

# Plasticisers in the terrestrial environment: sources, occurrence and fate

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**Environmental context.** Many human activities cause the release of plastic and associated plasticisers to land, where chemicals may persist for extended periods and be taken up by organisms. However, quantitative information of the terrestrial occurrence, fate and exposure of phthalate and non-phthalate plasticisers is lacking. Research into this field is needed, especially as society moves away from phthalates to the next generation of plasticisers which may themselves represent an emerging risk.

**Abstract.** Modern society is widely dependent upon plastic. Therefore, it is unsurprising that macro- and microplastic pollution is found in every environmental compartment on earth. Plasticisers are chemicals added to plastics to increase their flexibility. Like plastics themselves, plasticisers are also widely present in the environment. Plasticisers and plastic debris may undergo long-range transport in the atmosphere and the oceans, contaminating even the most remote areas of land. In addition, although plasticisers typically degrade in a matter of weeks–months, they can persist in soil for decades and have been shown to occur in all land uses studied. Some plasticisers are genotoxic and can be taken up by soil organisms, which may pose a risk to ecosystems and human health. To date the majority of data on plasticisers exists for phthalates. However, plasticisers are a diverse range of chemicals and with the increasing transfer to non-phthalate alternatives, research into the fate and effects of emerging plasticisers is required to determine their environmental risk and management options. Data on the occurrence and ecotoxicity of emerging plasticisers, in addition to the impacts of all plasticisers on terrestrial ecosystems, therefore, remain a key research need within the wider plastics debate.

**Keywords:** plasticiser, soil, phthalate, microplastic, nanoplastic, plastic, terrestrial, plastic pollution, hazard, litter.

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## Introduction

Ever since the mass production of consumer plastic goods began in the mid-20th century, large items of plastic litter have left a clear visible legacy in the landscape. However, the true extent to which plastic pervades the environment came with the recognition of the microscopic fraction of plastic waste that had, until the early 21st century, remained largely unstudied. The term ‘microplastic’ was coined in 2004 (Thompson et al. 2004), and formally defined as ‘plastic particles smaller than 5 mm’ (Arthur et al. 2009). The additional terminology of nanoplastics was later added, with a size range of 1–100 nm being recognised as a definition (Potočnik 2011; Koelmans et al. 2015). The usage of these terms has evolved as the diversity in chemical and physical properties of microscopic plastic waste has become apparent, and there is an ongoing debate about how best to define and categorise these contaminants (Hartmann et al. 2019; Rochman et al. 2019). This emerging additional fraction of plastic waste adds to the visible burden of macroplastics as sources of plasticisers in the environment.

Research has shown that almost every environmental compartment on earth is contaminated with plastic (Howell et al. 2012; Bergmann et al. 2017; Horton et al. 2017a; Hendrickson et al. 2018; Büks and Kaupenjohann 2020). The majority of research to date has focussed on aquatic, particularly marine, environments, whereas the occurrence of microplastics in soils has only recently started to generate research interest. The scarcity of soil studies is partly a result of the analytical challenges associated with the extraction and identification of microscopic particles from such a heterogeneous matrix (Möller et al. 2020). Despite this, there is growing evidence of widespread microplastic contamination of the terrestrial environment (Scheurer and Bigalke 2018; Zhang and Liu 2018; Zhou et al. 2020).

Assessment of the impact of plastic waste on the terrestrial environment has tended to focus on the ecotoxicology of microplastic particles themselves. Laboratory studies have reported that microplastics can adversely affect soil fauna. For example, negative impacts on survival (Huerta Lwanga et al. 2016; Cao et al. 2017), biomass (Cao et al. 2017; Zhu et al.

2018a; Boots et al. 2019) and reproduction (Zhu et al. 2018a; Kim et al. 2019) have been reported for terrestrial species including earthworms (*Eisenia fetida*, *Eisenia andrei*, *Lumbricus terrestris*), collembola (*Folsomia candida*) and nematodes (*Caenorhabditis elegans*). The exposure doses used in many studies likely represent ‘worst-case scenario’ environmental concentrations, although direct comparisons are uncertain owing to a relative scarcity of environmental concentration data and inconsistencies in how such data are reported (Büks and Kaupenjohann 2020). Some of these negative effects likely arise from the physical damage caused by the microplastic particles to dermal surfaces or within the organism itself. However, in addition to these physical impacts, the chemically inert nature of polymers (da Costa et al. 2016) means that some of the toxicity indicated may not result from the plastics themselves, but rather from plastic-associated chemicals such as additives, or other substances which may have become bound to the plastic following its release to the environment.

Plastics are typically composed of the base polymer doped with additives – compounds associated with, but not themselves part of, the polymer structure. These additives may be released from the polymer matrix over the lifetime of the plastic. Although there are many additives which may be incorporated in plastics, e.g. plasticisers, flame retardants, UV and heat stabilisers, pigments and colourants, biocides etc., plasticisers and flame retardants are used in the largest quantities. Plasticisers typically constitute 10–70% w/w of the plastic item in which they are used (Hansen et al. 2013), with flame retardants constituting 3–25% (Hahladakis et al. 2018). However, given that flame retardants are not exclusively used in plastics, which makes the attribution of their presence to plastics challenging, plasticisers are the focus of this review.

Given the vast and complex nature of the polymer industry, there are many plastics and additives which fill niches for very specific applications. As such, it is beyond the scope of this review to discuss every plasticiser produced on a commercial scale. Instead, we focus on the plastics and plasticisers that have environmental significance owing to e.g. high production volumes, occurrence, toxicity or likelihood for future use as replacements for legacy plasticisers such as phthalates. We begin by providing an overview of plasticiser compounds. This is followed by a discussion of the mechanics of plasticiser release into the terrestrial environment, in addition to the sources and occurrence of plasticisers in soils. We then discuss the persistence of plasticisers in soils, in addition to their uptake by soil fauna and flora. Finally, we discuss existing knowledge gaps and provide recommendations for future research.

## Overview of plasticisers

### *Introduction and physiochemical properties*

Plasticisers are substances added to plastics to increase their flexibility. In Europe, 1.35 million tonnes of plasticiser are consumed each year, and the global market is estimated to be 7.5 million tonnes (CEFIC 2018). Most (~85%) plasticisers are used in polyvinyl chloride (PVC) (CEFIC 2018), and typically account for 10–70% w/w of the plastic item (Hansen et al. 2013). Physiochemical data and chemical structures of six major classes of plasticiser are shown in Table 1 and Table S1 (Supplementary Material) respectively.

The most commonly used class of plasticiser are the phthalic acid esters (also known as phthalates, PAEs). In 2017, phthalates accounted for 60% and 65% of the European and global plasticiser markets respectively. However, the use of phthalates



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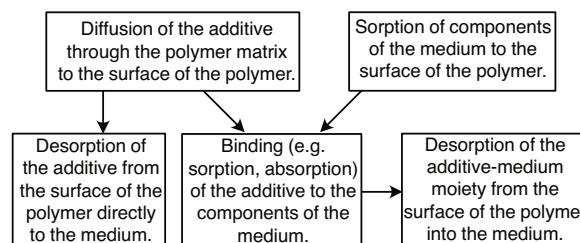
**Table 1.** Physicochemical data for the 21 phthalates and 5 emerging plasticisers discussed in this review; solubility in water is at 25 °C unless otherwise stated

Name and acronym	Class	Formula	Vapour pressure (Pa)	Log K <sub>OW</sub>	Log K <sub>OA</sub>	Log K <sub>AW</sub>	S <sub>H2O</sub> (mg L <sup>-1</sup> )	
Dimethyl phthalate	DMP	Phthalate	C <sub>10</sub> H <sub>10</sub> O <sub>4</sub>	2.63 × 10 <sup>-1</sup> <sup>A</sup>	1.61 <sup>A</sup>	7.01 <sup>A</sup>	-5.4 <sup>A</sup>	4000 <sup>C</sup>
Diethyl phthalate	DEP	Phthalate	C <sub>12</sub> H <sub>14</sub> O <sub>4</sub>	6.48 × 10 <sup>-2</sup> <sup>A</sup>	2.54 <sup>A</sup>	7.55 <sup>A</sup>	-5.01 <sup>A</sup>	1080 <sup>C</sup>
Dipropyl phthalate	DPtP	Phthalate	C <sub>14</sub> H <sub>18</sub> O <sub>4</sub>	1.74 × 10 <sup>-2</sup> <sup>A</sup>	3.40 <sup>A</sup>	8.04 <sup>A</sup>	-4.64 <sup>A</sup>	77 (nr °C) <sup>C</sup>
Di-iso-butyl phthalate	DiBP	Phthalate	C <sub>16</sub> H <sub>22</sub> O <sub>4</sub>	4.73 × 10 <sup>-3</sup> <sup>A</sup>	4.27 <sup>A</sup>	8.54 <sup>A</sup>	-4.27 <sup>A</sup>	6.2 (24 °C) <sup>C</sup>
Di-n-butyl phthalate	DnBP	Phthalate	C <sub>16</sub> H <sub>22</sub> O <sub>4</sub>	4.73 × 10 <sup>-3</sup> <sup>A</sup>	4.27 <sup>A</sup>	8.54 <sup>A</sup>	-4.27 <sup>A</sup>	11.2 <sup>C</sup>
Dimethoxyethyl phthalate	DMEP	Phthalate	C <sub>14</sub> H <sub>18</sub> O <sub>6</sub>	3.04 × 10 <sup>-2</sup> <sup>C</sup>	1.11 <sup>B</sup>			8500 (nr °C) <sup>C</sup>
Di-n-pentyl phthalate	DnPP	Phthalate	C <sub>18</sub> H <sub>26</sub> O <sub>4</sub>	1.28 × 10 <sup>-3</sup> <sup>A</sup>	5.12 <sup>A</sup>	9.03 <sup>A</sup>	-3.91 <sup>A</sup>	1.3 (nr °C) <sup>C</sup>
Diethoxyethyl phthalate	DEEP	Phthalate	C <sub>16</sub> H <sub>22</sub> O <sub>6</sub>		2.10 <sup>B</sup>			
Benzyl butyl phthalate	BBP	Phthalate	C <sub>19</sub> H <sub>20</sub> O <sub>4</sub>	2.49 × 10 <sup>-3</sup> <sup>A</sup>	4.7 <sup>A</sup>	8.78 <sup>A</sup>	-4.08 <sup>A</sup>	2.69 <sup>C</sup>
Diphenyl phthalate	DPhP	Phthalate	C <sub>20</sub> H <sub>14</sub> O <sub>4</sub>					
Dicyclohexyl phthalate	DCHxP	Phthalate	C <sub>20</sub> H <sub>26</sub> O <sub>4</sub>	1.16 × 10 <sup>-4</sup> <sup>C</sup>	6.20 <sup>B</sup>			4.0 (24 °C) <sup>C</sup>
Di-n-hexyl phthalate	DnHxP	Phthalate	C <sub>20</sub> H <sub>30</sub> O <sub>4</sub>	3.45 × 10 <sup>-4</sup> <sup>A</sup>	6.00 <sup>A</sup>	9.53 <sup>A</sup>	-3.53 <sup>A</sup>	0.159 (nr °C) <sup>C</sup>
Dimethylpentyl phthalate	DMPP	Phthalate	C <sub>20</sub> H <sub>30</sub> O <sub>4</sub>					
Di-n-heptyl phthalate	DnHpP	Phthalate	C <sub>22</sub> H <sub>34</sub> O <sub>4</sub>	2.76 × 10 <sup>-4</sup> <sup>C</sup>	7.56 <sup>C</sup>			1.83 × 10 <sup>-3</sup> <sup>C</sup>
Hexyl ethylhexyl phthalate	HEHP	Phthalate	C <sub>22</sub> H <sub>34</sub> O <sub>4</sub>		7.65 <sup>B</sup>			
Dibutoxyethyl phthalate	DBEP	Phthalate	C <sub>20</sub> H <sub>30</sub> O <sub>6</sub>	2.89 × 10 <sup>-1</sup> <sup>C</sup>	4.06 <sup>B</sup>			
Diethylhexyl phthalate	DEHP	Phthalate	C <sub>24</sub> H <sub>38</sub> O <sub>4</sub>	2.25 × 10 <sup>-5</sup> <sup>A</sup>	7.73 <sup>A</sup>	10.53 <sup>A</sup>	-2.8 <sup>A</sup>	0.27 <sup>C</sup>
Di-n-octyl phthalate	DnOP	Phthalate	C <sub>24</sub> H <sub>38</sub> O <sub>4</sub>	2.52 × 10 <sup>-5</sup> <sup>A</sup>	7.73 <sup>A</sup>	10.53 <sup>A</sup>	-2.8 <sup>A</sup>	0.022 <sup>C</sup>
Di-iso-nonyl phthalate	DiNP	Phthalate	C <sub>26</sub> H <sub>42</sub> O <sub>4</sub>	6.81 × 10 <sup>-6</sup> <sup>A</sup>	8.6 <sup>A</sup>	11.03 <sup>A</sup>	-2.43 <sup>A</sup>	0.2 (20 °C) <sup>C</sup>
Di-n-nonyl phthalate	DnNP	Phthalate	C <sub>26</sub> H <sub>42</sub> O <sub>4</sub>	6.81 × 10 <sup>-6</sup> <sup>A</sup>	8.6 <sup>A</sup>	11.03 <sup>A</sup>	-2.34 <sup>A</sup>	1.73 × 10 <sup>-5</sup> <sup>C</sup>
Di-iso-decyl phthalate	DiDP	Phthalate	C <sub>28</sub> H <sub>46</sub> O <sub>4</sub>	1.84 × 10 <sup>-6</sup> <sup>A</sup>	9.46 <sup>A</sup>	11.52 <sup>A</sup>	-2.06 <sup>A</sup>	0.28 <sup>C</sup>
Diethylhexyl adipate	DEHA	Adipate	C <sub>22</sub> H <sub>42</sub> O <sub>4</sub>	1.13 × 10 <sup>-4</sup> <sup>C</sup>				0.78 (22 °C) <sup>C</sup>
Diethylhexyl terephthalate	DEHTP	Terephthalate	C <sub>24</sub> H <sub>38</sub> O <sub>4</sub>	2.85 × 10 <sup>-3</sup> <sup>C</sup>				4.0 (20 °C) <sup>C</sup>
Acetyl tributyl citrate	ATBC	Citrate	C <sub>20</sub> H <sub>34</sub> O <sub>8</sub>	4.00 × 10 <sup>-2</sup> <sup>C</sup>				1.7 <sup>C</sup>
Di-iso-nonyl cyclohexanoate	DiNCH	Cyclohexanoate	C <sub>26</sub> H <sub>48</sub> O <sub>4</sub>					
Triooctyl trimellitate	TOTM	Trimellitate	C <sub>33</sub> H <sub>54</sub> O <sub>6</sub>	5.07 × 10 <sup>-4</sup> <sup>C</sup>				3.9 × 10 <sup>-4</sup> <sup>C</sup>

<sup>A</sup>Net et al. (2015).<sup>B</sup>Hu et al. (2020).<sup>C</sup>PubChem (2021).

in Europe is declining, with an estimated decrease in market share of over 25 % between 2005 and 2017 (CEFIC 2018). This change has been driven by concerns surrounding the potential endocrine disrupting properties of some phthalates, leading to restrictions on their use in the European Union (ECHA 2018). The increased regulatory pressure on phthalates is leading to diversification of the market. Terephthalic acid esters (aka. terephthalates), such as diethylhexyl terephthalate (DEHTP), were a minor part of the European market in 2005, but in 2017 accounted for 12 % of the European market (and 15 % globally) (CEFIC 2018). Other emerging classes of plasticiser include trimellitates (e.g. triooctyl trimellitate, TOTM), citrates (e.g. acetyl tributyl citrate, ATBC), adipates (e.g. diethylhexyl adipate, DEHA) and cyclohexanoates (e.g. di-iso-nonyl cyclohexanoate, DiNCH) (Table 1). Given the growing diversity in the classes of plasticiser being used in plastics, it is becoming increasingly important to assess not only the risk of phthalates in the environment, but the risk posed by emerging plasticisers, most of which have very little associated ecotoxicology or occurrence data.

Given that the function of all plasticisers is identical, these compounds have similar properties (Table 1). Plasticisers are generally viscous, lipophilic (and some highly lipophilic) and have low water solubility. For example, the phthalates diethylhexyl phthalate (DEHP), di-n-butyl phthalate (DnBP), di-iso-butyl phthalate (DiBP) and benzyl butyl phthalate (BBP) have logK<sub>OW</sub>, logK<sub>OA</sub> and logK<sub>AW</sub> in the range 4.27–7.73, 8.54–10.53 and -4.27 to -2.80 respectively (Net et al. 2015). Plasticisers have boiling points >300 °C, in addition to very low

**Fig. 1.** Migration and leaching processes for an additive into a medium.

vapour pressures, and therefore the gaseous release of these compounds from their liquid forms should be slow under environmental conditions.

### Plasticiser leaching

Plasticisers are not chemically bonded to the polymer matrix, and so can migrate over time to the surface of the plastic particle, where they can leach out to the environment. The ability of a molecule to migrate is a pre-requisite for some applications, e.g. the controlled release of drug molecules from a polymer matrix (Liechty et al. 2010). However, unwanted migration of additives also occurs over the lifetime of a plastic item, e.g. into food products from contact material or from medical use materials (Welle et al. 2005; Fankhauser-Noti et al. 2006). The process of plasticiser migration into foodstuffs has been conceptualised as a sequence of four steps (Ferrara et al. 2001), but this summary can be generalised for any environmental medium (Fig. 1).

Despite the apparent ubiquity of plastic in the terrestrial environment, there have been no published studies which assess the leaching kinetics of plasticisers directly into soil under controlled conditions, although attempts have been made to quantify their release in aqueous media. For example, the effects of plastic weathering on the leaching rates of a wide range of additives, including some plasticisers, from polyethylene, PVC and polyethylene terephthalate (polyester) into freshwater and saltwater were determined in one study (Suhrhoﬀ and Scholz-Böttcher 2016). Salinity and UV light were associated with a clear increase in the total additive release in only one instance (a printed polyethylene bag), with UV light appearing to reduce additive release in some cases. The effects of salinity were complex and appeared to be compound-specific, whereas the reduction of leached additive in some UV-exposed samples may have been caused by photodegradation of analytes. In contrast, turbulence significantly increased the total additive load in solution for all plastics, which was attributed to the continuous mixing of the solution preventing concentration gradients from forming at the plastic–solution boundary. Of all the target analytes, the plasticiser ATBC showed the highest leaching potential into ambient saltwater relative to its concentration in the test material (4.9% ATBC loss over 78 days from a sample of polyethylene). ATBC also appeared to be associated with printed plastic, underlining the composite nature of many items of plastic waste. In contrast to ATBC, the leaching of phthalates from PVC over 57 days was low, e.g. DEHP leached just 0.083% of its initial concentration, although further research is needed to understand leaching kinetics over longer timescales. Although it remains challenging to study the aqueous release of inherently hydrophobic plasticisers, recent work has demonstrated that an ‘infinite sink’ approach, which has been previously used to study aqueous release of polychlorinated biphenyls from plastic (Endo et al. 2013), could be applied successfully to the study of DEHP release from PVC (Henkel et al. 2019).

Despite the lack of environmental studies, there is a significant body of research on the leaching of plasticisers from food contact materials and medical use plastics. Given that the plastics and plasticisers used in these applications are likely to be similar, if not the same, as those entering terrestrial systems through e.g. mismanaged consumer waste or spent agricultural mulching films, the results of these studies can be used to predict and understand the dominant leaching processes relevant to the soil environment. In general, substances with lower molecular weights migrate from plastics at a faster rate as they can travel through the polymer matrix more freely to reach the polymer-matrix boundary (Hansen et al. 2013). In addition, plasticisers with a lower affinity for the polymer matrix are likely to have faster migration rates than those whose physiochemical properties are more similar to the polymer. Significant plasticiser migration occurs at higher lipid concentrations in the contact medium, whereas the leaching of plasticisers into aqueous media is very slow. This has been widely observed for phthalates in addition to emerging plasticisers (Goulas et al. 2000, 2007; Pedersen et al. 2008; Nara et al. 2009; Bueno-Ferrer et al. 2010; Kastner et al. 2012; Coltro et al. 2014; Choi et al. 2018). Despite the lack of published studies regarding the rates of plasticiser leaching to environmental media, the lipophilicity of plasticisers suggests that organic matter content and water holding capacity of a soil, in addition to soil water pH and ionic strength, will influence plasticiser leaching kinetics. Furthermore, this pattern has implications for the bioaccumulation and partitioning of plasticisers in animal tissues, although the fate of plasticisers

within soil fauna is largely unknown. Leaching also shows a clear positive correlation with temperature (Hanusová et al. 2013), thus plasticiser releases through leaching may show clear seasonal and regional climate-associated trends.

Determining the relative leaching rates of plasticiser classes is made challenging by the absence of reference plastics with known plasticiser formulations and percentage weight concentrations. In addition, concentrations of plasticisers in commercially available plastic items used in kinetic comparison studies are sometimes not reported, despite the fact that plasticiser content is a limiting factor determining leaching rate (Bueno-Ferrer et al. 2010). In the existing food contact and medical use literature, the phthalate DEHP is typically compared with one or more emerging plasticisers, with DEHP often reported as the fastest migrating substance (Tüzüm Demir and Ulutan 2013; Bernard et al. 2015; Eckert et al. 2016). DEHP may leach relatively quickly because it has a lower molecular weight than emerging alternative plasticisers, such as trimellitates (Table 1 and Table S1, Supplementary Material), although further work is required to fully understand which physiochemical properties play the most important role in determining plasticiser migration, and how the relative importance of these properties may be affected by the contact medium or polymer type. Nevertheless, multiple studies have found the high molecular weight trimellitate plasticiser TOTM to leach very slowly in comparison to other tested plasticisers (Welle et al. 2005; Bernard et al. 2015; Suhrhoﬀ and Scholz-Böttcher 2016), which suggests that molecular weight is indeed an important factor controlling migration rate. For example, TOTM leached just 0.23% of its initial content in PVC into a 1 : 1 ethanol : water solution after 10 days, and relative leaching rates were in the order DEHP > DiNCH > DEHTP > TOTM (Bernard et al. 2015). An earlier study reported rates in the order ATBC > DEHP > DiNCH > TOTM, with the citrate plasticiser ATBC detected at concentrations exceeding an order of magnitude higher than DEHP, in spite of the fact that ATBC was present in the lowest amounts in the test samples (Welle et al. 2005). In addition, ATBC was detected in a jarred foodstuff at 60 mg kg<sup>-1</sup>, despite being present in the lid at a low concentration (0.9% w/w), which implies high mobility (Fankhauser-Noti et al. 2006). Consequently, this plasticiser was more frequently detected than four phthalates in plastic-packaged foodstuffs (García Ibarra et al. 2018). Thus, these studies indicate that some of the emerging plasticisers may have significantly higher leaching capabilities than phthalates, even though the latter group are generally the only plasticiser class targeted in terrestrial occurrence and bioaccumulation studies.

In addition to plasticiser properties, the density and flexibility of the plastic polymer chains themselves can also affect plasticiser leaching rates (Hansen et al. 2013). Polymers with high glass transition temperatures generally have less migration, as the polymer chains are less free to move. In addition, plasticisers within crystalline polymer matrices or those with greater cross-linking and branching will migrate slower than those in polymers with amorphous structures, as there are smaller spaces between polymer chains through which molecules may travel. The effects of plastic properties on plasticiser migration rate have clear implications for understanding how plastics may act as plasticiser sources in the environment. For example, a crosslinked, branched polymer with a high glass transition temperature is more likely to act as a persistent, low-level input of plasticisers than a straight-chain, low glass transition temperature polymer which may produce a more rapid release.

The first step towards resolving the uncertainty surrounding plasticiser leaching in the terrestrial environment should be identifying the plastics which have the greatest potential for plasticiser release, in addition to the fates and sink areas of these plastics in the environment. This could be achieved through a systematic study of common polymers under controlled conditions to begin to understand the general trends that could lead to particularly high rates of leaching, thus allowing future work to target plastics or plasticisers which appear to be of particular concern. Most plasticisers are used in PVC, although this polymer is rarely reported in terrestrial occurrence studies (Büks and Kaupenjohann 2020). Therefore, the contribution of polymers such as polyethylene and polypropylene to total terrestrial plasticiser load may be greater than initially expected given their low plasticiser content. Leaching characteristics of a set of common polymers directly into a suite of soils with contrasting properties should be determined, in addition to the influence of plastic particle size and morphology (e.g. film, fibre etc.) on leaching rate. This data, in conjunction with data on environmental plastic waste occurrence, could be used to identify plastics of particular concern to gauge the nature and scale of potential plasticiser sources. In addition, experiments to determine leaching rates of different plasticiser classes and the effects that soil properties, e.g. pH, organic matter content, clay content etc., have on plasticiser leaching rate are critically needed, as the trends observed in the food contact and medical use literature may not hold true for more complex media such as soils. Weathering of polymers through climate (e.g. UV light) or organism-driven processes may also determine the release of plasticisers to the environment, although long-term field experiments are required to identify and quantify the nature of such effects.

### Sources of plasticisers in the terrestrial environment

Given the ubiquity of plastic waste in the environment, the leaching of plasticisers from plastic debris likely represents a common, diffuse source of plasticiser into the environment. For example, we estimate the total annual input of plastic waste to continental (terrestrial and freshwater) environments in the EU and the UK to be 130 kg of plastic per km<sup>2</sup> (Horton et al. 2017b; World Bank 2017). In addition to this widespread source, input of plasticisers may occur through intensive plastic use and management activities associated with point sources. The relative contribution of these sources to the total terrestrial plasticiser load has yet to be estimated, although given the levels of plastic litter often seen in terrestrial environments, it is reasonable to assume that the diffuse route may account for the majority of plasticiser occurrence in wider soils. Therefore, to understand the sources of plasticisers in the terrestrial environment, an understanding of plastic sources and occurrence is necessary given the likely intrinsic link between plastic presence and plasticiser inputs.

#### Urban areas

Plastic litter represents the most visible and obvious possible source of microplastics and plasticisers in soils. This waste will be generated and concentrated primarily in urban areas, particularly in countries without widespread formal waste disposal networks, and such areas have indeed been identified as hotspots of plasticiser concentration (Hongjun et al. 2013). The

weathering of plastic debris may indirectly increase the input of plasticisers to soils, e.g. consumer goods such as food packaging films become fragmented in the environment, increasing their surface area and leaching potential (Horton et al. 2017b). Although plastic items are primarily used and disposed of in urban areas, many items of large plastic waste, and especially primary or secondary microplastics, may be transported long distances to areas without a distinct plastic input by short- and long-range aerial transport, human activities and overland water flows (Horton and Dixon 2018). Therefore, diffuse plastic litter likely accounts for most of the widespread chronic input of plasticisers in soils at sites away from areas of immediate input (see below – *Multimedia transport as a route to land*).

The majority of plasticised items are used in urban areas. This includes those associated with consumer items and packaging and also larger-scale structural materials. The latter includes wires, cabling, flooring and wall coverings which together account for 45 % of plasticiser use in Europe, compared with only 13 % of plasticisers that are used in consumer goods and industry (CEFIC 2018). Many of these plasticised items will be used in a single location for a long period of time before disposal, and thus may provide a steady input of plasticisers into the environment. In addition, most plasticisers are used in PVC, which is widely used in e.g. building fittings and municipal water pipes, although many of these applications use rigid PVC which has a lower plasticiser content than more pliable PVC-based materials. Transport infrastructure can also act as a significant source of plastics, and therefore plasticisers. Soils in the vicinity of roads receive plastic contamination, e.g. mismanaged plastic litter or particles produced during the mechanical wear of tyres against the road surface, known as tyre-wear particles (TWPs) (Wagner et al. 2018; Knight et al. 2020). The compositions of TWPs are incredibly complex, although the potential for TWPs to act as sources of plasticisers in soils remains unexplored.

Multiple studies have linked urban activity with increased phthalate concentrations (Zeng et al. 2008; Hongjun et al. 2013; Zhang et al. 2014; Škrbić et al. 2016; Sun et al. 2016). However, not all studies have identified a clear link between urban activity and plasticiser loading. For example, an extensive survey of DEHP in Scottish soils from various land uses found no consistent link between DEHP contamination and distance from urban areas (Rhind et al. 2013a). Many urban areas will see a complex pattern of inputs from many sources, e.g. horticulture, waste disposal and industry. Further work is required to pull apart and enumerate the extent to which these inputs contribute to plasticiser occurrence in urban soils. Studies which measure diverse suites of plasticisers, in addition to exploiting continuing improvements in the ability to extract and identify the microplastic composition of soils (Möller et al. 2020), are needed to better establish the nature of these links.

#### Waste disposal sites

Landfills are the ultimate disposal site for a large proportion of plastic waste; however, the extent to which such sites act as a source of plasticiser contamination to the wider soil environment remains unclear. For example, the mean total phthalate concentration ( $\Sigma_{16}$  phthalates) of soils on a landfill site in China was 4.22 mg kg<sup>-1A</sup>, but the contamination in adjacent topsoils (0.68 mg kg<sup>-1</sup>) was significantly lower than many agricultural

<sup>A</sup>Plasticiser concentrations discussed in the text have, where possible, been quoted with either 'dw' or 'ww' to indicate whether they refer to dry or wet weight values respectively. However, this information was not reported in all studies.

and urban soils in similar study areas (Liu et al. 2010). Thus, plasticisers associated with the landfill seemed to be retained in situ. Total mean phthalate concentration ( $\Sigma_5$  phthalates) in soils in the vicinity of landfill in four Indian cities was also low, at  $0.093 \text{ mg kg}^{-1}$  (Chakraborty et al. 2019), although in contrast the adipate plasticiser DEHA was present at  $0.057 \text{ mg kg}^{-1}$ , which suggested that this substance may have higher mobility than some phthalates. In some instances, intense bacterial activity may cause landfills to act as sinks of plasticiser degradation, rather than being a significant source. For example, the phthalates DEHP, diethyl phthalate (DEP), DnBP and BBP were found in only 29–47% of leachates from primarily municipal landfills in four European countries, and dimethyl phthalate (DMP) was not detected at all (Jonsson et al. 2003), but there was widespread presence of phthalate monoesters in the leachates, at significantly higher concentrations. The results of one study suggest that downwards penetration of landfill leachates may act as a source of plasticisers in groundwater. Total phthalate concentration ( $\Sigma_{16}$  phthalates) in groundwater associated with a landfill site was over 3.5 times higher than in the surface water ( $6.35 \mu\text{g L}^{-1}$  versus  $1.78 \mu\text{g L}^{-1}$ ), despite the fact that the landfill was lined with a geomembrane (Liu et al. 2010). However, this difference may be driven by significantly slower biodegradation of phthalates in groundwater as opposed to surface water, as phthalate degradation is less rapid under anaerobic conditions (Shanker et al. 1985).

Sites of electronic waste (e-waste) processing, particularly in countries with informal e-waste recycling sectors, appear to represent a much greater phthalate input than standard domestic waste landfills in terms of both abundance and diversity of congeners. Mean total phthalate concentrations ( $\Sigma_5$  phthalates) were over 4 times higher at e-waste processing sites than at landfills in four Indian cities ( $0.396$  versus  $0.093 \text{ mg kg}^{-1}$ ). Plasticiser contamination was especially concentrated at sites where precious metal recovery occurred, which may be attributed to the fact that such sites could be associated with plastic incineration (Chakraborty et al. 2019). A further study of phthalates in soils near a large e-waste processing site by Zhang et al. (2019) also provided clear evidence of airborne transport of phthalates released during plastic-containing e-waste incineration. Soil potentially affected by these inputs exhibited a profile of phthalates not seen in agricultural soils, with the occurrence of dibutoxyethyl phthalate (DBEP,  $4.49 \text{ mg kg}^{-1} \text{ dw}$ ), dimethylpentyl phthalate (DMPP,  $2.88 \text{ mg kg}^{-1} \text{ dw}$ ) and dicyclohexyl phthalate (DCHP,  $1.66 \text{ mg kg}^{-1} \text{ dw}$ ), whereas common phthalates such as DnBP and DEHP were found at concentrations of  $1.92 \text{ mg kg}^{-1} \text{ dw}$  and  $1.05 \text{ mg kg}^{-1} \text{ dw}$  respectively ('dw' refers to dry weight value). Exceptionally high levels of phthalate contamination ( $\Sigma_5$  phthalates), ranging from 12.6 to  $46.7 \text{ mg kg}^{-1} \text{ dw}$ , were also found in soils from three e-waste processing sites in China, further indicating the potential of such sites as sources of plasticiser release (Liu et al. 2009).

#### *Plastic use in agriculture*

Most plastic used in agriculture is used in China (Espí et al. 2006), although use is also common in other regions. Of the 23 studies discussed by Büks and Kaupenjohann (2020) in their recent review of terrestrial microplastic occurrence, 11 were for Chinese soils, 6 European, 4 from the Americas, 1 from Iran and 1 from Australia. Of the 14 studies which conducted polymer identification, only 5 detected PVC, the polymer with the highest plasticiser loading. Furthermore, even when identified, PVC was at lower levels than other widely used consumer

polymers (Fuller and Gautam 2016; Ljung et al. 2018; Lv et al. 2019; Chen et al. 2020; Ding et al. 2020). The most commonly detected polymers in soils are polypropene and polyethylene, with these two polymers dominating microplastic profiles (Liu et al. 2018; Lv et al. 2019; Yu et al. 2021). A caveat on these results of polymer presence is that microplastic extraction procedures typically employ a density separation step to separate microplastics from soil particles. Of the terrestrial microplastic concentration studies, 30% employed a density separation in either water ( $\rho = 1.0 \text{ g mL}^{-1}$ ) or NaCl solution ( $\rho = 1.2 \text{ g mL}^{-1}$ ). Density separation can preclude the effective recovery of polymers such as PVC and polyester, as these polymers have  $\rho > 1.2 \text{ g mL}^{-1}$ . However, even in the studies that used density separation with a media theoretically capable of floating PVC, this polymer was still detected in only 3 out of 8 studies (Büks and Kaupenjohann 2020). Thus, as polyethylene and polypropene, and not PVC, appear to dominate soil microplastic profiles, the majority of plasticiser input from microplastics into agricultural soils will derive from widely dispersed but low-level leaching from these widely used polymer types.

A diverse range of plastic items are commonly used in agriculture, e.g. polytunnels, bale wrappings, irrigation tubing, twine etc, although plastic mulches have received the most interest as a source of microplastics and plasticisers in soils. Mulches often cover entire fields and may contain plasticisers in small proportions (Steinmetz et al. 2016). An individual sheet is typically used for a matter of months and may simply be ploughed back into the field at the end of its useful life. As such, plastics may accumulate in soils where mulch is reapplied on an annual or even sub-annual basis. Studies have reported positive correlations between the mass of plastic mulch used and microplastics recovered from agricultural soils, in addition to higher levels of microplastic abundance at sites with a longer history of mulch use (Huang et al. 2020, 2021; Yu et al. 2021).

Phthalate profiles of mulched agricultural soils are dominated by DEHP, and to a lesser extent DnBP, DiBP and di-n-octyl phthalate (DnOP), although the exact suite of target phthalates differs between studies (Hu et al. 2003; Kong et al. 2012; Wang et al. 2013a; Chai et al. 2014; Kaewlaoyong et al. 2018; Zeng et al. 2020). In addition to DEHP, DnOP in particular appears to be linked to agricultural plastic in some cases. For example, DEHP and DnOP were substantially more abundant than other targeted phthalates in agricultural plastic products on the Taiwanese market (e.g. DnBP was not present in mulch at all), and phthalate profiles of mulched soils resembled those found in the plastic products (Kaewlaoyong et al. 2018). In addition, DnOP was found to be almost 6 times as abundant in mulched non-vegetable soils than non-mulched, whereas concentrations of DMP, DEP and DEHP were only marginally higher (9.7–26.9%) (Kong et al. 2012). However, this pattern was reversed in mulched vegetable soils, which indicated that the type of mulch used may affect the profile of phthalates found in soils. This indicates the need for greater knowledge of the range and composition of mulched material, in order to develop an understanding of the potential inputs of different plasticisers into soils.

Based on widespread use and localised input potential directly to soil, there is strong evidence that plastic mulch is positively correlated with, and likely causes, increased phthalate concentrations in soils (Hu et al. 2003; Kong et al. 2012; Wang et al. 2013a, 2013b; Zhang et al. 2015; Sun et al. 2016; Zeng et al. 2020). For example, individual concentrations of all six targeted phthalates were between 62% and 6439% higher in mulched vegetable soils than in non-mulched soils in one

Chinese study (Kong et al. 2012). Similarly, total phthalate concentrations ( $\Sigma_{16}$  phthalates) were significantly higher in Chinese agricultural facilities where mulch was used ( $2.25 \text{ mg kg}^{-1} \text{ dw}$ ) than in mulch-free facilities ( $1.25 \text{ mg kg}^{-1} \text{ dw}$ ) (Zeng et al. 2020). The effect of seasonal mulch use was also investigated in one study (Zhang et al. 2015), with total phthalate concentrations ( $\Sigma_{15}$  phthalates) peaking in the summer. This was associated with a period of increased mulch use, and possibly coincided with greater leaching of phthalates owing to higher ambient temperatures. However, although there is a potential association of phthalate release with climate, it is not possible to attribute such seasonal variation entirely to higher temperatures, as summer also sees increased fertiliser application and greater irrigation, both of which may increase phthalate input.

Although there is reasonably good evidence linking mulch use to plastic load and soil plasticiser levels, the evidence for the role of other agricultural plastic products in determining plastic associated loading to soils is less well-established. For example, the use of polytunnels does not appear to increase either microplastic or plasticiser abundance in soils. For instance, a recent study found that polytunnel use was not associated with a significant difference in microplastic abundance when compared with non-polytunnelled sites (Yu et al. 2021). No significant difference was found between phthalate soil concentrations in greenhouses and open fields (Zeng et al. 2020). Sites which used a combination of mulch and polytunnels had significantly higher phthalate concentrations than sites which used polytunnels alone, although a shorter cultivation history in polytunnel-only sites likely accounted for some of this difference (Wang et al. 2013a).

#### *Wastewater treatment products in agricultural soils*

The wastewater treatment (WWT) process involves removing particulates and chemicals from the influent before releasing the treated effluent into water courses. Solids removed during this process are referred to as biosolids or sewage sludge, and it is here that plastics and plasticisers entering the WWT plant will generally become concentrated (Gibson et al. 2005; Simon et al. 2018; Ball et al. 2019; Lee et al. 2019a). A recent study of UK WWT plants found that less than 0.01 % of influent microplastic particles were released in the effluent. The remainder partitioned into sludge, with no clear difference in the extent of this separation between polymer types (Ball et al. 2019). Similar removal efficiencies (>99 %) of microplastics to sludge have also been reported in Danish WWT plants, which indicates the dominance of sludge as the sink for particles during treatment (Simon et al. 2018).

The removal of plasticisers during the WWT process is both better studied and potentially more complex than the partitioning of plastics. Although some modern WWT plants use a combination of processes which can degrade phthalates at levels between 80 % and 100 % (Gao and Wen 2016), plasticisers still become highly concentrated in sludge (Gibson et al. 2005; Tran et al. 2015; Lee et al. 2019a) and may also be present in effluent (Zolfaghari et al. 2014; Lee et al. 2019a). The partitioning of plasticisers to sludge is strongly influenced by their  $K_{OW}$  and molecular weight ( $M_W$ ), with the majority of low  $M_W$  plasticisers (e.g. DMP) released in effluent, but upwards of 90 % of higher  $M_W$  plasticisers (e.g. DEHP) bound to sludge (Lee et al. 2019a). This indicates that both effluent and sludge can act as a source of plasticisers to surface water and soil, although the extent may be congener specific.

In many countries, sewage sludge is applied to agricultural land as a soil enhancer in considerable quantities, e.g. 80 % of the UK's sewage sludge is spread on agricultural soil (DEFRA 2012). Hence, this route represents a potentially significant source of plastics and plasticisers in the soil environment (Tran et al. 2015; Büks and Kaupenjohann 2020). Owing to challenges and inconsistencies between extraction and analytical methodologies, estimates for microplastic concentrations in sewage sludge vary widely, from 10s to 1000s of microplastics per g dry sludge (Zubris and Richards 2005; Talvitie et al. 2017; Ball et al. 2019; Liu et al. 2019; Crossman et al. 2020). Polyethylene and polypropene, and to a lesser extent polyester and nylon, dominate the microplastic profiles of sewage sludges (Vollertsen and Hansen 2017; Ball et al. 2019; Crossman et al. 2020). The absence of PVC, which often has a very high plasticiser load, amongst the commonly detected polymers suggests that the plastic burden in sludge may result in lower plasticiser soil inputs than would be expected from a simple assessment based on production volume analysis (CEFIC 2018).

Historically DEHP and DnBP have been the dominant phthalates detected in sewage sludge. DEHP and DnBP were detected in every sludge sample investigated in a study of German WWT plants, at  $28\text{--}154 \text{ mg kg}^{-1} \text{ dw}$  and  $0.2\text{--}1.7 \text{ mg kg}^{-1} \text{ dw}$  respectively (Fromme et al. 2002), and accounted for 79 % and 18 % of the total phthalate concentration ( $\Sigma_{16}$  phthalates) of sewage sludge from Chinese WWT plants (Meng et al. 2014). DEHP was detected at  $62.5 \text{ mg kg}^{-1}$  and accounted for >95 % of total phthalates ( $\Sigma_6$  phthalates) in UK sewage sludge (Gibson et al. 2005) and was also detected at comparable levels ( $27.54\text{--}51.03 \text{ mg kg}^{-1}$ ) in five Chinese sewage sludge samples (Cheng et al. 2010). The three most abundant plasticisers detected ( $\Sigma_9$  phthalates) in sludge from a French WWT plant were the high  $M_W$  phthalates DEHP ( $\sim 12.5 \text{ mg kg}^{-1} \text{ dw}$ ), di-iso-nonyl phthalate (DiNP,  $\sim 10 \text{ mg kg}^{-1} \text{ dw}$ ) and di-iso-decyl phthalate (DiDP,  $\sim 5 \text{ mg kg}^{-1} \text{ dw}$ ) (Tran et al. 2015). These levels are consistent with those from both the UK and Chinese studies, which indicates a widespread and consistent nature of inputs of the dominant phthalate congeners into wastewater systems.

A recent study of sludge from WWT plants in South Korea ( $\Sigma_{16}$  phthalates,  $\Sigma_5$  emerging) found that emerging plasticisers are present in sludge in comparable levels to phthalates (Lee et al. 2019a). This demonstrates that sewage sludge may represent an input of emerging plasticisers into soils. DEHP had the highest mean concentration of any plasticiser ( $71 \text{ mg kg}^{-1} \text{ dw}$ ), followed by the terephthalate DEHTP ( $25 \text{ mg kg}^{-1} \text{ dw}$ ). Mean concentrations of other emerging plasticisers (DiNCH, ATBC, DEHA and TOTM) were considerably lower, ranging from  $0.34$  to  $1.2 \text{ mg kg}^{-1} \text{ dw}$ , although these substances were present in comparable quantities to commonly reported phthalates, such as BBP, DnBP and DiBP, which indicates their importance to wider plasticiser loading in sludge.

Despite considerable microplastic concentrations in sewage sludge, the extent to which repeated sludge application leads to long-term accumulation of plastics in soils remains unclear. A recent literature review noted that agricultural soils with a history of sludge application generally have microplastic concentrations approximately an order of magnitude higher than soils not receiving sludge inputs (Büks and Kaupenjohann 2020). However, limited data, analytical challenges and the potential for local-scale factors to influence results precludes a definitive conclusion on the link between repeat input and microplastic loads. For example, a recent study estimated that

over 99 % of the applied microplastic load was not retained in the soil, and only one out of three sites demonstrated a net gain of microplastics at ~6 months after the application of sewage sludge (Crossman et al. 2020). This was attributed to a prolonged period of unusually heavy rainfall following sludge spreading, which led to considerable losses of soil microplastics owing to e.g. surface runoff. Further work is required to elucidate the effects of climate and land management practices on the long-term accumulation of plastic in sludge-amended soils.

Sewage sludge application has been more clearly linked with both the accumulation and long-term persistence of plasticisers in soils, although evidence to date is limited to phthalates. DEHP levels in soils with a 13-year history of sludge application were ~4 times higher than controls ( $0.22 \text{ mg kg}^{-1} \text{ dw}$  v.  $0.06 \text{ mg kg}^{-1} \text{ dw}$ ) (Rhind et al. 2013b). The same study monitored DEHP concentrations in soils for 10 weeks after applications of sludge in spring and autumn. No clear seasonal difference was found, although DEHP concentrations began to increase ~3 weeks after sludge application and were typically around 3–6 times greater than control soils. Such a lag was not seen in a later study, which found DEHP concentrations in topsoil increased almost 8-fold immediately after application of sludge to agricultural land ( $0.03$  to  $0.24 \text{ mg kg}^{-1} \text{ dw}$ ) (Tran et al. 2015). After 6 months, the mean concentration of DEHP across four soil depths (0–20, 20–40, 40–60, 60–80 cm) was 21 % higher than after the initial sludge spreading, which was attributed to the release of the plasticiser from bound residues within the soil during re-ploughing. Vikelsoe et al. (2002) measured the concentrations of seven phthalates at a site with a history of high sludge application. Despite the fact that sludge application had ceased 6 years before sampling, individual plasticiser concentrations at this site were at least an order of magnitude higher than at neighbouring locations in which fertilisation was through manure, artificial fertiliser or a low–medium level sludge amendment. Such long-term persistence of phthalates in soil was also demonstrated in a study of soil amended with sewage sludge for 18 years, followed by a 12-year period without further addition. DEHP accumulated in the historically treated soil ( $1.47 \text{ mg kg}^{-1} \text{ dw}$  at year 15) and concentrations were over 6 times higher than background levels even after the 12-year application-free period (Patureau et al. 2007). In contrast, one study found no sustained increase in DEHP concentrations in soils amended with sludge for 2.5 years relative to control soils, and DEHP levels remained comparable between the soils for the duration of the experiment (Rhind et al. 2002). Nevertheless, despite some differences between studies that warrant further investigation, the weight of evidence suggests that sewage sludge application to soils can lead to high levels of phthalate contamination which persist long after sludge spreading has ceased, possibly owing to a reduction in bacterial bioavailability arising from strong partitioning of phthalates to soil organic matter.

#### *Fertilisers and agricultural soils*

A recent study has demonstrated that the use of mineral fertilisers in which the active ingredient is coated with plastic microcapsules may lead to significant plastic contamination in farmland soils ( $144 \text{ mg kg}^{-1}$  plastic,  $n = 19$  sites). Whether this also represents a route of plasticiser input currently remains unclear (Katsumi et al. 2021). Fertilisers themselves have been shown to contain considerable quantities of phthalates. Concentrations in organic fertilisers are higher than in inorganic

ones, although reported concentrations vary considerably from trace levels to over  $2.5 \text{ mg kg}^{-1} \text{ dw}$  (Mo et al. 2008; Kaewlaoyoong et al. 2018; Song et al. 2021). The mass and frequency with which fertilisers are applied, in addition to their water solubility and wide dispersal, has raised questions over their potential as a source of phthalates in agricultural soils and water courses. To date, there appears to be few systematic studies of plasticiser input from fertilisers into soils. Some studies have linked increased fertiliser use with higher phthalate concentrations, although additional inputs, such as plastic mulch and irrigation water, precluded definitive source apportionment (Wang et al. 2013b; Song et al. 2021). There is some indication that fertiliser use may not cause a sustained phthalate flux, e.g. increased phthalate concentrations in soils coincided with increased fertiliser application in summer, but by autumn contamination had generally returned to spring levels (Zhang et al. 2015). However, further work on these dynamics would be warranted in studies looking to establish the dominant route of plasticiser inputs into agricultural soils.

#### *Multimedia transport as a route to land*

Long-range aerial transport of persistent organic pollutants (POPs) is thought to occur by the process of global distillation, whereby vaporisation, aeolian transport and condensation allow for the movement of chemicals produced in lower latitudes towards the poles. Using an established framework for predicting how a POP travels in the atmosphere (Wania 2003), plasticisers will likely be subject to similar transport patterns as other types of organic pollutant such as polychlorinated biphenyls (PCBs), e.g. multiple distillations towards the high latitudes (Lohmann et al. 2007). In addition to the conventional transport mechanisms identified to date for organic chemicals, phthalates may also move by their association with plastics that travel in the atmosphere, leading to redistribution worldwide, which includes to remote soils (Feng et al. 2020). Therefore, long-range aerial transport of plasticisers and plastics of all size ranges can input these contaminants into soils at considerable distances from source areas. Plastics and plasticisers may also move locally within source areas. For example, wet and dry deposition from a nearby urban area was suspected to account for notable phthalate occurrence ( $\Sigma_{16}$  phthalates =  $1.67 \text{ mg kg}^{-1} \text{ dw}$ ) in woodland soil in a national park (Zeng et al. 2009), and occurrence of a distinct phthalate profile correlated with distance from an e-waste processing site (Zhang et al. 2019).

The transport of plastics and plasticisers from water courses may also lead to inputs into soil. Effluent from WWT plants contains a multitude of plasticisers (Lee et al. 2019a), and despite the fact that WWT plants can remove the majority of microplastic particles from influent (Simon et al. 2018; Ball et al. 2019), some microplastics will still be discharged in the effluent owing to the large volumes of influent that are processed. Flooding events or the extraction of riverine water for irrigation could therefore act as inputs of these contaminants to soils, although this remains largely unstudied. Food webs have also been identified as a route by which organic pollutants (Christensen et al. 2005; Previšić et al. 2021) and microplastics (Al-Jaibachi et al. 2018) can move from water to land, e.g. through predation of aquatic species by terrestrial species, or the emergence of insects from aquatic larval stages to terrestrial adult stages. To date, however, whether plasticisers can travel by these routes has yet to be definitively established.



## Occurrence of plasticisers in the terrestrial environment

Although an emerging area, there is already relatively well-developed and growing literature on the presence of plasticisers in soil. A review of published studies identifies over 50 papers that report the concentrations of one or more plasticisers in soil samples. This includes survey studies and studies, such as some of those discussed above, that reported the occurrence of plasticisers in an experimental setting, e.g. studies to assess the effect of sewage sludge additions in field-scale trials. Of the identified papers, 43 have extractable data on concentrations in soils. These data can be assembled to provide an overview of the range of concentrations found based on total measured plasticiser in addition to the number of detected plasticisers and concentration ranges for individual congeners.

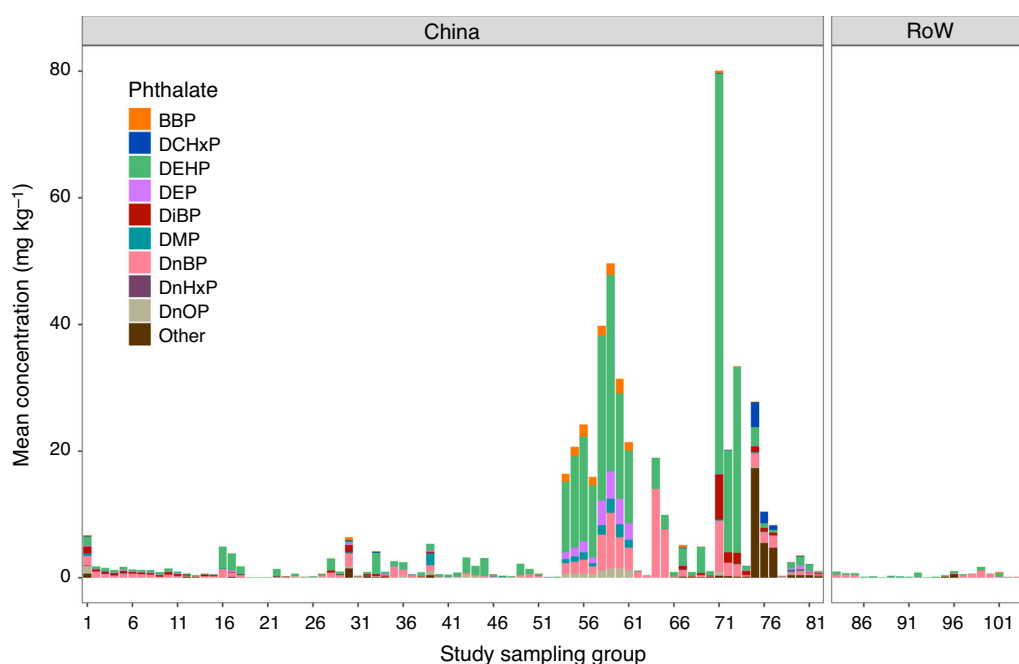
To date the large majority of studies of plasticiser occurrence have focussed on phthalates. Hence, the most robust comparative assessment of occurrence can be conducted for this group of substances. Of the studies that have measured phthalate concentrations in soil, 86% ( $n=37$ ) report data for phthalates in Chinese soils (79% of sampling groups,  $n=82$ ). The remaining studies came from Europe (12% of studies, 17% of sampling groups) and India (2% of studies, 4% of sampling groups). As many papers did not report median concentrations for individual phthalates, mean concentrations were used for comparative data analysis. The bias towards higher values associated with using the mean as opposed to the median may be mitigated by viewing the results from a risk assessment perspective, where one would focus on understanding the upper range represented by worst-case high-value measurements.

To obtain a higher resolution overview of the differences in plasticiser contamination between land uses, data within individual studies were split into discrete sampling groups as primarily urban, agricultural, landfill, rural or mixed. In a minority of cases it was necessary to calculate mean phthalate concentrations manually using the raw data for a sampling group. When such calculations were made, non-detect values

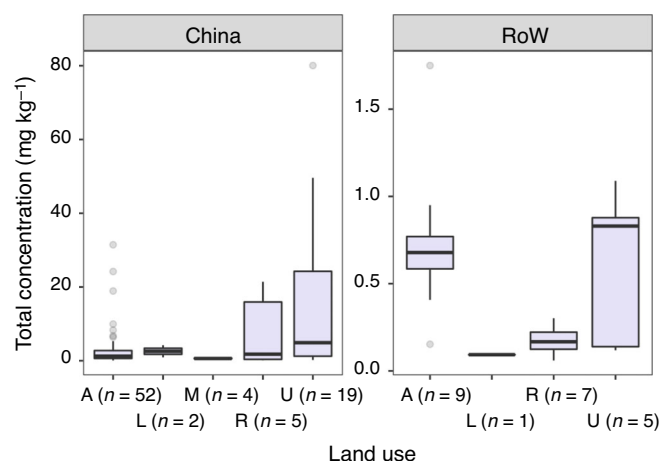
were assigned as half of the limit of detection (LOD) and included in the mean calculation. Two studies did not give any data for concentrations  $<1 \text{ ng g}^{-1}$ . In these cases, a value of  $0.5 \text{ ng g}^{-1}$  was assigned. If, for any substance, all analysed samples within a land use group reported concentrations below the detection limit, that group was given a null value for that analyte. A list detailing the data extracted from these 43 papers and the categories assigned to the sampling groups can be found in Table S2 (Supplementary Material), in addition to any notes on whether data had to be treated before analysis. Limits of detection for all 43 studies, where available, are detailed in Table S3 (Supplementary Material).

Soils with the highest reported total phthalate concentrations are exclusively found in China (Fig. 2). Such high levels in Chinese soil may arise from local or regional factors; however, the number of studies of soil plasticiser loads is greater in China than in other regions. Hence, until there is wider geographic investigation of phthalate occurrence, it is not possible to unequivocally establish whether the dominance of high values in China is simply a result of the greater focus on plasticiser measurement or relates to other aspects such as the degree of source intensity. No clear relationship was found between the number of phthalates that were targeted and total phthalate concentration in specific environments (Fig. S1, Supplementary Material). This indicates that some soils may have a limited range of phthalate inputs that are well captured by a specific analytical suite that focusses only on a small set, most often of the better known and characterised congeners.

The variation in total phthalate concentrations between different land uses is shown in Fig. 3. There is uncertainty in this analysis, as in addition to the variability in the number of phthalates targeted, assigning a particular site to a specific land use can be challenging based on the information available for some studies. Despite these challenges, some specific patterns are evident. For example, urban soils represent the land use groups that typically have the highest total maximum phthalate



**Fig. 2.** Total phthalate concentrations in all land use sampling groups extracted from published literature ( $n=43$  studies,  $n=104$  sampling groups). RoW = rest of world.



**Fig. 3.** Total measured phthalate concentrations in soils worldwide, broken down by land use. Here,  $n$  refers to the number of sampling groups in each box. Whiskers represent largest or smallest value less than or equal to the upper or lower hinge +  $1.5 \times$  interquartile range. A = agricultural, L = landfill, M = mixed, R = rural, U = urban. RoW = rest of world.

**Table 2.** Detection frequency (DF) and limits of detection (LODs) of phthalates in soils ( $n = 43$  studies,  $n = 104$  sampling groups)

LOD means, medians and ranges were calculated from available data (Table S3, Supplementary Material). Method LODs were used in calculations where possible, otherwise instrumental LODs were used. It should be noted that 60% of studies did not report LOD data for individual phthalates. As such, the LOD summary statistics in this table represent only a subset of studies

Phthalate	DF (%)	$n$	Non-detects	Detects	Maximum mean concentration ( $\text{mg kg}^{-1}$ )	LOD ( $\mu\text{g kg}^{-1}$ )		
						Mean	Median	Range
DEHP	100	104	0	104	63.20	22.93	10	0.01–130
DnBP	100	98	0	98	14.06	7.96	1.8	0.01–50
DiBP	100	51	0	51	7.15	2.07	2	0.01–4.69
DnHpP	100	3	0	3	0.05	7.79	7.79	-
DMPP	96	26	1	25	3.72	2.16	1.72	0.83–4.34
DMP	94	88	5	83	2.17	5.15	1.6	0.01–30
DEP	93	88	6	82	4.41	15.15	2.5	0.01–110
DiNP	92	13	1	12	0.50	0.26	0.26	0.01–0.5
DnOP	89	72	8	64	1.49	14.18	1.98	0.01–110
BBP	87	71	9	62	2.36	4.03	1.8	0.01–20
DEEP	83	24	4	20	0.20	1.7	1.16	0.67–3.8
DMEP	81	21	4	17	0.25	1.69	1.42	0.59–3.31
DnHxP	81	31	6	25	0.09	1.33	1.08	0.59–2.3
DCHxP	81	31	6	25	3.96	2.39	1.43	0.89–5.8
DBEP	75	24	6	18	12.54	1.77	1.52	0.84–3.2
DPhP	75	12	3	9	0.24	2.63	1.43	1.25–5.2
DiDP	75	4	1	3	0.07	0.01	0.01	-
DnNP	68	22	7	15	0.95	4.83	4.83	3.05–6.6
HEHP	67	18	6	12	0.07	1.02	1.02	-
DnPP	58	26	11	15	0.10	1.58	1.58	0.46–2.7
DPrP	33	3	2	1	0.05	-	-	-

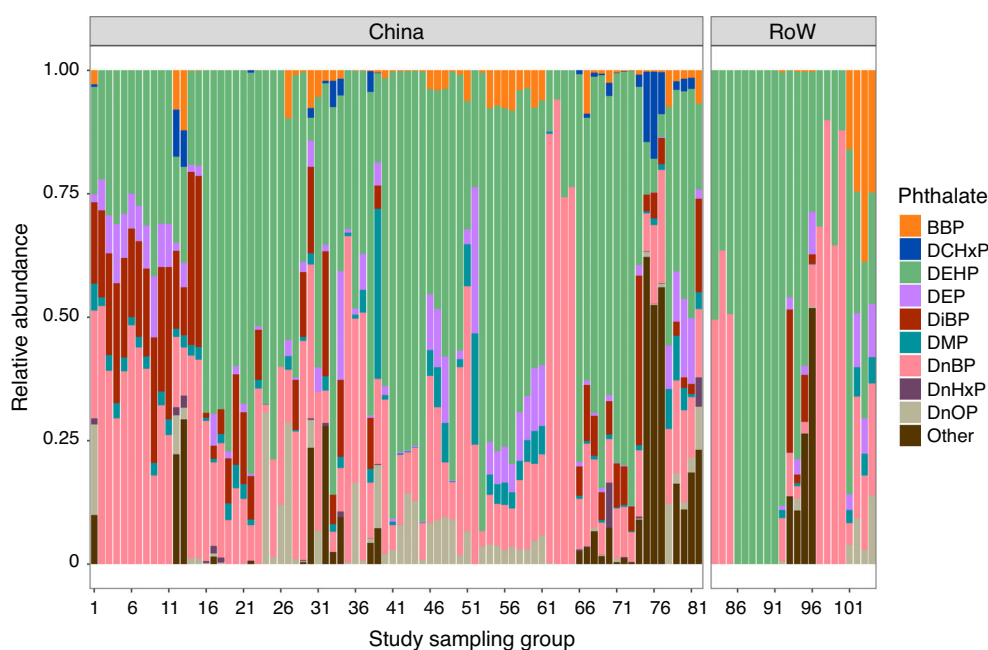
concentrations. Within China, there is relatively low variability of total phthalate concentrations in agricultural compared with urban and rural soils. This may arise from a less complex set of source inputs in agricultural environments that may be subject to similar land management, and to the reduced potential for point source contributions compared with urban sites.

DEHP was the only phthalate that was targeted in every study. A detection frequency (DF) of 100% indicated that this substance is ubiquitously present in soil samples across all studies and land use categories. Other phthalates detected at 100% frequency across all sampling groups, although not

necessarily always targeted, were DnBP, DiBP and di-n-heptyl phthalate (DnHpP) (Table 2). Indeed, in the case of DnHpP this positive detection related only to a single study (Zhang et al. 2015). Of the 21 phthalates that have been quantified in soils, 17 had a detection frequency of 75% or more, which indicated that diffuse sources and long-range transport have caused significant dispersion of these contaminants widely in the terrestrial environment. The LODs for individual phthalates tend to fall in the range  $0.5\text{--}5 \mu\text{g kg}^{-1}$  (Table 2), which is generally far below reported soil concentrations, and indicates that the results obtained between studies are

**Table 3.** Pearson correlation coefficients (*r*) of the mean concentrations of the nine most commonly targeted phthalates in soil study sampling groups  
\* =  $P < 0.05$ , \*\* =  $P < 0.01$ 

	DMP	DEP	DiBP	DnBP	BBP	DEHP	DnOP	DnHxP
DEP	0.895*							
DiBP	0.075	0.035						
DnBP	0.702**	0.750**	0.952*					
BBP	0.835**	0.900**	0.571**	0.683**				
DEHP	0.485**	0.521**	0.946**	0.608**	0.544**			
DnOP	0.874**	0.845**	0.418*	0.742**	0.830**	0.535**		
DnHxP	0.017	0.230	-0.043	-0.029	0.011	-0.106	0.351	
DCHxP	-0.001	-0.114	0.044	0.203	-0.054	-0.052	0.063	-0.036

**Fig. 4.** Relative abundances of phthalates in all land use sampling groups extracted from published literature to date ( $n = 43$  studies,  $n = 104$  sampling groups). RoW = rest of world.

broadly comparable. However, only 40% of studies reported LOD data for individual phthalates, and 12% gave no LOD information at all. Thorough reporting of LOD data in future studies will increase the confidence with which results from different studies can be compared, thereby improving assessments of the risk of phthalates to terrestrial environments.

Statistically significant positive correlations exist between the concentrations of some of the most commonly targeted phthalates in study sampling groups (Table 3), which indicates similar sources of these congeners in soils. A group of 7 phthalates (DMP, DEP, DiBP, DnBP, BBP, DEHP and DnOP) appear to share common sources, as these compounds show a high degree of correlation across the different sampling groups. Particularly strong relationships are evident between DEHP and DiBP, DnBP and DiBP, and DMP and DEP. DMP and DEP represent the lowest molecular weight phthalate congeners and as such share similar uses, e.g. in cosmetics (Hansen et al. 2013), and potentially similar drivers of post-release fate and transport. Although plastic formulations tend to contain only 1–2 plasticisers, DEHP, DiBP and DnBP share very similar properties so may be used interchangeably, which indicates a common potential source through consumer use.

When considered as a proportion of total phthalate load, DEHP and DnBP tend to dominate the soil plasticiser profiles (Fig. 2 and Fig. 4). Additionally, DiBP, DnOP, BBP and DEP are all commonly present, albeit it in lower concentrations than is the case for DEHP and DnBP. Given that DEHP constitutes 40% of global plasticiser consumption (CEFIC 2018), its high prevalence and dominance is unsurprising.

Although the profile of phthalate contamination in many sampling groups appears to follow similar patterns, phthalate soil signatures can in some cases be specific to a certain site. For example, notably high concentrations of DBEP ( $4.49 \text{ mg kg}^{-1} \text{ dw}$ ), DMPP ( $2.88 \text{ mg kg}^{-1} \text{ dw}$ ) and DCHP ( $1.66 \text{ mg kg}^{-1} \text{ dw}$ ) were detected in soils in an e-waste processing area, whereas DnBP and DEHP were found at these locations only in concentrations of  $1.92 \text{ mg kg}^{-1} \text{ dw}$  and  $1.05 \text{ mg kg}^{-1} \text{ dw}$  respectively (Zhang et al. 2019). The patterns of phthalate contamination seen in soils (Fig. 2 and Fig. 4) not only highlight the importance of diffuse inputs in determining the profiles on a wide scale, but also the significant impact that point sources of phthalates can have on local sites. E-waste sites in particular appear to have distinct impacts on local phthalate profiles, possibly owing to the specialised use of certain phthalates in electronics. Hence, at any individual site the total

burden of phthalates and composition of congeners may be a combination of local, regional and global drivers.

Despite the extensive and widespread analysis of phthalates across land use types, there is to date only one study that the authors are aware of that has quantified the presence of an emerging plasticiser in soil. Although it was found in relatively low levels ( $0.057 \text{ mg kg}^{-1}$ ), DEHA had the highest mean concentration of any of the studied plasticisers in landfill soils in Indian cities (Chakraborty et al. 2019). Emerging plasticisers have been detected in air and dust in Europe, the USA and Japan (Takeuchi et al. 2014; Fromme et al. 2016; Larsson et al. 2017; Subedi et al. 2017), and in sewage sludge at comparable levels to phthalates (Lee et al. 2019a). Although limited, the data suggest the potential for the occurrence of emerging plasticisers in soil at concentrations on a par with those for phthalates. As use of these chemicals increases in the future, the potential for further emission and release is evident.

Analytical challenges are very important to take into account when considering plasticiser occurrence data, as they underpin the conclusions that can be drawn from published studies. These challenges are not limited to the issues surrounding the reporting of LODs and under-reporting of sum concentrations owing to limited analysis suites discussed above. Some studies did not report whether concentrations were reported as dry or wet weights. Such inconsistencies may lead to under-reporting of concentrations in soils that measured the concentrations as wet weights, and therefore to aid comparability between studies it is preferable that future work focuses on reporting dry weight concentrations. At present, there is no established method for quantifying the relative contribution of microplastic-associated plasticisers compared with leached plasticisers. As such, microplastics present in soils may lead to an over-reporting of free plasticiser concentrations. The extraction of plasticisers from microplastics will be related to the extraction method used, with less harsh methods likely to lead to less extraction from microplastics but may cause lower extraction efficiencies for targeted phthalates from the soil. Accurate assessments are important for understanding the potential for biological effects of phthalate contamination, as the bioavailability of plastic-interred plasticisers is likely much less than those free in the soil. Additionally, as for other organic chemicals, phthalates can occur as bound residues that can be difficult to extract and analyse, and thus may be under-reported (Tran et al. 2015). This has further implications for the reliability of measured values as an assessment of total plasticiser loads.

## Fate

### *Persistence of plasticisers in soils*

Several studies have demonstrated that biodegradation is one of the dominant processes controlling the persistence of plasticisers that reach the soil environment (Xie et al. 2010; Hurtado et al. 2017). The degradation rates of plasticisers are controlled by several factors including environmental conditions (e.g. temperature, moisture, UV light), soil properties and the microbial communities present in soils, sludges and waste amendments. It is well established that anaerobic degradation of plasticisers in soils is significantly slower than aerobic degradation, e.g. half-lives of DMP and DnBP were found to be  $\sim 8$  times longer under anaerobic conditions (Shanker et al. 1985). Most studies of plasticiser degradation in soils are aerobic, thus the discussion in this section focuses primarily on aerobic degradation rates, although many of the underlying factors

controlling degradation will be the same in both aerobic and anaerobic environments. These studies have focussed exclusively on phthalates (Table S4, Supplementary Material), with no quantitative data available for emerging plasticisers.

It has been clearly demonstrated that phthalates with longer chain lengths have greater persistence in the environment than shorter chain ones (Cartwright et al. 2000; Xie et al. 2010; Zhu et al. 2018b; Tang et al. 2020). This is attributed to the greater hydrophobicity of long chain molecules resulting in stronger soil organic matter (SOM) binding, which reduces their bioavailability (Murillo-Torres et al. 2012). In addition, bacterial degradation tends to occur more slowly for larger, more complex organic molecules (Kanaly and Harayama 2000). No degradation of the more complex, highly hydrophobic DEHP was observed in over 65 days at  $20^\circ\text{C}$  in a field soil, whereas the simpler, more hydrophilic DEP had a half-life of just 0.75 days (Cartwright et al. 2000). Phthalates with branched chains have also been found to degrade more slowly than straight-chained congeners. For example, DnOP degraded more rapidly than DEHP in a marine sediment (DnOP  $t_{1/2} = 172$  days, DEHP  $t_{1/2} = 347$  days) (Kickham et al. 2012). In addition, significantly faster biodegradation of unbranched maleate plasticiser congeners than the equivalent branched congeners has been demonstrated (Erythropel et al. 2015). Conversely, the straight-chain phthalate DnBP was found to degrade to a marginally lesser extent than its branched counterpart (DiBP) in two agricultural soils after 42 days (9.6–18.2% DnBP versus 4.6–6.2% DiBP remaining) (Zhu et al. 2018b). A further complexity to understanding phthalate fate in soils is that rates determined in the laboratory may not necessarily reflect those under field conditions, with some phthalates significantly more persistent than any experimentally determined half-life would suggest. For example, DEHP concentrations in a polluted soil decreased from  $1.47 \text{ mg kg}^{-1} \text{ dw}$  to  $0.88 \text{ mg kg}^{-1} \text{ dw}$  over 15 years, a reduction of only 40%, and no significant increases were observed in a control soil (Patureau et al. 2007).

The rate of phthalate degradation in soils has been found to be positively correlated with temperature (Fig. 5), possibly owing to increased bacterial activity combined with more rapid physical processes. Thus, even modest temperature changes can have a significant impact on degradation rate. For example, an early study reported a consistent decrease in DEHP soil half-life with increasing temperature ( $t_{1/2} = 158, 86$  and  $55$  days at  $5, 10$  and  $20^\circ\text{C}$  respectively) (Madsen et al. 1999). DnBP was also degraded quicker at higher temperatures (Cheng et al. 2018). Conversely, other studies have found that overall degradation rates of phthalates actually begin to decrease at higher temperatures. Soil half-lives of DnBP and DEHP consistently reduced between  $5$  and  $30^\circ\text{C}$ , but rose at  $40^\circ\text{C}$  (Chang et al. 2009). The complexities in the effects of temperature on phthalate degradation may be explained by the distinct profiles of native soil bacterial communities and the temperature sensitivity of metabolic processes controlled by these microbial taxa.

The effects of soil properties (e.g. pH, SOM, clay content, water holding capacity) on plasticiser degradation are complex. Recent results suggest that the principle soil properties affecting degradation of phthalates in soils may differ from those which determine long-term retention. Microbial-associated carbon was the most important factor influencing the half-life of DnBP in agricultural soils, followed by pH then the total SOM (Cheng et al. 2019). This was in contrast to the relative importance of factors affecting the residual DnBP content in the soil, where

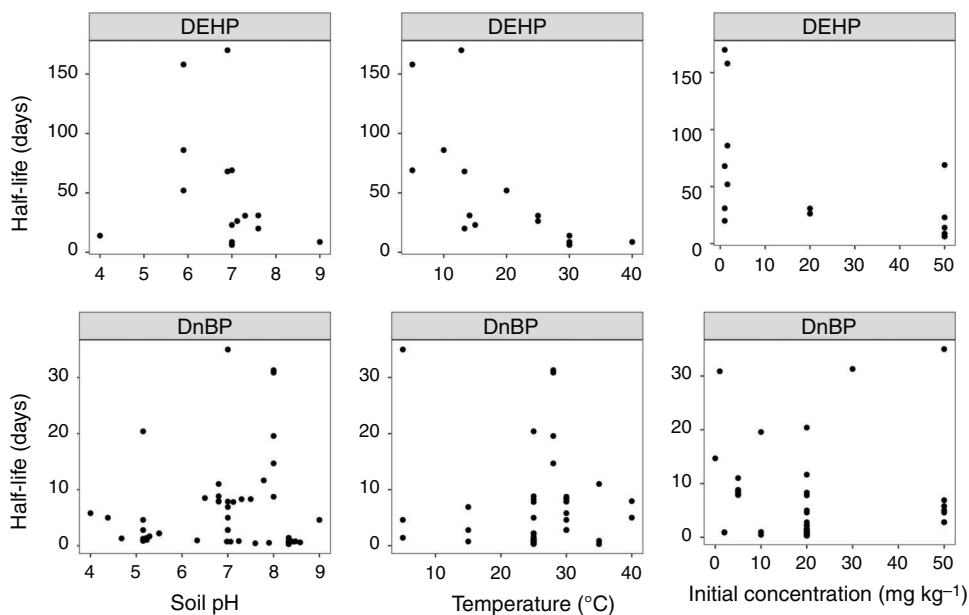


Fig. 5. Half-lives of the phthalates DEHP and DnBP in soils reported in the literature. Details of all phthalate soil half-life data extracted from the literature can be found in Table S4 (Supplementary Material).

organic carbon was most influential, followed by microbial carbon. These results compliment conclusions from studies discussed above, where a fraction of phthalate contamination remains persistent in soil for long periods despite the presence of microorganisms.

SOM can reduce the bioavailability of phthalates in soil, which in turn can reduce biodegradation rates as molecules are not accessible to microorganisms for catabolism. This influence is, however, complex as some constituents of SOM, e.g. humic acids, have been shown to increase phthalate degradation rates in soils by providing a nutritional source that can prime bacterial communities to promote phthalate degradation (Tang et al. 2020; Tao et al. 2020). The overall effect of SOM on phthalate degradation, therefore, can have counteracting influences. For example, a recent study observed no significant correlation between phthalate degradation and soil adsorption capacity, and therefore the overall effect of SOM was to increase the degradation rate (Tang et al. 2020). In contrast, a pronounced effect of increasing chain length on the phthalate persistence in marine sediment has been observed, which was attributed to organic matter binding (Kickham et al. 2012). The half-lives of DnBP and DEHP in two contrasting agricultural soils indicated that phthalates degraded slower in the soil with higher organic matter (Xu et al. 2008). Such results suggest that the effects of SOM levels on phthalate degradation rates may be congener specific.

Values towards the extremes of the pH values found in soil have been shown to reduce phthalate degradation rates compared with those found in more neutral soils (Fig. 5). For example, DnBP and DEHP half-lives were longer at pH 4 and pH 9 than at pH 7 (Chang et al. 2009), and faster DEHP degradation occurred in soils with pH 6–8 (Zhu et al. 2018c), values that are within the ranges typical for lowland soils in temperate regions. Phthalate sorption to organic matter was negatively correlated with pH for four phthalates (Yang et al. 2013). This finding pre-empted the result of a later study that found DnBP degradation to increase with pH, which was attributed to higher DnBP bioavailability at higher pH owing to ionisation of organic groups in SOM that increased

hydrophilicity, and potential greater bacterial abundance in higher pH soil (Cheng et al. 2019). In addition, increasing moisture from 20% to 50–80% WHC significantly increased DnBP degradation rates in soils (Cheng et al. 2018), indicating that soil moisture content could be an important controlling factor in some temperate environments.

#### Uptake of plasticisers by soil organisms

There has been increased interest in the study of the effects of plastics, and in particular microplastics, on soil fauna in recent years, e.g. laboratory and field studies have demonstrated that soil organisms are capable of ingesting microplastics (Huerta Lwanga et al. 2016, 2017; Prendergast-Miller et al. 2019). In addition, laboratory studies have shown that soil microplastic pollution has the capacity to negatively impact growth (Cao et al. 2017; Zhu et al. 2018a; Boots et al. 2019), reproduction (Zhu et al. 2018a; Lahive et al. 2019) and mortality (Huerta Lwanga et al. 2016; Cao et al. 2017), in addition to altering gut microbiota (Zhu et al. 2018a) of soil fauna, although owing to limited data on the occurrence of microplastics in soils, whether these effects are seen at realistic field levels remains unclear. Polymers are inherently inert, and therefore many of the chemical effects of microplastic pollution could result from plastic additives such as plasticisers. For instance, a recent study found that the acute effects caused by microplastics on the soil nematode *Caenorhabditis elegans* were not observed again following the extraction of additives from the plastics (Kim et al. 2020). Indeed, phthalates themselves have been shown to exhibit genotoxicity to the earthworm *Eisenia fetida* (Du et al. 2015; Ma et al. 2016, 2017; Wang et al. 2018a; Song et al. 2019), although the impacts on typical sublethal endpoints, such as growth and reproduction, are currently less clear (Hu et al. 2005; Feng et al. 2016).

Despite the evidence of the toxicity of plasticisers in soil organisms, only one study has assessed uptake and bioaccumulation of plasticisers in soil fauna (Hu et al. 2005). The earthworm *E. fetida* was exposed to five phthalates (DMP, DEP, DnBP, DEHP and DnOP) at 5, 10, 20, 40 and 50 mg kg<sup>-1</sup>

dw soil for up to 30 days. DEHP and DnBP accumulated in worm tissues, although DMP, DEP and DnOP were not detected. At environmentally relevant concentrations of  $5 \text{ mg kg}^{-1}$  dw in soil (Hongjun et al. 2013; Chai et al. 2014; Ma et al. 2019), DnBP and DEHP in worm tissue reached a steady-state after approximately 10 days to  $\sim 1\text{--}1.5 \text{ mg kg}^{-1}$  ww and  $\sim 0.4\text{--}1.2 \text{ mg kg}^{-1}$  ww respectively ('ww' refers to wet weight value). Bioconcentration factors (BCFs) after 15 days of exposure to DnBP were in the range of 0.23–0.30 for an agricultural soil (pH 8.3, SOM 1.35 %) and 0.18–0.23 for a forest soil (pH 7.6, SOM 4.53 %), whereas BCFs for DEHP were in the range of 0.13–0.20 for the agricultural soil and 0.06–0.08 for the forest soil. There was lower phthalate accumulation in the soil with a higher SOM content, possibly owing to strong partitioning of phthalates to the organic material in soil, which reduced their bioavailability for uptake through either dermal or oral ingestion following patterns observed for other organic chemicals (Sijm et al. 2000; Kraaij et al. 2001).

Phthalates are also capable of inducing ecotoxicity in terrestrial flora, e.g. oxidative stress and negative impacts on growth and germination in multiple vegetable crop species including cucumbers (*Cucumis sativus*), common beans (*Phaseolus vulgaris*) and radishes (*Raphanus sativus*) (Kumari and Kaur 2020). Phthalate uptake by plants has been demonstrated, although the extent of reported accumulation varies between studies. A recent study reported BCFs of DEHP in *Brassica chinensis* in the range 0.03–2.00, with the highest BCFs found at environmentally relevant DEHP concentrations (Yuan et al. 2020). The concentration of DEHP in rice cultivars in DEHP-spiked soil ( $20 \text{ mg kg}^{-1}$  dw) was monitored over the course of the plants' development (Cai et al. 2015). The DEHP concentration of the roots and shoots was very high in some cases, varying in the range of  $0.26\text{--}11.8 \text{ mg kg}^{-1}$  dw and  $0.40\text{--}7.58 \text{ mg kg}^{-1}$  dw respectively, with no obvious change over the course of the experiment. Although the exposure concentration of  $20 \text{ mg kg}^{-1}$  dw used in this study is significantly higher than most concentrations of DEHP reported in soils, it is representative of the pollution reported in several studies (Zeng et al. 2008; Teng et al. 2015; Wu et al. 2015; Wang et al. 2018b). Very high BCFs (5.8 to 17.9) of six phthalates were reported in an earlier field study of plant species cultivated near an e-waste processing site, although the high levels of some phthalates in aboveground tissues may have arisen from direct atmospheric exposure rather than through soil uptake and translocation to shoots (Ma et al. 2013). In addition, the composition of sampling and storage materials were not reported, thus it is unclear whether samples may have been contaminated following collection. Nevertheless, the weight of evidence suggests that phthalate uptake by terrestrial flora occurs, although further work is required to determine the relative contributions of different exposure routes.

Despite evidence of bioaccumulation of phthalates in soil fauna and flora, transfer of plasticisers through trophic networks has yet to be clearly demonstrated. There is evidence from both marine and freshwater aquatic studies to suggest that plasticisers can be transferred to species occupying higher trophic levels, e.g. eight phthalates were detected in 18 species across four trophic levels in a marine aquatic food chain (Mackintosh et al. 2004). A more recent study assessed the concentration of 14 phthalates in multiple freshwater fish species and found total phthalate concentrations ranged from n.d. to  $1081 \mu\text{g kg}^{-1}$  dw, with DEHP and DnBP again the most commonly detected phthalates (Lee et al. 2019b). However, the presence of plasticisers in higher trophic levels in these studies

could come through direct uptake of phthalates or ingestion of microplastics.

Metabolism likely represents the primary removal pathway of plasticisers within organisms. Although the nature of the various relevant metabolic pathways is complex and beyond the scope of this review, known rates of elimination of phthalates in biota appear rapid (Frederiksen et al. 2007). The nature of these main biotransformation reactions is well studied in some mammals, e.g. humans and rats (Frederiksen et al. 2007; Domínguez-Romero and Scheringer 2019), and to a lesser extent marine organisms (Stalling et al. 1973; Hu et al. 2016). In contrast, elimination in soil organisms is less understood. However, given that enzymatic metabolic pathways are relatively well conserved between species, pathways for metabolism in soil fauna may reflect those seen in higher organisms. Studies in soil species may therefore initially seek to identify the presence of known metabolites in organisms and the activities of enzymes in families such as the cytochrome P450s, esterases and glutathione-S transferases to begin to elucidate the biotransformation pathways for plasticisers in these species.

### Knowledge gaps and future research recommendations

To date, research into the sources, occurrence and fate of plasticisers in the terrestrial environment has focussed on phthalates, with a distinct lack of data for emerging classes of plasticisers. The profiles of phthalates in soils generally reflect their production volumes, with DEHP dominating soil profiles, although knowledge of phthalate occurrence in soils outside of China is limited. Half-lives of phthalates in soils are on the scale of weeks, although in some cases contamination is significantly more persistent. This persistence may result in the prolonged presence of bioavailable phthalate residues that can potentially be taken up by soil organisms. Based on currently available knowledge of the sources, fate, presence and exposure of plasticisers, it is possible to identify several key knowledge gaps that remain for the relevant classes of chemicals and to recommend productive areas for future research.

- The influence of soil properties and weathering of plastics on plasticiser leaching rates are largely unknown, and the relative soil leaching potentials across different plastic and plasticiser classes are yet to be determined. Establishing the leaching rates from different plastic types in soils of contrasting properties should be a priority. Longer-term field experiments to establish the effects of weathering and determine environmental leaching rates are especially needed.
- Source apportionment of plasticisers in soils remains unclear, particularly in urban areas. Future work should take a holistic approach by determining not only the occurrence of plasticisers in different land uses, but the presence of plastic pollution both above and below ground, to establish the links between polymer and plasticiser soil loadings, and the resulting polymer and plasticiser profiles. In addition, studies of sewage sludge amendment as a source of plasticisers in agricultural soils are mainly limited to DEHP, and further work is required to establish whether the long-term accumulation and persistence seen for DEHP is also observed for other phthalates and emerging plasticisers.
- Knowledge of the occurrence of phthalates in soils is limited in areas of the world outside of China. The occurrence of phthalates and emerging plasticisers across different regions should be determined, particularly in areas such as south-east Asia and Africa, where rapid urban expansion may be causing

particularly high inputs of plasticisers in the environment as a result of the expansion of plastic use without the development of adequate waste management and recycling networks.

- Inconsistencies in analytical suites and reporting of key data (e.g. LODs) are hampering comparisons of plasticiser occurrence between studies. A coordinated approach is required through the use of a standard suite of commonly detected plasticisers and developing requirements for data reporting. A body of consistent and directly comparable data would be suitable for a meta-analysis approach, which would further address uncertainties in environmental plasticiser distributions and allow for more rigorous analysis of trends in both phthalate and emerging plasticiser occurrence and fate.
- Although the degradation of phthalates in soils is well studied, the persistence of emerging plasticisers in soils is unknown. Degradation kinetics of emerging plasticisers in soils with contrasting properties should be investigated to determine whether the factors underpinning phthalate degradation rates are consistent across all plasticisers.
- The fate of phthalates in soil organisms is understudied and is still unstudied for emerging plasticisers. Controlled studies across multiple species are required to assess the rates and routes of uptake and elimination, in addition to the potential for bioaccumulation and trophic transfer. Further, the potential for interactive effects of plasticisers with commonly used agricultural chemicals, e.g. pesticides, in terrestrial fauna and flora is a priority, given the abundance of plasticisers in agricultural soils.

## Conclusion

Plasticisers have many sources in the terrestrial environment. These may be diffuse, chronic sources (e.g. microplastics present in soils through the degradation of agricultural plastic film) or point sources (e.g. incineration of electronic waste). Any plastic item which contains plasticisers has the potential to act as an input of plasticiser into the environment, through leaching and migration of additives from the polymer matrix. The ubiquitous occurrence of phthalates in the terrestrial environment has been demonstrated, and these compounds are now distributed across all land uses. However, given the rapidly increasing use of emerging plasticisers as replacements for phthalates, a dearth of studies investigating the occurrence of emerging plasticisers represents a significant knowledge gap. Some plasticiser contamination is degraded in a matter of days or weeks, although local effects can lead to some plasticisers acting as persistent organic pollutants at high levels of contamination, and thus represent a potential threat to terrestrial fauna and flora. Despite this, the terrestrial ecotoxicology of plasticisers is relatively understudied, despite evidence of cytotoxicity, oxidative damage and endocrine disruption. The impacts of emerging plasticisers and the ecotoxicology of plasticisers in higher vertebrates are unstudied. Plastic is ubiquitous in modern society, and there are increasingly intensive patterns of agriculture requiring even greater use of plastic products. Given this fact, the determination of the impacts of plasticiser contamination on the terrestrial environment is urgently needed from the perspective of environmental management and human and ecosystem health.

## Supplementary material

Further information on the publications and data discussed in this review are available on the Journal's website.

## Data availability statement

Data sharing is not applicable as no new data were generated or analysed during this study.

## Conflicts of interest

The authors declare no conflicts of interest.

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