relocation into deeper soil layers are very limited. Our analysis shows that desorption from nanoplastics is fast for most polymercontaminant combinations and nanoplastics bound contaminants will reach equilibrium with the soil matrix within the first meter of soil. Only larger microplastics with very slow contaminant desorption rates might add to the relocation of hydrophobic organic contaminants in addition to the plethora of natural mobile soil particles, provided that fast flow regimes and preferential flow paths prevail, and that the microplastic particles are mobile. However, in farmland soils, few preferential flow paths extend beyond 1 m in depth³⁶, and by the time microplastics reach deeper soil layers the organic contaminants will be desorbed. Therefore, MNP are not expected to enhance the vertical mobility of most organic contaminants in farmland soils to an extent that endangers underlying groundwater resources. This conclusion stresses the need to focus further research activity on the potential risks associated with the presence of MNP derived organic contaminants in the root zone, plant uptake, or adverse effects on the soil microbiome.

Methods

Particle-facilitated transport. The Damköhler number (Da) describes the ratio between the transport time of MNP particles and the desorption time of the released contaminants. Thus, Da can be used to evaluate the influence of the MNP particle mobility and the diffusion of non-intentionally added substances and additives within these particles on the transport of contaminants in soil and to identify if MNP particle facilitated contaminant transport can occur. For Da <0.01, the MNP contaminant transport is decoupled, and MNP are expected to relocate contaminants in soil, while Da >100 indicate equilibrium transport and MNP facilitated contaminant transport can be neglected. While at Da = 1 contaminant transport can only be explained by kinetic transport modeling, the deviation from decoupled or equilibrium transport decreases towards Da = 0.01 or 100^{66} . As the deviation from equilibrium modeling is small for Da from 0.1 to 0.01 and from 10 to 100^{31} , Da numbers of 0.1 and 10 will be used to define the boundaries of kinetic to decoupled and to equilibrium transport, respectively.

The Damköhler number (Da) [-] can be expressed as

$$Da = \lambda * \tau \tag{1}$$

where λ [s⁻¹] is the reaction rate constant and τ [s] is the mean MNP particle travel time^{53,66,67}.

Considering spherical MNP particles the diffusion rate $\lambda_{\rm IPD}$ for IPD limited mass transport can be described by

$$\lambda_{\rm IPD} = \ln \left[\frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp \left(-n^2 \pi^2 \tau \frac{D_{\rm app}}{r^2} \right) \right] / \tau \tag{2}$$

where $D_{\rm app}$ [m² s⁻¹] is the apparent diffusion coefficient of a contaminant in an MNP particle and r [m] is the particle radius⁶⁷. $D_{\rm app}$ can be expressed as

$$D_{app} = \frac{D_{eff}}{\varepsilon + K_{PW}(1 - \varepsilon)\rho} \tag{3}$$

where $D_{\rm eff}$ [m² s⁻¹] is the effective diffusion coefficient of a contaminant in an MNP particle, $K_{\rm PW}$ [L kg⁻¹] is the partitioning coefficient of a contaminant between the MNP particle and water, and ε [-] and ρ [kg L⁻¹] are the porosity and density of the MNP particle, respectively.

Considering ABLD limited mass transfer of contaminants from spherical MNP particles, the approach described by Bold et al⁶⁷. was extended and the diffusion rate λ_{ABLD} can be described by⁶⁸:

$$\lambda_{ABLD} = \frac{3D_w}{r\delta\rho K_{PW}} \tag{4}$$

where D_w [m² s⁻¹] is the aqueous diffusion coefficient of the contaminant, r [m] is the particle radius and δ [m] is the ABL thickness and ρ [kg L⁻¹] is the density of water. The ABL thickness is an experimental parameter and decreases with the

turbulence of the flow³⁰. To estimate the ABL thickness of the MNP particle the flow conditions in soil were evaluated by calculating the Reynolds number Re $(Eq. 5)^{30}$.

$$Re = \frac{\nu d_p}{\nu_{\text{aq}}} \tag{5}$$

where $\nu=$ flow velocity [m s^-1], $d_p=$ grain diameter [m], $\nu_{\rm aq}=$ kinematic viscosity of water [m^2 s^-1].

The aqueous diffusion coefficient D_w [m² s⁻¹] can be approximated after Worch (Eq. 6)⁶⁹.

$$D_{w} = \frac{3.595 * 10^{-11} T}{n m_{c}^{0.53}} \tag{6}$$

where T[K] is the temperature, η [g m⁻¹ s⁻²] is the dynamic viscosity of water and m_s [g mol⁻¹] is the molecular weight of the diffusing contaminant.

Da of 0.01 (fully decoupled) or 0.1 (mainly decoupled) and Da of 10 (mainly equilibrium) or 100 (full equilibrium) were calculated for particle sizes between 100 nm and 1 mm, adopting the categorization by Hartmann et al. to subdivide plastic particles into microplastics (1 to <1000 μ m) and nanoplastics (1 to <1000 μ m)⁵.

To calculate the diffusion rates for IPD and ABLD, $\log D_{\rm app} - 8$ to -30 and $\log K_{\rm PW}$ 0–12 corresponding to a wide range of contaminant polymer combinations were considered. $\log D_{\rm app}$ and $\log K_{\rm PW}$ values used to calculate Da of 0.01 and 0.1 (decoupled transport) and 1 and 10 (equilibrium transport) were compared to values reported in the literature for sorption and desorption experiments for a broad combination of polymers and contaminants. Since the film stacking method provides the most reliable values to experimentally determine $D_{\rm app}^{30}$, only $D_{\rm app}$ acquired through this method were considered.

Parameter range. To estimate the ABL thickness, the flow conditions were evaluated by calculating the Reynolds number (Eq. 5) for the two proposed flow conditions: slower flow through the soil matrix of 1 m a-1 and faster flow through preferential flow paths of 1 m h⁻¹. To account for varying d_p and v_{aq} at different temperatures in soil, the Reynolds number was calculated for possible minimum and maximum values for d_p and v_{aq} (Table 1). For both flow conditions, slow flow (Re = $1.6*10^{-6} - 1.8*10^{-5}$) and fast flow (Re = 0.14-0.62), the Reynolds number over the given range of d_p and v_{aq} did not exceed a value of 1, indicating laminar flow for all conditions. Based on the flow conditions and a thorough comparison of our model with reported experimental set-ups, an ABL thickness δ of 300 and 100 μm was estimated, for flow velocities of 1 m a⁻¹ and 1 m h⁻¹, respectively. For comparison, δ of 500 and 30 µm were reported for nearly stagnant and turbulent flow conditions, respectively⁶⁸. To calculate the possible variability of D_w (Eq. 6), a range of m_s covering most organic contaminants in soils and η at different temperatures were evaluated. The range of D_w was found to be $2.6*10^{-10}$ m² s⁻¹- $1.2*10^{-9}$ m² s⁻¹. Using these highest and lowest realistic D_w values, at a specific Damköhler number a maximum deviation of 7% of the $log K_{PW}$ values were calculated. A constant D_w of $5*10^{-10} \,\mathrm{m^2 \, s^{-1}}$ corresponding to the D_w of pyrene at 15 °C was used for calculations of λ_{ABLD} .

Our calculations are based on an average soil water flow velocity of 1 m a⁻¹, which can be derived from many different combinations of annual rainfall (P), evapotranspiration (ET), and effective soil porosity (n_e), e.g., P=800 mm, ET = 600 mm and $n_e=20\%$ in temperate climatic zones. The soil porosity can be influenced by the grain size distribution and taxonomy of the soil, e.g., soils with more uniform grain sizes have a higher effective porosity and soils with a higher share of clay are less permeable⁷⁰. We did not consider slower flow regimes since the relevance of MNP-facilitated transport would decrease further. Our fast flow regime under preferential flow conditions assumed very fast MNP relocation of

Table 1 Parameter range for the calculation of D_w and Re in soil. Given are the variability of the grain diameter d_p the kinematic viscosity of water $\nu_{\rm aq}$, temperature T, dynamic viscosity of water η , and the molecular weight of the diffusing contaminant m_s .

		Used	Min	Max
Reynolds number (Eq.4)	d _p soil matrix [m]	2.0*10 ⁻⁴	5.0*10 ⁻⁵	5.0*10 ⁻⁴
	d_p pref. flow [m]	1.0*10 ⁻³	5.0*10 ⁻⁴	2.0*10 ⁻³
Aqueous diffusion coefficient (Eq.5)	$ \nu_{\rm aq} [{\rm m^2 s^{-1}}] $ $ T [{\rm K}] $ $ \eta [{\rm g m^{-1} s^{-2}}] $ $ m_{\rm s} [{\rm g mol^{-1}}] $	1.1*10 ^{-6a} 288.15 1.14 ^a 202 ^d	8.9*10 ^{-7b} 276.15 0.89 ^b 78 ^e	1.0*10 ^{-6c} 298.15 1.62 ^c 395 ^f

Water at a15 °C, b25 °C, c3 °C; molecuar weight of dpyrene, ebenzene, fPCB180

 $1~\rm m~h^{-1}, i.e.,$ roughly 10,000 times faster than the average scenario. The two model scenarios frame a very large range of naturally expected flow conditions.

Data availability

The datasets analyzed during the current study are available in their supporting information files and at https://doi.org/10.5281/zenodo.5180642.

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Author contributions

S.C., T.Hü and T.Ho designed the research. S.C. conducted the calculations, the compilation of literature data, prepared the graphs, and wrote the first draft. C.H. substantially contributed to the compilation of literature data, discussion of data and results, and the preparation of the paper. T.Hü and T.Ho contributed with discussions of data and results and their revisions substantially improved the paper.

Competing interests

The authors declare no competing interests.

Additional information

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