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1 Fate and occurrence of micro(nano)plastics in soils:

2 knowledge gaps and possible risks

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8 Abstract

- 9 The majority of micro(nano)plastic research has been concentrated on the marine environment.
- 10 Whilst the ocean represents an ultimate sink for contamination, this focus overlooked key processes
- 11 and pathways of micro(nano)plastics in the terrestrial environment that are of critical importance for
- 12 their global environmental budget and exposure of humans and biota. Lack of robust analytical
- 13 methods for the isolation of these materials from complex, organic-rich soil matrices represent a
- 14 major hindrance. Regardless, soils in agricultural and urban areas are expected to represent major
- 15 environmental reservoirs of micro(nano)plastics, possibly comprehensively larger than the marine
- 16 one. Additionally, soils exhibit several potential exposure pathways for micro(nano)plastics to
- 17 organism and human health, including contamination of groundwater aquifers.

18

19 Keywords: microplastic, nanoplastic, soil, sewage sludge, groundwater

20 1. Introduction

21 Microplastics research is a rapidly evolving domain. Traditionally, studies have focused on the 22 marine environment; however, recent research has identified the significance of microplastics in 23 terrestrial ecosystems [1]. This has included work on freshwater systems, such as rivers and lakes, 24 and has recorded high microplastic concentrations [2–4]. Very little work has dealt with the 25 presence, fate, or impact of microplastics in soils [5–7]. Despite recent efforts to establish effective 26 analytical procedures [e.g. 8,9], detection of nanoplastic in environmental substrates is not yet 27 possible. Hence, no studies have thus far investigated the occurrence or fate of nanoplastics in soil 28 systems. This review brings together the existing research on soil micro(nano)plastics and draws 29 upon wider material to infer potential sources and fate of small plastic particles within soils. We 30 focus primarily on research published in the last two years with the purpose of identifying recent 31 advances relevant to the soil micro(nano)plastics research domain.

32

33 2. Existing research

34 Early studies identified synthetic fibres in soils treated with sewage sludge [10,11] and the potential 35 for soil microplastic contamination was first reviewed by Rillig [5]. Recently, a large portion of soil 36 microplastic research has concentrated on interactions with biota. Several studies have investigated 37 the effects on soil organism health and behaviour [12–18*]. Key findings reveal variable responses to 38 microplastic ingestion by earthworms, where histological damage in the gut trait was related to 39 exposure [12*,13]. Fauna are capable of moving microplastic within soil systems, including vertical 40 transfer [17,18*]. However, thus far, no studies have recorded this under environmental conditions 41 and realistic exposure scenarios.

42 Agricultural practices are relevant to soil microplastic contamination. The application of sewage
43 sludge on farm soils has been theoretically estimated as one of the largest sources of microplastics

44 to the environment $[19^{**}, 20]$. It is proposed that 125-850 tons microplastic million inhabitants⁻¹ are 45 added each year to agricultural soils in Europe, with an annual total of 63,000-430,000 and 44,000-46 300,000 tons of microplastic added to European and North American farmlands respectively [19]. 47 The broad confidence intervals of these estimates stem from the uncertainties regarding the fate of 48 microplastics deriving from car tire debris and surface runoff, for which efficiency of sewer collection 49 and fate in wastewater treatment plants (WWTPs) are unknown. These figures suggest that soil 50 systems may represent a larger environmental reservoir than the global ocean. Very little data are 51 available on the ecological implications of such an exposure. One study has linked the effect of 52 microplastics debris from plastic mulching to changes in organic matter cycling and nutrient 53 dynamics in Chinese loess soils [21]. However, the influence of soil type is unknown. Several studies 54 have examined methodologies for microplastic analysis in soils and other complex organic-rich 55 environmental matrices such as sewage sludge [22,23*]. Although, no standardised approach has 56 yet emerged.

Finally, urban soils are contaminated by microplastics. Soils close to industrial areas in Sydney were
found to be composed of 0.03-6.7% microplastic [23*]. However, details of the type of microplastic
contamination observed were not presented.

60

61 **3.** Sources of micro(nano)plastics to soils

The sources of micro(nano)plastics to soil have recently been reviewed by Bläsing and Amelung
[24**]. The key sources can be separated into three categories: inputs from agricultural practices,
the influence of runoff and deposition, and the fragmentation of larger plastic debris. Figure 1
depicts these inputs to soil systems.

Sewage sludge is often used as an agricultural fertiliser. Approximately 50% of sludge is recycled in
this way in Europe and North America [19**]. Several studies have investigated the fate of

68 microplastics within WWTPs. The findings consistently point to a high trapping efficiency, where the 69 majority of microplastics are believed to be captured in the solid sludge phase [25–27]. 70 Micro(nano)plastics entering WWTPs include microbeads from cosmetic products and industry, 71 fibres from washed synthetic garments, and tire debris and fragmented plastics from road/urban 72 runoff. Inputs may also include nanoplastics from cosmetics [28*]. The addition of polymeric 73 flocculants during wastewater treatment has to be accounted as a source, too. While analytical 74 methods mainly track polymeric particles at millimetre to micrometre scales, no protocol has been 75 established so far to quantify car tire debris and nano-scale materials in sludge or recipient soils. This 76 is a major hindrance for any accurate exposure assessments. On paper, both nanoplastics and tire 77 debris must represent a major and, thus far, unquantifiable proportion of micro(nano)plastics in the 78 terrestrial environment.

Micro(nano)plastic debris may also derive from plastic mulching in agriculture. This technique is widely applied to increase crop yields and reduce pests. Regularly, fragments of the plastic films are left behind after use [29,30]. This debris may accumulate within soils and further fragment to produce particles down to a nano-scale or beyond [31]. Although, where biodegradable plastics are used, ecotoxicological effects may be controlled [32]. Furthermore, polymers such as polystyrene and polyurethane are sometimes added to compost to improve soil properties [6,33]. These may integrate with natural soils and disperse micro(nano)plastic components across a wide spatial area.

Runoff from roads or urban areas that is not captured by sewer systems can contaminate
surrounding soils. Moreover, atmospheric transport has the potential to move plastics in the
smallest size classes over long distances and likely contributes a proportion of micro(nano)plastic in
soils. Atmospheric deposition has been demonstrated in urban environments [34] and the transport
of particles from landfill sites to soils has also been discussed [5,35]. Additionally, overbank
deposition likely enriches alluvial soils with micro(nano)plastic particles. Fluvial sediments have been
shown to contain high concentrations of microplastics [2,4], which are mobilised during flooding

[36]. Inundation of riparian zones will lead to the deposition of plastic particles, which will then
become incorporated into soils. This likely represents a significant, albeit localised, source of
microplastics.

Finally, micro(nano)plastics may be produced in-situ through the breakdown of larger plastic debris.
This may derive from discarded plastic litter that degrade into micro- and nano-sized components.
This fragmentation has been confirmed for both micro- [37] and nanoplastics [38]. In fact, the
process of degradation is considered to be the major source of *nano*plastics in the environment
[28*,39*].

101

102 4. Fate of micro(nano)plastic particles in soils

103 *4.1. Storage*

104 Figure 1 identifies mechanisms related to the fate of micro(nano)plastics in soils. Incorporation of 105 plastics into soil aggregates may promote long term storage. Aggregation may limit exposure to soil 106 fauna and hinder the transport of plastic particles. Nanoparticles aggregate rapidly in aquatic 107 environments [40,41] and hetero-aggregation has been noted as an important control on 108 nanoplastic fate [28*,39*]. Aggregate stability is associated with soil system health [42], so the role 109 of aggregates as micro(nano)plastic stores is likely to be dynamic and environment-dependent. 110 Accumulation may also occur through burial where successive flood events bury contaminated 111 layers in alluvial soils. A theoretical assessment of microplastic transport and erosion, based on the 112 frame of a hydrological/sediment transport catchment model, suggested the potential for soils to 113 effectively retain, and therefore store, micro(nano)plastics [43]. However, experimental data to 114 confirm these findings are not yet available.

Microplastics are preserved in marine and lacustrine sediment profiles [44,45]. Particle burial limits
 degradative forces and thus increases preservation potential [45]. It is probable that a similar effect

117 will occur in soil systems, establishing soils as a sink for contamination. Although, the role of soil 118 characteristics, such as pH and microbial communities in maintaining degradation must be assessed. 119 Furthermore, disturbance of buried layers may remobilize stored micro(nano)plastics. For example, 120 alluvial soils may be reworked and agricultural practices such as tilling can bring buried particles back 121 to the surface. The accumulation of plastics in soils must also be examined in the context of 122 ecological risk through long term exposure. While the discussion of the state of ecotoxicological 123 research for microplastics in soils is not the primary focus of this paper, the increasing number of 124 publications in this area demonstrates international interest [12*-18*].

125 4.2. Translocation and erosion

126 Erosion by water and wind will transport particles across soil systems and eventually towards 127 streams and rivers [43]. The dynamics of these processes have not yet been investigated; however, 128 inferences can be drawn from the wider domain of microplastics research. Recent work examining 129 the sinking velocities of microplastics in the marine realm has established particle shape as a 130 dominant control [46,47]. It is likely that shape is also relevant for the erosion of micro(nano)plastics 131 by water in soils. While the effect of erosion and entrainment of microplastics mediated by size and 132 density has been assessed through a theoretical model [43], the influence of shape on translocation 133 over soils or sediments as well as on their hetero-aggregation has not yet investigated. However, 134 these processes will lead to a winnowing effect based on particle morphology and properties, such 135 as that seen for natural soil particles.

The irregular shape and low mass of particles such as fibres lead to a preferential entrainment by *wind* erosion [34,48]. Hence, the scale of wind and water erosion is significant in determining enrichment or depletion of specific particle types. Furthermore, micro(nano)plastics do not necessarily represent inert polymers upon entry to soil systems. Particles that have been through wastewater treatment or have been exposed to the environment may have become significantly biofouled or gained a surface charge. This can alter the nature of particle mobilisation and erosion.

142 Soil fauna also contribute to the transport of microplastics within soil systems. Plastic particles may 143 adhere to the organism exterior or be transported internally through intake and subsequent 144 egestion. These mechanisms have been shown to contribute towards the dispersal of plastic 145 particles from a point source [16]. Bioturbation also results in vertical transport. This occurs through 146 the process of burrowing, which establishes biopores in the soil matrix and incorporates 147 microplastics into burrow walls and casts [17,18*]. This process significantly increases the downward 148 translocation of plastic [18*]. Micro(nano)plastic ingestion may represent a removal from soils 149 resulting in systemic translocation or trophic transfer. The uptake of microplastics by plants is 150 unlikely but may occur for nano-sized particles [49]. However, data are insufficient to establish the 151 significance of this process for the overall budget of particles in a soil.

152 4.3. Degradation

Environmental degradation has been discussed for aquatic and sedimentary environments
[37,50,51] and many of these processes also occur in soils. The topsoil likely represents a key
degradative environment, due to the direct exposure to UV radiation, increased oxygen availability,
and higher temperatures [52]. Soil microbial communities and terrestrial organisms may accelerate
biodegradation of brittle plastics [5]. Furthermore, agricultural processes such as tilling may
fragment plastic debris. All these processes contribute to the progressive fragmentation of plastic
from macro- to nano-scale.

160 *4.4. Leaching to groundwater*

Leaching is an important process driving contaminants with certain properties to groundwater. Micro(nano)plastics have not yet been analysed in groundwater samples but transport through biopores has been identified as a possible mechanism for groundwater contamination [12*,18*]. Theoretically, assuming plastics as mainly inert materials, the potential for leaching will be modulated by soil texture properties and particle size, density, and shape [24**]. Additional soil properties such as zeta potential and ionic strength may, in principle, influence transport of non-

167	inert particles [53]. Fundamental speculative reasoning suggests that nano-scale inert plastics with a
168	density higher than water may be effectively leached to groundwater. This can represent a potential
169	pathway to human exposure.

170 **5.** Future research

- 171 This paper summarises existing studies on soil microplastics and outlines the potential mechanisms
- 172 for soil micro(nano)plastic dynamics. Further work is crucial to elucidate sources, behaviour, and
- 173 fate. The following steps in soil micro(nano)plastics research should be prioritized:
- 174 Filling the methodological/technological gaps hindering an accurate assessment of
- 175 micro(nano)plastics in soil samples, including methods for car tire debris and nano-scale
- 176 materials.
- 177 Delivering baseline studies on soil exposure along a gradient of land uses and soil management.
- 178 This will establish the scale of contamination and can point towards potential source
- apportionment: for example, fibres and microbeads as indicators of sludge application or tire
- 180 dust as an indicator for road runoff.
- Unravelling the processes controlling budgets of microplastics in soil environments, including the
- assessment of microplastic transfer from soil to humans through the uptake in foodwebs and
- 183 through leaching to the groundwater.
- 184 Developing a solid experimental and conceptual framework to characterize risk and impacts from
- soil micro(nano)plastics for humans and the environment.
- 186 Timely translating of findings to stakeholders (i.e. industry, wastewater utilities, and farmers) and
- 187 governance endorsing knowledge-based decision making.

188

189 6. Conclusions

- 190 Despite a lack of analytical evidence, it is highly likely that soils are significant, possibly dominant
- 191 environmental reservoirs of micro(nano)plastic. Potential sources to soil systems are numerous and
- 192 likely exceed primary inputs to freshwater or marine environments. Furthermore, the dynamics of
- 193 micro(nano)plastics fate within soil is complex. Unravelling the controlling process will be critical for
- 194 risk and impact assessment, as well as for management.

195 Figure caption

- 196 Processes affecting the concentration of micro(nano)plastics in soil systems, including sources and
- 197 fate processes.

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